

COLLECTED PAPERS

ON

NANO/ATOM PHOTONICS

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PREFACE

In order to realize the ultimate status of light and matter, M. Ohtsu tries to control the spatial properties of light. The approach he has used is through the research of near-field optics and its application to nano-structure fabrication and atom manipulation. This will open up a new era of nano/atom photonics. It should be pointed out that the nano/atom photonics is closely related to quantum optics, atom optics, high density optical storage, nano-structure fabrication technology, and so on (see the figure on the next page). And for this relationship of near-field optics with a variety of fields, nano/atom photonics exhibits rapid progress. Further research will be done to realize novel nanometric materials and devices.

A "PHOTON CONTROL" project, which was executed in Kanagawa Academy of Science and Technology(KAST) and directed by Ohtsu, has terminated at the end of March,1998. KAST is transferring the results of the research of this project to industry so as to produce commercial near field optical spectrometers, optical memories, fiber probes, and so on. Ohtsu still has an extended project group in KAST for this technical transfer(*).

Results of the recent research carried out by Ohtsu are reviewed in this issue of the COLLECTED PAPERS.

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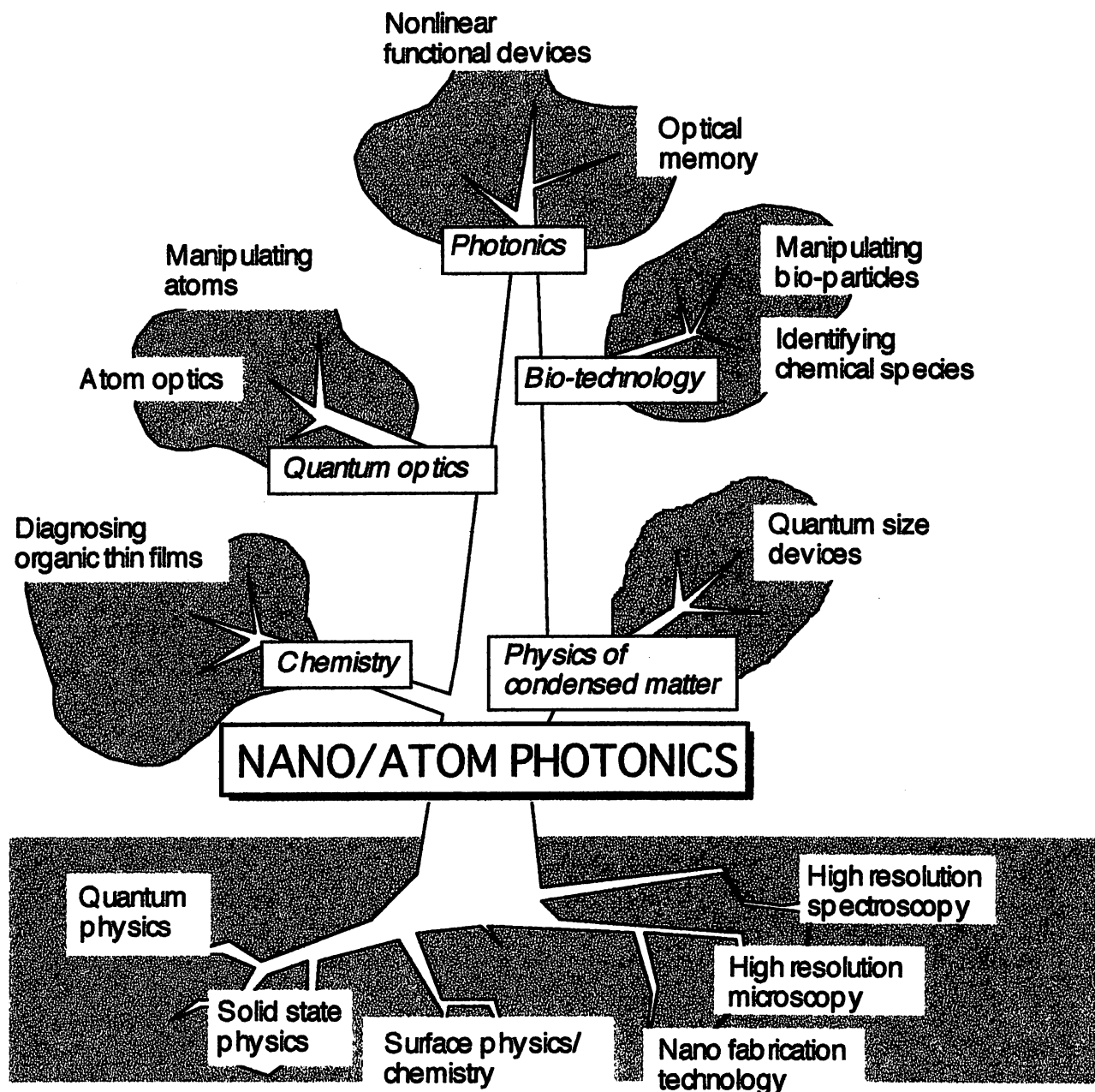
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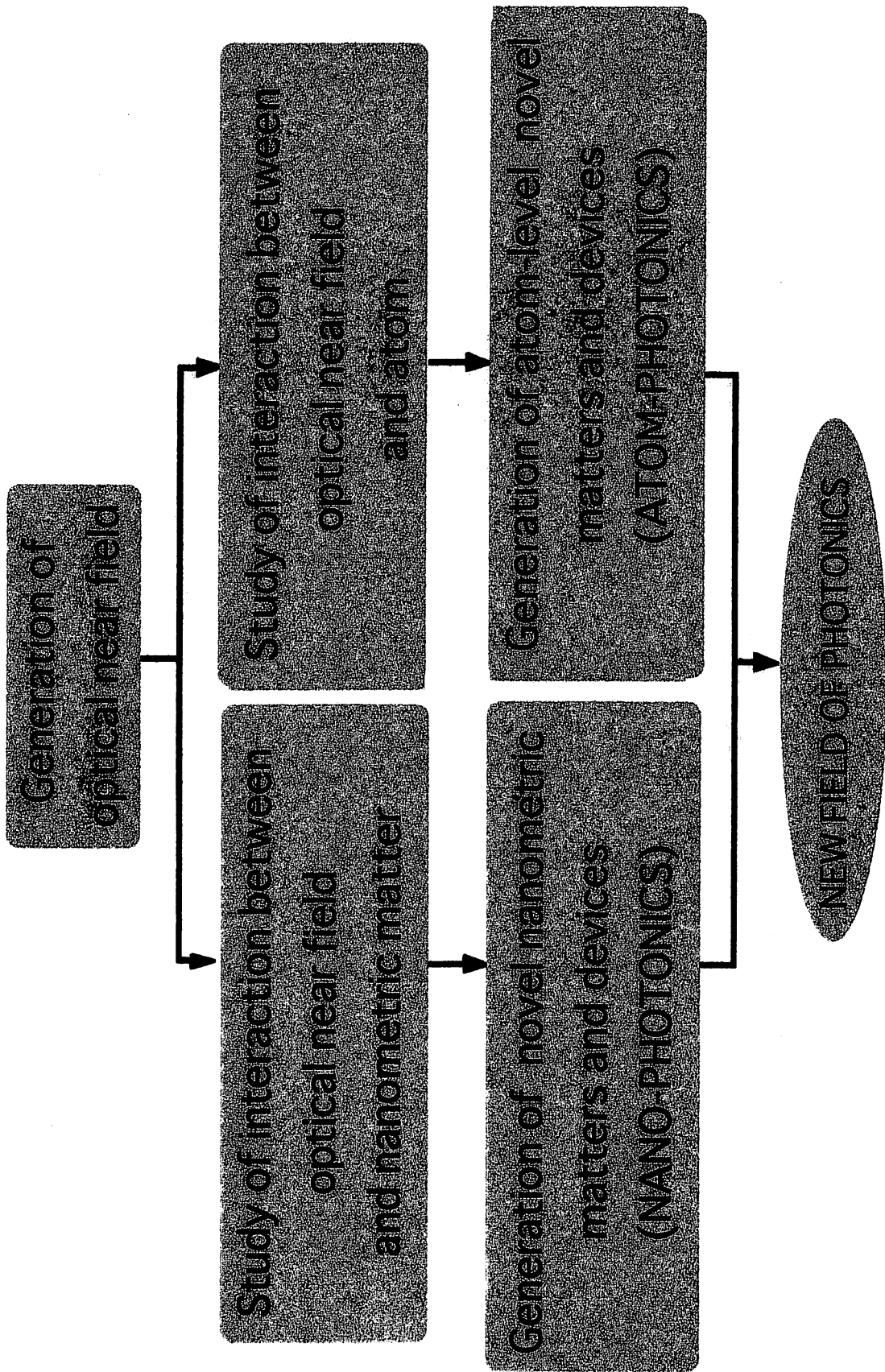
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Nano/atom photonic tree with its roots and leaves covering a wide range of fields



MEMBERS

(From April 1, 1998)

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LIST OF PAPERS

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[I-1] ATOM PHOTONICS

(a) Journal Papers

[1] H. Ito, K. Sakaki, W. Jhe, and M. Ohtsu, "Evanescent-light induced atom-guidance using a hollow optical fiber with light coupled sideways", Opt. Commun., vol.141, August 1997, pp.43-47

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[I-2] NANO PHOTONICS

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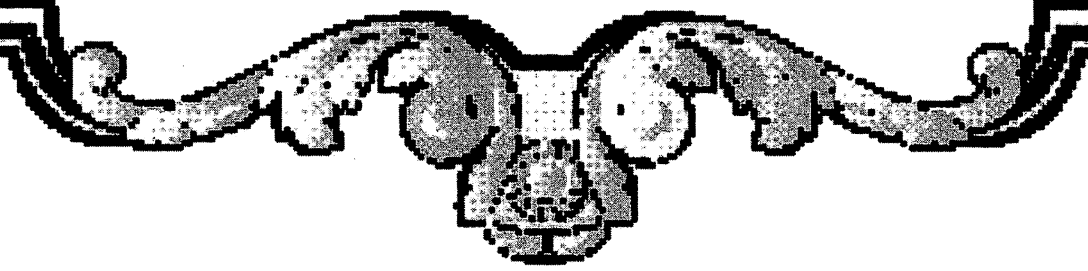
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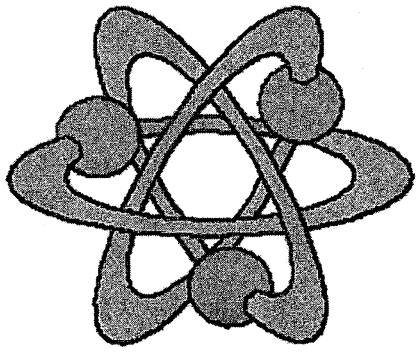
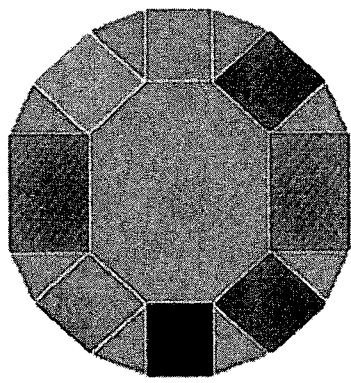
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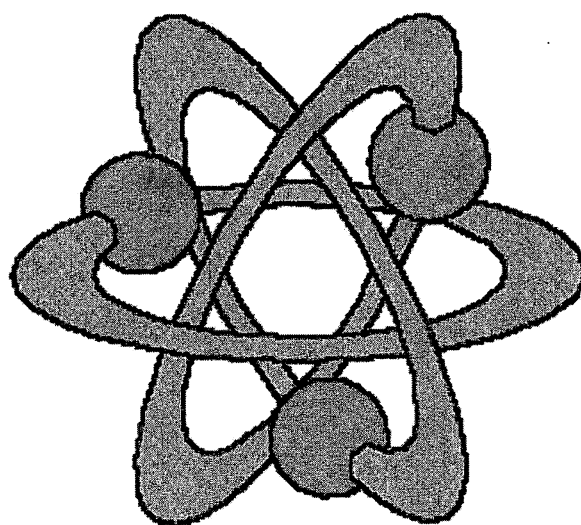
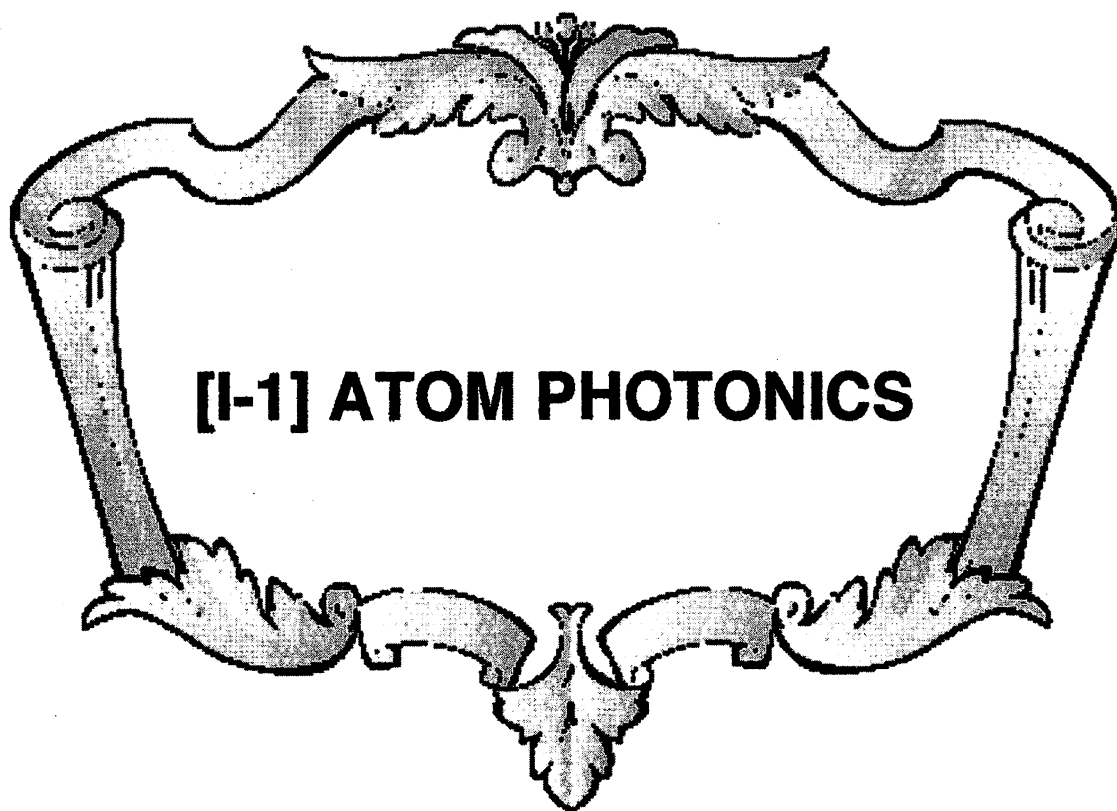
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**[[I] PAPERS IN
NANO/ATOM PHOTONICS**







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OPTICS
COMMUNICATIONS

Optics Communications 141 (1997) 43–47

Evanescent-light induced atom-guidance using a hollow optical fiber with light coupled sideways

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Abstract

This work presents a new way useful for guiding of cold atoms with a hollow optical fiber escorted by blue-detuned evanescent light. To induce an atomic mirror-tunnel, we couple a guide-laser beam via total internal reflection from the side to the core of a hollow fiber with a facet ground at an angle of 45°. The guiding of Rb atoms is demonstrated with complementary detection by means of surface-ionization and two-step photoionization. The efficiency of the lateral light-coupling is also estimated from both experiment and numerical analysis. © 1997 Elsevier Science B.V.

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Keywords: Atom guidance; Evanescent light; Hollow optical fiber; Lateral light coupling

1. Introduction

Evanescent light offers a most effective means of atom manipulation with high spatial accuracy because the strong dipole interaction between atoms and evanescent light occurs in a region below the wavelength. Reflection of atoms with evanescent light induced over a surface of a prism [1–3] gave the first evidence. Then, one-dimensional guiding of atoms with blue-detuned evanescent light has been demonstrated in a glass capillary [4] and a micron-sized hollow optical fiber [5]. This technique of atom guidance can be applied to interesting studies on quantum-mechanical or quantum-optical phenomena, for example, quantized atomic motion in a gravitational trap composed of a hollow fiber [6] and cavity quantum electrodynamic (QED) effects [7–9] in a cylindrical dielectric.

On the other hand, there is now enhanced interest in atom holography [10], lithography and deposition [11–14] with methods of atom optics. We have recently demonstrated that the evanescent-light guiding of atoms through a micron-sized hollow fiber is useful as a novel form of optical atomic deposition with nanometric accuracy [15]. In this case, one can precisely control the deposition rate by adjusting the frequency of a guide laser. In addition, the use of laser-cooled atoms will be useful for atom-by-atom deposition. For example, if a cold atom is guided to the tip of a sharpened optical fiber [16], the cold atom can be caught by an evanescent field produced on the nanometric tip [17] and carried to an arbitrary point on a substrate. Atom lasers have attracted theoretical or experimental attention [18,19] since Bose-Einstein condensation has been achieved with elaborate laser-cooling techniques [20]. The minute-hollow optical fiber escorted by blue-detuned evanescent light has the possibility of working as a waveguide of atomic de Broglie waves [21], and furthermore as a cavity of the atom laser [22].

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This work develops a new method useful for guiding cold atoms as well as studies of quantum-mechanical or quantum-optical phenomena. Introduction of cold atoms into a hollow fiber can be efficiently made by means of an atomic funnel composed of a conical or pyramidal hollow prism [23,24]. When a hollow fiber is combined with other atom-optical elements such as an atomic funnel, the coupling of a guide-laser beam to the hollow fiber is a crucial problem. In our previous work [5], a guide-laser beam has been introduced from a front facet of a hollow fiber via a pinhole mirror placed upstream of the fiber. This scheme, however, is inconvenient for attachment of an atomic funnel. In order to settle this problem, we use a hollow optical fiber with a facet ground slantwise. In this case, a guide-laser beam is coupled to the hollow fiber directly from the side via total internal reflection. The scheme of lateral light-coupling greatly increases the degree of freedom on the fiber configuration because there is no extra optics. In addition, this scheme removes such undesirable light as propagates through the hollow region. One can therefore apply the atom-guidance technique straightforward to various studies mentioned above.

2. Experiment and discussion

Total internal reflection is utilized for the side coupling of a guide-laser beam to a hollow fiber. For a germanium-doped silica glass fiber with a refractive index of 1.45, the critical angle of total internal reflection at the core-vacuum interface is estimated to be 43.5° . Taking into account this condition, we have ground a facet of a germanium-doped silica glass hollow fiber at the slant. Fig. 1 shows a microscope image of a $7\text{-}\mu\text{m}$ -hollow optical fiber polished at an angle of 45° . In this fiber, light-waves propagate through the cylindrical core with a thickness $d = 4\text{ }\mu\text{m}$ around the hollow region. The fabrication process of the

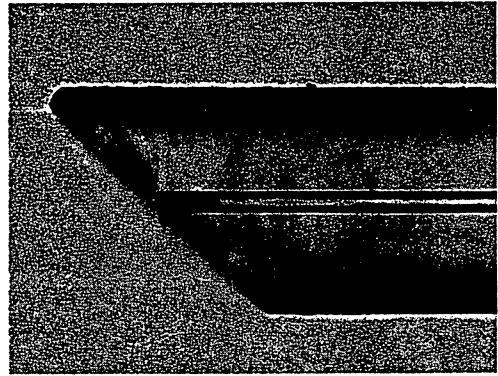


Fig. 1. Microscope image of a $7\text{-}\mu\text{m}$ -hollow optical fiber with a facet polished at an angle of 45° . A guide light propagates through the $4\text{-}\mu\text{m}$ -thick cylindrical core around the hollow region. The outer diameter of the fiber is $125\text{ }\mu\text{m}$.

45° -cut hollow fiber is as follows: (1) fixing the hollow fiber to a device for keeping the fiber at an angle of 45° against a plane, (2) rough grinding, (3) fine grinding and polishing of three steps, and (4) washing and baking for removal of any remnants. The mean roughness of the polished core-surface is estimated to be below 50 nm with an electron microscope.

The experimental setup is schematically shown in Fig. 2. A guide-light beam from a Ti:sapphire laser is coupled to the core from the side of the 45° -cut hollow fiber with a hollow diameter $2a = 7\text{ }\mu\text{m}$. Measuring the coupling efficiency of the guide-laser beam to the hollow fiber, we get 38%. A collimated thermal Rb atomic beam is introduced into the hollow fiber at a pressure below 10^{-6} Pa . The typical fiber-length is 4 cm . This fiber becomes multimode for a wavelength (780 nm) of the Rb D_2 -line: the fundamental LP_{01} mode is required for guiding of atoms [25]. Monitoring the light mode-pattern on the exit

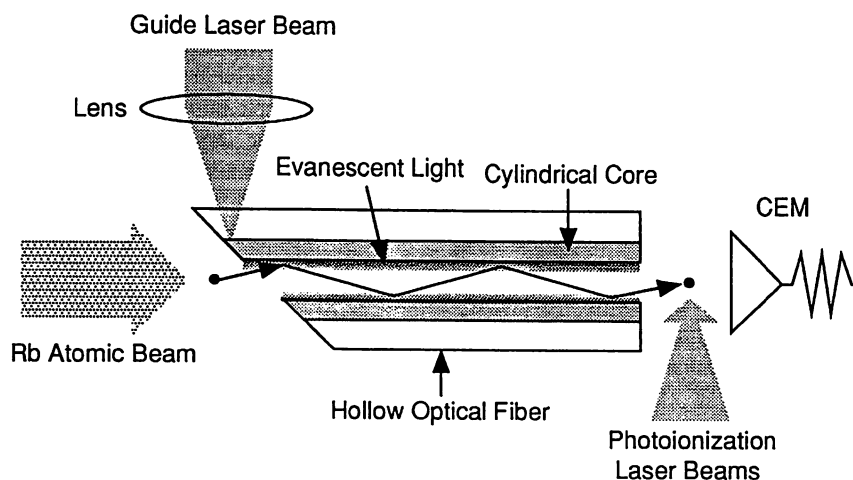


Fig. 2. Sketch of the experimental setup. A guide-laser beam is coupled to a cylindrical core from the lateral direction of a hollow optical fiber via total internal reflection at the slantwise facet. A collimated Rb atomic beam is introduced into a straight hollow fiber. The guided Rb atoms are detected by a channel electron multiplier (CEM) via surface-ionization with a Pt hot wire or Doppler-free two-step photoionization with a diode laser and a high power Ar-ion laser.

facet of the hollow fiber with a CCD camera, we selectively excite the LP_{01} mode by adjusting the incident angle of the guide-laser beam. The guided Rb atoms are detected by a channel electron multiplier (CEM) via ionization downstream of the fiber.

In order to check if the guiding of atoms is successfully made, we have carried out ionization of the guided atoms in two ways: (a) surface ionization with a hot wire, and (b) two-step photoionization with a diode laser tuned to the Rb D_2 -line and an Ar-ion laser whose wavelength is 476.5 nm. The surface-ionization detection gives the net number of the guided atoms if the ionization efficiency is given. On the other hand, the photoionization detection gives spectroscopic information on the guided atoms: in particular, the photoionization detection makes clear the state-selectivity of the evanescent-light guiding of atoms. Since the dipole force becomes repulsive when the frequency ω of the guide laser is higher than the atomic resonant frequency ω_0 , atoms can be guided under the blue-detuning condition. This is seen by examining the dependence of the atom flux guided through the hollow fiber on the frequency detuning $\delta = \omega - \omega_0$.

First, we have made surface ionization using a Pt hot wire with a diameter of 10 μm . Fig. 3(a) shows the surface-ionization signal obtained at a distance of 12 mm downstream from the exit facet in the case that the coupling power of the guide-laser beam is 100 mW. Here, the guided atom flux is measured above a background level, which is due to the atoms flying ballistically through the hollow region and the atoms from the residual gas in the vacuum chamber. It should be noted that the surface-ionization scheme detects simultaneously the four $5S_{1/2}$ ground states of two Rb isotopes: the $F = 2$ and 3 hyperfine states of ^{85}Rb , and the $F = 1$ and 2 hyperfine states of ^{87}Rb . In Fig. 3(a), the blue-detuning δ is measured with respect to the resonant frequency of the ^{85}Rb atom in the $F = 3$ upper ground state. Increase in the number of the guided atoms is clearly observed in the blue-detuning region. This corresponds to the fact that the ^{85}Rb atoms in the $F = 3$ upper ground state are dominant in the atomic beam. Considering the incident flux of about 10^6 atom/s, we get the guiding efficiency of more than 20% from the maximum net flux at an optimal detuning of +3 GHz although the surface-ionization efficiency is uncertain.

Then, we have made laser-photoionization spectroscopy. The two-step photoionization scheme has the advantage of detecting the guided atoms state-selectively with an extremely high signal-to-noise ratio. Fig. 3(b) shows the Doppler-free photoionization spectrum on the guided ^{85}Rb atoms in the $5S_{1/2}$, $F = 3$ upper ground state. Here, we select this state by tuning the diode laser to the $5S_{1/2}$, $F = 3 \rightarrow 5P_{3/2}$, $F = 4$ transition. In Fig. 3(b), we see that the guided ^{85}Rb atom flux is maximum at the same blue-detuning of +3 GHz as the case of surface-ionization. From similar photoionization experiments on

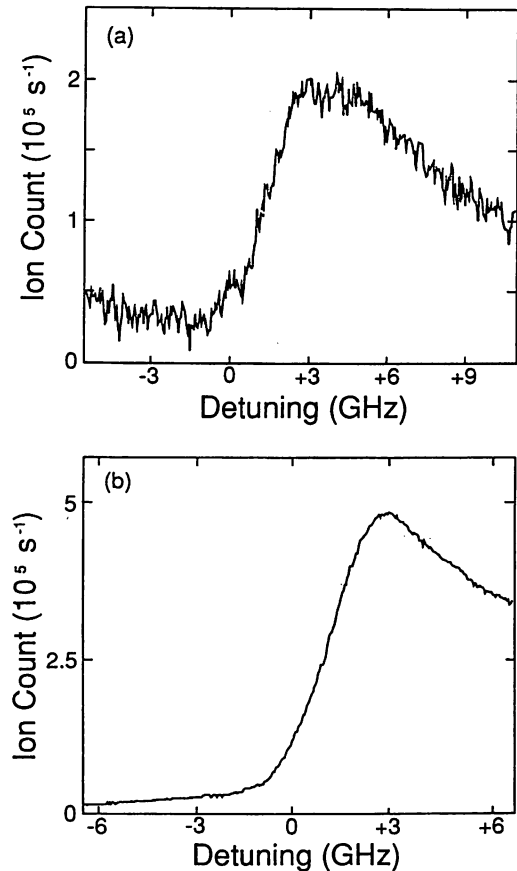


Fig. 3. Ionization signals of the Rb atom flux guided through a 4-cm-long hollow fiber with a hollow diameter of 7 μm as a function of the frequency detuning δ : (a) surface-ionization signal obtained by means of a Pt hot wire with a diameter of 10 μm placed at a distance of 12 mm downstream from the exit facet, and (b) state-selective photoionization spectrum of the guided ^{85}Rb atoms in the $5S_{1/2}$, $F = 3$ upper ground state.

the ^{85}Rb atoms guided in the lower ground state and the guided ^{87}Rb atoms, we also see that the position where the guided atom flux is maximum shifts to higher blue-detuning for those Rb atoms. The abundance ratio of ^{85}Rb to ^{87}Rb is about 7 to 3. In addition, measuring the detection ratio of the Rb atoms guided in the upper ground state to those in the lower ground state, we get the ratio of 10 to 1 for the Rb atomic beam used here. Consequently, comparing the surface-ionization signal with the state-selective photoionization signal, we can confirm that the surface-ionization signal principally comes from the ^{85}Rb atoms guided in the upper ground state.

For the two-level atoms, the optimal detuning depends on the light intensity [26]. However, it is not clear what determines the optimal detuning for the multilevel atoms because optical pumping between the hyperfine ground states can be caused by absorption of photons of evanescent light inside the hollow fiber. The lateral light-coupling scheme has the advantage of improvement in removing undesirable lights, for example, copropagating light through

the hollow region. This scheme, therefore, is useful for the study of optical pumping effects. The details of the optical pumping effects on atom guidance will be discussed elsewhere.

3. Lateral light-coupling efficiency

Let us estimate the coupling efficiency of the guide-laser beam in this lateral coupling scheme. We assume that the electric field of an incident light beam focused on a spot $2r_0$ has a Gaussian profile as

$$G(r, \theta) = G_0 \exp\left\{-\left[(r \cos \theta - x)^2 + (r \sin \theta)^2\right]/r_0^2\right\}, \quad (1)$$

where x is the displacement from the center $r = 0$ of the hollow region in a circular coordinate system (r, θ) . The coupling efficiency $\eta(x, r_0)$ of the guide-laser beam can be approximately obtained from the following formula:

$$\eta(x, r_0) = \frac{\left| \int_0^\infty \int_0^{2\pi} F_{01}(r) G(r, \theta) r dr d\theta \right|}{\sqrt{\left| \int_0^\infty \int_0^{2\pi} F_{01}(r)^2 r dr d\theta \right| \left| \int_0^\infty \int_0^{2\pi} G(r, \theta)^2 r dr d\theta \right|}}. \quad (2)$$

The electric field $F_{01}(r)$ of the LP_{01} mode with a wave number k and a propagation constant β is written in terms of the 0th Bessel functions $J_0(ur)$ and $N_0(ur)$ of the first and second kinds, and the 0th modified Bessel functions $I_0(wr)$ and $K_0(wr)$ of the first and second kinds as [25]

$$F_{01}(r) = \begin{cases} A I_0(vr) & (r < a), \\ B J_0(ur) + C N_0(ur) & (a \leq r \leq a + d), \\ D K_0(wr) & (r > a + d). \end{cases} \quad (3)$$

The three characteristic constants u , v and w are given by [25]

$$u = \sqrt{k^2 n_1^2 - \beta^2}, \quad v = \sqrt{\beta^2 - k^2}, \quad w = \sqrt{\beta^2 - k^2 n_2^2}, \quad (4)$$

where n_1 and n_2 are the refractive indices of the core and the cladding, respectively. The constants A , B , C and D are determined from the boundary conditions and the conservation law of the light power along the fiber.

In the current experiment, the guide-laser beam is focused on a spot of $40 \mu\text{m}$ via a convex lens. From the above equations, assuming that a Gaussian-light beam with $2r_0 = 40 \mu\text{m}$ is focused on the center of the core at $x = 5.5 \mu\text{m}$, we estimate the coupling efficiency η to be 45%. Moreover, taking into account the loss of 20% due to the

Fresnel reflection at the outer surface of the hollow fiber, we estimate the net efficiency of the lateral light-coupling to be 36%. This is in good agreement with the experimental result of 38%.

Since the focusing spot is larger than the hollow diameter and the incident angle is near the critical angle of total internal reflection, part of the incident light beam goes through the cladding near both sides without being reflected. Although this simple scheme of light-focusing with a convex lens is sufficient for guiding of a thermal atomic beam, we will need tight focusing of the guide-laser beam for guiding of cold atoms to avoid heating effects due to the leaked guide-laser beam. The severe focusing on the core will be made with an object lens.

4. Conclusion

We have demonstrated a new form of evanescent-light atom-guidance using a small hollow optical fiber with a slantwise facet. The results obtained from complementary detection via surface-ionization and photoionization show that the lateral light-coupling scheme effectively works for the guidance of thermal atoms. In addition to removal of the light propagating through the hollow region, the lateral light-coupling scheme enables one to introduce the guide-laser beam from the downstream of the hollow fiber. The lateral light-coupling scheme, therefore, will be very useful for studies on the influence of spontaneous emission and Doppler shift on the atom-guidance mechanism. Moreover, this scheme will be useful for studies of the cavity QED effect in a cylindrical dielectric and atom laser. The improved scheme of the atom guidance presented here will open a new way to precise atom-manipulation with cold atoms.

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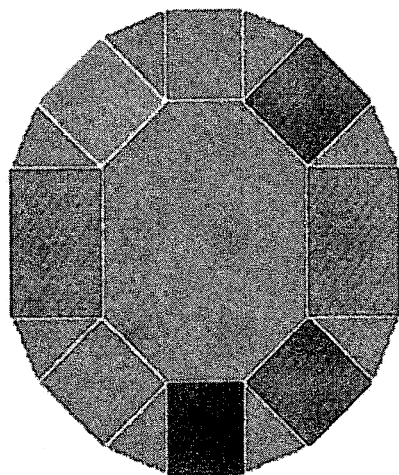
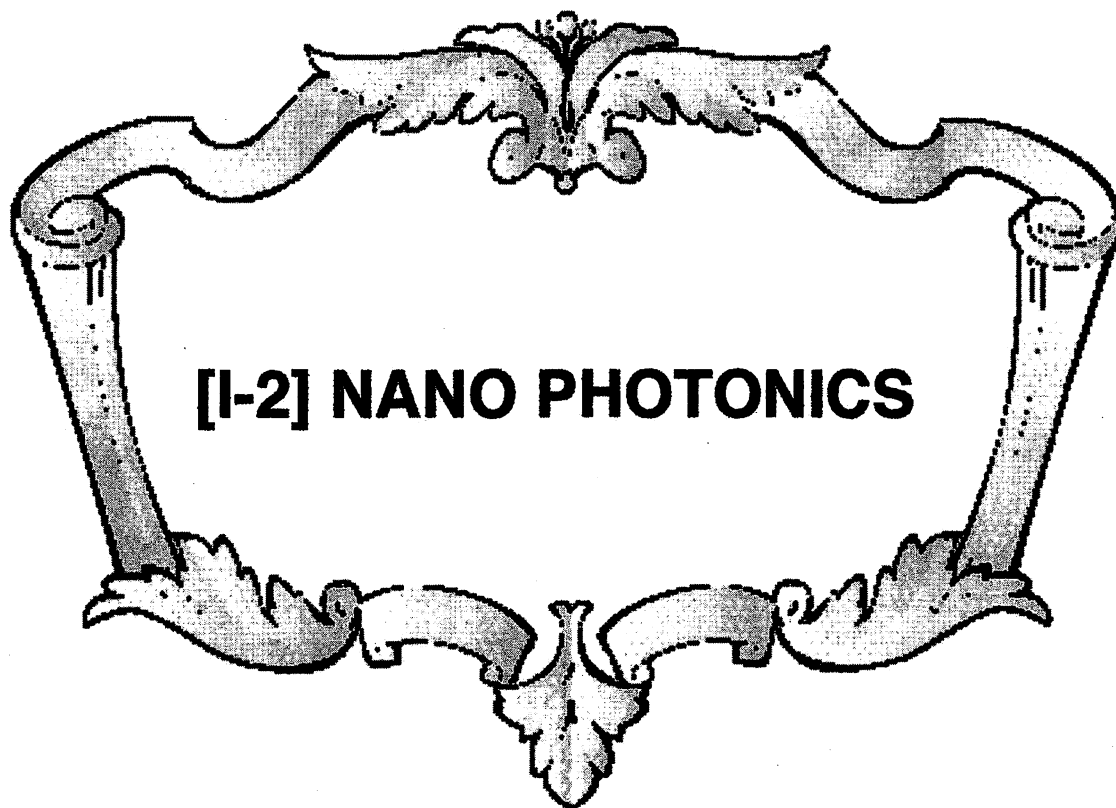
ATOM GUIDANCE AND MANIPULATION BY OPTICAL NEAR FIELD FOR ATOMIC-LEVEL DEPOSITION

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We review our recent experimental results on atomic quantum wire, i.e., guiding atoms by a dipole force of a cylindrical optical near field on the inner surface of a hollow fiber. Magnitude of cavity potential on the surface is estimated by guiding rubidium atoms, and in-line isotope separation is demonstrated. To present the capability of atomic-level deposition, deposition time for one nanometer thick layer and the coefficient of discriminating the number of guided atoms per unit change of optical frequency are estimated. To carry atoms to the desired position, atomic funnel is proposed to couple cold atoms to a bent hollow fiber. Further, possibility of forming an atomic quantum dot, i.e., trapping a single atom by the optical near field on the top of a metal coated nanometric protruded fiber probe tip, is discussed.

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Fabrication of a triple tapered probe for near-field optical spectroscopy in UV region based on selective etching of a multistep index fiber

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Abstract

We propose a method to fabricate a novel triple tapered probe for highly sensitive and highly resolved near-field optical microscopy in a near ultraviolet region. The method is based on selective etching of a multistep index fiber. The obtained fiber probe has a triple tapered structure with three taper angles of 60°, 120°, and 60°. With near-field optical microscopy employing the probe, we succeeded in obtaining a photoluminescence image of polysilanes excited by an ultraviolet laser with a wavelength of 351 nm. © 1998 Elsevier Science B.V.

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Keywords: Near-field optics; Ultraviolet spectroscopy; Scanning probe microscopy; Optical fiber; Vapor-phase axial deposition; Selective etching; Buffered hydrogen fluoride solution

With a near-field optical microscope [1] employing a tapered fiber probe with metal coating, some researchers have performed spectroscopic studies of semiconductor devices and materials excited by a visible or near infrared light at a subwavelength-sized portion. However, there is, to our knowledge, no report on spectroscopic near-field imaging in the ultraviolet (UV) region because it is difficult to employ tapered probes having a germanosilicate (GeO₂) doped core due to the optical absorption by GeO₂ at around 363 nm. Once the near-field UV probe is fabricated, one can investigate local optical response of UV-emitting devices and wide gap materials such as polysilanes [2] having one-dimensional electronic structure and high quantum efficiency.

In order to realize highly spatially resolved imaging under near UV operation, one has to fabricate the tapered probe having a subwavelength apex size and the pure silica core with a high transmittance in the near UV region. Further, to avoid thermal damage of the sample, the probe

should be sensitively tailored and used with an input power as low as possible. The transmission efficiency defined as the relative output power to the input power is generally enhanced by increasing the taper angle of the probe. However, employing the sensitive probe [3] with an enlarged taper angle, one must bear with the limited resolution affected by optical leakage out of the metal around the apex of the probe. In this paper, we propose a novel probe having a triple taper structure, a nanometric apex, and pure silica core for highly sensitive and highly resolved near-field optical microscopy in a region from near UV to near infrared.

Fig. 1(a) illustrates the metallized fiber probe with a triple taper structure. Here, λ is the optical wavelength in the fiber. α , β , and γ are the three taper angles of the triple taper. d_1 and d_2 are the two base diameters of the tapers with α and β , respectively. L is the length from the portion with cross sectional diameter λ to the apex. The light entering the fiber is strongly attenuated by the coated

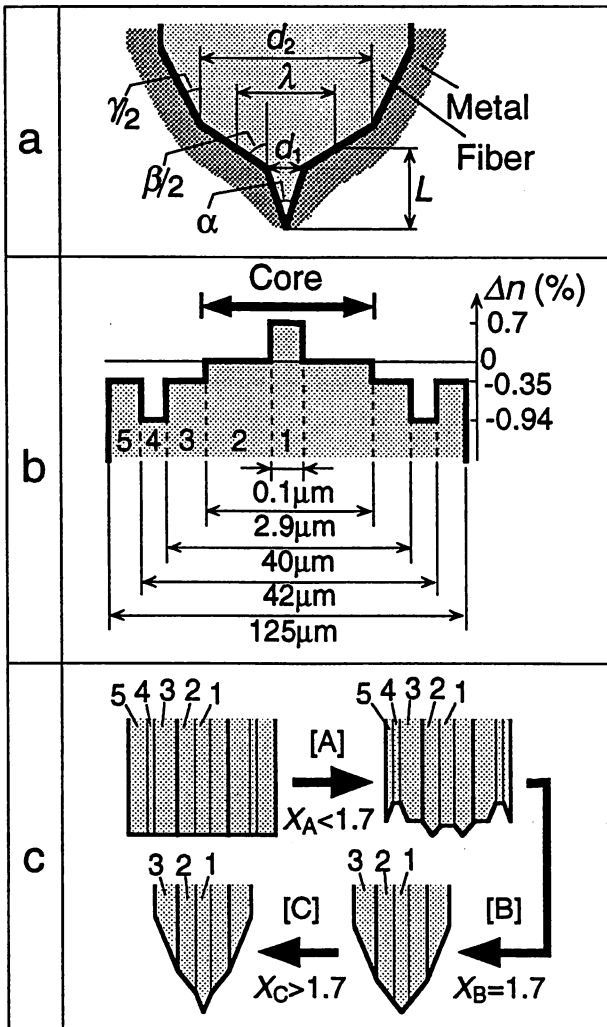


Fig. 1. (a) Schematic illustration of the triple tapered fiber. Here, α , β , γ , λ , d_1 , d_2 , and L are the three taper angles of the triple tapered probe, the wavelength in the fiber, the two base diameters of the cones with α and β , the length from the cross sectional diameter λ to the apex, respectively. (b) Cross sectional profile of the relative refractive index difference of a multistep index fiber used for fabrication of the probe. Here, Δn is the relative refractive index difference to pure silica. Sections 1–5 are made of GeO_2 doped silica, pure silica, low fluorine doped silica, high fluorine doped silica, and low fluorine doped silica, respectively. The core consists of sections 1 and 2. Section 2 covers 99.9% of the area of the core. (c) Schematic illustration of the process for fabricating the probe based on selective etching of the fiber in buffered hydrogen fluoride solution (BHF) with a volume ratio of NH_4F aqueous solution (40 wt.%): HF acid (50 wt.%): $\text{H}_2\text{O} = X:1:1$. Here, sections 1–5 are defined as in Fig. 1(b). The process involves three steps: [A] hollowing of sections 1 and 4 in $X_A:1:1$, [B] tapering of sections 2 and 3 in $X_B:1:1$, [C] sharpening of section 1 in $X_C:1:1$. The values of X_A , X_B , and X_C are X of BHF used in steps [A]–[C], respectively.

metal from the portion with diameter λ to the apex [3]. To reduce the optical attenuation, one has to decrease the length L . In this triple tapered probe, we can decrease the length L by increasing β so as to enhance the transmis-

sion efficiency of the probe. Further, we can obtain a highly spatially resolved image using the probe with a nanometric apex diameter and a sharp taper angle α which is much smaller than β . To employ the probe in a near UV region, the core region has to be made of pure silica.

To realize such a probe, we propose a method based on selective etching [4] of a multistep index fiber as schematically shown in Fig. 1(b). Here, the core region consists of sections 1 and 2 made of GeO_2 doped silica and pure silica, respectively. The refractive index difference between sections 1 and 2 is 0.7%. Section 2 covers 99.9% of the cross sectional area of the core. The cladding is made of fluorine doped silica and involves sections 3 and 5 made of low fluorine doped silica with an index difference of -0.35% and section 4 is made of high fluorine doped silica with an index difference of -0.94% . The diameters of sections 1–5 are 100 nm, 2.9 μm , 40 μm , 42 μm , and 125 μm , respectively. We fabricated a preform glass rod having a diameter of 25.5 mm by combination of vapor-phase axial deposition [5] and plasma-activated chemical vapor deposition and then, drew the 125 μm -diameter fiber at a speed of 150 m/min by heating the preform with a temperature of 2106°C while maintaining the drawing tension of the fiber as high as 75 g.

Fig. 1(c) shows the schematic illustration of the etching process for the fabrication of the triple tapered fiber. Here, we denote a buffered hydrogen solution (BHF) with a volume ratio of NH_4F solution (40 wt.%): HF acid (50 wt.%): H_2O as $X:1:1$. The process involves three steps: [A] hollowing of sections 1 and 4 in $X_A:1:1$, [B] tapering of sections 2 and 3 in $X_B:1:1$, [C] sharpening of section 1 in $X_C:1:1$. The values of X in three steps are defined as $X_A < 1.7$, $X_B = 1.7$, and $X_C > 1.7$. The etching time and X_A strongly affect both the total etching time and the diameter of the fabricated triple tapered fiber. In step [C], the taper angle α is controlled by varying X_C .

To obtain the triple tapered fiber with a diameter of about 35 μm , the fiber of Fig. 1(b) was consecutively etched by $X_A = 0.6$, $X_B = 1.7$, and $X_C = 10$ for 40 min, 20 min, and 10 min. All steps are performed at 25°C. Figs. 2(a), 2(b), and 2(c) show scanning electron micrographs of the triple tapered fiber, the magnified top region, and the apex region, respectively. The triple tapered fiber has an apex diameter of less than 10 nm (Fig. 2(c)). The values of the apex diameter d_1 , the base diameter d_2 , the first taper angle α , the second taper angle β , and the third taper angle γ are 100 nm, 2 μm , $60^\circ (\pm 1^\circ)$, $120^\circ (\pm 1^\circ)$, and $60^\circ (\pm 1^\circ)$, respectively. After completing the etching process, we coated the triple tapered fiber with an aluminium film with a radial thickness of 200 nm by vacuum evaporation. Fig. 2(d) shows the scanning electron micrograph of a top region of the metallized probe. The apex region of the probe was covered with aluminium with a few tens of nanometers.

Next, we discuss the etching rates of the multistep index fiber to tailor the triple tapered probe. From here on,

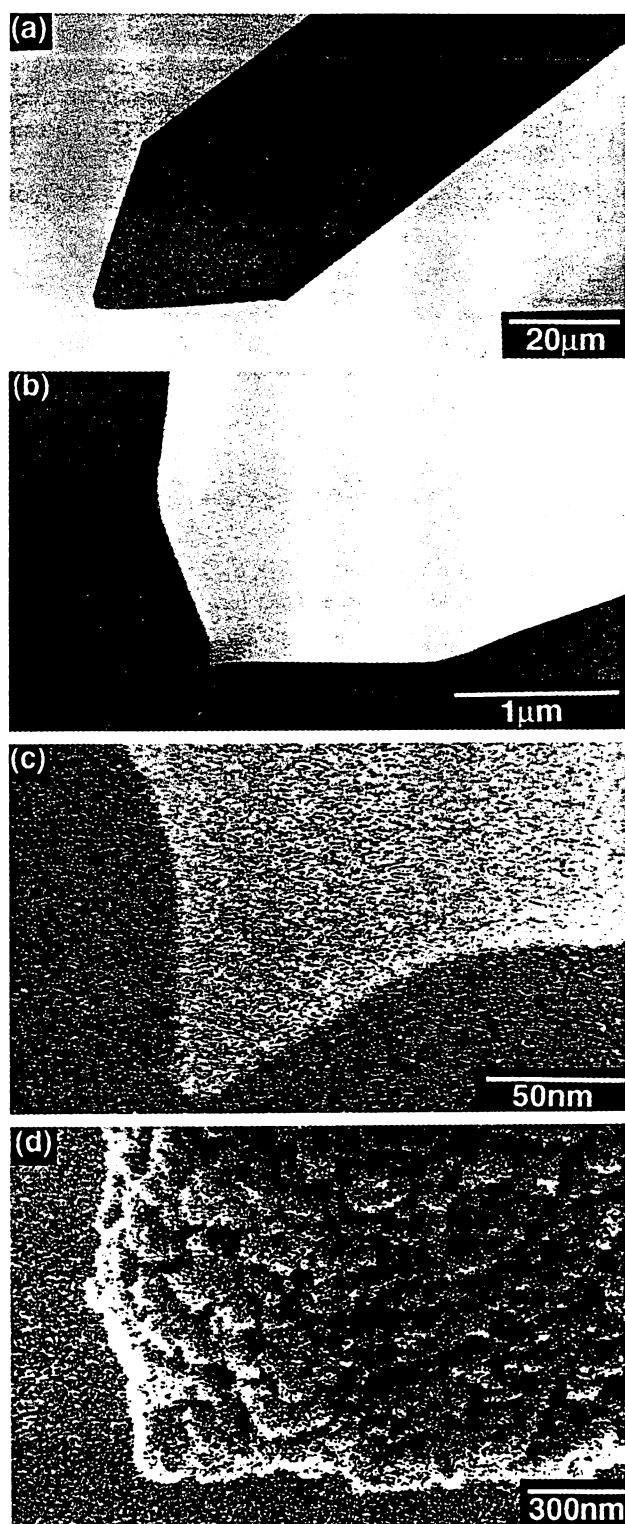


Fig. 2. Scanning electron micrographs of (a) the fabricated triple tapered fiber, (b) the magnified top region, (c) the magnified apex region. Here, the fiber is coated with gold having a thickness of 3 nm. The three angles of the triple tapered fiber are $\alpha = 60^\circ$, $\beta = 120^\circ$, and $\gamma = 60^\circ$. The apex diameter is less than 10 nm. (d) Scanning electron micrograph of the triple tapered probe coated with an aluminium thickness with a radial thickness of 200 nm.

the etching rates of sections 1–5 in step $[i]$ (where $i = A, B, C$) is defined as R_{1i} , R_{2i} , R_{3i} , R_{4i} , and R_{5i} ($= R_{3i}$), respectively. Fluorine doped silica has a smaller etching rate than pure silica which decreases with increasing doping ratio. Therefore, the etching rates satisfy $R_{3i}/R_{2i} = R_{5i}/R_{2i} < R_{4i}/R_{2i}$. On the other hand, the ratio of GeO_2 doped silica to pure silica decreases with increasing X value of BHF and converges to a constant value at $X = 10$ –30. At $X = 1.7$, the ratio is equal to unity. So, for $X_A < 1.7$, $X_B = 1.7$, and $X_C > 1.7$, the etching rates satisfy the conditions $R_{1A} > R_{2A} < R_{3A} = R_{4A} < R_{5A}$, $R_{1B} = R_{2B} < R_{3B} = R_{5B} < R_{4B}$, and $R_{1C} > R_{2C} < R_{3C} = R_{5C} < R_{4C}$, respectively. Based on our geometrical model [4], the taper angles α , β , and γ are expressed as

$$\sin(\alpha/2) = \sin(\beta/2)(R_{1C}/R_{3C}) \quad (1)$$

$$\sin(\beta/2) = R_{2i}/R_{3i} \quad (i = A, B, C) \quad (2)$$

$$\sin(\gamma/2) = R_{3i}/R_{4i} \quad (i = A, B, C) \quad (3)$$

respectively. For the fiber of Fig. 1(b), the right-hand side of Eq. (1) is 0.5 at $X = 10$ for an index difference of 0.7% and is diminished by increasing the index differences. The right-hand sides of Eqs. (2) and (3) take constant values of 0.87 and 0.50 in all steps using different concentrated BHF. To obtain the probe with a sharp angle $\alpha = 20^\circ$, we fabricated a preform in which sections 1 and 2 had a high index difference of 2.0%. However, the drawn fiber had a decreased index difference of 0.7%. The decrease of the refractive index can be attributed to diffusion of GeO_2 occurring during drawing of the fiber.

To evaluate the transmission efficiency of the triple tapered probe, we define a transmission coefficient given by the ratio of the detected intensity of light collected by a 0.4 NA objective lens to the input intensity. It was experimentally confirmed that the aluminized fiber had a transmission coefficient of about 1×10^{-4} at 633 nm wavelength which is as high as that of our single tapered probe having a taper angle of 20° and an aperture diameter of 100 nm. Employing the triple tapered probe having both such high transmission efficiency and a small apex diameter less than 10 nm, highly sensitive and highly spatially resolved imaging can be performed in the visible region or the near infrared region. To obtain higher transmission efficiency, we will fabricate the triple tapered probe with a protrusion [6] emerged from metal in the near future.

Finally, to demonstrate the imaging capability of the probe in the UV region, we measured the spatial distribution of photoluminescence (PL) of polydihexylsilanes, excited by an argon ion laser with a wavelength of 351 nm, by an illumination mode near-field optical microscope with shear force feedback control [7]. The sample was prepared by dropping a diluted solution of polydihexylsilanes with trans-planar-type conformation on a silica substrate. The typical length of polydihexylsilanes is a few hundreds of nanometers. Polydihexylsilanes have absorption and PL peaks at a wavelength of 370 nm and 380 nm,

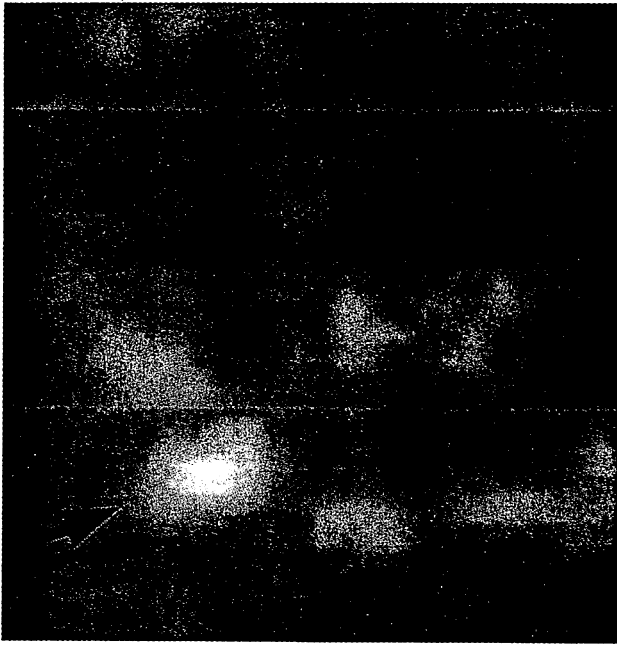


Fig. 3. Near-field photoluminescence image of polydihexylsilanes at 360 nm wavelength. Here, the scan area is $2 \mu\text{m} \times 2 \mu\text{m}$. Polydihexylsilanes were excited by an UV Ar^+ laser with a wavelength of 351 nm through the triple tapered probe. The sample–probe distance was controlled under shear force feedback. The bright portions in this figure represent UV-emission from polydihexylsilanes. The full width at half maximum of the portion indicated by the arrow is 150 nm.

respectively. For the detection of PL, we employed a band pass filter having a center wavelength of 380 nm and a line width of 9 nm, a photomultiplier, and a photon counter. Fig. 3 shows the photoluminescence image. Here, the scan area is $2 \mu\text{m} \times 2 \mu\text{m}$. To suppress bleaching of the photoluminescence, the excitation power was kept at less than 100 pW. The bright portions in this figure represent polydihexylsilanes emitting UV light. The width of the portion indicated by the arrow is as small as 150 nm. The contrast is very high. Further, we performed transmission imaging at 351 nm to demonstrate the resolution capability and obtained a near-field image of a particle with a diameter of 20 nm [8]. We found that the nanometric probe tip works near-field high spatial frequency components of

the optical near field. The details of polysilane images, will be discussed elsewhere.

In conclusion, we fabricated a triple tapered fiber probe for near-field optical spectroscopy in the UV region. The probe had an apex diameter of less than 10 nm of the triple tapered structure with angles of 60° , 120° , and 60° . Employing the probe, we obtained a photoluminescence image of polysilanes excited by a laser with a wavelength of 350 nm.

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Fabrication and evaluation of a localized plasmon resonance probe for near-field optical microscopy/spectroscopy

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A technique that excites a localized plasmon on the apex of probe for near-field optical microscope and spectroscopy is presented. Vapor deposition and heating of several tens nm thick gold films on 1 nm thick germanium-covered tapered optical fiber probe make highly adherent and smooth overlayers. A "tunneling-photon-mode" experimental system combined with a dye laser is used to excite a localized plasmon. The enhancement of near-field intensity was measured varying incident photon energies with its maximum occurring at 2.11 eV. This local field enhancement could be estimated to generate only in the close proximity of the substrate. © 1998 American Institute of Physics. [S0003-6951(98)00511-7]

The localized collective oscillation of electrons, i.e., "localized" plasmons excitations in small particles or protrusions of noble metals cause the strong enhancement of local-field, which plays an important role for the optical properties of rough or nanostructured surfaces of such metals.¹⁻⁴ Especially, in the context of surface enhanced Raman scattering, the electromagnetic enhancement by "localized" plasmons excitations drew considerable attention, and is considered to be the dominant origin of its phenomenon in noble metals.⁵⁻⁷ This surface-enhanced nonlinear-optical process was proposed to be used as a probe of a scanning microscope.⁸ Furthermore, by using small metallized polystyrene particles, as probes of near-field optical microscope,⁹ a large increase in detected light intensity compared to conventional probes, has been observed.¹⁰

In near-field optical microscope, various configurations, i.e., illumination, collection, and reflection mode^{9,11-13} and tunneling-photon mode,^{9,14,15} have been developed. In the former three modes, the probe consists of a sharpened tapered optical fiber with metal-clad and a narrow aperture at its front end acting as a generator or a detector of near-field in the proximity of samples. High-resolution beyond the diffraction limit can be achieved by the sharpness of the apex and small size of the aperture. In the tunneling-photon mode, on the other hand, the probe works as a scattering center for image formation. Since the sample modulation of the near-field, generated under total-internal-reflection (TIR) configuration, strongly depends on the distance from samples, high-resolution imaging is possible in close proximity of the samples. The negative dielectric constant of the sample and/or probe is also known to contribute to higher spatial-resolution owing to both the classical electrostatic (Rayleigh) and surface plasmon excitation effects.¹⁶⁻¹⁸

Local-field enhancement between sample and probe is not only expected to attain high spatial resolution and contrast in near-field microscopy, but also high efficiency of

excitation or sensitivity of detection in near-field spectroscopy, especially near-field Raman spectroscopy.¹⁹ In the present study, we have fabricated a smoothly metal-coated fiber probe with sharpened taper, and excited the "localized" plasmon at the probe apex in the tunneling-photon-mode configuration. We report here the first achievement of local-field enhancement that strongly depends on both the incident photon energies and probe-substrate separations. In the illumination mode, the light confinement and local-field enhancement at the probe apex is theoretically investigated.²⁰ As our fabricated probe is analogous to the treated model in Ref. 20, the localized plasmon excitation and light confinement in other configurations, e.g., illumination mode, will be also suggested.

The sharpened optical-fiber probe for the near-field microscope was prepared by selective etching the fiber end by dip into a mixture solution of NH_4F , HF , and H_2O . Such an etching technique makes a sharpened core whose apex diameter is far less than the wavelength with high reproducibility.²¹ After this process, the "bare" fiber probes were set on holders having rotary machinery inside a vacuum chamber. By rotating axially at $\sim 5-6$ rpm, germanium was initially evaporated on the etched surfaces for 0.4-0.5 min with 2-3 nm/min rate at $\sim 5 \times 10^{-4}$ Pa vacuum, and gold was consecutively evaporated for 2-3 min at 10-20 nm/min. During and after (for nearly 3 h) depositions, the probe was heated by radiation.

The scanning electron microscope (SEM) images of the fabricated probe are shown in Fig. 1. Figure 1(a) shows the whole view and Fig. 1(b) is the magnification of the apex. The grain size of the gold overlayer was increased by radiation heat, whereas roughness remained smaller. A schematic model of the fabricated probe is depicted in Fig. 1(c). The cone angle of the fiber core was $\sim 19^\circ$, and that of the coated one was $\sim 20^\circ$. The apex diameter was ~ 10 nm, and the coated one was 60-70 nm. The overlayer thickness can be estimated to be 25-30 nm at the apex and ~ 80 nm at the root of the sharpened core. This configuration has been formed by conventional annealing. The surface morphology of the annealed metal-coated probe, however, almost becomes heterogeneous since the adhesion of the metal over-

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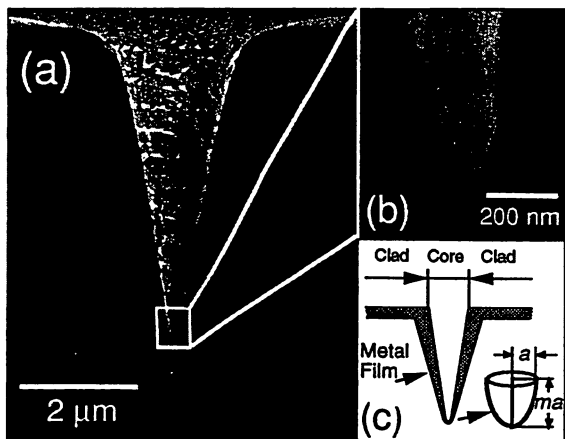


FIG. 1. Scanning electron microscope (SEM) images of the fabricated probes: (a) whole view of the probe with gold film deposited on a germanium-coated surface, (b) the magnified image of apex of (a), (c) a schematic cross-sectional model of (a).

layer on the glass surface is so weak.

We have measured the adhesive force (Benjamin-Weaver force: F_{BW}) of gold film deposited on a glass plate under almost the same conditions as those of the tapered fiber using the Scratch test.^{22,23} The critical load W_c of the stylus was ~ 0.3 mN on ~ 30 nm thick gold film deposited on the glass substrate at room temperature. On the other hand, the W_c was ~ 6.2 mN for ~ 30 nm thick gold film deposited on Ge(1 nm)/glass under radiation heat as mentioned before. The F_{BW} of each of the above mentioned gold films were ~ 4.5 and ~ 23 MPa, respectively. This result shows the considerable increase of the adhesive force by the insertion of only thin Ge layers. An atomic force microscope analysis also shows that the surface roughness of the annealed gold film on the Ge(1 nm)/glass is much less than that on the bare glass. This is because the Ge layer works as a binder between the gold film and the glass substrate. The increase of adhesive force, consequently makes the annealed

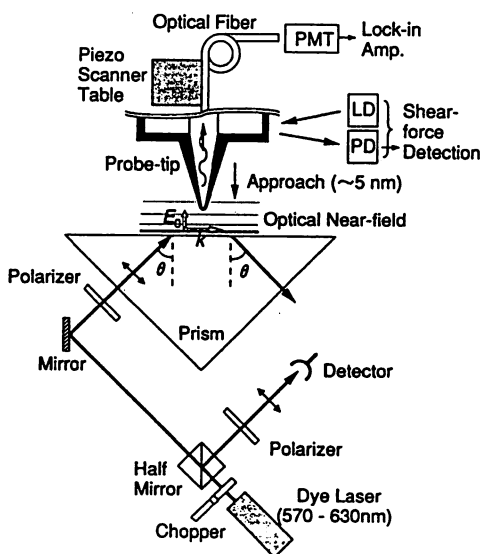


FIG. 2. Schematic diagram of the experimental setup for excitation of a localized plasmon on the front end of the probe

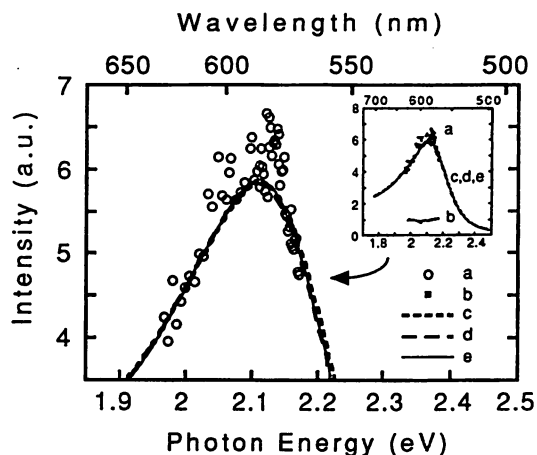


FIG. 3. Dependence of the detected intensity on incident photon energy. Inset shows the full scale variation: (a) the detected intensity for the “plasmon” probe, (b) that for the uncoated probe, (c)–(e) fitting curves given from Eq. (1) using parameters in Table I. The vertical axis is normalized by the value for the uncoated probe at 2.11 eV.

gold film on the sharpened fiber smooth and mechanically strong.

We examined the generation of localized plasmons on the fabricated probes in a tunneling-photon-mode configuration as shown in Fig. 2. A p -polarized and unfocused dye(R6G) laser was totally reflected on one interface of prism(BK7)/air keeping the incident angle at $45^\circ \pm 0.01^\circ$ ($> \theta_c = 41.24^\circ$; critical angle for TIR). The probe apex was maintained at ~ 5 nm separation above the interface by shear force feedback control. The radiation picked up by the probe was detected by the photomultiplier tube at the fiber end, and was converted into electrical signals. The incident light power was controlled to be $\sim 150 \mu\text{W}$ by detecting the split light with a half mirror. The excitation photon energy of the dye laser with ~ 40 GHz linewidth was varied from 1.97 to 2.18 eV (from 570 to 630 nm in wavelength). The signal intensities picked up by the “plasmon” probe and by an uncoated fiber probe were plotted together with fitting curves in Fig. 3. The plots for the “plasmon” probe indicate a clear dependence on the incident photon energy compared with the little dependence for the uncoated probe. The fitting curves have a peak at ~ 2.11 eV. The maximum intensity for the “plasmon” probe was about 5.8 times that for the uncoated.

The fitting curves as shown in Fig. 3 were obtained by adjusting the fabricated probe configuration to the surface local-field theory²⁴ including the damping effect.²⁵ In this theory isolated hemispheroids on a perfect conductive plane and incident electromagnetic field E_0 were assumed. The local-field enhancement factor E_{tip}/E_0 is given as follows:

TABLE I. Parameters to give fitting curves in Fig. 3 and corresponding magnitude of local-field enhancement. Parameter “a” is fixed from SEM image of fabricated probe.

	ϵ_s	m	a (nm)	$ E_{\text{tip}}/E_0 ^2$
curve c	1.00	2.61	31	385
curve d	1.53	1.92	31	202
curve e	2.07	1.52	31	128

$$\frac{E_{\text{tip}}}{E_0} = \frac{\epsilon_m / \epsilon_s}{1 + [(\epsilon'_m / \epsilon_s) - 1]A + i(\epsilon''_m / \epsilon_s)A + R\epsilon''_m \sqrt{\epsilon_s} + iR(1 - \epsilon'_m) \sqrt{\epsilon_s}}, \quad (1)$$

with

$$R = \frac{4\pi V}{3\lambda^3} = \frac{16\pi^3 m}{9} \left(\frac{a}{\lambda}\right)^3,$$

$$A = \frac{1}{m^2 - 1} \left[\frac{m}{2\sqrt{m^2 - 1}} \ln \left(\frac{m + \sqrt{m^2 - 1}}{m - \sqrt{m^2 - 1}} \right) - 1 \right],$$

where ϵ_m and ϵ_s are dielectric constants of the hemispheroid and the surrounding medium, respectively, a and m are one semiaxis of the hemispheroid and the aspect ratio [refer to Fig. 1(c) for the definition], $V = 4\pi m a^3 / 3$ is the full spheroid volume, and λ is the wavelength of a driving field. The tunneling-photon-mode setup in our experiment can be compatible with this model on the assumption that the incident field E_0 in Fig. 2 is uniform without dependence on the distance from the prism/air interface. The fitting curves in Fig. 3 are given by substituting three sets of values of a , m , and ϵ_s as shown in Table I for $|E_{\text{tip}}/E_0|^2$ derived from Eq. (1), where the dielectric constant of gold in Ref. 26 was used as ϵ_m . The effect of glass fiber core was neglected because the SEM observation of the ‘‘plasmon’’ probe shows that the diameter of the metal-coated apex is 6–7 times as large as the uncoated.

It has been claimed that the dielectric constant ϵ_s of the space surrounding the metallic probe becomes larger when the probe gets closer to the substrate.¹⁰ The fitting curves are almost the same in spite of the change of parameters in Table I. The decay length of the near-field intensity in our experiments was 177.6–197.8 nm. The assumption $E_0 \sim \text{const.}$ can be effective if the dimension of the probe apex is much smaller than incident wavelength. In SEM images of the fabricated probe, the length ‘‘ $m \times a$ ’’ from the cross section of radius ‘‘ a ’’ in Table I to the top of probe, as depicted in Fig. 1(c), shows that the aspect ratio ‘‘ m ’’ is between 1.5 and 2.0. Hence the dielectric constant ϵ_s is 1.5–2.0, and the expected magnitude of enhancement is 100–200. However, in Fig. 3 the observed magnitude was, at the largest, about six times as large as that of the uncoated probe. This can be because the background intensity, e.g., collection of scattering radiation from the prism surface by the tapered core, was dominant for the uncoated probe or the metal overlayer thickness of our fabricated probe was too large to transmit to the fiber core.

In addition, we also measured the probe-substrate separation dependence of the detected intensities at nearly resonant photon energies (~ 2.11 eV) in order to investigate the contribution of near-field to the localized plasmon excitation. In the case of the fabricated ‘‘plasmon’’ probe, within ~ 20 nm from the sample surface, there is an *additional*

*strong narrow peak*¹⁰ that corresponds to the generation of local-field enhancement between the probe apex and the substrate. The resonance condition of this ‘‘plasmon’’ probe can be satisfied only in close proximity to the substrate.

In summary, we report the first achievement of localized plasmon enhancement that strongly depends on both the incident photon energies and probe-substrate separations. The deposited gold film on 1 nm germanium-covered sharpened tapered fiber probe is smooth and mechanically strong. The magnitude of the near-field enhancement is expected to be 100–200 times as large as the incident electromagnetic field.

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Low Temperature Near-Field Photoluminescence Spectroscopy of InGaAs Single Quantum Dots

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We investigate InGaAs single-dot photoluminescence spectra and images using a low-temperature near-field optical microscope. By modifying the commonly used near-field technique, a high spatial resolution and high detection efficiency are achieved simultaneously. Local collection of the emission signal through a 500 nm ($\lambda/2$) aperture contributes to the single-dot imaging with a $\lambda/6$ resolution, which is a significant improvement over the conventional spatially resolved spectroscopy. Tailoring the tapered structure of the near-field probe enables us to obtain the emission spectra of single dots in the weak excitation region, where the carrier injection rate is $\sim 10^7$ excitons/s per dot. By employing such a technique, we examine the evolution of single-dot emission spectra with excitation intensity. In addition to the ground-state emission, excited-state and biexciton emissions are observed for higher excitation intensities. By a precise investigation of the excitation power dependences of individual dots, two-dimensional identification of their emission origins is obtained for the first time.

KEYWORDS: near-field optical microscope, quantum dot, photoluminescence, exciton, biexciton

1. Introduction

With the progress in the fabrication techniques of self-assembled quantum dots (QDs),¹⁻³ the unique features of semiconductor nanostructures have received increasing attention in the last few years. One of the most attractive properties of the zero-dimensional confined structure is the narrow and strong optical transition due to its atomic-like discrete density of state, which is favorable for optoelectronic devices, such as lasers with a low threshold current density. Photoluminescence (PL) spectroscopy is the most commonly used method for obtaining information regarding the electronic structures and relaxation processes in QDs. Spectroscopy of a single QD is essential for the precise evaluation of the intrinsic capability of the QD since the conventional far-field studies measure an ensemble of dots with size and shape fluctuations, which result in inhomogeneously broadened spectral features. In order to realize single QD spectroscopy, we need to restrict the observation area by applying novel microscopic techniques.

Here we describe several techniques for single-dot PL spectroscopy. The first involves the use of local electron injection methods, such as a high-energy electron beam from a scanning electron microscope (cathodoluminescence)^{4,5} and low-energy electron injection through a scanning tunneling microscope tip (tunneling luminescence).⁶ As the injected electrons or generated electron-hole pairs can diffuse before recombination, the spatial resolution is limited by the diffusion area as long as the PL signal is collected by a lens in the far-field configuration. The second method involves the use of micro-PL techniques, where the observation area is reduced by employing micro-fabrication techniques, such as the fabrication of mesa structures^{7,8} and a metal mask with small windows.⁹⁻¹¹ In the metal-mask method, as shown in Fig. 1(a), since the PL signal is also collected through the window, we can achieve the spatial resolution determined by the aperture size free from the carrier diffusion effect. Recently, the metal-mask technique has been regarded as the most successful method for the realization of single QD spectroscopy. Its only disadvantage is the lack of the scanning ability of the metal mask. Compared with these conventional techniques,

a near-field optical microscope¹²⁻¹⁴ has achieved both merits simultaneously, that is, the high spatial resolution and the scanning ability, as the near-field probe, operating in the illumination and collection hybrid mode,^{15,16} can be considered as a scanning mask with a small aperture. (see Fig. 1(b)).

In this letter, we report on the experimental results of single-dot PL spectroscopy using a low-temperature near-field optical microscope with the illumination and collection hybrid mode. By employing a highly sensitive near-field probe with a large aperture of 500 nm ($\lambda/2$) and an optimized shape at the apex, we have successfully measured the PL spectra and images of single QDs with a resolution of $\lambda/6$ and a considerably high sensitivity. The power dependent features of single QD emissions are also investigated in order to determine the states from which these emissions originate.

2. Near-Field Fiber Probe

The heart of the near-field optical microscope is the fiber probe, which has a small aperture at its apex (see Fig. 2(a)). The aperture corresponds to the small window in the metal-mask technique. After tapering the end of an optical fiber, the aperture is made by coating its sides with a metal, such as aluminum or gold. Other than the aperture size, the most important factor which affects the sensitivity of the near-field probe is the shape and the modal occupation of its tapered

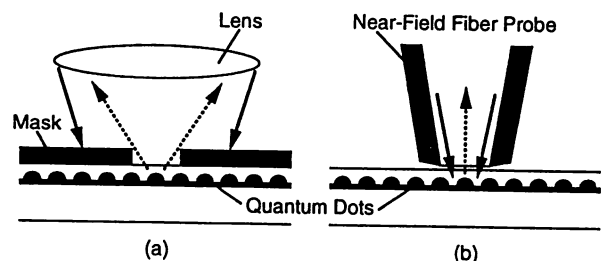


Fig. 1. (a) Micro-photoluminescence technique with sub-micron sized hole in the metal mask and (b) near-field optical microscope operating in illumination and collection hybrid mode. The near-field probe is considered as a scanning metal mask coupled with an illumination and collection lens.

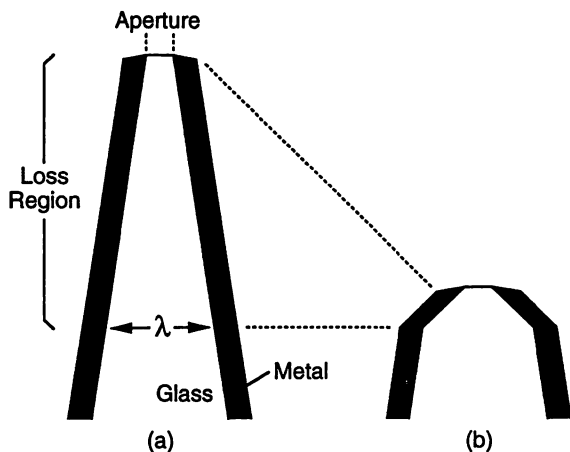


Fig. 2. (a) Standard-type near-field fiber probe and (b) a double-tapered one optimized to shorten the optical loss region and to have a large cone angle at the apex, which contributes to an efficient illumination and collection of light.

region. The tapered region of the probe can be considered as the metal-cladding optical waveguide, whose propagation properties are characterized by the cutoff diameter and the absorption coefficient of the cladding metal. Due to the poor propagation property in the narrow tapered region, it is generally said that the sensitivity of the near-field probe is lower than the metal-mask technique combined with the high numerical aperture (NA) far-field optics. Therefore, the optimization of the structure of the near-field probe is the most significant factor to improve its sensitivity.^{17,18)}

To tailor the shape of the probe, it is necessary to determine the most influential factor in the transmission of the metal-cladding waveguide. By evaluating the transmission efficiency of the probe for a wide range of the aperture diameter, it is confirmed that the efficiency decreases in the region where the core diameter is smaller than the wavelength of the propagating light. On the basis of these experimental results, the shape of the probe is optimized by shortening the optical loss region as shown in Fig. 2(b).¹⁷⁾ The double tapered structure is easily realized using a multi-step chemical etching technique. It is demonstrated that the transmission efficiency is improved by one order of magnitude as compared to the standard type near-field probe (Fig. 2(a)).

3. Experimental

Figure 3 shows a schematic of the QD sample structure and the experimental configuration. In_{0.5}Ga_{0.5}As self-assembled QDs were grown on a (100) GaAs substrate by gas-source molecular beam epitaxy with a density of $\leq 10^{10}$ dots/cm².³⁾ A typical dot diameter of around 30 nm and a height of 15 nm were observed using an atomic force microscope. These dots were covered with cap layers composed of GaAs and AlGaAs with a total thickness of 80 nm. The wide gap layers of AlGaAs were introduced in order to prevent carrier leakage from the GaAs layer to the substrate or surface.

We employed a chemically etched fiber probe¹⁹⁾ with an aperture²⁰⁾ of 500 nm diameter, which was around half the PL wavelength of QDs. The shape of the tapered part was optimized to attain a high sensitivity as mentioned in §2. For the near-field excitation and detection, the aperture was con-

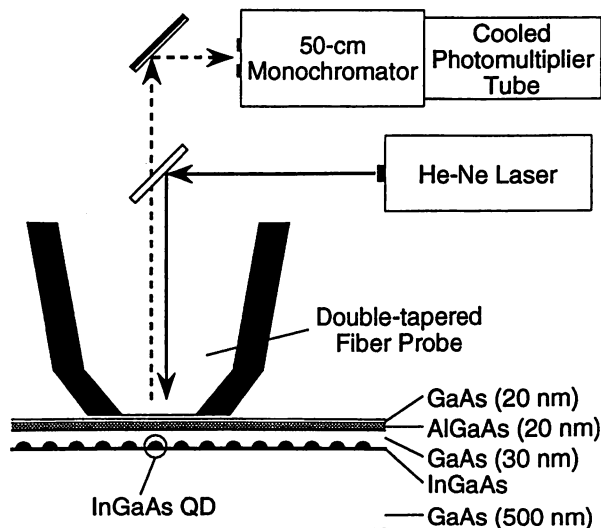


Fig. 3. Schematic of a sample structure, optical near-field configuration and block diagram of the measurement.

trolled in close proximity to the sample surface (~ 10 nm) by employing the shear-force feedback technique.²¹⁾ The QD sample on a scanning piezotube was illuminated with a He-Ne laser light ($\lambda=633$ nm) through the aperture. Carriers were generated not only in the InGaAs QDs but in the barrier layers of GaAs and AlGaAs. Most of the photoexcited carriers migrated in the barrier layers and were captured by the confined states of the QDs. In order to achieve a high spatial resolution free from the carrier diffusion effect, the resultant PL signal was collected by the same aperture.^{15,16)} In the case of signal collection through the aperture, the resolution was determined by various factors of the probe rather than the aperture diameter, as detailed in §4. After rejecting the excitation light by means of a long pass filter, the signal was focused on a 50 cm monochromator and was detected by a cooled photomultiplier tube (Hamamatsu R5509-41, ≤ 10 dark counts/s, 0.2–1% quantum efficiency at the detection wavelength of 0.92–1.0 μm) by a photon counting technique. The sample, the scanner, and the fiber probe were mounted in a continuous He-gas flow optical cryostat and all the measurements were performed at 5–10 K.

4. Basic Performance of Near-Field Optical Microscope

Figure 4(a) shows a near-field PL spectrum with a weak excitation power density of 0.2 W/cm². By restricting the observation area with near-field excitation and collection, we obtained a sharp emission line from a single quantum dot, which was buried in the inhomogeneously broadened far-field spectrum. The linewidth of 0.5 meV was determined by the spectral resolution of the monochromator. Within an excitation area of $\pi(0.25 \mu\text{m})^2$, the excitation light of 0.4 nW through the aperture generates 1×10^9 electron-hole (e-h) pairs per second. From the value of dot density, the injection rate of the e-h pairs into the individual dots was estimated to be $< 1 \times 10^8$ e-h/s. In this estimation, we did not take into account the effect of carrier diffusion. Since the injection rate of the e-h pairs was much smaller than $1/\tau$, where τ is the ground state emission lifetime of about 1 ns, we concluded that the

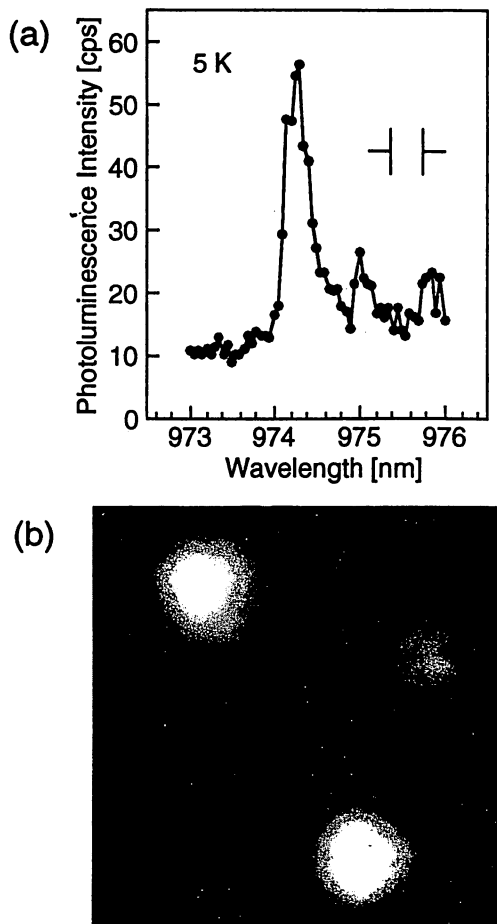


Fig. 4. (a) Photoluminescence spectrum of InGaAs single quantum dot detected in near-field configuration at 5 K. The linewidth of 0.5 meV is limited by the spectral resolution of the monochromator. (b) Monochromatic photoluminescence image of single dots with an emission wavelength of 965 nm and a spectral resolution of 1 nm. The scanning area is $0.8 \mu\text{m} \times 0.8 \mu\text{m}$.

observed emission lines originated from the recombination of ground-state excitons.

Here we roughly estimate the collection efficiency of the PL signal of the near-field probe. When the injection rate of the excited carriers per single dot was 1×10^8 e-h/s, the resultant photon counting rate of the PL signal was around 50 counts/s. By using the apparatus parameters of the detection system, such as filters, monochromator, and photomultiplier tube (0.5% quantum efficiency at 970 nm), the product of the quantum efficiency of the dots (ϕ_{qe}) and the collection efficiency of the probe (ψ_{ce}) could be evaluated as $\phi_{qe} \times \psi_{ce} \sim 10^{-3}$. If ϕ_{qe} is equal to 10%, ψ_{ce} reaches 1%, which was as high as the collection efficiency in the high NA far-field configuration.

Figure 4(b) shows a monochromatic PL image constructed by fixing the detection wavelength at 965 nm and scanning the probe in $0.8 \mu\text{m} \times 0.8 \mu\text{m}$ area. Each circular spot corresponds to a single QD with an emission wavelength of around 965 nm. The minimum and average sizes of the spot images at full-width at half maximum are 160 nm and 220 nm, respectively. A significantly high spatial resolution of $\lambda/6 - \lambda/5$ was achieved.²²⁾ It should be noted that the obtained spot size

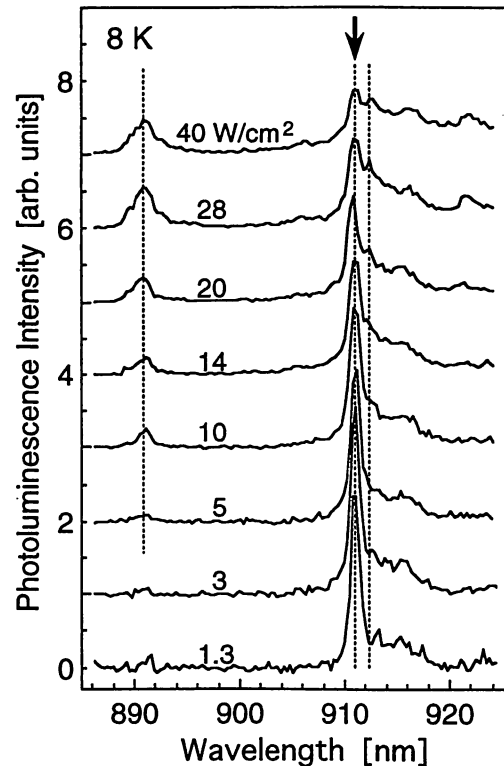


Fig. 5. Single-dot photoluminescence spectra observed at 8 K with various excitation densities. The emission wavelength of the ground-state single exciton is indicated by an arrow.

was much smaller than the aperture diameter of 500 nm ($\lambda/2$). Since the carriers generated in the barrier layers of GaAs diffuse into the extended region, the spot size is determined only by the collection area of the PL signal through the aperture. For the apertures with diameters of 500 nm–1 μm , it was demonstrated that the resolution was not strongly dependent on the aperture diameter; even when a rather large aperture of 1 μm diameter was utilized, a similar resolution of around 250 nm was obtained. The mechanism of achieving such a high resolution has not been fully understood. We assume that the close proximity of the glass apex to the sample, a large cone angle of the tip, and the modal occupation of the tapered region contribute to the realization of something like a “high NA lens”.

5. Identification of Various Emission Origins

The evolution of the single-dot PL spectrum with excitation intensity yields information on the intrinsic physical properties of a zero-dimensionally confined system, such as the quantization energy of electrons and the binding energy of electron-hole pairs. Figure 5 shows the excitation power dependence of the PL spectra of a single quantum dot. The peak intensity of each spectrum is normalized by the excitation power density. For an excitation power lower than 5 W/cm², the PL spectra consisted of a single line. As mentioned in §4, with this power density, the single PL line originates from the recombination of single excitons from the ground state. With increasing excitation power, other elementary features were observed in the PL spectra. After the saturation of the ground-state emission, a new PL line appeared at 30 meV

above the ground-state transition. This line corresponds to the emission from the first excited state, which arises from the state filling of the ground state.⁹⁾ Furthermore, Fig. 5 shows another feature 2.2 meV below the single-exciton emission. The emission intensity increases super-linearly with the excitation intensity. From the energetic position and the excitation power dependence, this emission line can be associated with the recombination of biexcitons,²³⁾ the rate of which increases quadratically with the generation rate of the single exciton in a single dot.²⁴⁾

Figure 6 shows the monochromatic PL images in the same scanning area of $3\ \mu\text{m} \times 3\ \mu\text{m}$ for various excitation power densities. The detection wavelength is fixed at 975 nm with the bandwidth of 1 nm. The number of emission spots from the individual dots changes drastically with an increase in the

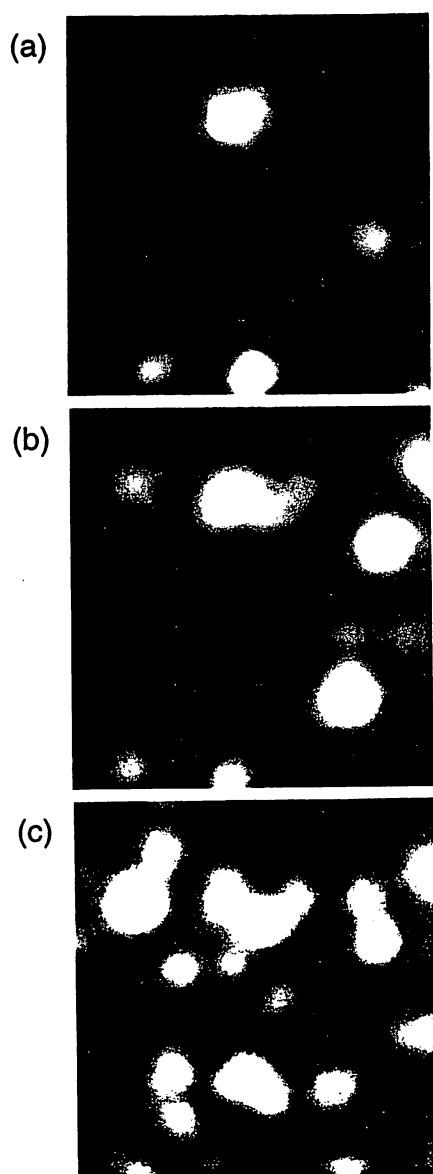


Fig. 6. Monochromatic photoluminescence images obtained at 8 K with various excitation densities of (a) $3.9\ \text{W}/\text{cm}^2$, (b) $60\ \text{W}/\text{cm}^2$ and (c) $700\ \text{W}/\text{cm}^2$. The detection wavelength is 975 nm with a spectral bandwidth of 1 nm and the scanning area is $3\ \mu\text{m} \times 3\ \mu\text{m}$.

excitation intensity, while the spot sizes remain unchanged. Such a power-dependent behavior shows the appearance of emissions from the excited states and the biexciton states, which are also observed in the PL spectra in Fig. 5.

In order to determine the origin of the individual emissions, Fig. 7 shows the plot of the spot intensity as a function of the excitation power density for each spot observed in Fig. 6. Specific power dependent behaviors can be classified in three types as shown in Fig. 7(a)–7(c). The plot in Fig. 7(a) shows a linear increase in the weak excitation region. The beginning of saturation at the excitation intensity of around $10\ \text{W}/\text{cm}^2$ is consistent with the result obtained in Fig. 6. Such an evolution of the emission intensities confirms that these emissions originate from the recombination of the ground-state excitons. The decrease in the emission intensity for higher excitation is due to an increase of the generation rate of biexcitons and to the change in the injection rate of the e-h pairs into the individual dots. Another behavior shown in Fig. 7(b) depicts quadratic dependence on the excitation density, which is one of the most characteristic features of biexciton recombination. The other emissions observed in Fig. 7(c) are classified with respect to the threshold intensities in the appearance of individual spots and their saturation intensities. These plots display the appearance of the higher excited states due to the state filling of the lower excited and ground states. Based on these considerations, we can identify the origins of the individual emission spots, which are summarized in Fig. 8. This two-dimensional

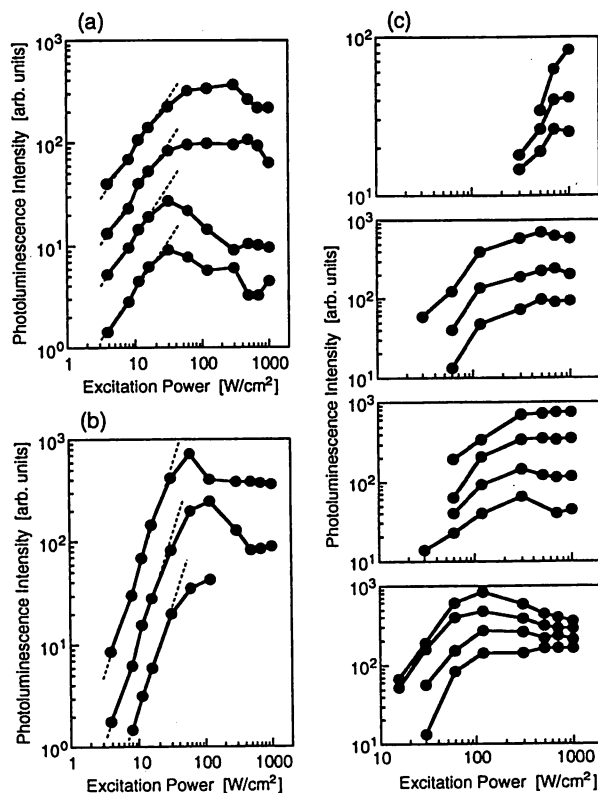


Fig. 7. Plots of emission spot intensities as a function of excitation densities for each spot observed in Fig. 6. (a) linear increase, (b) quadratic increase in weak excitation region and (c) having threshold in the appearance of emission. Plots in (c) are classified according to the threshold intensity and saturation intensity.

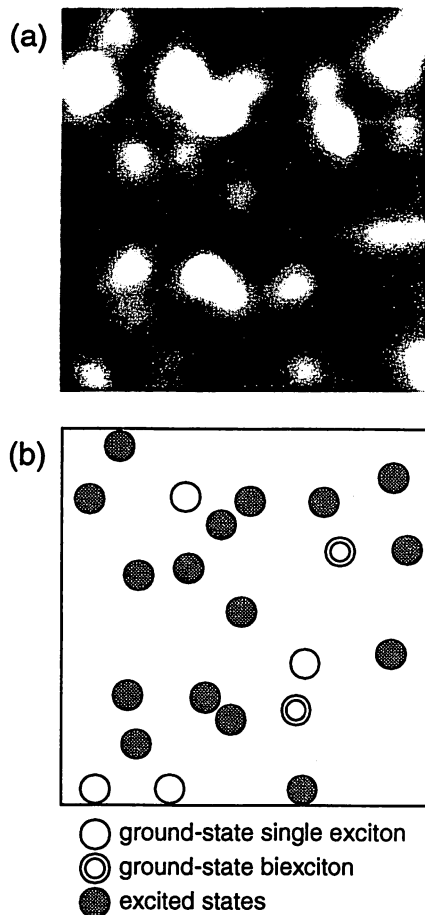


Fig. 8. (a) Monochromatic photoluminescence image with excitation density of 500 W/cm^2 in the same scanning area shown in Fig. 6. (b) Corresponding two-dimensional map indicating the emission origins of individual spots in (a).

specification is especially important in the investigation of the lasing mechanism of QD lasers.

6. Summary

We have performed the single QD spectroscopy with sufficient spatial resolution and detection sensitivity using the low temperature near-field optical microscope. The operation in the illumination and collection hybrid mode realizes considerable high spatial resolution of $\lambda/6$, whereas a rather large aperture of $\lambda/2$ diameter is employed in order to achieve a high collection efficiency of the PL signal through the aperture. The problem of the poor propagation property of the tapered region of the probe is solved by the optimization of the probe to have a large cone angle at the apex, which also appears to contribute to the narrow collection area of the PL signal. We examined the evolution of the single-dot PL spectra and images with excitation power density. In addition to the ground-state single-exciton emission, some other features were clearly observed. From their energetic positions in the

spectra and their power dependent behaviors, we relate them with the excited-state emission and biexciton emission. This successful first acquisition of the two-dimensional identification of emission origins is attributed to the scanning ability of the near-field optical microscope. The measurement techniques reported here have a variety of applications for the spatially resolved spectroscopy of semiconductor nanostructures. The most significant advantage of this system in actual operation is the high reproducibility of spatial resolution, which is relatively insensitive to the aperture diameter.

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Development of a Fiber Used for Fabricating Application Oriented Near-Field Optical Probes

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Abstract—We propose and develop a new fiber for application oriented probes for near-field optical microscopy. Based on a geometric model of selective etching, we design the fiber having the germanosilicate-doped silica core, an inner pure silica clad, and an outer fluorine doped silica clad. The fiber is fabricated by vapor-phase axial deposition. By selective etching of the fiber in buffered hydrogen fluoride solutions, we fabricated three probes having high-resolution capability and/or high-transmission efficiency for applications to high-resolution imaging and local spectroscopy. The apex diameters of the probes were less than 10 nm.

Index Terms—Etching, microscopy, near-field optics, optical fibers, optical fiber (device) fabrication, optical fiber measurement applications.

NEAR-FIELD optical microscopy (NOM) [1]–[3] employing a tapered fiber probe is applied to optical imaging of nanometric biological samples and locally spectroscopic study of optical devices in a nanometric region. In imaging applications requiring high-spatial resolution, a tapered probe with a small taper angle and a nanometric apex diameter has to be fabricated because the resolution capability of the probe is determined by its parameters such as the taper angle, the apex diameter, etc. However, the transmission efficiency of the probe is diminished on decreasing the taper angle [4]. Further, in spectroscopic applications where one must bear with extremely low detected power, it is required that the probe has high-transmission efficiency in illumination mode NOM¹ and is highly sensitive in collection mode NOM.² In illumination mode, to avoid thermal damage of the sample and the probe, the probe should be used with an input power as low as possible. Therefore, the resolution capability and the transmission efficiency of the probe have to be optimized depending on the application of NOM to imaging or spectroscopy. This has to be done through tailoring of the probe to an optimized shape. In this letter, we propose a fiber used for fabricating application oriented near-field optical probe. We also describe methods to fabricate three types of

probe, a pencil-shaped probe with high resolution capability, a probe with high transmission efficiency, and a triple tapered probe with high resolution capability and high transmission efficiency. For details of pencil-shaped probe and the triple tapered probe, refer to [5] and [6], respectively.

Fig. 1(a) shows the cross-sectional profile of the refractive index difference Δn of the proposed fiber. Here, the fiber is composed of three sections; (1) the germanosilicate (GeO_2) doped core, (2) the pure silica clad, and (3) a fluorine doped silica support. The value of Δn of sections (1) and (3) with respect to section (2) are 1.2% and -0.7% , respectively. The diameters of sections (1)–(3) are $[2r_1] = 1.3 \mu\text{m}$, $[2r_2] = 27 \mu\text{m}$, and $[2r_3] = 125 \mu\text{m}$, respectively. Fig. 1(b) shows a schematic diagram of the etching process. The process consists of three steps [A], [B], and [C]. By denoting the etching rate of section (i) in step [j] as R_{ij} (where $i = 1, 2, 3$; $j = A, B, C$), we assume that the fiber has the relative etching rates $[R_{ij}/R_{3j}]$. Their profiles are as shown in Fig. 1(c). The relative etching rate of GeO_2 doped glass to pure silica glass depends on the concentration of buffered hydrogen fluoride solution (BHF) mixed with a volume ratio of NH_4F solution (40 wt.%): HF acid (50 wt.%): H_2O being $X:1:Y$. From here on, we denote BHF as $X:1:Y$. To obtain the etching profiles, we can use for example, 1.7:1:1, 10:1:1, and 1.7:1:5, in which the etching rates of sections (1) and (2) are experimentally found to satisfy the relation

$$\frac{R_{1B}}{R_{2B}} = 0.29 < \frac{R_{1A}}{R_{2A}} = 1.0 < \frac{R_{1C}}{R_{2C}} = 1.48 \quad (1)$$

respectively. On the other hand, the relative etching rates of fluorine doped silica glass to pure silica glass are independent of the concentration of BHF. Therefore, the relative etching rates of section (3) to pure silica take a constant value of

$$R_{2j}/R_{3j} = 0.51 \quad (\text{where } j = A, B, C) \quad (2)$$

which is smaller than unity. Based on the geometrical model [7], we consider the etching process of using 1.7:1:1, 10:1:1, and 1.7:1:5, respectively, in steps [A], [B], and [C]. In step [A], the fiber is tapered to an angle of θ_A given by

$$\sin(\theta_A/2) = R_{2j}/R_{3j} \quad (\text{where } j = A, B, C). \quad (3)$$

Assuming the fiber diameter to be equal to $[2r_2]$ after step [A], the etching time T_A is given by

$$T_A = (r_3 - r_2)/R_{3A}. \quad (4)$$

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¹In the illumination mode, the probe is used as a generator of optical near field, the sample is illuminated by the probe, and the transmission light is detected.

²In the collection mode, the sample is illuminated under total internal reflection, the probe is used as a scatterer of the optical near field over the sample, and the scattered field is detected.

If T_A is larger than the etching time τ_A where τ_A is the time required for making an apex diameter of $[2r_1]$ and expressed as

$$\tau_A = \frac{r_2 - r_1}{R_{3A}} \sqrt{\frac{R_{2A} + R_{3A}}{R_{3A} - R_{2A}}} \quad (5)$$

the tapered fiber will have an apex diameter less than $[2r_1]$. So, it is straightforward to find that the radius r_2 of section (2) must be smaller than the critical radius r_{2P} expressed as

$$r_{2P} = \frac{r_3 + \xi \cdot r_1}{1 + \xi}$$

where

$$\xi = \sqrt{\frac{R_{2A}/R_{3A} + 1}{1 - R_{2A}/R_{3A}}} \quad (6)$$

In step [B], section (1) is sharpened with a different angle θ_B given by

$$\sin(\theta_B/2) = R_{1B}/R_{3B} \quad (7)$$

When the etching time T_B is larger than τ_B expressed as

$$\tau_B = (r_1/R_{3B})[(R_{1B} + R_{3B})/(R_{3B} - R_{1B})]^{1/2} \quad (8)$$

we obtain a pencil-shaped probe with a zero apex diameter and the taper angles θ_A and θ_B . The taper angle θ_B is controlled by varying the right-hand side of (7). Further, to obtain the triple tapered probe, we have to perform step [C]. When the three taper angles are defined as θ_{C1} , θ_{C2} , and θ_A , respectively, the large taper angle θ_{C2} is represented by

$$\sin(\theta_{C2}/2) = R_{1C}/R_{3C}$$

where

$$R_{1C} > R_{3C} \quad (9)$$

The taper angle θ_{C1} is increased from θ_B by increasing the etching time T_C and equal to θ_{C2} at $T_C > \tau_C$ where τ_C is represented by

$$\tau_C = (r_1/R_{3C})[(R_{1C} + R_{3C})/(R_{3C} - R_{1C})]^{1/2} \quad (10)$$

Therefore, we can obtain a triple tapered probe and a pencil-shaped probe with the taper angle $\theta_B = \theta_{C2}$ at $0 < T_C < \tau_C$ and at $T_C > \tau_C$, respectively.

Next, we discuss the designed fiber based on the above geometrical model. To decrease the taper angles of θ_A and θ_B represented by (3) and (7), we have to increase the absolute values $|\Delta n|$ of the relative refractive index differences of sections (3) and (1) (or the doping ratios of fluorine and GeO_2), respectively. To obtain a small taper angle of $\theta_A = 62^\circ$ (which is estimated from (3) and (7)), we design the section (3) with a high value of $|\Delta n| = 0.7\%$. To obtain a cutoff wavelength of around 400 nm, section (1) is tailored with a relative refractive index difference of 1.2% and a core diameter of $[2r_1] = 1.3 \mu\text{m}$. Then, we obtain an estimated value of $\theta_A = 17^\circ$ from (1) and (7). The probe having the small angle can be employed for the high resolution imaging. Further, in the case of making the outer diameter $[2r_3]$ a standard value of $125 \mu\text{m}$, we obtain a critical diameter of $[2r_{2P}] = 46 \mu\text{m}$ from (2) and (6). We make the diameter $[2r_2]$ a value of $27 \mu\text{m}$, which is smaller than the critical diameter.

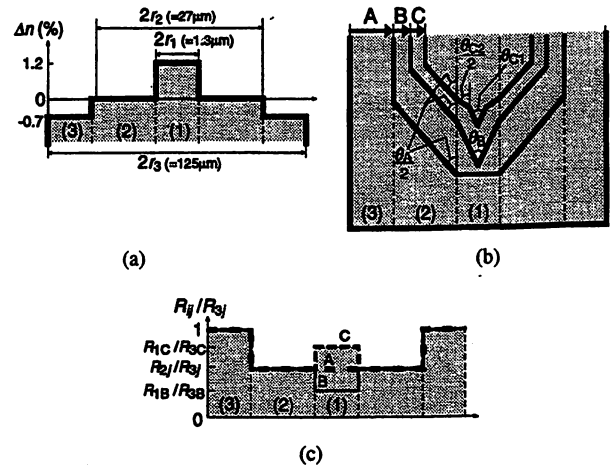


Fig. 1. (a) Cross-sectional profile of relative refractive index difference of a fiber used for fabrication of near-field optical probes. Here, sections (1)–(3) are the GeO_2 doped silica core, the pure SiO_2 clad, and a F doped SiO_2 support, respectively; (b) Etching process for fabricating the probes. The process involves steps [A], [B], and [C]. The pencil-shaped probe having the taper angles of θ_B [defined by (3)] and θ_A [defined by (7)] is obtained by step [A] and [B]. Further, by step [C], the triple tapered probe is fabricated with three taper angles of θ_{C1} and θ_{C2} [defined by (9)] and θ_A ; (c) Etching rate profiles in steps [A]–[C]. Here, R_i ; is defined as the etching rate of section (i) (where $i = 1, 2, 3$) in step [j] (where $j = A, B, C$).

To realize the designed fiber of Fig. 1(a), we produced a preform glass rod by vapor-phase axial deposition [8] and drew the fiber using the preform. To suppress the diffusion of GeO_2 and fluorine, the drawing tension should be as high as possible. However, in the case of drawing the fiber with a high tension of 60 g, we could not reproducibly cleave the fiber for obtaining a flat end of the fiber with a commercial fiber cleaver. We consider that the low reproducibility attributes to the remained stress between sections (2) and (3). To suppress the remained stress, we kept a low tension of less than 30 g during the drawing.

Finally, to demonstrate the tailoring capability of the different type probes, we actually perform the etching process using the fabricated fiber. We prepared thirty samples with flat ends. The fibers are consecutively for $T_A = 40$ min in 1.7:1:1 and for $T_B = 20$ min in 10:1:1. We obtained a high resolution type of the pencil-shaped probe with a small taper angle. Fig. 2(a)–(c) shows scanning electron micrographs (SEM) of the probe, the magnified top region, and the magnified apex region, respectively. The taper angles are $\theta_A = 62^\circ (\pm 0.5^\circ)$ and $\theta_B = 17^\circ (\pm 1^\circ)$. They agree with the estimated values from (3) and (7). The apex diameter is less than 10 nm. For the thirty samples, the probe was fabricated with 100% reproducibility. This reproducibility is much higher than those of previous techniques [5], [9]–[11]. Further, by etching the pencil-shaped probe in 1.7:1:5 for $T_C = 2$ min, we obtained a triple tapered probe having high-resolution capability and high transmission efficiency. Fig. 2(d) is a SEM image of top region of the probe. The probe has three taper angles of $\theta_{C1} = 50^\circ$, $\theta_{C2} = 85^\circ$, and $\theta_A = 62^\circ$, and an apex diameter less than 10 nm. The two base diameters of tapers with $\theta_{C1} = 50^\circ$ and $\theta_{C2} = 85^\circ$ are about 250 and $1.28 \mu\text{m}$, respectively. At $T_C = 2.75$ min, we obtained another type of the pencil-shaped probe with a large obtuse angle near the apex region. It

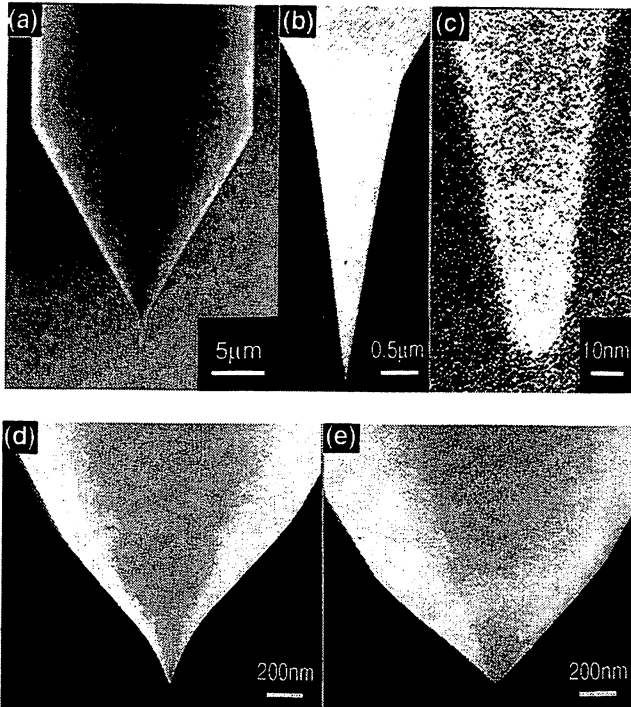


Fig. 2. Scanning electron micrographs (SEM) of (a) a pencil-shaped fiber probe with a small taper angle near the apex, (b) the magnified top region, and (c) magnified apex region. The taper angles are $\theta_B = 17^\circ$ and $\theta_A = 62^\circ$. The apex diameter is less than 10 nm; (d) SEM image of the triple tapered probe. The taper angles are $\theta_{C1} = 50^\circ$, $\theta_{C2} = 85^\circ$, and $\theta_A = 62^\circ$; (e) SEM image of a pencil-shaped probe with a large obtuse taper angle. The taper angles are $\theta_B = 85^\circ$ and $\theta_A = 62^\circ$.

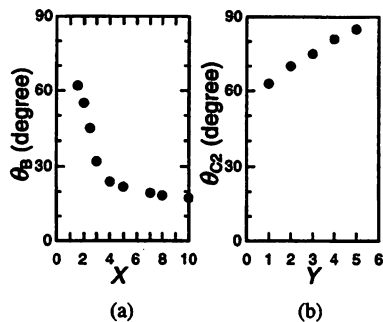


Fig. 3. Dependencies of the taper angles of (a) θ_B and (b) θ_{C2} on X and Y in the etching process. Here, buffered hydrogen fluoride solutions mixed with volume ratios of NH_4F solution (40 wt.%): HF acid (50 wt.%): $\text{H}_2\text{O} = 1.7:1:1$, $X:1:1$ and $1.7:1:Y$ are used for steps [A]–[C] respectively.

becomes just suitable for fabricating a metallized probe having high transmission efficiency. The obtained result is shown in Fig. 2(e). Here $\theta_A = 62^\circ (\pm 0.5^\circ)$ and $\theta_B = 85^\circ (\pm 1^\circ)$. The same probe can also be obtained by etching the fiber with a flat end for $T_A = 40$ min in $1.7:1:1$ and in $1.7:1:5$ for $T_B = 2.75$ min. Fig. 3(a) and (b) are dependencies of θ_B and θ_{C2} on the concentrations of BHF. Here, $1.7:1:1$, $X:1:1$, and $1.7:1:Y$ are used in steps [A]–[C]. The taper angles θ_B and θ_{C2} are controlled by varying X and Y , respectively. However, at around $X = 1.7$ (where $R_{1B}/R_{2B} \approx 1$), the apex diameter increased up to several tens of nanometer. We consider that the increase attributes to the geometrical eccentricity between sections (1) and (2). Actually, the fabricated fiber has an

eccentricity radius of about 200 nm. To produce the probe having $\theta_B = 62^\circ$ and apex diameter less than 10 nm, we apply the hollowing-sharpening technique developed in [7] to this fiber. We obtained a probe having $\theta_B = 62^\circ$ apex diameter less than 10 nm with almost 100% reproducibility by the technique consisting of hollowing in $1.2:1:1$ and sharpening in $3:1:1$. The taper angle θ_B is represented by

$$\sin(\theta_B/2) = R_{1B}/R_{2B} \quad (11)$$

and is independent of the geometric structure of section (3). So, the apex diameter is not affected by the eccentricity radius of the fiber. The three types of probe are coated with aluminum or gold with a thickness of a few hundreds nanometer by a vacuum evaporation unit and employed for the near-field optical imaging and spectroscopy. For details on metallizing technique, refer to [5]. For applications of the probe, we will discuss elsewhere.

In conclusion, we proposed and developed a novel fiber consisting of three sections to fabricate application oriented near-field optical probes. We designed the fiber based on a geometrical model and fabricated it by the vapor-phase axial deposition method. Using the fiber, we fabricated three types of probe with apex diameters of less than 10 nm for high resolution imaging and local spectroscopy with almost 100% reproducibility.

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Solution to the bistability problem in shear force distance regulation encountered in scanning force and near-field optical microscopes

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The bistability problem, common to scanning microscopes employing lateral dithering of the probe for image formation (i.e., shear force microscope) or probe-sample distance control (i.e., near-field optical microscope) is shown to stem from the two nearly degenerate vibration degrees of freedom possessed by a laterally dithered fiber. Controlling the fiber vibration direction by means of a four-sectioned piezo was found to be a simple and effective solution of the problem. An image of a microtubule is presented to demonstrate the improved imaging ability. © 1997 American Institute of Physics. [S0003-6951(97)03843-6]

The shear force and near-field optical microscopes (NOMs) are members of the family of scanning probe microscopes. The former is a type of noncontact scanning force microscope (SFM)¹ [the atomic force microscope (AFM) is a type of contact SFM] distinguished by its use of shear force to map the topographic surface of a sample. It is especially desirable for biological samples, because it exerts far less force on the sample than the competing AFM techniques. The latter exploits the spatial localization² of the optical near-field in order to achieve subwavelength resolution without harming a sample. In many NOM systems shear force control is used to keep the nanometric sized probe close to the sample surface.

This letter will discuss the origin of the problem of bistability in the shear force feedback signal, present a simple solution and its incorporation in a mechanically and thermally stable system, and illustrate the improved resolution with a topographic image³ of a soft (i.e., low mechanical compliance) biological sample (microtubule).

Shear force control of probe-sample separation is implemented as follows: A sharpened fiber tip is glued on a piezoelectric transducer (PZT). For shear force purposes, it has one fixed and one free end and fiber is vibrated laterally by applying a dithering voltage to the PZT at a frequency close to a fiber eigenfrequency [Fig. 1(a)]. (A typical 1.5-mm-long, $\phi 40 \mu\text{m}$ glass fiber has its first harmonic resonance at 20 kHz with a quality factor of 200). When the fiber comes within $\sim 10 \text{ nm}$ [c.f. Fig. 1(b)] of a surface, the tip enters the range of attractive Van der Waals and/or capillary forces, F_w ,¹ which are a function of the probe-sample separation. The component of this force perpendicular to the fiber axis $F_r \cong -|F_w|\varphi e_\varphi$ acts to oppose the motion of the fiber, causing a shift of the fiber's resonance to higher frequency and an accompanying reduction of the amplitude of the tip vibration. This tip vibration can be detected by a variety of methods,⁴⁻⁸ among which the stray light detection method⁹⁻¹¹ has proved to be simple, robust, and sensitive. As described in Fig. 1(a), a laser beam is focused onto the fiber tip, and the diffracted light detected by a photodiode. This signal, containing information on the tip vibration frequency

and amplitude, is heterodyned with the dithering frequency applied to the fiber. At a great distance from the surface the fiber's vibration frequency is equal to the applied dithering frequency; close to the surface the change in the fiber's resonance frequency results in a sharp drop in lock-in amplifier output. A proportional integration controller compares the lock-in output with a preset value and feeds back the difference to a Z-positioning piezo, thus keeping the tip at a predetermined height above the sample. This works provided

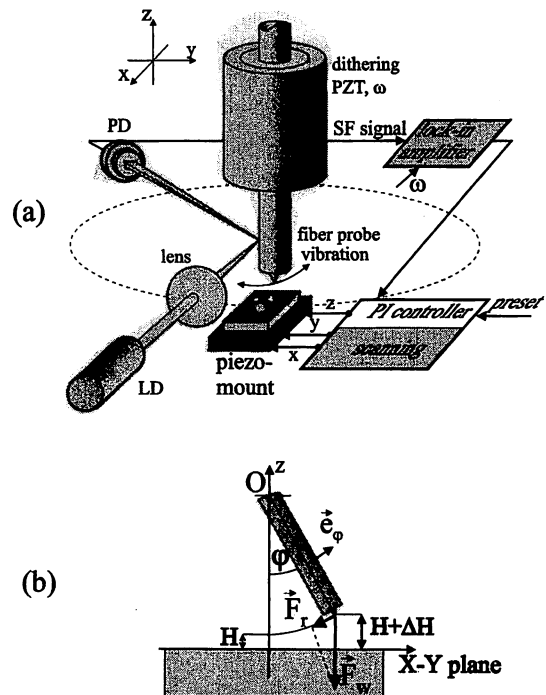


FIG. 1. Explanation of the shear force probe-sample separation regulation as employed in a shear force or near-field optical microscope. (a) LD represents a laser diode ($\lambda = 1.3 \mu\text{m}$ wavelength in our case) and PD a photodiode. The fiber probe is dithered at its near resonant frequency, ω , by means of a voltage applied to the PZT. (b) Force diagram of a fiber coupled to the surface. The XY plane coincides with the sample plane, Z is the fiber vibration axis in the direction perpendicular to the sample surface, e_φ is angular unit vector defined in the plane where the fiber vibration takes place, φ is total displacement angle, F_w is Van der Waals and capillary attractive forces, F_r is restoring force, H is altitude of the fiber tip, $\delta H = o(\varphi^2)$, and therefore is neglected.

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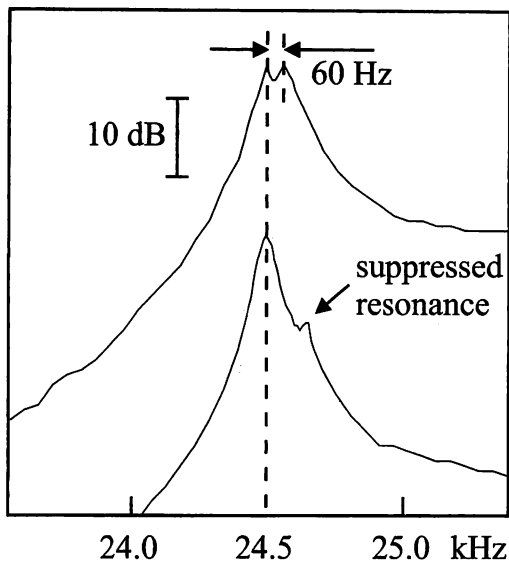


FIG. 2. Frequency spectrum of the fiber under lateral dithering. The two resonance peaks are separated by ~ 60 Hz. The network analyzer resolution bandwidth = 10 Hz. In the upper trace the amplitude of the two resonant peaks are equal, while in lower trace the high frequency peak has been suppressed by greater than 10 dB using the scheme illustrated in Fig. 3.

the dependence of the sample-probe separation on the vibration amplitude is monotonic.⁴

Unfortunately, the dependence of the sample-probe separation on the vibration amplitude is not necessarily monotonic, due to the two degrees of freedom possessed by a laterally vibrated fiber. Let us consider the vibrating system in a coordinate system [defined in Fig. 1(b)] in which the origin, O , is placed at the fiber clamping point. The motion of the fiber's tip thus takes place equidistantly with respect to the origin, and can be analytically described by a two-dimensional vector $\varphi = \varphi_e \mathbf{e}_\varphi = \varphi_x \mathbf{e}_x + \varphi_y \mathbf{e}_y$, with $\mathbf{e}_x, \mathbf{e}_y$ being the elementary orthogonal angle unit vectors in the XOZ and YOZ planes, respectively; φ_x, φ_y are the fiber deflection angle projections onto the XOZ (YOZ) plane. The fiber tip motion is governed by a set of differential equations:

$$\begin{cases} \ddot{\varphi}_x + 2\gamma\dot{\varphi}_x + (\omega_{0x} + \Delta\omega_w)^2 \varphi_x = f_x \cos \omega t \\ \ddot{\varphi}_y + 2\gamma\dot{\varphi}_y + (\omega_{0y} + \Delta\omega_w)^2 \varphi_y = f_y \cos \omega t \end{cases} \quad (1)$$

where γ is a damping constant of the fiber tip; $\omega_{0x}(\omega_{0y})$ is eigenfrequency of the fiber tip along the $x(y)$ axis; $\Delta\omega_w \equiv \Delta\omega_w(H)$ is the separation dependent frequency shift in the fiber tip's eigenfrequency due to coupling the fiber to the surface through Van der Waals and/or capillary forces; ω is the applied dithering angular frequency (near the fiber's resonant frequency); and $f_x(f_y)$ is the vibration amplitude along the $x(y)$ axis of the fiber dithering. This equation is only of a representative order of magnitude due to the complicated structure of forces involved in the probe-sample interaction. Ideally these two lateral vibrations are degenerate ($\omega_{0x} = \omega_{0y}$). Unfortunately, due to uncontrolled factors, such as oblique fiber attachment to the dithering piezo and/or slight asymmetries in the fiber probe itself, this degeneracy is removed. Figure 2 (upper trace) shows a double resonance structure in which the two peaks are separated by 60 Hz. To illustrate the serious consequences of this doublet structure for shear force control, let us consider a fiber being dithered

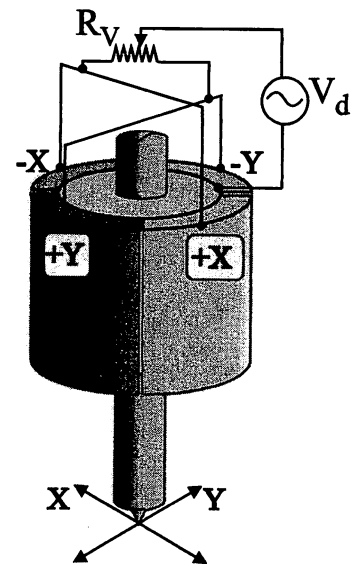


FIG. 3. Schematic diagram of the fiber lateral vibration direction regulation for the suppression of one fiber resonance. The fiber is attached to a four-sectioned piezo tube to which a dithering voltage V_d is applied. $+X$ denotes the PZT segment which shrinks upon applying positive voltage between outer and inner common electrode, while $-X$ denotes the expanding segment. Y is similarly denoted.

at the higher frequency resonance. When it enters the attractive force range, along with a fall in vibration amplitude, the fiber's resonance frequency shifts in the positive direction as it is no longer a "free end" [c.f. Eq. (1)], and hence the dithering frequency ω applied to the fiber is no longer at a fiber's resonance. By the time the amplitude has dropped to half of its original magnitude, due to the surface-probe interaction, the fiber's resonance frequency has shifted by $\omega/2Q = +2\pi \times 60$ rad/s as a simple estimation based on Eq. (1) shows. The applied dither is now resonant to the lower frequency resonance. This results in severe oscillations in the shear force servo system. As these two peaks often lie very close together, this doublet structure may not be recognized when choosing a frequency to dither the fiber (due to inadequate resolution of the spectrum analyzer being used to find the peaks), only showing itself in the form of unexplained system instabilities. Among a variety of empirical methods to obtain a stable shear force signal, we are aware of the following: trying successive harmonics of the fiber resonance frequency until one giving a stable approach curve is found; choosing an off-resonance frequency on which to vibrate the fiber; or when two peaks were seen, operating on the lower frequency peak.¹² The first two techniques are quite time consuming and although the third technique, dithering the fiber at the lower frequency peak, is to be preferred over vibrating at the higher frequency peak it cannot halt servo loop oscillations caused by external perturbations or/and sample topography.

We have succeeded in removing this doublet structure simply and effectively by using a four-sectioned dithering piezo (shown schematically in Fig. 3). The dithering voltage is distributed by means of a variable resistor (R_v). By varying the value of R_v from 0 to its maximum value one can gradually switch the fiber vibration from along the y axis to

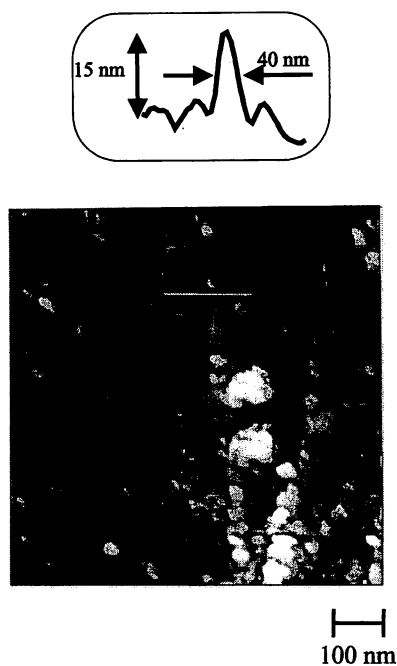


FIG. 4. Shear force image of protein stabilized microtubules. Three filaments spread radially from the right bottom corner of the image. The bright round structures also visible in this image are protein aggregates. The top inset shows profile of a microtubule taken along the line marked in the main figure.

along the x axis. This allows either of the two fiber resonances to be suppressed or enhanced. Figure 2 demonstrates the suppression of an undesirable high frequency peak by 10 dB by adjusting the variable resistor. This allows stable operation of the shear force servo loop in the middle of the force curve (where it is the steepest and thus most sensitive).

This improved shear force detection system has been incorporated into a highly compact and stable combination shear force, collection-mode NOM. The kinematic part of the setup consists of two coaxial piezo tubes. The outer tube supports a wagon on which the prism and sample are mounted and is used for both lateral scanning and fine vertical positioning. In addition, it operates as a motor for the vertical piezoelectric inertial slider¹³ used for coarse vertical positioning. During scanning, the servo loop Z voltage as well as the X - Y scanning voltage are applied to the outer PZT. The inner tube holds and dithers the gold coated sharpened optical fiber. The topographic shear force image is created by recording the shear force feedback signal, and the optical NOM image by recording the intensity of light trans-

mitted through the fiber as it is scanned laterally across the surface of the sample. All components were mounted on an invar plate, placed on a damping stage.

To confirm the improved imaging resolution obtained by eliminating this bistability, Fig. 4 presents a topographic image of a soft biological specimen (microtubule with associated proteins) chosen for its well identified nanometric size (25–30 nm in diameter and microns in length) and importance biologically. The observed width (40 nm) and height (15 nm) are in agreement with previous observations that dried and fixed microtubules take a squeezed tubular, rather than circular tubular shape,¹⁴ and agree with NOM data obtained using this instrument.³ To the best of our knowledge, this image exhibits the best spatial resolution of a microtubule obtained using a laterally dithered scanning force microscope.¹⁵

In summary, oscillations of the shear force control servo system can be caused by the nonmonotonic dependence of the tip lateral dithering amplitude on the probe-sample separation. This bistability, at least partly due to the existence of two near degenerate resonances of the fiber tip, can be removed simply by redistributing the voltage between terminals in a four-sectioned dithering PZT so as to match the applied voltage to one of the nearly degenerate modes.

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Fabrication of a pencil-shaped fiber probe with a nanometric protrusion from a metal film for near-field optical microscopy

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Abstract: We propose a method to fabricate a probe with a nanometric protrusion for near-field optical microscopy. The method involves a tapering process based on selective etching of a GeO₂-doped fiber in a buffered hydrogen fluoride solution and a metallizing process by vacuum evaporation and chemical polishing. We fabricated a tapered probe which has a protrusion emerging from a metal film. The protrusion has an apex diameter less than 10nm and a foot diameter less than 20nm. Employing the probe, we succeeded in obtaining a highly resolved image of 20nm gold particles.

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Recently, super high resolution optical imaging has been performed beyond the diffraction limit by near-field optical microscopy [1-4] employing a tapered fiber probe with a nanometric apex and a small taper angle. We previously produced a probe with a taper angle of 20° , an apex diameter less than 10nm, and a nanometric protrusion emerging from a metal film [5] by selective resin coating (SRC) [6]. Employing the probe, we succeeded in obtaining nanometric images [7,8] of biological samples. Such a probe is found to be effective both as a scatterer (and/or a generator) of high spatial frequency evanescent components responsible for high resolution capability and as a suppresser of the low spatial frequency components and the propagating components. However, the SRC method can not be applied to a tapered probe such as a pencil-shaped probe without a flat end portion [6]. In this letter, we propose a method to fabricate a pencil-shaped probe which has a nanometric protrusion emerging from a metal film using a commercial GeO_2 -doped fiber.

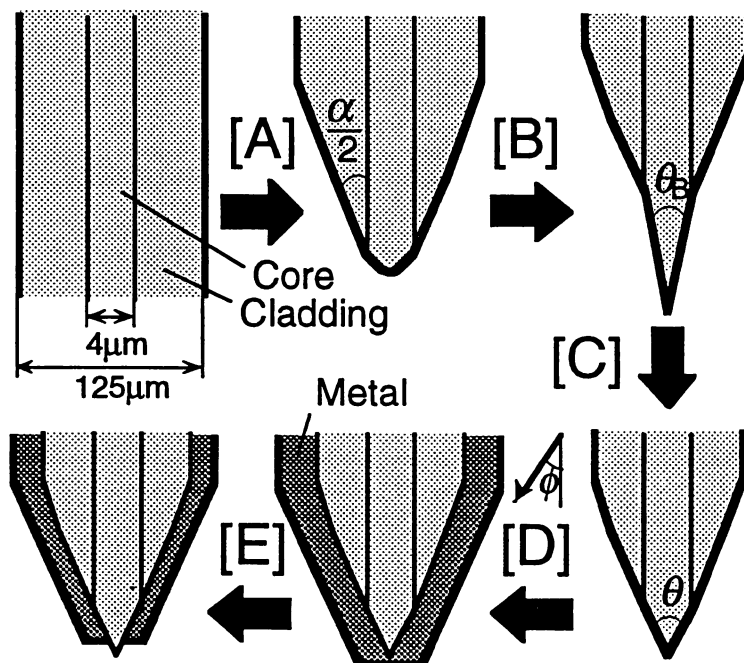


Fig. 1 Fabrication method of a pencil-shaped fiber probe with a protrusion from a metal film. Here, the method involves five steps; [A] tapering the cladding by meniscus etching in hydrofluoric (HF) acid with a surface layer of organic solution, [B] sharpening the core by selective etching in buffered hydrogen fluoride solution (BHF), [C] increasing the taper angle in HF-containing aqueous solution [D], coating a metal film by vacuum evaporation, and [E] removing the metal that covers the apex region by chemical polishing. α is the taper angle of the cladding. θ and θ_b are the taper angles of the core. In vacuum evaporation, the fiber is tilted with the angle ϕ and rotated.

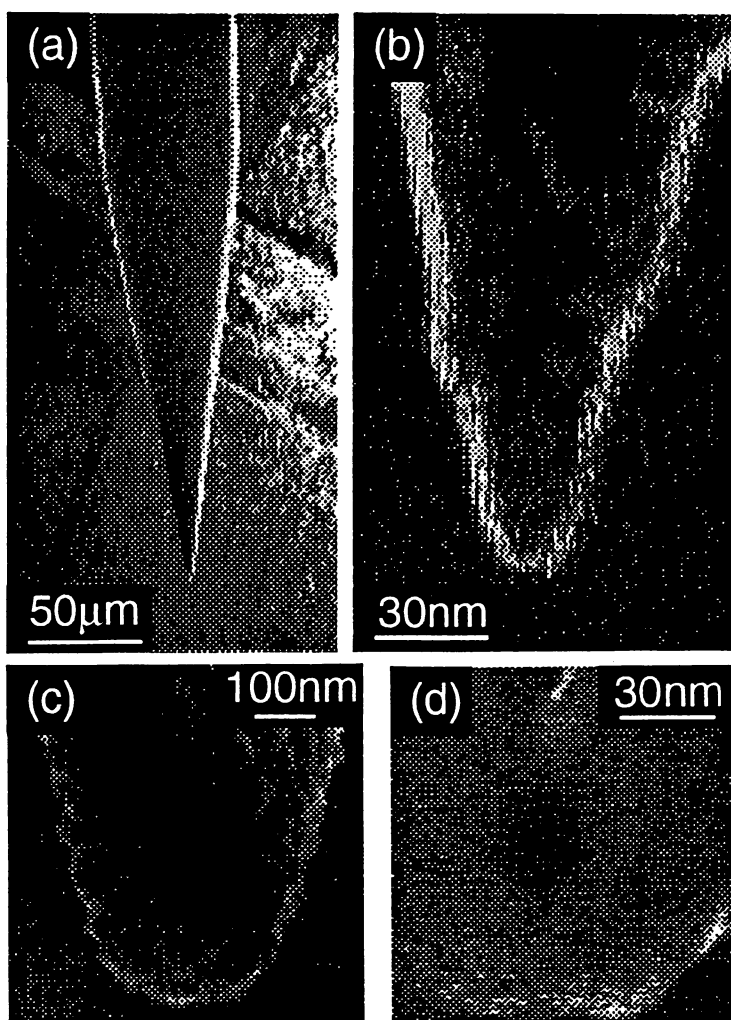


Fig. 2 (a) Scanning electron micrographs (SEM) of (a) pencil-shaped fiber with a taper angle of 20° , (b) the magnified top region. Here, the fiber is coated with gold having a thickness of 3nm. The apex diameter is less than 10nm; SEM images of (c) the top region and (d) the apex region of the metallized probe which has a protrusion from a metal film. Here, the metal is gold. The radial thickness of the gold film is 150nm. The foot diameter of the protrusion as shown by the dark portion is less than 20nm. Because the length of the protrusion is less than 10nm, nearing the limit of SEM, it is difficult to get a visualization in (d).

Figure 1 schematically explains the method involving five steps; [A] tapering the cladding, [B] sharpening the core, [C] increasing the taper angle, [D] coating a metal film, and [E] removal of metal covering the apex region. Here, the method is divided into the tapering process of steps [A]-[C] and the metallizing process of steps [D] and [E]. The symbols α , θ , and θ_b are defined as the taper angles of the pencil-shaped fiber. The notations of Oil/HF, BHF, HF are HF acid with a surface layer of an organic solution, a buffered hydrogen fluoride solution with a volume ratio of 40wt.-%- NH_4F aqueous solution: 50wt.-%-HF acid: $\text{H}_2\text{O} = X:1:Y$, and an aqueous solution containing HF. Throughout this letter, BHF is denoted as $X:1:Y$. Figures 2(a) and 2(b) show scanning electron micrographs (SEM) of the pencil-shaped fiber obtained with taper angles

of $\theta=10^\circ$ and $\alpha=20^\circ$ by steps [A]-[C]. The taper angle and the apex diameter are 20° and less than 10nm, respectively. Figures 2(c) and 2(d) are, respectively, SEM images of the magnified top region of a fabricated probe with a protrusion and of the apex region. The dark portion in Fig. 2(d) shows a fabricated probe with a protrusion. The radial thickness of gold film is about 150nm. The foot diameter of silica tip protruding from the metal film is less than 20nm. In the following, we describe the method for fabricating the probe.

In step [A], a dispersion-shifted fiber (Sumitomo Electric Industries, DS1) having the germanosilicate (GeO_2) doped core was immersed for 50min in a mixed solution of a volume ratio of 50wt. %-HF acid: 95wt. %-sulfuric acid= 4:1. Here, the fiber has a diameter of 125 μm , a core diameter of 4 μm , and a GeO_2 doping ratio of 9mol%. To suppress the vapor phase evaporation of $\text{HF-H}_2\text{O}$, the surface of the solution was sealed with a layer of dimethylsilicone oil as was done in meniscus etching of our previous method [9]. Sulfuric acid is added to maintain the symmetry of the conical taper. The fiber has a taper angle of $\alpha=20^\circ$ by this meniscus etching. In step [B], we selectively etched the fiber for 120min in 10:1:1 of BHF. We obtained a pencil-shaped fiber with taper angles of $\theta_b=10^\circ$ and $\alpha=20^\circ$. Step [C] was performed by immersing the fiber for 90 seconds in HF-containing solution mixed with a volume ratio of 50wt. %-HF acid: H_2O = 1:3. We obtained the pencil-shaped fiber with taper angles of $\theta=20^\circ$ and $\alpha=20^\circ$. In step [D], the fiber was coated with chromium and gold films of thickness 3nm and 200nm, respectively, by a vacuum evaporation unit. Here, the pressure was maintained at 10^{-6} - 10^{-7} Torr. We used electron-beam evaporation for coating with gold. The fiber is tilted with an angle $\phi=50^\circ$ and rotated. In step [E], the metallized fiber was immersed for 15min in a KI-I_2 aqueous solution mixed with a weight ratio of $\text{KI:I}_2:\text{H}_2\text{O}=20:1:80000$ at 25°C ($\pm 0.5^\circ\text{C}$).

Next, we discuss the reproducibility of the etching process. In step [B], by denoting the etching rate of the core and cladding as R_1 and R_2 , respectively, the taper angle θ_b is represented by [10: Eq. (A1)]

$$\sin(\theta_b/2)=(R_1/R_2)\sin(\alpha/2).$$

The right side is controlled by varying the GeO_2 doping ratio and the concentrations of BHF (or X and Y). The pencil-shaped probe with taper angles of $\theta_b=20^\circ$ and $\alpha=20^\circ$ is fabricated by the etching with a ratio of 1.7:1:1 where $R_1/R_2=1$. However, it is difficult to produce a probe with an apex diameter less than 10nm with high reproducibility. We consider that this difficulty is attributed to geometric eccentricity of the taper formed by step [A]. The taper has an eccentric radius of around 1 μm between the core and the apex of the taper. To improve the reproducibility, we add steps [C]. As a result, we succeeded in obtaining the probe with a taper angle of $\theta=20^\circ$ ($\pm 3^\circ$) and an apex diameter of less than 10nm with 80% reproducibility or more.

Futhermore, we discuss the metallizing process using vacuum evaporation. We performed step [D] with various angle ϕ in a region of 55° - 90° and observed the probe by SEM. In the case of a thin coating of 120nm and a tilted angle of $\phi=50^\circ$, we evaluated a metal thickness of 30nm covering the fiber apex from the SEM photographs before and after the chemical polishing. For fabricating a probe with a protrusion, step [E] or another removal technique is required. At $\phi=50^\circ$, we obtained a protrusion-type probe with a foot diameter of 20nm ($\pm 15\text{nm}$) with about 75% reproducibility or more using 12 fiber samples with apex diameters less than 10nm.

Finally, to demonstrate the imaging capability of the fabricated probe, we observed gold particles of size 20nm fixed on a glass substrate by a near-field optical microscope (NOM). The NOM is operated under illumination mode with the probe illuminating the sample substrate by an argon ion laser with a 488nm wavelength. Details of the experimental system can be found in Ref. 11. The size of the gold particles are well calibrated by TEM to be 20nm ($\pm 1\text{nm}$) and they are just suitable for the high resolution imaging capability of the probe. The obtained image is shown in

Fig. 3. Here, the scan area is $200\text{nm}\times 200\text{nm}$. The dark portions (as marked by the arrow) correspond to single gold particles. Based on power spectral analysis of the image, the estimated size of the single particles is found to be around 22nm . For details of the analysis, refer to Ref. 12. This closely agrees with the actual size of the particle, thus demonstrating the effective functioning is high resolution imaging of the proposed NOM probe.

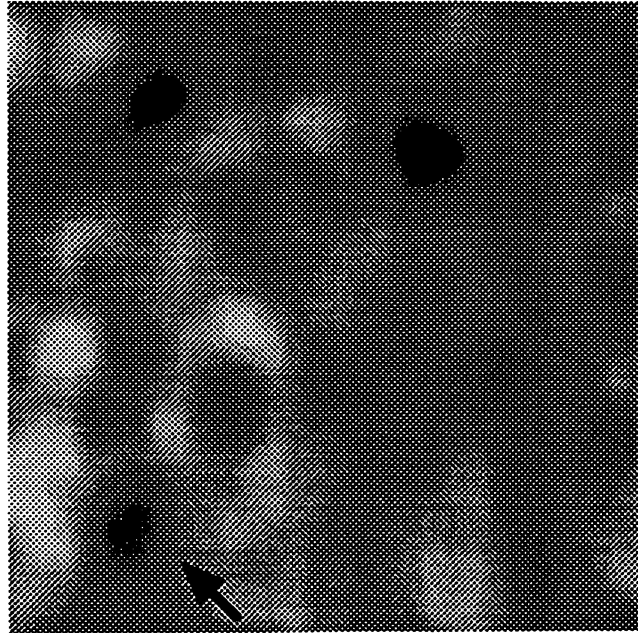


Fig. 3 Near-field optical image of 20nm -gold particles illuminated by a 488nm argon ion laser. Here the scan area is $200\text{nm}\times 200\text{nm}$. The dark portions as indicated by the arrow show single gold particles.

In conclusion, we proposed a method to fabricate a pencil-shaped fiber with a nanometric tip protruding from metal film. We obtained a probe which has a silica protrusion with an apex diameter less than 10nm and a foot diameter less than 20nm . Employing the probe, we obtained a high-resolution image of 20nm -sized gold particles.

Highly efficient excitation of optical near-field on an apertured fiber probe with an asymmetric structure

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We propose and demonstrate a novel method to enhance the near-field optical intensity on the apertured probe with its foot removed asymmetrically by using a focused ion beam. The spatial distribution of the near-field optical intensity on the asymmetric probe was observed by scanning another sharpened symmetric probe over the aperture. The observed spatial distribution profile was in good agreement with a numerical result corresponding to the HE_{11} mode. Furthermore, compared with the symmetric apertured probe, a 10 times enhancement of the near-field optical intensity on the asymmetric apertured probe was observed. © 1997 American Institute of Physics.
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Recent development of near-field optical microscope has made it possible to yield optical images with nanometric spatial resolution by scanning a metal coated probe with a subwavelength aperture.^{1,2} For improvement in the spatial resolved spectroscopy, the most serious problem of using the apertured probe is its low efficiency, i.e., throughput ratio (=scattered light power from the aperture/input light power from the another end of the fiber) of the near-field excitation on the probe. The throughput ratio rapidly decreases by decreasing the aperture diameter (D).^{3,4} For example, it is less than 10^{-5} with $D=100$ nm.⁵ Thus the scanning speed of the probe is limited quite low. One typical example suffering from the low efficiency of the near-field excitation is optical near-field recording/readout. Even though an ultrahigh recording density of over 10 Gb/in.² has been demonstrated,⁶⁻⁸ the recording/readout speed is much lower than that of conventional optical memory.

To increase the near-field optical intensity on the probe, one must consider the method to decrease the propagation loss in the probe. For example, Saiki *et al.*⁵ have demonstrated an over 10 times decrease of the propagation loss in the probe using a two-step tapered probe. For further enhancement of the near-field optical intensity, it is found by the present analysis that the efficient excitation of the HE_{11} mode is also promising method. Since the near-field optical intensity on the probe depends on the excited mode in the probe, we have approximated the probe as a dielectric core [refractive index (n)=1.53] surrounded by an infinitely thick gold ($n=0.17+i5.2$) cladding for a mode analysis at the wavelength of 830 nm.⁹ As a result, we found that the HE_{11} mode can propagate even at core diameter as small as 30 nm. The HE_{11} and EH_{11} modes represented in Ref. 9 converge on surface plasmon mode and the lowest mode of the optical fiber, respectively, by increasing core diameter. Therefore it can be considered that the HE_{11} mode is hardly excited in the conventional gradual tapered probe from the lowest mode of the optical fiber.

We propose and demonstrate here a novel method to

excite the HE_{11} mode efficiently by breaking the shape of the gradual tapered probe to be an asymmetric probe in order to introduce the capability of converting the propagation mode in the optical fiber to the HE_{11} mode in the metal-coated core. To confirm this mode conversion capability, the spatial distribution of the near-field optical intensity on the asymmetric apertured probe was observed by scanning another sharpened symmetric probe over the aperture to scatter and detect the optical near-field. Finally, we present experimental results demonstrating that the near-field optical intensity of the asymmetric apertured probe increased drastically.

Figure 1 shows a novel experimental setup to measure the spatial distribution of the near-field optical intensity on the apertured probe. Linearly polarized light from a laser diode (830 nm) was coupled into the single-mode fiber. The direction of incident light polarization was changed by a half wave plate to examine the dependence of the image on the direction of incident light polarization. The optical near-field generated on the apertured probe (probe A) was observed by scanning another sharpened probe (probe B) and detecting the scattered light with a photomultiplier, while the separation between the two probes was maintained within several nanometers with shear-force technique. Note that probe B was symmetric.

We compared the near-field optical intensity of symmetric and asymmetric apertured probes (probe A) with $D \sim 500$ nm (cut-off diameter of the EH_{11} mode ~ 450 nm) and

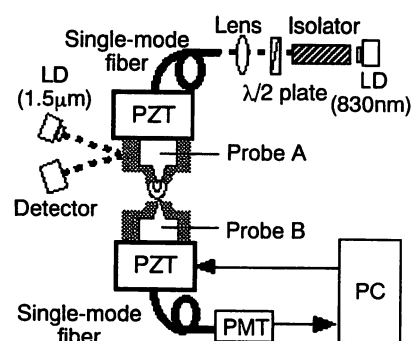


FIG. 1. Experimental setup: Probe A: apertured probe, probe B: sharpened probe for scattering the optical near-field on probe A.

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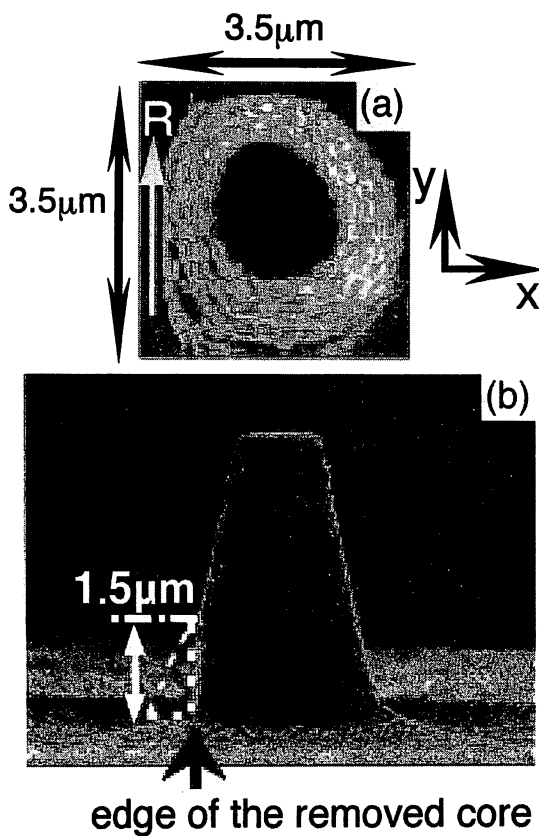


FIG. 2. SEM images of an asymmetric apertured probe: (a) top view, (b) side view. x and y axes are the directions perpendicular and parallel to removed direction R [along the black arrow in (a)].

$D \sim 100$ nm ($\lambda/8$). Although both EH_{11} and HE_{11} modes can propagate in the probe with $D = 500$ nm, note that the EH_{11} mode cannot exist in the probe with $D = 100$ nm. Figure 2 shows a scanning electron microscopic image of a fabricated asymmetric apertured probe with $D = 500$ nm. The foot of the core was removed [represented by a dashed triangle in Fig. 2(b)] to make the core asymmetric, where the height of the removed part was $1.5 \mu\text{m}$. This probe was fabricated by the following four steps:

- (i) By selective chemical etching technique,¹⁰ the GeO_2 doped core of fiber was tapered, where the cone angle was 20° .
- (ii) The foot of the core was removed by using a focused ion beam (FIB).
- (iii) The core was coated with 500-nm-thick gold film.
- (iv) The top of the core was removed to form an aperture with FIB.

Note that apertured probes with D as small as 30 nm were realized by using the same fabrication procedure.

First, we checked whether the asymmetric structure of the probe led to efficient excitation of the HE_{11} mode. We observed the spatial distribution of the near-field optical intensity on the asymmetric apertured probe with $D = 500$ nm. Figures 3(a) and 3(b) show the observed distributions, where the direction of incident light polarization was orthogonal to each other. A protruded probe¹¹ with an apex diameter of 10 nm and a foot diameter of 60 nm was used as probe B. Figures 3(c) and 3(d) show calculated distributions corre-

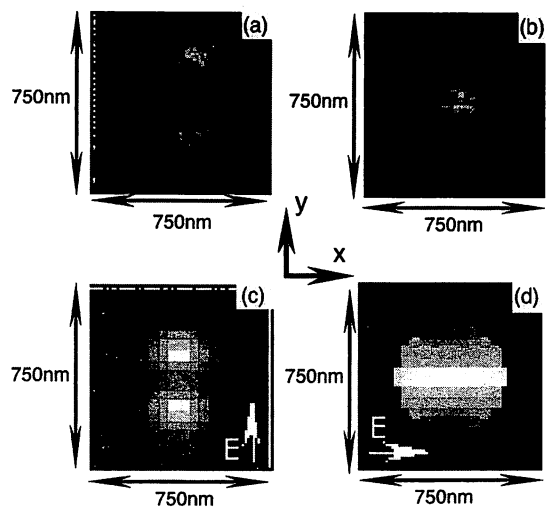


FIG. 3. Spatial distribution of the near-field optical intensity ($\lambda = 830$ nm) on an asymmetric apertured probe with $D = 500$ nm. (a) EH_{11} mode: measured, (b) HE_{11} mode: measured, (c) EH_{11} mode: calculated, (d) HE_{11} mode: calculated. The incident light for (b) was taken with linear polarization oriented 90° relative to that for (a).

sponding to EH_{11} and HE_{11} modes for $D = 500$ nm, respectively. One can find that Figs. 3(a) and 3(b) are in good agreement with Figs. 3(c) and 3(d), respectively, on the number of peaks. Because the direction of incident light polarization rotates in the fiber, we could not determine experimentally the direction of polarization at the foot of the probe. However it is reasonable to consider that the directions of the polarization of Figs. 3(a) and 3(b), correspond to those indicated in Figs. 3(c) and 3(d), respectively. We also observed the spatial distribution on the symmetric apertured probe [fabricated without step (ii)] with $D = 500$ nm. In this case, only the double-peaked distribution corresponding to the EH_{11} mode was observed, where the direction of the peaks was rotated according to the rotation of that of the incident light polarization. These results indicate that an asymmetric structure helps to excite the HE_{11} mode, where its excitation efficiency depends on the direction of incident light polarization.

Second, we checked whether the near-field optical intensity on the apertured probe is enhanced due to the asymmetric structure. We compared the spatial distributions of both symmetric and asymmetric apertured probes with $D = 100$ nm. To enhance the efficiency of light scattering, we used a sharpened probe coated with 30-nm-thick gold as probe B.¹² Figure 4 shows these observed cross-sectional distributions. Only the single-peaked distribution corresponding to the HE_{11} mode was observed. Curves A and B are for the asymmetric apertured probe, where the direction of incident light polarization was orthogonal to each other. Curve C is for the symmetric apertured probe. One can find that intensity for curve A is 10 times larger than that for curves B and C. It is calculated by the present analysis that the full width at half maximum of the intensity distribution of the HE_{11} mode for $D = 100$ nm is 120 nm, which is comparable to that of curve A (~ 150 nm). These results indicate that effective excitation of the HE_{11} mode due to the probe asymmetry led to the enhancement of the near-field optical intensity.

The efficient excitation of the HE_{11} mode realized here

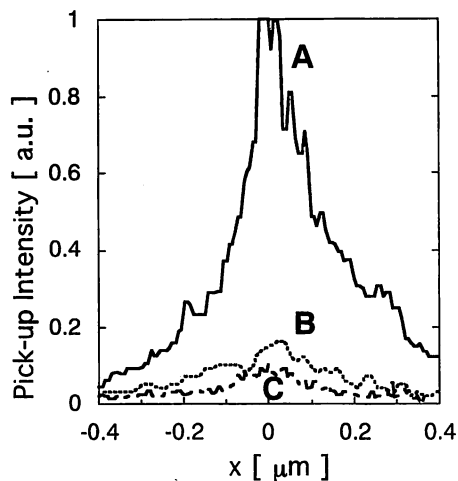


FIG. 4. Cross-sectional spatial distribution of the near-field optical intensity on asymmetric (A and B) and symmetric (C) apertured probes with $D = 100$ nm, at $\lambda = 830$ nm. The incident light for B was taken with linear polarization oriented 90° relative to that for A.

can be attributed to the scattering coupling.¹³ Since an asymmetric apertured probe has a sharp edge at the foot of the probe [shown by a black arrow in Fig. 2(b)], a part of the radiated power can be scattered and converted to the HE_{11} mode. A more detailed mechanism will be presented by investigating the dependence of the efficiency of exciting the HE_{11} mode on the direction of incident light polarization. It is quite reasonable to consider that the HE_{11} mode was excited when the direction of polarization at the foot of the probe was parallel to the x direction in Fig. 2. The oscillations of the electric field perpendicular to the edge induced charges on the edge of the removed core, and these induced charges generate strong electric field and the associated scattering.¹⁴ It is expected that these mechanisms of enhanc-

ing near-field optical intensity due to the asymmetric structure can be applied to the apertured probe with D as small as 30 nm. Detailed results will be reported in a future publication. It should be also noted that further enhancement of the near-field optical intensity can be expected by optimizing the probe structure.

In conclusion, we have demonstrated the efficient excitation of the HE_{11} mode using an apertured probe with an asymmetric structure. The highly efficient excitation of the HE_{11} mode resulted in 10 times the enhancement of the near-field optical intensity of the apertured probe with $D = 100$ nm. By use of such an asymmetric structure it will be easy to realize the ultrahigh speed and high density optical recording/readout.

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Near-field optical study of semiconductor photonic devices

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Abstract

A novel-structured semiconductor photonic device is investigated using a near-field scanning optical microscope. By tailoring the shape of the fiber probe, high transmission and collection efficiencies are successfully achieved. Employing optimized fiber tips, multi-diagnostics of lateral p-n junctions is performed with the AFM operation. Measuring the spatially resolved photoluminescence spectra, we precisely examine the carrier distribution in the transition region of the p-n junctions. Electroluminescence imaging reveals the width and the position of the active region. The slant angle of the p-n interface is determined by applying the multiwavelength near-field photocurrent measurement. We also clarify the mechanism of the mode conversion due to the interaction between the evanescent light on a small aperture and optically dense semiconductors. © 1997 Elsevier Science S.A.

Keywords: Near-field scanning optical microscope; Optical fiber probe; Evanescent light; Near-field spectroscopy; Lateral p-n junction

1. Introduction

Near-field optical techniques [1–3] have made a remarkable contribution to subwavelength resolution imaging and spectroscopy in the diagnostics of semiconductor devices including laser diode [4], photodetector [5–8], and light emitting diode [9]. So far, a near-field scanning optical microscope (NSOM) has been employed in two different manners. Firstly, spatially resolved spectroscopy has been successfully implemented to determine optical structures and responses of photonic devices with quantum structures [10,11]. Secondly, NSOM works as a probe for local excitation and collection to observe individual spectra of inhomogeneously broadened system, such as self-assembled quantum dots [12–14].

For these advanced experiments, we have serious problems which are special to the optical investigation of semiconductors. One of the problems is concerned with the conversion of evanescent modes into propagating ones due to the interaction with the samples. Since the semiconductor has a large refractive index, as compared to the glass and air, the evanescent modes are easily coupled into the propagating modes in close

proximity of the semiconductor surface. In order to retain the evanescent field in the optical dense semiconductors, the aperture diameter should be much smaller than the wavelength of the excitation light [7], which causes low excitation efficiency. The diffusion of photoexcited carriers brings about another problem. In the detection of photoluminescence images and spectra, an illumination-collection hybrid mode [9] is an essentially important technique in order to avoid the deterioration of the resolution, limited by the carrier diffusion length, which is of the order of 1 μm . In this operation, however, the detected signal is very small due to the low excitation and collection efficiencies. To overcome these difficulties, it is necessary to improve the efficiency of transmission to the aperture by tailoring the shape of fiber tip.

In Section 2, the transmission efficiency of the fiber probe is evaluated as a function of the aperture diameter. We determine the most influential factor in the transmission property of the metal-cladding tapered waveguide. A long tip with high efficiency is developed so as to be suitable during actual scanning operation [15]. In Section 3, multi-diagnostics of lateral p-n junctions is performed using a highly efficient fiber probe. By measuring the spatially resolved photoluminescence spectra, we precisely determine the position and the width of the transition region of p-n junctions [9]. The

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illumination-collection hybrid mode is also employed to map the two-dimensional emission efficiency [9]. Through the electroluminescence measurement with collection mode, we examine the active region of the junctions by comparing with the results of photoluminescence spectroscopy. Near-field photocurrent measurement with propagating modes into the sample is applied in order to probe the internal structure. By systematically varying the penetration depth over a wide range, the slant angle of the p-n interface is quantitatively determined [8]. In Section 4, the properties of evanescent modes on a small aperture is characterized through the near-field photocurrent detection. We clarify the aperture-size-dependent feature of mode conversion, lateral resolution, and decay length of evanescent field. Section 5 is devoted to the summary of the present paper.

2. Tailoring high-throughput fiber probe

For the preparation of sharpened fiber probe, we utilize a chemical etching technique with high reproducibility [16,17]. In Fig. 1(a), a schematic of the fiber probe with metal coating is shown introducing our terminology. The metal-coated fiber probe, which is regarded as a metal-cladding tapered waveguide, has complex loss mechanisms. The existence of cutoff diameter, even in the lowest mode, is the most specific feature of the metallic waveguide. With respect to the transmission efficiency, the distance between the aperture and the cutoff diameter is expected to be one of the significant aspects.

To observe the dependence of transmission efficiency on cone angle, two types of fiber tips (tip A and tip B) are prepared. The cone angle θ and the tip length l of

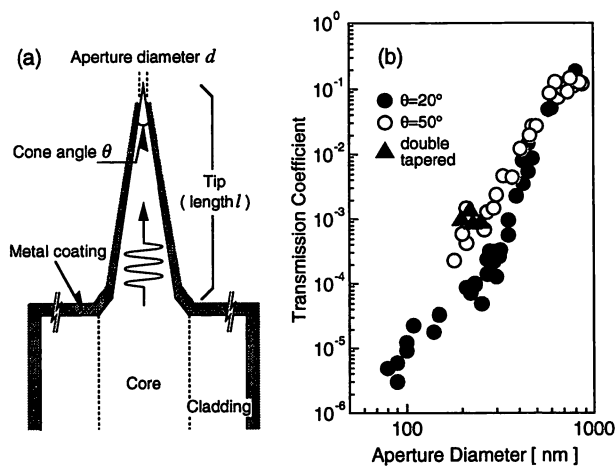


Fig. 1. (a) Schematic of chemical-etched fiber probe with metal coating. (b) Transmission coefficients as a function of the aperture diameter: tip A (●), tip B (○), and double tapered tip (▲).

the tip A are 20° and $6 \mu\text{m}$, respectively, and those of the tip B are 50° and $2.5 \mu\text{m}$. For the estimation of transmission coefficient, light from a He-Ne laser (633 nm) of $130 \mu\text{W}$ is coupled into the fiber probe. The far-field light ejected from the aperture is collected with a 0.4 NA objective lens. The output power is measured with an optical power meter. The transmission coefficient is defined as the ratio of the collected output power to the input power. The geometrical size of the aperture is estimated by using scanning electron microscope (SEM) after the throughput measurement.

In Fig. 1(b), transmission coefficients are plotted as a function of the aperture diameter d for the tip A (●) and the tip B (○). For the diameter d that is larger than the wavelength of He-Ne laser light in glass d_c ($\sim 400 \text{ nm}$), the transmission coefficients of the tip A and the tip B take almost the same values when the aperture diameter is the same. This result shows that the efficiency for delivering light into the region $d > d_c$ is not so strongly dependent on the length and the cone angle of the metallic tapered waveguide, which is $5 \mu\text{m}$ long at most. On the other hand, in the region $d < d_c$, the differences between the two types of probes are remarkable under the influence of metal coating. For the case of aperture diameter being 200 nm , transmission efficiency of the tip A is lowered by 10 times to that of the tip B. The dependence of transmission power on the cone angle is thought to be coming from the evanescent propagation of light into the metallic-cladding waveguide with the diameter less than the cutoff. In order to increase the transmission efficiency, it is reasonable to shorten the distance between the aperture and the cutoff diameter by increasing the cone angle [18]. In theoretical consideration, assuming perfect conductivity of the cladding, the cutoff diameter of the lowest bound mode in a metallic cylindrical waveguide is 250 nm for 633 nm light [19]. The discrepancy between d_c and the theoretical cutoff may be explained by considering the tapered structure and a complex dielectric permeability of metal cladding.

Although highly efficient, a short tip with large cone angle leads to contact between its cladding and the bumpy surface of the sample during actual scanning operation. To avoid this inconvenience, it is necessary to lengthen the tip still maintaining the large transmission coefficient. For this purpose, we have successfully developed a fabrication technique of a double tapered tip (Fig. 2(a)). As shown in the SEM image of Fig. 2(b), the resultant top cone angle is about 90° . To demonstrate the high efficiency of this tip, we measure again the transmission coefficients of some tips with diameters of around 200 nm . The results are plotted in Fig. 1(b) (▲). Higher transmission efficiency by an order than that of the tip A (●) is achieved. The ability of this optimized tip is demonstrated in the following section.

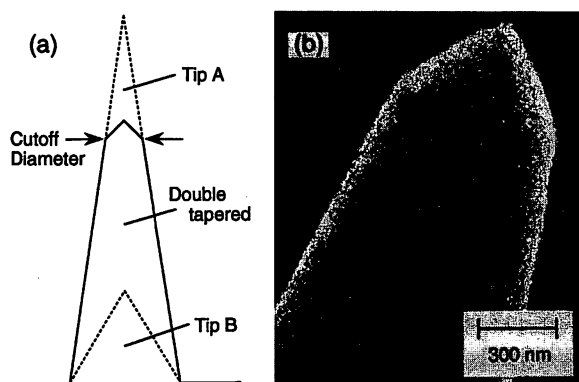


Fig. 2. (a) Schematic structure of double tapered probe compared with tip A and tip B. (b) Scanning electron microscope image of double tapered apex region of the tip.

3. Near-field optical diagnostics of lateral p-n junctions

3.1. Sample and fundamental setup

Fig. 3(a) shows the schematic structure of the lateral p-n junctions [20–22]. This device is expected to be applied to the lateral carrier confinement structures, such as surface-emitting laser diode. The conduction type of GaAs layers with Si dopant depends on growth conditions and the orientation of the substrate. By using this amphoteric nature of Si, both n- and p-type regions can be grown simultaneously on a patterned substrate, and lateral p-n junctions are formed at the boundary of the two regions. The luminescence peak wavelength strongly depends on the conduction type and the carrier concentration of GaAs layers.

Schematics of the experimental configurations are shown in Fig. 4. A gold-coated fiber probe with 200 nm aperture is employed as a tip. In the illumination mode, as an excitation light source, 0.5 mW of He-

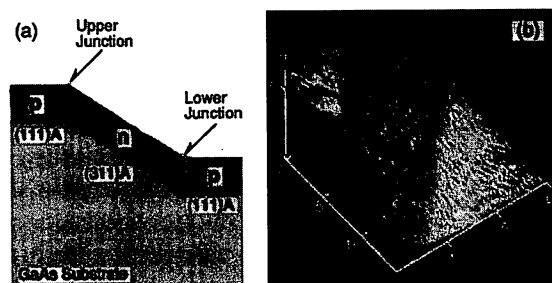


Fig. 3. (a) Cross-sectional profile of lateral p-n junctions grown on a patterned GaAs (111)A substrate. (b) Perspective view of shear-force image in the vicinity of the slope.

Ne laser (632.8 nm) is typically coupled into the fiber probe. For the regulation of tip-sample distance, shear-force feedback technique is employed. To monitor the amplitude of the vibration, we use a 1.55 μm laser diode, whose photon energy is far below the absorption edge of GaAs. By positioning the tip < 10 nm above the sample, the excitation region is restricted to $(100 \text{ nm})^2$ in lateral area and 200 nm in depth, which are determined by the aperture radius and the absorption coefficient of GaAs.

The shear-force image of the sample in the vicinity of the slope is shown in Fig. 3(b). The width and the height of the slope are approximately 10 and 6 μm , respectively. From the top surface to the slope, we see some bumps, which may be produced in the etching process of the substrates.

3.2. Spatially resolved photoluminescence spectroscopy

In order to determine the position and the width of the transition region of p-n junctions, spatially resolved photoluminescence (PL) spectra are measured in the illumination mode of NSOM. The experimental setup is depicted in Fig. 4(a). The PL from the sample is collected on the same side of the sample with a 0.4 NA objective lens, and transported to a 20 cm monochromator with an avalanche photodiode for photon-counting detection.

In Fig. 5, the peak wavelength and total integrated intensity of the PL spectra are plotted along the slope. The peak wavelength at the top surface and at the bottom surface shows the same value of 870 nm. This peak wavelength corresponds to that of flat (111)A surface with the same Si concentration. On the (311)A slope, the PL peak shifts to the higher-energy side. Its value of 855 nm is also in agreement with that of n-type flat (100)A surface. At the lower junction, the PL peak shows graded changes with a transition width of 5 μm . Since the conduction type and carrier concentration vary with tilt angle of the substrate from (111)A surface, the transition width is closely relevant to the structure of the junction. The gradual change of tilt angle at the lower junction, as shown in Fig. 5(a), causes the wide transition width of the carrier concentration. At the upper junction, on the other hand, the top surface and the slope make a clear ridge. This abrupt change results in the narrower transition width of 1 μm .

The total PL intensity in the transition region is much lower than the other regions. This is mainly due to the separation and the drift of photoexcited electrons and holes by the internal electric field in the transition regions. In the lower junction, some anomalous optical structures, plateau in the change of the PL peak and increase of PL intensity, are observed as indicated by arrows in Fig. 5.

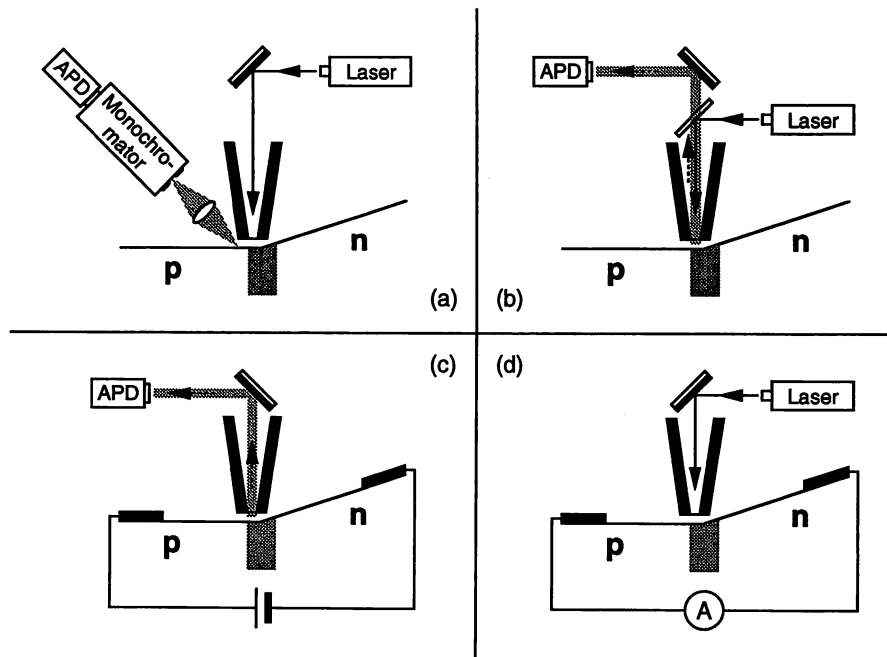


Fig. 4. Block diagrams of experimental configurations for the measurement of (a) photoluminescence spectra, (b) integrated photoluminescence intensity, (c) electroluminescence intensity, and (d) photocurrent spectroscopy: APD, avalanche photodiode.

3.3. Two-dimensional mapping of PL intensity

It is important to investigate local optical properties in the vicinity of the junctions in detail since the distribution of defects and strains, and nonuniformity of dopants in this active region will affect the emission efficiency as a light-emitting device. To measure the two-dimensional distribution of the PL intensity, the illumination-collection hybrid mode was employed (Fig. 4(b)). In this mode, as the probe tip locally collects the emission in addition to being a local excitation source, we can achieve higher resolution (< 200 nm), which does not depend on the diffusion length. The PL intensity image is shown in Fig. 6. In the transition region, as shown previously, the PL intensity decreases a lot, compared to the other regions. Some bright areas, where the PL intensity locally increases, appears in the lower junction. These signals indicate essentially optical properties since no corresponding structure is found in the shear-force image (Fig. 3(b)). A nonuniform distribution of Si dopants and of the resultant internal electric field, or that of defects and strains will affect the local optical responses.

3.4. Collection-mode imaging of electroluminescence

In the operation of lateral p-n junctions as the surface-emitting diode, the quality of the electroluminescence (EL) is the most important feature. Using the collection-mode imaging of EL [4], the properties of the active region of the junction is determined compared to those of transition region investigated in Section 3.3.

Fig. 4(c) depicts the configuration of the collection-mode operation. Injecting current through the electrodes, EL from the active region is collected through the aperture. The line scans of EL signal from the upper and lower junctions are shown in Fig. 7 with the cross sectional view of the slope. The full widths at half-maximum of EL profile are $1.1 \mu\text{m}$. At both the junctions, the emission peaks are shifted to the p side in the transition region. Comparing with the results of PL spectra measurement, these results imply that the active regions exist at the point where PL peak wavelength has the same value in the transition regions. We also observe the slower rise of EL signal in the p side than in the n side. This asymmetry should be caused by the difference of the diffusion length of electrons and holes or by the internal structure of the p-n interface.

3.5. Multiwavelength near-field photocurrent measurement

So far, NSOM has been employed for the observation of small structures on the surface or optically thin materials. In the study of bulk devices, however, their internal optical and transport properties should also be precisely examined. For this purpose, near-field photocurrent (PC) measurement with propagating modes into the sample is expected to make an important contribution. Though propagating modes do not have any resolving power, 'tomographic' information of the investigated material can be obtained by systematically varying the optical penetration depth over a wide range.

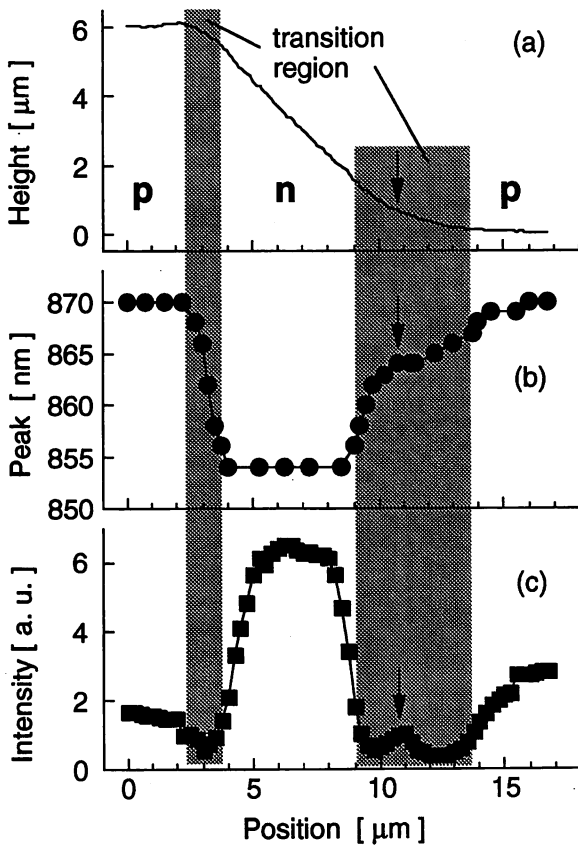


Fig. 5. (a) Cross-sectional view of the slope. (b), (c) Plots of photoluminescence peak wavelength and integrated emission intensity, respectively.

Schematic diagram of experimental setup is shown in Fig. 4(d). As multi-wavelength light sources, Ar⁺ laser ($\lambda = 488$ nm), He-Ne laser (633 nm) and Ti:Sapphire laser (780 and 830 nm) are utilized. By using these

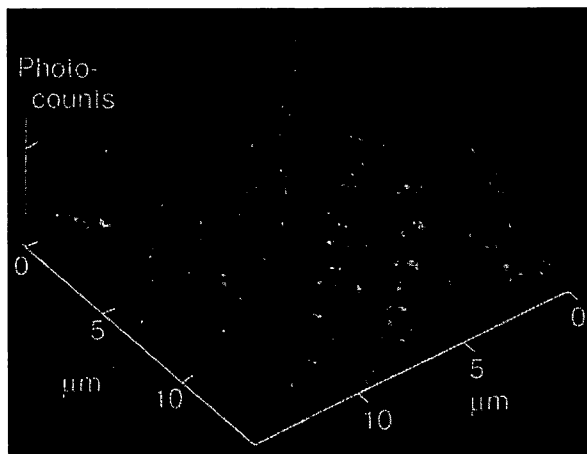


Fig. 6. Perspective view of the photoluminescence-intensity image with the illumination-collection hybrid mode. The scanned area is the same as that of image (b) in Fig. 3.

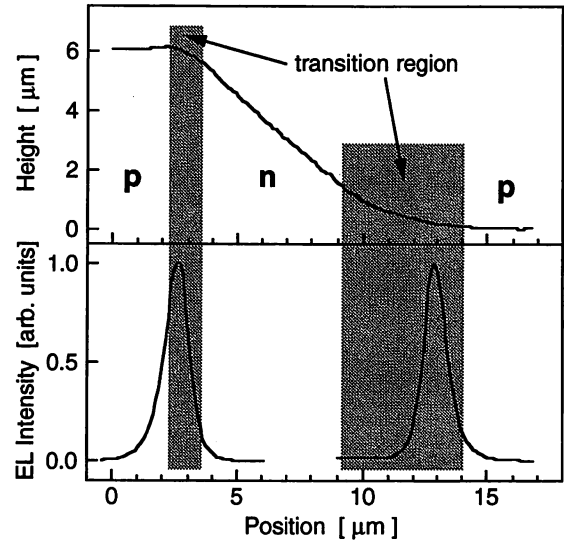


Fig. 7. Cross-sectional view of the slope and electroluminescence line scans at the upper and lower junctions.

lights, the optical penetration depths in GaAs can be tuned from 80 nm to 1.0 μm . For the case of the 200 nm aperture in the proximity to the GaAs surface, almost all the evanescent modes in air are coupled into propagating ones for the entire wavelength range. In this experimental situation, the resolution is limited not only by the aperture size but also by the penetration depth [6,8].

Fig. 8(a) shows the cross-sectional profiles of PC intensities. At the excitation wavelength of 488 nm, due to the shallow penetration depth, the resolution is determined by the aperture size and the diffusion length of photoexcited carriers. The difference of diffusion

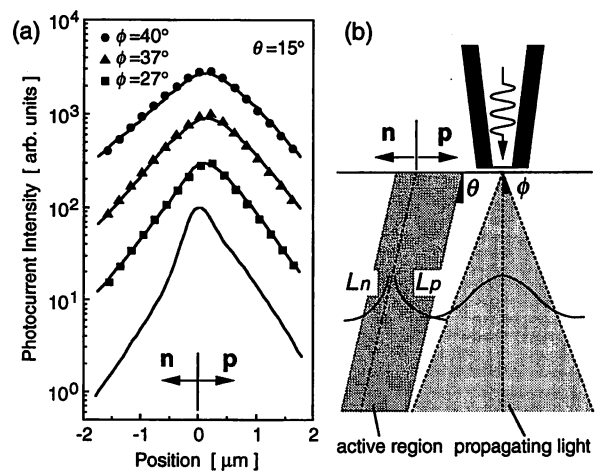


Fig. 8. (a) Near-field photocurrent line scans at the excitation wavelength $\lambda = 488, 633, 780, 830$ nm from the bottom to the top. (b) Schematic of the slant p-n interface and propagating light into the GaAs. The slant angle θ and the beam spread angle ϕ are the fitting parameters in the calculation.

length of electrons and holes can be observed through the slower rise of PC signal in p-region than in n-region. The increase of penetration depth results in the decrease of the resolving power. Moreover, on increasing the penetration depth, the decay length becomes longer and the asymmetric behavior reverses. We believe that the longer decay length in n-region than in p-region at larger penetration depth can be explained by the slant of the p-n interface (shown as θ in Fig. 8(b)).

Here, we make an analysis of the asymmetric signal behavior using a one-dimensional model as shown in Fig. 8(b). Since the detailed procedure of this analysis is described in [7], we briefly show the results here. The experimental curves (b)–(d) are perfectly fitted as shown in Fig. 8(a). From this calculation, we obtain the slant angle of the p-n interface $\theta = 15 \pm 8^\circ$. A total slant of the p-n interface of $30 \pm 8^\circ$ to the p side, which is the sum of the observed slant angle θ and the intended tilt angle of 15° in the experimental setup, can be explained by the crystal orientation dependence of the growth nature. The most significant origin is that the growth rate of GaAs on (311)A is faster than that of (111)A, which causes the shift of n-type region to p side on growth [23].

4. Characterization of evanescent light on small aperture

In this chapter, through the near-field PC measurement, the aperture-size-dependent feature of evanescent field is discussed. By employing apertures with the diameter of 100–200 nm, we clarify the mode-conversion process, lateral resolution, and contribution ratio of the evanescent light to the propagating one.

Fig. 9(a) shows the PC line scans in the vicinity of the upper junction using 100, 150, 200 nm diameter apertures. Since, in the case of 100 nm, a large part of the excitation light through the aperture remains evanescent in the sample [7], the resolution is determined only by the aperture diameter. With increasing diameter, the propagating light into the sample becomes dominant and the resolution is affected by the penetration depth as pointed out in Section 3.5. Even if we use 100 nm aperture, the propagation light partially generates the PC signal as a background, which lowers the contrast. In order to evaluate the essential nature of the evanescent field, we measure the position-dependence of decay length of the PC signal as shown in Fig. 9(b). When the tip is positioned aside the active region, only the propagating light induces PC signal. When put just above the active region, on the other hand, not only the propagating light but also the evanescent one, generates the signal. Higher spatial frequency components (wavevector $k_{//}$ parallel to the aperture plane) of the evanescent

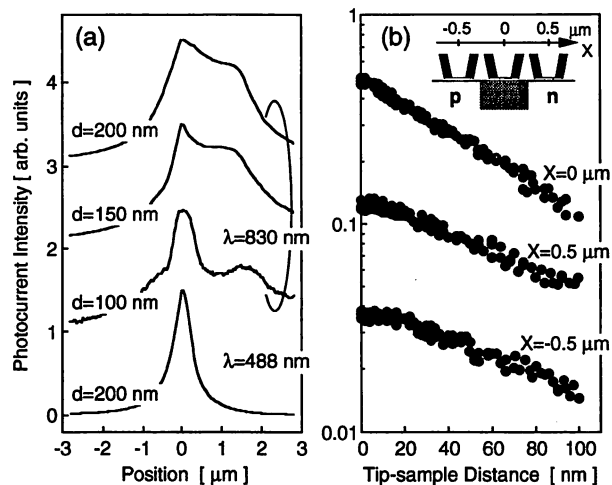


Fig. 9. (a) Near-field photocurrent line scans using the apertures with the diameters of 100, 150, 200 nm at the excitation wavelength $\lambda = 830\text{ nm}$. The bottom line scan is the result of the measurement with the 200 nm aperture and $\lambda = 488\text{ nm}$ excitation. (b) Near-field photocurrent intensity as a function of tip-sample distance at the positions indicated in the inset.

modes than of the propagating ones result in a shorter decay length of 60 nm. The upper limit of $k_{//}$ should be determined by the depth of the surface depletion layer. By comparing these curves, we can deduce the intrinsic behavior of evanescent modes.

5. Summary

Near-field scanning optical microscope is applied for the diagnostics of semiconductor photonic devices. To overcome the serious problem of the low sensitivity, we have tailored the shape of the fiber probe. Evaluating the transmission efficiency over a wide range of aperture diameters, the strong optical loss region in the metal-cladding waveguide is quantitatively determined. By shortening this part as much as possible, the transmission coefficient is enhanced by an order. The capability of highly sensitive measurement is demonstrated by the imaging and spectroscopic analysis of novel-structured lateral p-n junctions. Through the photoluminescence and electroluminescence observations, the carrier distribution and dynamics in the transition and the active regions are investigated. Multiwavelength photocurrent measurement reveals the internal structure of the sample, the slant angle of the p-n interface. The intrinsic nature of evanescent field on a small aperture is clarified by examining the aperture-size-dependent behavior of the photocurrent signal. The amount of evanescent field retained in the sample is estimated from the lateral resolution and the decay length of the signal.

Acknowledgements

We are grateful to S. Mononobe and Y. Yokoyama for helping us fabricate fiber probes.

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Nano-Fabrication and Atom Manipulation by Near-Field Optics

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Recent progress of our activities on near-field optical technology is reviewed. Topics to be presented are:

- (1) High throughput probe fabrication and evaluation,
- (2) Nano-fabrication and application to the high density optical storage/read-out, and
- (3) Atom guidance and application to deposition.

ULTRA-HIGH SPATIAL RESOLUTION MAPPING
MEASUREMENT OF POLYDIACETYLENE USING
A NEAR-FIELD RAMAN SPECTROMETER

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The Near-Field Optical Microscope (NOM) is a new optical system designed for ultra-high spatial resolution measurement using visible light. By combining this NOM technique with NRS-2000 Raman spectrometer, the 100 nm resolution measurement of Raman spectra can be achieved. The optically excited area of the sample is determined not by the wavelength of light, but by the aperture size (around the 150 nm) of the probe tip.

This system can simultaneously measure the topography of a sample surface and the near-field Raman spectrum. A single tabular Polydiacetylene (PDA) crystal is prepared for mapping the 1 μm X 1 μm area in 100 nm steps. The two spectra (separated by 100 nm) show different spectral features, which suggests differences in the *cis*- and *trans*- bonds distribution in the crystal. The mapping results also suggest that we can characterize the spatial differences of a sample using a Near-Field Raman Spectrometer, and that the spatial resolution is at least one order higher than a conventional micro Raman spectrophotometer.

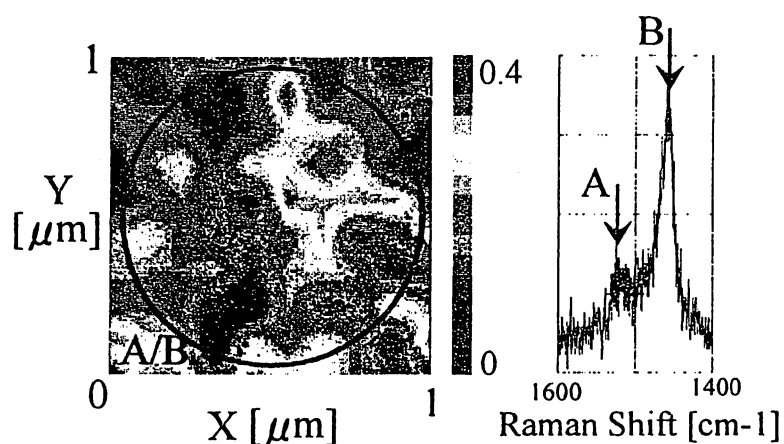


Figure 1. Near-field Raman spectrum of polydiacetyrene and its mapping results. The circle indicates the spatial resolution of conventional Raman spectrometer.

**HIGH SPEED NEAR FIELD OPTICAL TECHNOLOGY
FOR ULTRAHIGH DENSITY STORAGE****Motoichi OHTSU**

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Requirements on near-field optical technology[1] are presented to realize a high density optical storage. They are: (1) software architecture, (2) system design, (3) optical head for write/read, and (4) optical storage media. Among them, we review recent progress of our works on (2) and (3).

For designing the system, we have proposed a tracking-less scanning of an apertured planar probe array in order to maintain a data transmission rate as high as 100 Mb/s with 10^{-9} shot noise limited bit error rate.

For fabrication of an optical head, we have proposed and demonstrate a novel method to enhance the near-field optical intensity on the apertured probe with its foot removed asymmetrically by using a focused ion beam(Fig.1). Observed spatial distribution profile of a generated optical near field on the probe was in good agreement with a numerical result corresponding to the HE-plasmon mode. Compared with the symmetric apertured probe, ten times enhancement of the near field optical intensity on the asymmetric probe was observed(Fig.2).

In order to apply the principle of asymmetric probe, a novel Si subwavelength planar apertured array was fabricated utilizing the microfabrication technique of semiconductors for near field optical head(Figs. 3, 4). The spatial distribution of near field optical power shows a single peak with a high throughput. For further increase in the throughput, a ball lens was fixed on each aperture for focusing an far field incident/picked up light(Figs.5, 6). Experimental results and future problems to be solved will be presented at the session.

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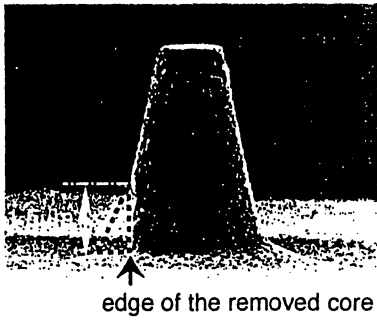


Fig.1 SEM image of an asymmetric fiber probe.

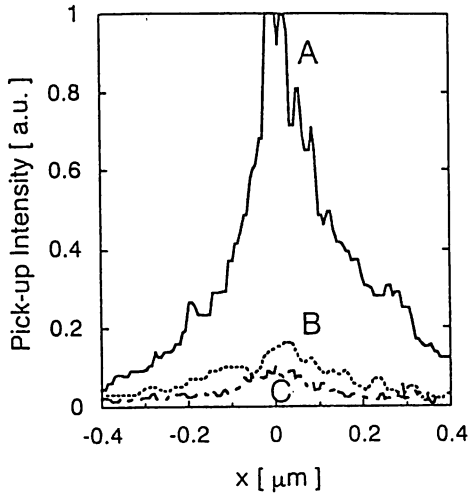


Fig.2 Cross-sectional spatial distribution of the near field optical intensity on an asymmetric (A and B) and a symmetric (C) probes with aperture diameter of 100 nm

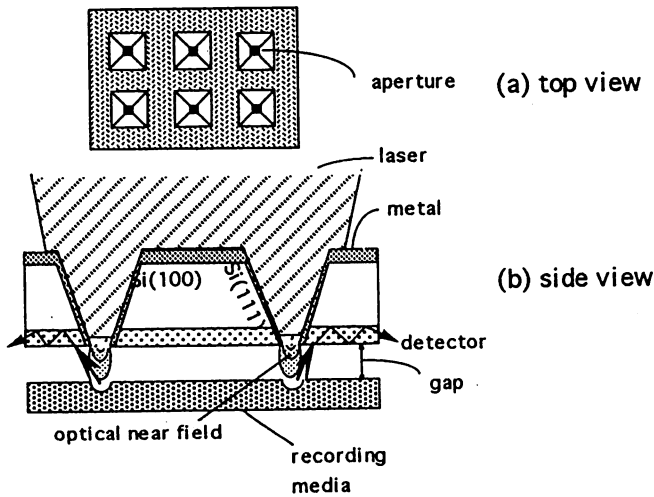


Fig.3 Schematic explanation of a planar apertured probe array.

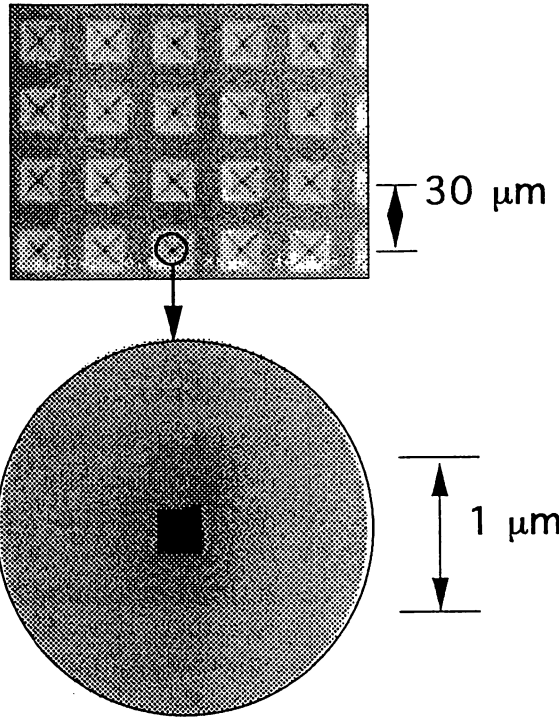


Fig.4 Top view of a fabricated planar apertured probe array.

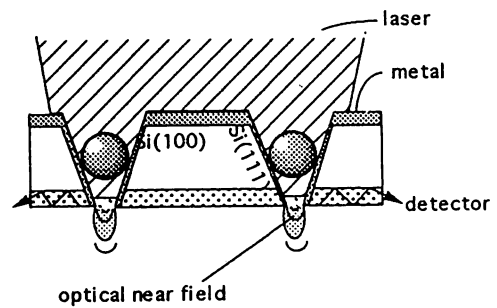


Fig.5 Schematic explanation of a planar apertured probe array with a ball lens.

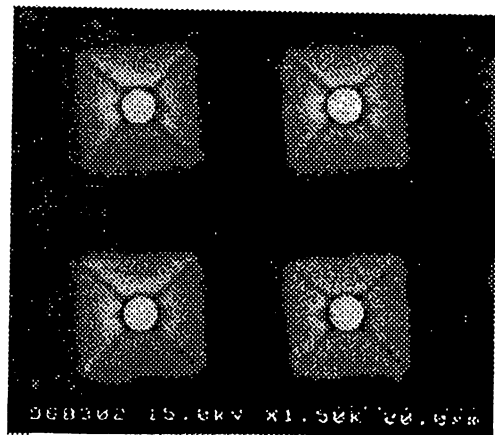


Fig.6 Top view of a fabricated planar apertured probe array with a ball lens.

Near Field Optical Technology for Nano/Atom Photonics

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We review recent progress of our works on near field optics. We focus on presenting the results of

1. Spatially resolved photoluminescence spectroscopy of a single quantum dot of FaAs at liquid-He temperature, quantum wire of polysilanes in UV region, and Raman spectroscopy of Si.

2. Application to high density optical storage: We present technical problems to be solved by the national project which was recently founded under supervision of MITI, Japan. They are for the software architecture, nanomechanic driving systems, optical pickup devices, and storage media. Among them, we have proposed the two dimensional apertured planar probe array for the optical pickup for fast writing/reading. Results of its fabrication are presented.

3. Atom manipulation: Toward atom-level deposition and crystal growth, we have succeeded in guiding Rb atoms through a hollow fiber by using a dipole force of the optical near field. Recent experimental results and future outlook are presented.

Based on these results, we try to demonstrate that near field optical technology can be used to a variety of applications in the field of photonics dealing with nano-materials and/or atoms, which can be called nano/atom photonics.

Near-Field Optical Spectroscopy of Single Quantum Dots and Related Materials/Devices

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1. Introduction

In the last years zero-dimensionally confined semiconductor quantum structure have received much attention because of the rapid progress in the growth of quantum dots (QDs) using the Stranski-Krastanow mechanism. One of the most attractive optical properties of QD is its narrow and strong transition probability due to atom-like level structure with discrete density of states. This specific feature of quantum structure is reflected in the single-QD photoluminescence (PL) spectrum [1-3], which provides us with important information on the quantum confined energy and on the scattering and relaxation processes of excitons. In early studies, the sharp PL peaks of single dots are hidden in the inhomogeneously broadened spectrum of a large number of dots with statistical size fluctuation. In order to extract the individual QD contribution from a dot ensemble, observation number of dots must be restricted by applying novel techniques with high spatial resolution. In this paper, we report the low temperature near-field PL spectroscopy and imaging of InGaAs single QDs in a wide excitation power region.

2. Experimental

Figure 1 shows a schematic of the QD sample structure and the experimental configuration. In_{0.5}Ga_{0.5}As self-assembled QDs were grown on (100) GaAs substrate by gas-source molecular beam epitaxy with a density of $<10^{10}$ dots/cm² [4]. Typical dot diameter of around 30 nm and height of 15 nm were observed with atomic force microscope.

We employed chemically etched fiber probe having an aperture with the diameter of 500 nm at the apex part. The shape of the fiber probe was optimized to attain high sensitivity by the method detailed in ref. 5. By tailoring the apex part to have a large cone angle, the excitation and collection efficiencies are practically improved. For the near-field excitation, the QD sample on a scanning piezotube was illuminated with the 633-nm

line of a He-Ne laser light through the aperture. Carriers were generated not only in the InGaAs QDs but in the barrier layers of GaAs and AlGaAs. Most of the photoexcited carriers migrated in the barrier layers and were captured by the confined states of QDs. The resultant PL signal was collected by the same aperture. After rejecting the excitation light with long pass filter, the signal was focused into a 50-cm monochromator and was detected by a cooled photomultiplier tube using photon counting technique. Shear-force feedback was employed to control a probe-sample distance of less than 10 nm. The temperature of the sample was kept at 5 K in a cryostat.

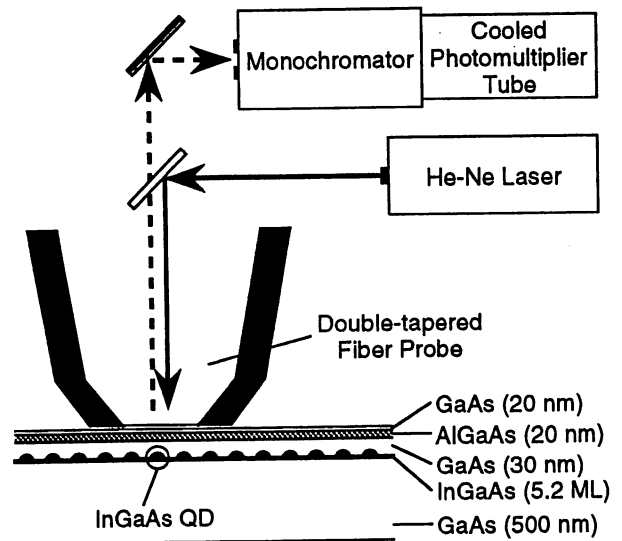


Fig. 1 Schematic of sample structure, optical configuration, and block diagram of the measurement.

3. Results and Discussion

Figure 2 shows near-field PL spectra of a single quantum dot for different excitation power densities. At the weak excitation (<3 W/cm²), PL spectrum consists of a single peak. By taking into account the carrier injection rate of $\sim 10^8$ e-h/s per dot and the lifetime of

~1 ns, we demonstrate that the single line originates from the recombination of the ground-state single exciton. With increasing excitation power, additional features are observed in the spectra. After the saturation of the ground state, an emission from excited state appears 30 meV above the ground-state emission. Moreover, 2.2 meV below the single-exciton emission, another line grows super linearly with the increase of excitation intensity. From the energy position and power dependence, we associate this feature with the biexciton emission.

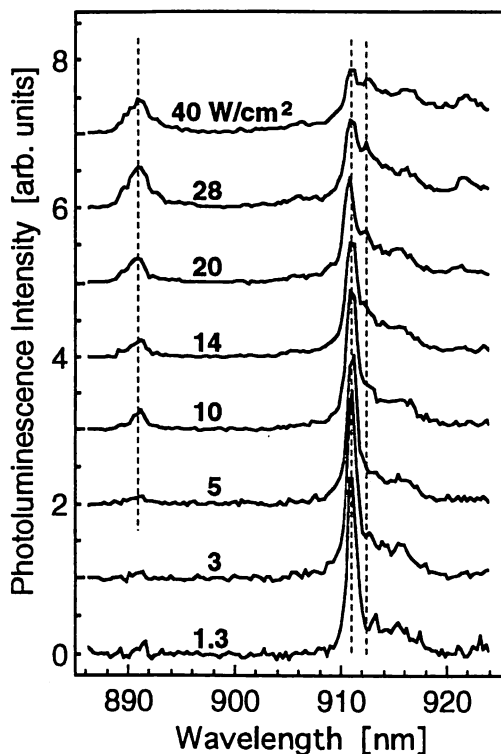


Fig. 2 Near-field PL spectra of single quantum dot for different excitation power densities.

In Fig. 3, we construct the monochromatic PL images by fixing the detection wavelength and by scanning the probe in $4 \mu\text{m} \times 4 \mu\text{m}$ area. The bright circular spots display the spatial distributions of single QDs having the PL wavelengths of 975 nm for various excitation powers. With increasing the excitation power, the number of spots increases drastically due to the appearance of additional emissions of excited states and biexciton states. The minimum size of the spot image at FWHM is 160 nm. Since the photoexcited carriers diffuse into extensive region ($>1 \mu\text{m}$), the spot size is determined only by the collection area of PL signal through the aperture of the fiber probe. From

these results obtained, considerably high resolution of $\lambda/6$, much smaller than aperture diameter of 500 nm, is found to be attained.

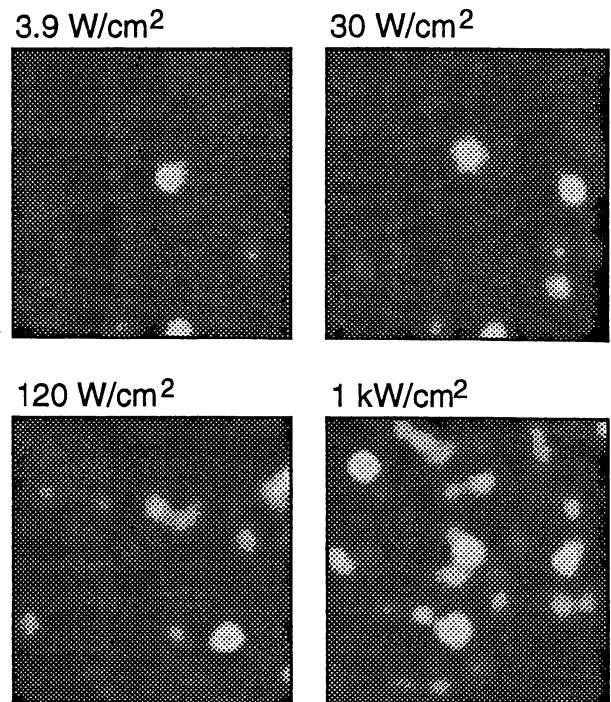


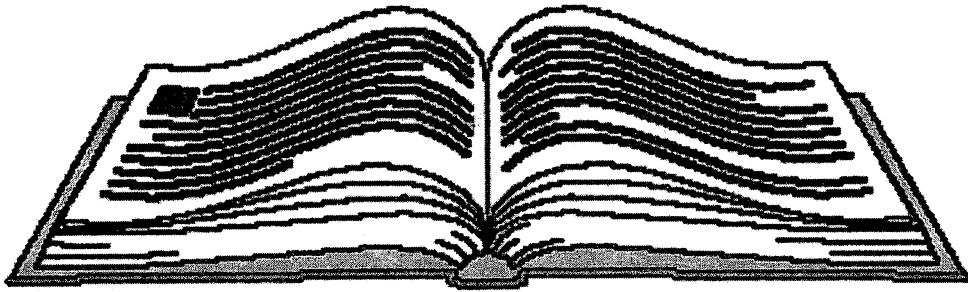
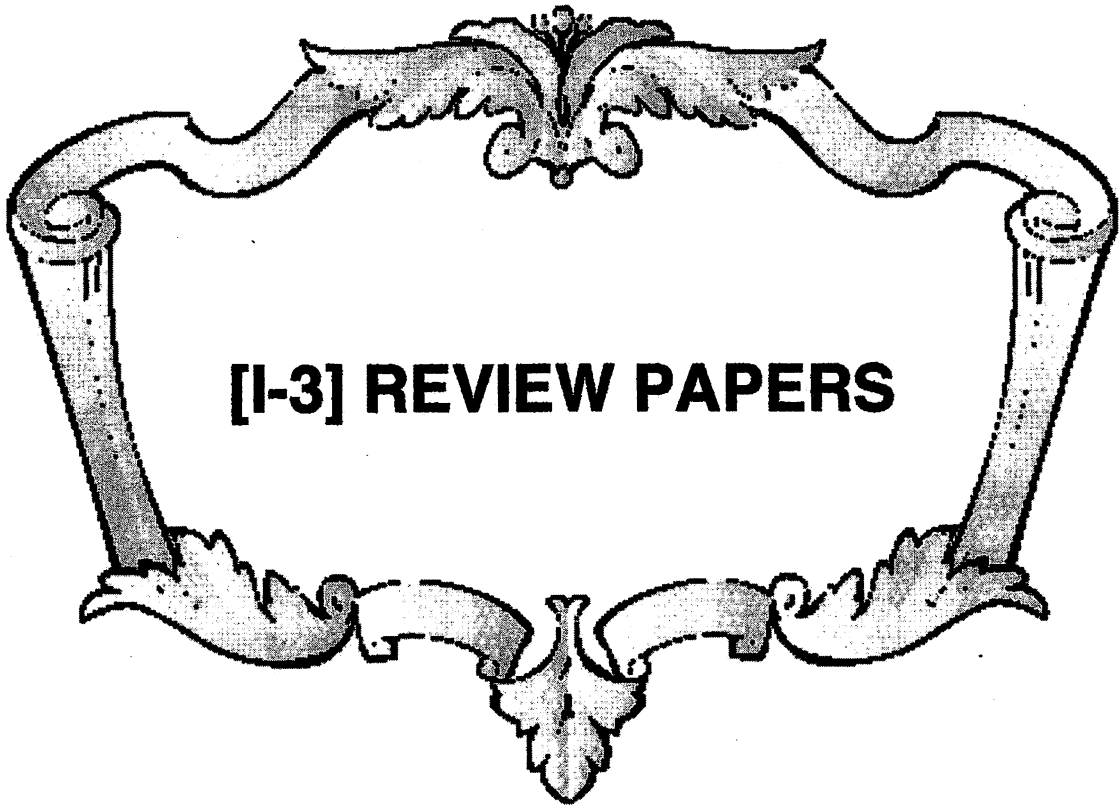
Fig. 3 Monochromatic PL images for various excitation power intensities. The scanning area is $4 \mu\text{m} \times 4 \mu\text{m}$.

4. Conclusion

We study PL spectroscopy of single QDs in high density structure by using near-field optical microscope. For the enhancement of the spatial resolution, the illumination and collection hybrid mode is utilized with double-tapered fiber probe. We precisely examine the excitation-power dependent feature of PL spectra and monochromatic PL images. Besides the single-exciton emission, additional emission lines from excited states and biexciton states are also observed. A single QD is imaged as a circular spot of 160-nm at minimum. The reported technique can be widely applied to the single particle observation of the inhomogeneously broadened systems prepared on opaque substrates, where the signal cannot be detected in the transmission configuration.

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近接場光学の固体光物性への応用

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§1 はじめに

固体光物性の研究分野において、「できるかぎり狭い領域からの発光を観測したい」、「局所的な透過スペクトルの情報が欲しい」など、顕微分光技術の重要性はかなり以前から認識されていた。殊に最近、半導体量子構造の個々の特性や、デバイスと直結した光機能材料の均一性の評価などを目的に、その必要度が年々高まりつつある。これは、光機能部分、さらにはそれらを構成要素とするデバイス自体のサイズが著しく小型化へ向かっていることと密接に関係していると思われる。

従来の光学顕微鏡(ここでは走査レーザー顕微鏡を例にとる)の場合、高い倍率(大きな開口数)をもつレンズを使用することにより、レーザービームはその波長の半分程度のスポットに絞り込むことが可能である。第1図(a)に示すように、たとえばこのレーザー光が試料の蛍光を励起する役割を果たすとすれば、ビームの集光位置を走査することにより、上記のスポット径の分解能をもつ蛍光画像が得られることになる。しかしこのようなレンズによる集光技術にとどまる限り、その分解能は光の回折現象のために、半波長を越えることは原理的に不可能である。

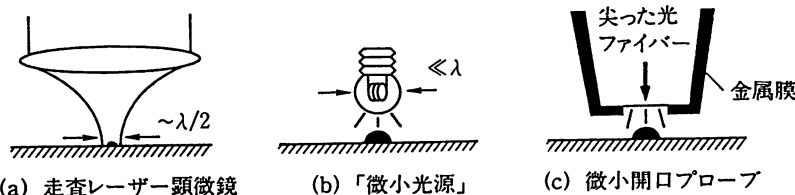
では、この限界を打破するにはどのようにすれば良いのであろうか? 答は簡単に思いつくのだ

が、波長よりもずっと小さな光源を作り、それを試料のごく近傍で照らしてやれば良いのである(第1図(b))。この単純な原理を実現したものが、まさに近接場光学顕微鏡である^{1~3)}。最も典型的な微小光源をもつ探針(プローブ)の形態を第1図(c)に示す。光に対して強い遮光性をもつ金属膜に、波長よりも十分小さな穴(以下では微小開口とよぶ)を開け、そこから漏れ出てくる光を試料に照射する。さらにこの微小開口を試料の近傍で走査することにより、開口の大きさ程度の分解能をもつ画像計測が可能となる。

このような近接場顕微鏡技術は、ここ10年あまりで急速に研究が進み、各国において市販品が出回るまでに発展した。その背景として、以下の3つの技術進歩が大きく貢献していると思われる。

(1) 光ファイバーの先鋭化を主とする、プローブ作製のための微細加工技術、(2) STMやAFMに代表されるプローブ走査技術、(3) 微弱な信号(蛍光やラマン散乱)に対する光検出技術。このようなバックグラウンドと、はじめに示した研究ニーズに後押しされ、近接場顕微鏡はその性能を高めながら、固体物性にとどまらず、生体観察など広い分野にその応用範囲を獲得しつつある。

このように近接場顕微鏡の測定対象は多岐にわたり、それぞれに応じてプローブをはじめとする各要素の最適形状、構造がある。本稿では、固体



第1図
従来の光学顕微鏡から
近接場光学顕微鏡へ。

光物性への応用に的を絞る、実際の測定例を通して、近接場の概念、技術的な現状、近接場固有の物理、今後の進展の方向などについて述べる。特に後半では、いわゆる「微小開口型」とは趣を異にする「散乱型」の近接場顕微鏡について触れる。これは、微小開口型をはるかに凌ぐ分解能が期待され、その動作原理も分極の制御という点に集約されており、光物性の観点からも大変興味深い。できる限り誌面を割いて詳述したいと思う。

§2 微小開口の光学

2-1 開口近傍の電場分布

近接場光学顕微鏡の心臓部である微小光源、すなわち波長よりも小さな金属開口の光学について簡単に説明する⁴⁾。無限に広い理想的な金属(誘電率の実部が負の無限大、虚部がゼロ)に波長よりも小さな開口(現実的な大きさとして、50~100 nm,あるいは $\lambda/10$ 程度を想定していただきたい)が開いている。そのような金属スクリーンに対し、裏面から直線偏光(x 方向)をもつ平面波を照射したときに、面内に生じる電場分布を計算した結果が第2図(a)である。金属の鋭いエッジを反

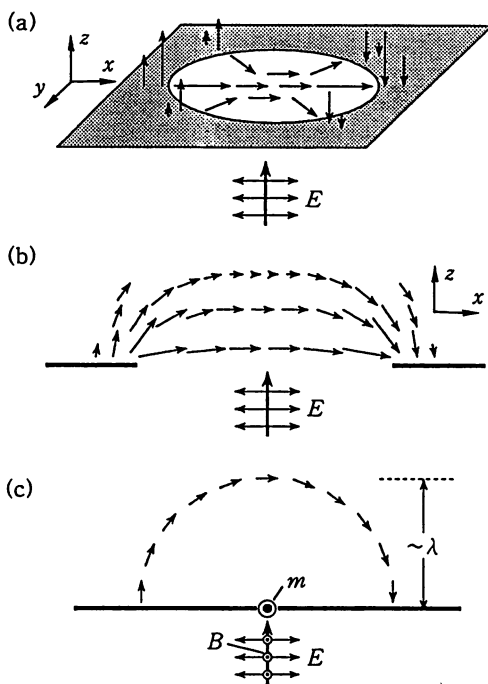
映して、開口の縁近傍には非常に強い電場が発生する。 x 方向と y 方向での大きな違いは、直線偏光の光を照射したこと起因しており、直感的には以下のように説明される。 x 方向の縁では、金属の自由電子が集中し、強い電場をもたらす。一方、 y 方向の縁では自由電子は接線方向に素通りするため、電場は発生しない。また、金属面内では特に縁近傍において z 方向に強い電場分布をもつ。

続いてこの開口面内での分布をもとに、開口近傍でどのような電場が生じているかを計算する。ここでは近接場の様子を最も象徴的に反映した xz 平面での分布を第2図(b)に表示する。幸いこの面内に限れば y 方向の電場成分はゼロである。開口面内の分布から容易に想像されるように、開口の中心近傍では x 方向の電場のみであるのに対し、開口の縁近傍では z 方向の電場も発生している。 x 方向、 z 方向いずれに対しても、開口から遠ざかるに従い、急速に電場強度が減衰していることがわかる。まさに開口が「微小光源」として機能する所以である。

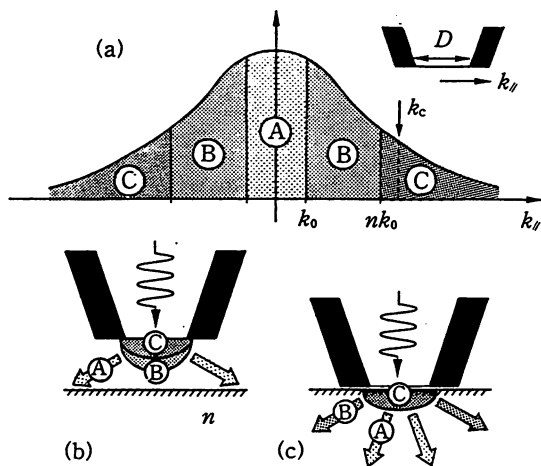
では、開口から十分遠ざかってしまったら、もはや電場はまったく存在しないのであろうか?第2図(c)は開口から波長程度離れた位置における電場の様子を図示している。このような電場はいくら開口が小さい場合でも必ず存在し、開口からの距離に対して -2 乗で減衰する。つまりどこまでも伝播する放射(伝搬)成分である。この成分は、入射光の磁場が開口面内に作る実効的な磁気双極子モーメント(m)から放射される場と完全に等価であることが理論的に示されている⁴⁾。

2-2 エバネッセントモードと伝搬モード

上で示したように微小開口からの回折波は、無限遠まで伝播していくモード(伝搬モード)と開口近傍にのみ存在する非伝搬モード(エバネッセントモード)の2成分からなることがわかる。第3図はそのモード占有の様子を概念的に示したものである。開口面内の波数を $k_{||}$ 、それに対して垂直方向の波数(z 方向)を k_{\perp} とした場合、 $k_{||}^2 + k_{\perp}^2 = k_0^2$ という関係が成り立つ。 k_0 は真空中の波数($2\pi/\lambda$)である。 $k_{||} < k_0$ の場合、 k_{\perp} は実数であり、



第2図 (a) 開口面内、(b) 開口近傍、(c) 開口から波長程度離れた位置での電場分布。



第3図 微小開口による回折波のモード分布.

伝搬モードとして振る舞う。一方、 $k_{\perp} > k_0$ の場合、 k_{\perp} は虚数となってしまい、電場強度は開口からの距離に対し指数関数的に減衰するエバネッセントモードとなる。第2図(b)に示した開口のごく近傍の電場分布には、両方のモードが混在しているのに対し、第2図(c)の電場は伝搬モードによるものである。両モードの占有比は、開口の大きさに依存し、第3図の回折波スペクトルの拡がり、つまりカットオフ波数 k_c (およそ π/D であることが実験的に確認されている。§4も参照)によって決まる。開口が大きくなるに従い、伝搬モードの占有率が大きくなっていく。

このような波数空間におけるモード表示は、真空中における微小開口の光学を理解する上では、さほど本質的ではない。しかし、固体物理への応用、つまり非常に屈折率の高い媒質、あるいは波数の概念が重要な意味をもつ電子系との相互作用を理解し、現象を予測する上では、見通しの良い表示方法である。具体的には§4で述べる。

§3 近接場光学顕微鏡技術のキーワード

3-1 プローブと分解能・感度

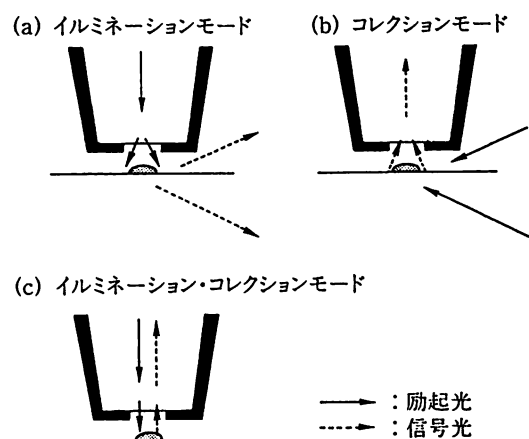
現在の近接場顕微鏡の微小開口型プローブは、すべて光ファイバーをベースに作製されている。化学エッチング法⁵⁾、あるいは加熱引っ張り法⁶⁾により、光ファイバーの先端部分を数nm～数百

nmのサイズに尖らせる。その後、先端部を除いた全体に遮光用の金属(アルミまたは金)を蒸着し、微小開口を作製する。典型的には50～200nmの直径をもつ開口が多いが、必要に応じて500nm～1 μ mという大きな開口を使用することもある。

§2で述べたように、強い電場は開口のごく近傍にのみ存在するので、原理的には開口径が小さいほど、高い分解能が得られる。しかし現実には、開口径を小さくした場合、励起、集光の効率が著しく低下し、特に分光測定においては深刻な問題となる。その大きな理由は、プローブ全体がテーパ形状をもつ金属クラッドの光導波路となっており、波長以下の細い部分において、強い光損失を受けるためである(カットオフによる遮断と金属による吸収という2つの要因がある)。最近では、この問題点を克服するため、テーパ部分の形状の工夫や⁷⁾、導波モードとして表面プラズモンを利用する⁸⁾などの工夫が積極的になされている。

3-2 測定モードと距離制御

励起光の照射方法、信号光の集光方法によって、測定モードは多種多様である。第4図にそれらのうちの代表的なものを示す。まず、頻繁に用いられるイルミネーションモード(第4図(a))では、微小開口を通して励起光を照射し、外部で信号光を検出する。透明なガラス基板やホストに分散させた試料を観察する場合に、照射と反対方向において、大きな開口数をもつ光学系で集光が



第4図 近接場光学顕微鏡のさまざまな測定モード.

可能であるため、高い感度が得られるというメリットがある。続いて、信号光を開口を通して検出するのが、コレクションモード(第4図(b))である。光励起を必要としない、電流注入による発光の検出などでよく用いられる。最後に第4図(c)に示すのがこれらのハイブリッドモードであり、励起と集光を同一の開口を用いて行う。これは特に半導体試料の観察において、光励起キャリアの拡散に起因する分解能の低下を回避する際に、大きな威力を発揮する。

近接場顕微鏡において、開口径程度の分解能を再現性良く達成するためには、常に試料のごく近傍から光を照らす必要がある。そのための距離制御として、開口と試料との間に働く力(シアフォース)を検出する方法が広く利用されている⁹⁾。これにより、試料との距離は5~10 nmにおいて、Åの精度で保持される。これはまた、試料表面の凹凸構造を画像化するAFMとしての役割を果たしている。

3-3 既存技術との組合せ

励起光源として短パルスレーザーを用いることにより、時間・空間分解分光が可能となる。その際に最も問題となるのは、光ファイバー、およびプローブの先端部における強い波長分散(正のチャージング)である。これを解決するため、ファイバーへの導入の前にあらかじめ負のチャージングをかけておき、プローブの開口部で最適のパルス幅となるように調整を行う。この方法によれば、50~100 fs程度のパルスを開口近傍で発生させることが可能である¹⁰⁾。

また、発光の量子効率やその遷移幅が測定温度に強く依存する半導体試料などでは、低温における観測が不可欠となる。近接場顕微鏡のヘッド部分をコンパクトに設計することにより、クライオスタット中において、液体ヘリウム温度下で動作を行った測定が数多く報告されている¹¹⁾。

§4 実際の測定例

近接場顕微鏡の固体・表面物性分野におけるこれまでの応用方法を振り返ってみると、その用途

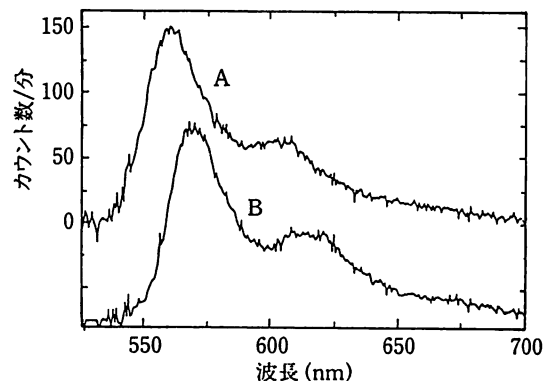
は大きく2つに分けられる。まず1つは、単一粒子分光である。半導体量子構造や色素分子など、環境やサイズのばらつきをもつ粒子群の中から、ただ1つを選び出し、個別の特性を評価するものである。もう1つは空間分解分光であり、結晶中での不純物や欠陥の存在やその均一性を観察するための手段として用いられている。

以下では、このような用途の分類に従って、実際の応用例を紹介しながら、近接場顕微鏡に固有の物理現象についても触れていく。

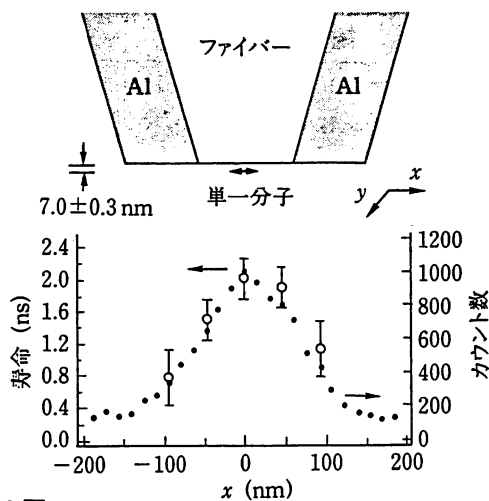
4-1 単一粒子分光

ここではまず、最も典型的な観測例として、単一色素分子の観測を取り上げる。色素分子の発光特性は分子自身の向きやそれがおかれている環境に強く依存するため、単一の分子観測には大きな意義が見いだされていた。1993年のBetzigらによる成功¹²⁾を皮切りに、これまでに世界の数多くのグループから報告がなされている。測定手法はほぼ共通しており、直径100 nm程度の開口を用い、ガラス基板上または薄膜中に分散させた色素分子をイルミネーションモード動作によって観察している。

第5図に単一色素分子からの発光スペクトルを示す¹³⁾。AとBは互いに150 nm離れた色素分子から得られたものである。周囲の環境の違いを反映して、そのスペクトルのピーク位置が大きく異なっていることが確認できる。単一分子観測ならではの重要な結果である。また、光ビーム照射の測定ではみられない興味深い現象として、プロ



第5図 150 nm 離れた2つの単一色素分子からの発光スペクトル¹²⁾。



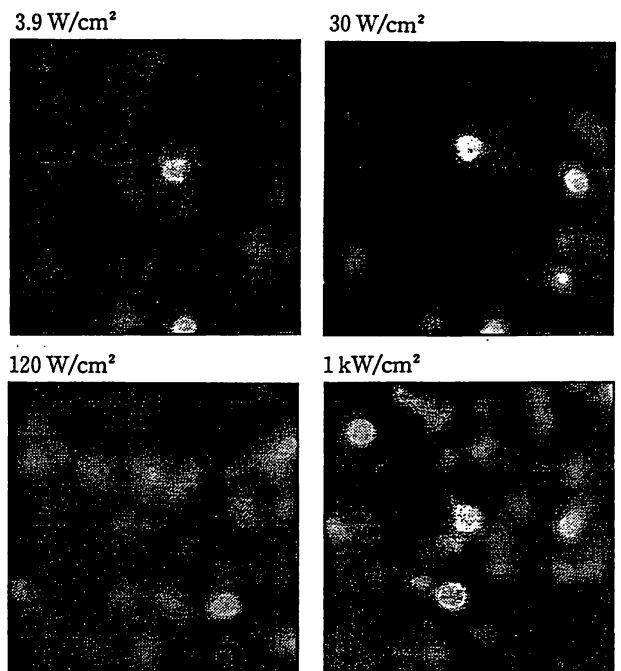
第6図 蛍光強度と寿命のプローブ位置に対する依存性¹³⁾。

プローブの接近による色素分子の発光特性の変化が議論されている。第6図に示すように、分子が開口の中心部(ファイバー部)にいる場合に比べ、縁(金属部)の近傍においては、発光寿命が非常に短くなっていることがわかる¹⁴⁾。これは現在のところ、分子から金属への無輻射エネルギー移動が強起しているためであると解釈されている。

上に挙げた結果以外にも、以下のような主だった成果がこれまでに得られている。(1)発光像のスポット形状を解析することにより、分子(双極子)の向きを個々の分子について決定する¹²⁾。(2)単一分子の観測を裏付ける、発光強度の不連続な変化(退色)¹⁵⁾。(3)ドナー分子とアクセプター分子間のエネルギー移動の観測¹⁶⁾。その他の測定例については文献¹⁷⁾を参照されたい。

続いて、単一粒子分光のもう1つの観測対象である、半導体量子構造について述べる。量子ドットを例にとると、その離散的な(原子的な)準位構造は非常に鋭く強い光学遷移をもたらすため、光デバイス、とりわけ半導体レーザーへの応用が期待されている。このようなドットが本来もつ光学特性を正しく評価するにあたっては、単一ドット分光が不可欠である。なぜなら、通常のマクロなスペクトル測定では、ドットのサイズのばらつきを反映した不均一ひろがりしか情報として得られないからである。

量子構造の観測においても、もっぱらフォトル



第7図 単一量子ドットからの発光像の励起強度依存性。

ミネッセンス測定が主流である。先に述べたようにこれらの測定はすべてヘリウム温度のクライオスタット中で行われている。量子井戸中の層厚のゆらぎによる島構造からの鋭い発光特性¹⁸⁾や単一の量子細線¹⁹⁾やドットからの発光スペクトル測定^{20,21)}などがこれまでに行われている。

第7図に筆者らが試みている低温における単一ドットからの発光観測の結果を紹介する²²⁾。試料はGaAs基板上に成長した自己形成InGaAs量子ドットである。検出波長を975 nmに固定し、プローブを $4 \mu\text{m} \times 4 \mu\text{m}$ の範囲で走査することにより、発光画像を得ている。1つ1つのスポットが単一のドットからの発光に対応している。ここでは、直径30 nmのドットが約150 nmのスポットとして観測されている。励起強度を上げるとともに、スポット数が急激に増加しており、基底準位のみからではなく、さまざまな高次準位からの発光が検出されていることがわかる。発光スペクトルについても同時に測定しており、均一幅の評価だけでなく、電子間の相互作用や準位構造に関する情報も得られ始めている。

4-2 空間分解分光

光機能材料の表面近傍の欠陥や発光センターな

第1表
近接場光学顕微鏡の空間
分解分光への応用例.

観測対象	観測信号	文献
Ga _x In _{1-x} P 中の規則構造(ドメイン)	フォトルミネッセンス	23)
CaN 薄膜表面の不純物	フォトルミネッセンス	24)
GaAs 上のパッシベーション膜	フォトルミネッセンス	25)
ポーラスシリコンの発光特性	フォトルミネッセンス	26)
Ge _x Si _{1-x} 中の線状格子欠陥	光起電力	27)
Pt/GaP 界面のショットキーバリア	光電流	28)
Co/Pt 多層膜中の磁区構造	磁気カー回転	29, 30)
Pb ドープ KTP 中のストレス	ラマン散乱	31)

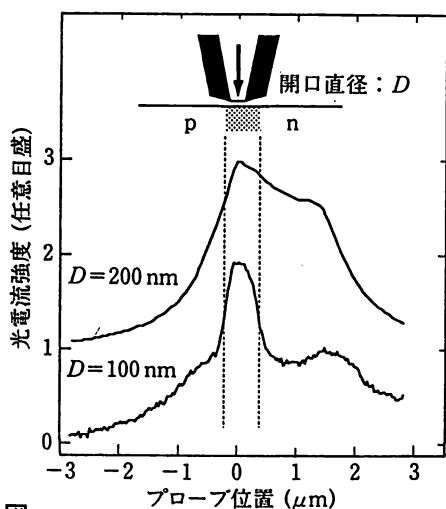
どの空間分布, 膜の均一性などを開口径程度(100~200 nm)の分解能で観察するという実験が数多くなされている. やはりフォトルミネッセンスの強度変化やスペクトル変化を信号として検出している例が大半である. このような近接場顕微鏡の応用方法については, この4,5年の間におびただしい数の論文が報告されている. それらについて1つ1つをここでは取り上げることはできないので, 第1表に主なものをまとめる.

その他の重要な観測例として, p-n 接合³²⁾や半導体レーザー^{33,34)}など, 実際の光デバイスの評価への応用がある. 特に p-n 接合を光検出器として用いた場合, 微小開口近傍の光強度分布や高屈折率媒質との相互作用を明らかにする上で, 有用な情報が得られている^{35~37)}. GaAs($n=3.5$)などの高い屈折率をもつ半導体試料に対して微小開口が接近したときの状況を考える. 第3図(b),(c)にもう一度戻っていただきたい. §2 で述べたよう

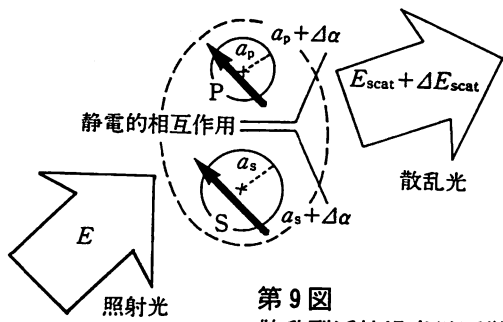
に, 真空中では $k_{||} < k_0(A)$ のみが伝搬モードとして振る舞い, 残りの B と C の成分がエバネッセントモードとして開口近傍に局在する. ところが, 開口が試料に十分接近した場合, $k_{||} < nk_0(A$ と B) が試料中への伝搬を許されるため, わずかな成分(C)だけがエバネッセントモードとして残る.

このようなモード変換に関して, 実験的に確認を行った結果を第8図に示す³⁸⁾. GaAsの p-n 接合上でプローブを走査し, 光電流測定を通して, 結晶中での光の挙動を探る方法を用いている. 波長 830 nm の光(この波長では, GaAsの吸収係数が小さいため, 結晶中への侵入距離は 1 μm 程度ある)を照射する場合, 開口直径が 100 nm の場合と 200 nm の場合で, その分解能が著しく異なっていることがわかる. この違いは以下のように説明される. 直径が 200 nm のときには, 回折スペクトルのカットオフ波数 k_c が小さい(広がり狭い)ために, 伝搬モードが支配的となる. したがって, プローブが接合から横方向に離れた位置で光照射を行っても, 伝搬モードの光が接合部まで達してしまうため, 光電流信号が検出されてしまう. 他方, 開口径が 100 nm の場合, スペクトルの広がりが大きくなるので, 試料との相互作用下においても有意なエバネッセントモードが存在する. プローブが接合部の真上にきたときに, このモードが強い信号を発生させるので, 高い分解能が得られる.

このように微小開口が生成する特異なモードと結晶中の電子との相互作用は, 近接場特有の興味深い現象である. 上述したような振舞いの他にも, 大きな波数成分を利用した, 2次元電子系での波束発生³⁹⁾やファーフールドでは禁制された



第8図
エバネッセントモードによる光電流信号の発生.



第9図 散乱型近接場光学顕微鏡の原理.

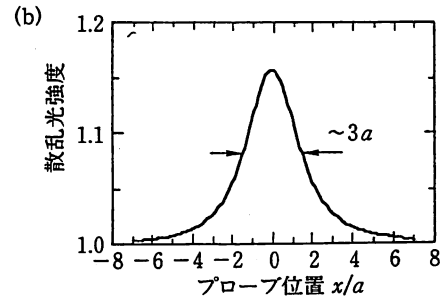
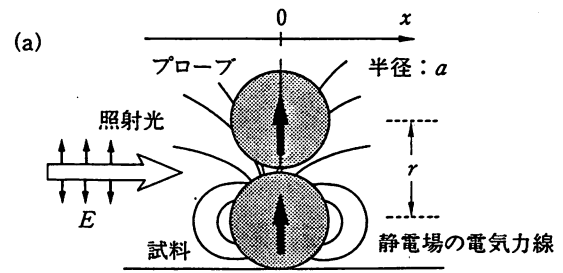
光学遷移の許容化なども検討されている.

§5 散乱型近接場光学顕微鏡

波長よりも十分小さなサイズの2つの微小物体(PとS)に外部から光を照射し,そこからの散乱光を検出する状況を考える(第9図).ここでPとSの距離もまた波長よりも十分に近いとする.散乱光の強度は,2物体間の微小な距離(相対的な位置関係)の変化によって大きく変わる.その理由は以下のように説明される.Pの感じる電場は,外部から照射した光電場そのものと,それによってSに誘起された分極が生成する電場の和となっている.P,S間の距離が波長よりも短い場合,PはSの生成する電場のうち,特にSの中心からの距離の-3乗で減衰する静電場成分を感じる(Pに誘起された分極がSに及ぼす作用についても同様のことがいえる).この強い距離依存性のために,2物体間の距離がわずかに(物体のサイズ程度)変化するだけでも,PとSに誘起される正味の分極の大きさが変わり,検出される散乱光強度にそれが反映される.ここで2物体を微小試料とプローブに見立てると,一般に散乱型と呼ばれる近接場光学顕微鏡の動作として応用できることがわかる^{1,40)}.この方法においても,開口型と同様に分解能は波長には一切依存していないことが最大の特徴である.

5-1 分解能と基本動作

上で述べたように,2つの物体に誘起された分極がお互いに及ぼし合う(強い距離依存性をもつ)静電的な相互作用が散乱型近接場顕微鏡の基本原



第10図 散乱型近接場顕微鏡の分解能.

理である.ここでは,簡単な状況を仮定し,その分解能について議論する.

試料(添字s)とプローブ(添字p)を半径 a_s, a_p をもつ球と仮定する.それぞれの球は,体積に比例した分極率,

$$\alpha_i = g_i a_i^3, \quad g_i = (\epsilon_i - 1) / (\epsilon_i + 2) \quad (i = s, p) \quad (1)$$

をもつ.この2つの球が十分に接近した状況で(中心間の距離 $r \ll \lambda$),第10図(a)示すような配置で光(電場 E)を照射する.試料とプローブはお互いの分極が影響し合い,実効的な分極率の変化,

$$\Delta\alpha = 2\alpha_s \alpha_p / r^3 \quad (2)$$

が生じる.2球からの散乱光強度(信号)は,

$$(\alpha_s + \alpha_p + 2\Delta\alpha)^2 |E|^2 \quad (3)$$

によって計算される.ここでプローブを試料に対して走査したときに期待される信号のプロファイルを第10図(b)に示す.プローブが試料のごく近傍まできたとき(試料のサイズ程度)に初めて,大きな信号の変化が観察されている(プロットの縦軸に注意.プローブと試料が十分離れているときに比べ,すれすれまで近づいたときに約15%の散乱光強度の増加が期待されると解釈する).言い換えれば,散乱型の近接場顕微鏡は,プローブ球のサイズ程度の分解能をもつことがわかる.ただし,観測対象のサイズに比べて必要以上に小さな

プローブを用いてしまうと、コントラスト(何%の散乱光強度の増加が得られるか)の大幅な低下を招いてしまう。理想的には、試料とプローブサイズが同程度であることが望ましいことは、簡単に示すことができる。

実際の散乱型近接場顕微鏡では、プローブ球単体を自由に操ることはできないので、鋭く尖った針の先端部を利用することになる。光ファイバーを化学エッチングによって先鋭化すると、先端曲率半径を数 nm まで小さくすることが可能である。その他にも、感度(散乱効率)を高めるために、金属の針⁴¹⁾(たとえば STM の探針そのもの)やファイバープローブ全体を金属膜で覆った針⁴²⁾を利用することも行われている。

5-2 今後の発展の方向

微小な金属は一般の誘電体とは異なり、散乱型近接場顕微鏡において非常に有利な特徴をもつ。金属中の自由電子のダイナミクスがこのような特性を生み出すのである。たとえば鋭く尖った針の先端部では、自由電子の集中が起こるので、局所的に強い電場を発生させることが可能である。また、孤立した金属微小球の場合も考えてみる。分極率を表わす式(1)の中で、物質の誘電率に依存する因子 g に着目する。一般に金属は負の誘電率をもつため、 g の値は常に 1 以上であり、誘電体と比べ(ガラスの場合、 g は約 0.3 である)、大きな分極を生成することができる。特に誘電率 ϵ が -2 となるような波長の光を照射した場合、 g は共鳴的に大きな値に達し、高い散乱効率をもたらすことになる(共鳴が起こる ϵ の値は形状に強く依存する。 -2 はあくまでも球形の場合である)。

このような金属特有の増大効果は、「表面増強分光法」としてその威力がひろく実証されている^{43,44)}。単一の金属微粒子による強いラマン信号の観測などもその一例である。これまでの一連の成果をヒントに、金属プローブを走査可能な「信号増幅器」として利用することができれば、高分解能(プローブ先端の大きさ程度)と高感度をかねそなえた近接場顕微鏡としての応用が期待される。さらに、このような観察手段にとどまらず、分極の制御という観点からの展開も今後重要であ

ろう。たとえば、大きな非線形性をもつ微小物質と結合させることにより、双安定性などの新しい光機能を発現させることもできると考えられる。

§6 まとめ

従来の光学顕微鏡が長らく抱えてきた回折限界という障害を打破すべく、近接場光学顕微鏡は登場した。その原理はシンプルであり、金属のスクリーンに小さな穴をあけ、そこから漏れ出した光を観察したい試料に照射するというものである。本稿ではまず、この「微小光源」の特徴を理解していただくために、微小開口の光学について簡単に述べた。開口の出口近傍で非常に強い光電場が発生していることを示し、このような電場が伝搬モードとエバネッセント(非伝搬)モードから成り立っていることに言及した。続いて、このような微小開口を用いた近接場顕微鏡の固体分光への応用例をいくつか紹介した。その多くは、従来のレンズ系による顕微分光の延長という色合いが濃い研究であったといえる。分光測定においては高い感度を得ることを優先するため、分解能としては 100~200 nm が現実的な値であった。しかし、表面形状との対応付けが可能であるという付加価値もあり、これまでにない分光技術として成功を収めてきたと思われる。今後、微小開口型の展開としては、次の2つが考えられよう。(1) プローブの高感度化をすすめ、分解能の向上(より小さな開口による観察)をはかる。(2) エバネッセントモードの特徴を活かした固体中の電子励起を行う。(2)に関連して、§4-2では高い屈折率をもつ半導体試料との相互作用によるモード変換について触れた。

§5では、散乱型の近接場顕微鏡について紹介した。試料とプローブに誘起された分極が及ぼし合う、静電的な相互作用がその動作原理である。分解能に関してはプローブ先端のサイズ自体がそれを決定するので、現実的には開口型よりもすぐれている。ただし感度とコントラストをいかに向上させるかがこれからの大きな課題である。感度の問題を克服する最有力の手段として、金属による高い散乱効率を利用することを挙げた。高感度

化という枠にとどまらず、微小な「光機能材料」とこのような「増幅媒質」との結合は、極微デバイスにとって大変魅力的である。近接場顕微鏡による観察を通して、そこで繰り広げられる相互作用に対して新たな知見が得られれば、それは願ってもないことである。散乱型をベースとした研究が今後、観察という領域を越え、材料探索・設計のスタンダードな手法としての地位を築いていくことを期待する。

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近接場光学とその光メモリへの応用

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Near Field Optics and Its Application to Optical Memory

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あらまし 近接場光学の原理について概説し、この応用の一つとして1 Tbit/inch²の記録密度が期待される高密度光メモリの問題点を列挙した。更に筆者らの試みとして、平面プローブアレーによる記録再生、特に再生の場合のトラッキング回避の可能性について紹介し、プローブアレーの開発状況を紹介した。

キーワード 光メモリ、近接場光学、ファイバ、トラッキング

1. まえがき

自由空間を伝搬する光の特性を表す物理量としてパワー、波長、周波数、偏光等があり、これらは制御可能である。この制御可能性を利用して光は通信、情報処理等へ広く応用されてきた。しかし、波動としての光のもつ性質の一つである回折のために光を波長以下の寸法の空間に閉じ込めることはできなかった。従って光デバイスの寸法は光波長以下にはならず、光デバイスの微小化には限界があった。

回折限界を超えた微小化をするための有力な方法として近接場光学がある。この学術分野が扱う近接場光とは微小物質表面に発生し、そのエネルギーが物質表面に局在し、非伝搬の光である。従ってこの光を発生させるには光の波長より小さな微小物質を加工製作する必要があるので、近接場光学にはナノ加工技術が不可欠である。

歴史的には1928年に高倍率の光学顕微鏡の可能性が示唆されたが[1]、この時点では近接場光の局在性、ナノ加工の必要性等、本質的な議論はなされていない。その後、ナノ加工技術が可能となる1980年代になってようやく実験結果が報告されるに至り[2]、[3]、近接場光学が創始された。

1990年代に入って近接場光学は計測から加工へと進

んでいる。更には原子操作[4]および原子堆積[5]へと発展しており、ナノ/アトムホトニクスとでも称すべき新分野が開拓されつつある。これらの現状を踏まえ、本論文では特に加工の応用としての光メモリの現状と将来について解説する。なお、近接場光学において必須素子であるプローブおよびその作成法、更にこれを用いた計測等についての研究開発の現状は文献[3]、[6]、[7]等を参照されたい。

2. 近接場光とその特性

近接場光とは物質に光を入射したときに発生する誘起分極のうち、表面付近で互いに隣接する誘起分極間の近距離電磁相互作用を媒介する場である。それは表面プラズモン、表面ポラリトン等と同様に表面素励起の一種であると考えられている[8]。これは物質表面近傍に局在する非伝搬光である。すなわち光照射の際、物質の寸法、形状によらず近接場光が伝搬光と同時に発生する。特に物質の寸法が入射光の波長より小さいときには近接場光の局在する体積は物質寸法と同程度であることが散乱理論により示されている[9]。但し、近接場光は非伝搬光なので光波長以上の遠方では検出できない。従って図1(a)に示すようにプローブと呼ばれる第2の微小物質を近接場光の場の中に挿入し、近接場光を散乱して伝搬光に変換し、検出する。ここで、プローブの寸法が、近接場光を発生している物質寸法と等しいとき、散乱効率が最大となるので[9]、近接場光の発生と同様、検出にはナノ加工技術によるプローブ製作が不可欠である。

計測のためにはプローブを物質表面に沿って走査し

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ながら、散乱光のパワーを測定してプローブの位置の関数として表示する。これにより近接場光パワーの空間分布、すなわち物質表面の形状が測定でき、物質の形状を観察する顕微鏡の画像が得られる。その分解能はプローブの寸法に依存し、光の波長とは無縁である。従って光波長以下の寸法をもつプローブを用いれば、回折限界を超えた計測が可能となる。

なお、図1(b)に示すように、プローブ表面に近接場光を発生させ、微小物質によりこれを散乱させる検出法も可能である。筆者らは図1(a)をコレクションモード、図1(b)をイルミネーションモードと呼んでいる。なお、図1(a),(b)のどちらの場合も、物質とプローブとにより近接場光は多重散乱される。この局所的な光散乱にかかわる光のエネルギー密度は決して小さくなく、時によっては物質そのものが変形、構造変化する場合もある。むしろこれが近接場光の本質であり、近接場光学は新しいナノ加工技術を提供する。この加工の場合にも回折限界を超えることができるのは上記の計測の場合と同様である。

3. 光メモリへの応用

3.1 光メモリの高密度化に関する現状

従来の光メモリは伝搬光をレンズで集光する方式に基づいており、その記録密度の伸びの平均値は1年ごとに約30%と言われ、例えばDVD-RAMでは1997年現在、約2Gbit/inch²に達している(トラック幅0.74 μ m, 半導体レーザー光源の波長650nm)[6]。しかし近い将来に記録寸法が光の回折限界に到達する。更に記録密度を増加させるために青紫色半導体レーザー等の短波長光源を使うこと、体積ホログラム記録、多値変調記録等が提案されている。これらは既存の概念を組み合わせることで実現する近未来型の技術であり、現状の数倍以上の記録密度が期待される。これらに対し、近接場光学による方法は新しい概念に基づく技術である。

この近接場光学による方法に対して、混乱を招きやすいものに屈折率の大きな材料で作った固浸レンズ(Solid immersion lens; SILと略記)を用いる方法がある[10]。SILは開口数(NA)が大きいので回折による焦点のぼけの寸法が従来のレンズに比べて小さく、従ってより高い記録密度が得られる。このレンズの焦点距離は短いので記録媒体をレンズに近接して置く必要があるため近接場光学技術と呼ばれている場合もあるが、これは伝搬光を使っていることには違いなく、従来の回折限界内での技術である。なお、この方法により典型

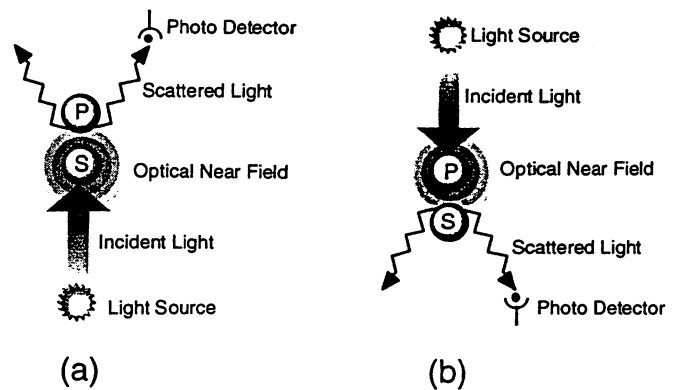


図1 近接場光の発生と検出の原理。(a)微小試料(S)の表面に近接場光を発生させ、それを微小プローブ(P)で散乱させて検出。(b)微小プローブ(P)の表面に近接場光を発生させ、それを微小試料(S)で散乱させて検出。

Fig.1 Principle of generation and detection of optical near field. (a) Generating the optical near field on the surface of a sample(S), and scattering by the probe (P) for detection. (b) Generating the optical near field on the surface of a probe(P), and scattering by the sample (S) for detection.

的な光磁気記録媒体であるTbFeCoに直径約350nmのビットの記録再生が行われている[11]。しかし従来の回折限界の枠組みの中で動作する方法はSIL用材料の屈折率増加に伴い、開口数上限が自ずと制限される上、焦点深度も厳しくなることがわかっており、早晚限界に達する。

近接場光学による手法はこれらの回折限界を超え、記録密度の原理的上限はプローブ先端の寸法の値によって決まり、1Tbit/inch²に達し得る可能性をもつ。図1(b)に対応するイルミネーションモードを用い、光磁気記録媒体の一つであるPt/Co多層膜に対して直径約60nmの磁区の記録再生が最初に行われた[12]。対応する記録密度は170Gbit/inch²に達する。これは光による局所加熱を利用しているため従来の光記録の分類では熱モード記録と言える。同時期に筆者らはホトクロミック材料の有機超薄膜に近接場光を照射し、トランス異性体からシス異性体へと構造変化を誘起することにより記録するフォトンモード記録を行い[13],[14]、薄膜の透過率変化を計測することにより再生し、長波長の光を照射することにより消去可能であることを示した。得られた1ビットの記録寸法は上記の熱モード記録と同様50nm程度である。この方法の利点は局所加熱が不要なことである。

更に最近では熱モード記録に関し、より低温での記録を可能にするために相変化型記録媒体である

Ge₂Sb₂Te₅を用いた試みがなされている[15], [16]. 最小記録寸法は約80nmであり, 100Gbit/inch²の記録密度に相当する. また, より感度の高いフォトクロミック材料を用いた光子モード記録も進展している[17].

但し, 現状はガラスファイバを先鋭化して作成したプローブ(ファイバプローブと称する)によって記録再生の実験を行い, 高い記録密度が実証されているのみであり, 実用化に向けての方向が示されているとはいえない。

3.2 問題点の抽出

民生品としての光メモリおよびその周辺機器は廉価性, 信頼性等に関する問題を解決した後に実現されることは言うまでもない. 以下ではこれらの問題点を明らかにし, 解決するための手掛りについて概説する. なお, 以下の議論は筆者らのグループでの検討結果であり, 必ずしも絶対的かつ一般的な要件ではないが, 将来の一つの方向を示唆するものと考えられる.

近接場光学による光メモリを実用化するための問題はソフトウェアとハードウェアにかかわっており, かつこれらは相互に関連している. これらの問題点のうち的主要なものを以下に列挙する.

[A] ソフトウェアに関して: (1)ユーザに提供する情報(個人の医療情報, 図書館の文献情報, 気象情報, 動画像等)の設定. これによりROM,消去可能, 等のメモリ形態が決まる. これがハードウェアの開発に影響する. (2)記録媒体表面のナノメータ寸法の凹凸等の形状特異点を読み飛ばす再生方式.

次にハードウェアに関し, 3部に分けて記す.

[B] システムに関して: (1)記録媒体の形状(CDのような円盤型か磁気テープのような線型か). これは記録情報の種類に依存する. (2)再生ヘッドの走査. (3)プローブと記録媒体との間の距離制御. (4)再生ヘッドのトラッキング制御. (5)記録媒体の脱着. これにより光メモリの可搬性が可能になる. (6)再生時の微弱光の高速検出法.

[C] デバイスに関して: (1)高効率および必要な速度で走査可能なプローブ. (2)光源, プローブ, 光検出器をつなぐインタフェース用デバイス. (3)プローブ走査用のフライングヘッド, アクチュエータ等.

[D] 記録媒体に関して: (1)ナノメータ寸法の微粒子と近接場光との間の高感度な相互作用の機構の探索と, そのための材料. (2)小さな粒径, 磁区をもつ材料. (3)ナノメータ領域での物理量, 熱力学的概念の検討. すなわち統計力学的な物理量である熱, 温度の概

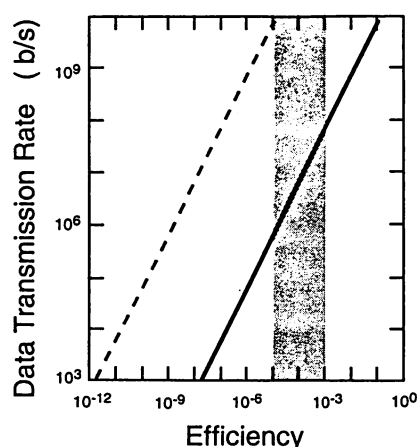


図2 プローブの近接場光発生効率[≡(発生する近接場光パワー)/(プローブへの入射光パワー)]とデータ伝送速度との間の関係の計算結果. 網掛けを施した領域は現在のファイバプローブで実現し得る近接場光発生効率の値. 実線は受信光のショット雑音によるビット誤り率を 1×10^{-12} に保つためのデータ伝送速度. 破線は 100×100 個のプローブアレイを用いる場合.

Fig.2 Dependence of data transmission rate on the efficiency of generating the optical near field. The efficiency is defined as the ratio of the generated optical near field power to the incident light power to the probe. Meshed area represents the value of the efficiency realized by the existing fiber probe. A solid line represents the data transmission rate to maintain the shot noise-limited bit error rate as low as 1×10^{-12} . A broken line is by using the probe array with 100×100 elements.

念, およびその拡散の理論モデルの適用可能性の検討. (4)ナノメータ膜厚の保護膜.

3.3 問題の解決に向けて

前節で列挙した問題点のいくつかを解決するための筆者らの試みについて紹介する(前節の問題[B](2)~(4), [C]に関連).

[A] 再生ヘッドの構成について: まず再生の際のデータ伝送速度の限界を推定する. 光源からの光をプローブに入射し, 先端に発生した近接場光を記録媒体表面に照射し, 散乱光を受信して信号再生する場合を考える. プローブの近接場光発生効率[≡(発生する近接場光パワー)/(プローブへの入射光パワー)]とデータ伝送速度との間の関係の計算結果を図2の実線で示す[18]. この場合, 計算の仮定は次のとおりである. (1)再生方式はPCM方式. (2)ビット誤り率を 1×10^{-12} に保つ[19]. (3)誤り率を与えるものは受信光のショット雑音. (4)プローブへの入射光パワーは金属膜蒸着したファイバプローブ[20], [21]の破損しきい値以下とし, 1mWの値を使用. (5)光源の波長は600nm. (6)記録媒体からの散乱光のコントラスト(すなわちビット1からの

反射光量とビット0の反射光量との比)の値は0.1 (これは筆者らのフォトンモードの記録実験の結果をもとにしている[13], [14]. 熱モード記録の場合にもこの値を採用できる).

筆者らがガラスファイバを化学エッチングにより先鋭化し, 金属膜を蒸着して作成したプローブ[20], [21]では近接場光発生効率の値は $1 \times 10^{-5} \sim 1 \times 10^{-3}$ (図2中の網掛けの領域)であるので, この図の実線より約50Mbit/sに達するデータ伝送速度が期待できることがわかる. $1 \text{Tbit}/\text{inch}^2$ に達する高密度記録に対応させて, 更にこの伝搬速度の値を増加させ, 例えば1Gbit/sの値を実現するにはプローブの改良が必要となる. 改良法の第1として, より近接場光発生効率の高いファイバプローブを開発することであるが, その場合にもこのプローブの走査可能性が問題である. すなわち, データ伝送速度 r , 記録ビットの間隔 s , 走査速度 v との間の関係 $v=sr$ に目標とする数値($r=1 \text{Gbit}/\text{s}$, $s=25 \text{nm}$)を代入すると $v=25 \text{m}/\text{s}$ となる. しかしPZT等の従来のアクチュエータを用いて記録媒体とプローブとの間の距離を数十nm以下程度に保ちながらこの速度で走査することは容易ではない. そこで第2の改良法として, 2次元アレイ状に配列した複数個のプローブによる再生が有望である. 2次元アレイ中のプローブ数が n のときアレイの走査速度は $v=sr/n$ となる. ここで100個 \times 100個のプローブを用いると($n=1 \times 10^4$), $v=2.5 \text{mm}/\text{s}$ となり, これは従来技術で実現可能な低速値となる.

このような2次元アレイを実現するために筆者らは図3に示すように半導体平面プロセス技術を用いてシリコン基板上に2次元微小開口列を作り, 上面から照射した光によって微小開口に近接場光を発生させるデバイスを提案している. このデバイスは次の利点をもつ. (1)底面が平たんなので記録媒体との間の距離をアレイ全体にわたって一定に保つことができる. (2)コンタクト型のハードディスクの走査と同様, 記録媒体に塗布した潤滑剤薄膜[22]を介して滑らせ, 記録媒体との距離を一定に保ったまま上記の走査速度を実現することが可能である. (3)下記のトラッキング不要な走査が可能である. (4)スラブ導波路および光検出器アレイ等との集積化が可能である.

[B] プローブアレイによる記録: 記録媒体の形状としてはディスク状平面とする. 図4(a)に示すようにプローブアレイの上面に光を入射し, 開口に近接場光を発生させる. プローブアレイと記録媒体との間に潤滑剤を塗布し, 表面張力で近接する程度(約10nm)まで両

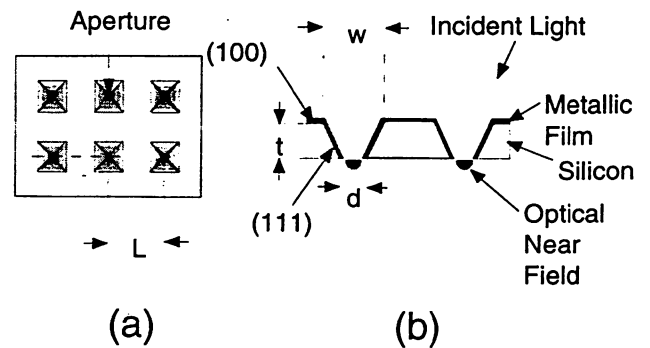


図3 シリコン基板上に形成された2次元微小開口からなるプローブアレイの形状. (a)上面図. (b)断面図.

Fig.3 Profile of the probe array with two dimensional apertures fabricated on a silicon substrate. (a) Top view. (b) Cross-sectional view.

者を近づけて掃引する. 入射光を空間変調器を通した後に入射光強度を記録情報に応じて独立に変調して記録することができる. ここで, 光源, 空間変調器, プローブアレイ上面までの光の伝送形態は空中伝搬である. これにより再生ヘッドとしてのプローブアレイの低重量化を図る. ここで, アドレス間の干渉を防ぐために, プローブアレイ中の隣り合う開口間の距離 L は光波長以上の必要があるが, これはアレイの製作用のホトマスクの準備の際に特別な微細加工技術を必要としないので有利である.

[C] 再生時のトラッキングの回避: 従来の光メモリでは再生ヘッドが記録ビットの上を走査するように位置決め制御, すなわちトラッキング機能が必要であった. しかし上記のプローブアレイを用いるとそれが不要になる[18].

すなわち図4(b)に示すように, 記録ビット列に対してプローブアレイの列を角度 θ 傾けて走査する. プローブアレイは矢印の方向に走査する. この際, プローブアレイ中の各開口は間隔 $D(=L \sin \theta)$. L は開口間隔の破線を通して通過する. ここで従来の光学系を用いれば記録媒体面での散乱光は, 開口面の像をレンズ系によってプローブアレイ上空にて光検出器アレイ上に結像して独立に検出することができる. これは開口間隔 L は入射光波長以上の値であることにより可能である. こうして検出した光の強度を時間軸上の電気信号として記憶しておけば, 掃引速度が既知であるため, 記録媒体上の任意の位置からの散乱光強度の情報を通過時間から逆算して求めることができる. これにより開口間隔 L より狭い間隔で並んだ記録ピットを観測できる.

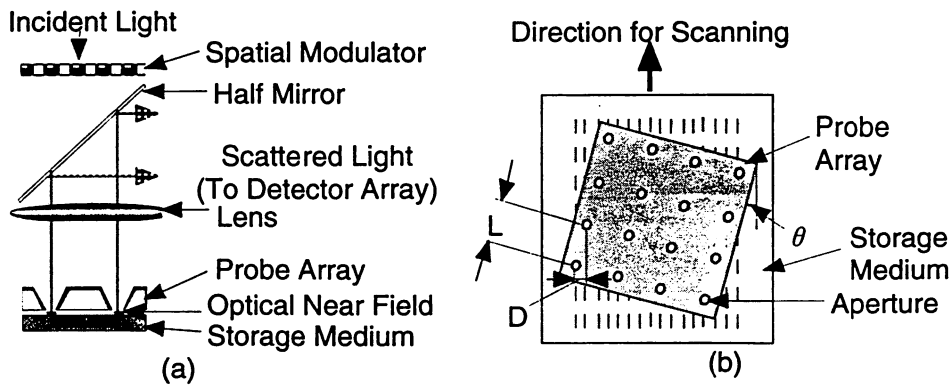


図4 記録、再生の説明。(a)空間的に変調された空中伝搬光およびプローブアレイを用いた記録。この図には記録媒体からの散乱光をプローブアレイ、半透鏡を用いて光検出器アレイへ伝送し再生するための系も描いている。(b)記録媒体に対しプローブアレイを角度 θ 傾けて走査する再生。破線列はプローブアレイの走査時の各開口の軌跡。

Fig.4 Schematic explanation of storage and read out. (a) Storage by using a spatially modulated propagating light and a probe array. (b) Read out by scanning a probe array with the tilted angle of θ . Broken lines represent the trajectory of each aperture.

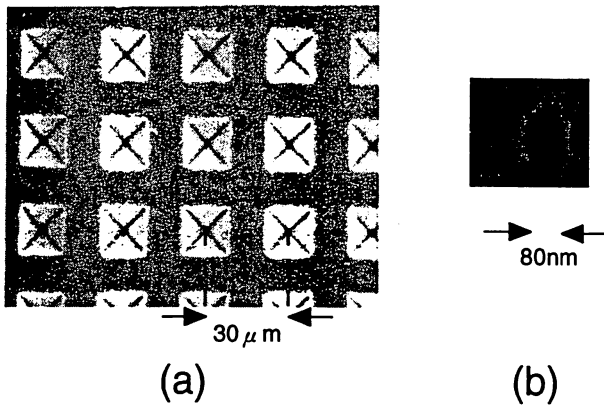


図5 製作したプローブアレイの走査電子顕微鏡写真。(a)上面図。(b)最小開口寸法(1辺80nm)をもつプローブアレイの一開口の下面拡大図。

Fig.5 Scanning electron micrograph of a fabricated probe array. (a) Top view. (b) Bottom view of the smallest aperture.

この方法によりプローブアレイの走査方向には連続的に検出可能である。ここでプローブアレイの各開口の寸法 d は記録媒体のメモリ寸法 d_m 以下なので、プローブアレイにより $1/d_m$ によって決まる空間フーリエ周波数成分以下の散乱光強度を総て観測でき、従って記録媒体からの散乱光強度の走査方向の空間分布は完全に復活できる。また走査方向と横方向に対しては離散的であるが、 $D \leq d_m/2$ であれば標本化定理によりこれも完全に復活可能である。なお、同様の提案は面発光レーザーアレイを再生ヘッドとして用いる方法でもなされている[23]。ここで[A]のように記録ビットの間隔 $s=25\text{nm}$ 、開口数 $n=100\text{個} \times 100\text{個}$ とするとプローブア

レイ寸法は $250\mu\text{m} \times 250\mu\text{m}$ 程度の小型であり、また走査の際の機械的位置決め精度を数十 μm 以内にする事は従来の安価なシステムでも可能である。従って横方向の位置座標を補助記憶装置に蓄えておけば、記録媒体上の各ビットに対するプローブアレイのトラッキング制御機構がなくとも再度機械的に場所を選択して検索することができる。更に、走査方向およびその横方向ともに誤り訂正符号を付加し、またインタリーブも同時に行えばプローブアレイや記録媒体の欠損等にも対処できると考えられる。

[D] プローブアレイの製作：筆者らが提案するプローブアレイはシリコン基板の異方性エッチングにより製作する。すなわちシリコン結晶の(100)軸方向と(111)軸方向とのエッチング速度の違いを利用して図3に示す凹部を形成し、その底部を開口とする。この場合凹部のテーパ角度は(111)軸の方向によって決まり70.5度となる。また開口寸法 d はシリコン厚 t 、凹部の上面寸法 w とにより $d=w-\sqrt{2}t$ となる。従って、 d は w および t によってその大きさと精度が決まる。 w はリソグラフィの精度に依存する。一方、 t の均一性と精度を確保するためには、 SiO_2 層が埋め込まれた SOI (Silicon on Insulator) 基板の利用が有効である。埋込み SiO_2 層を裏面からのエッチングの停止層として利用して表面 Si 層のメンブレンを残し、ここに凹部形成の窓開けパターンを形成する。そして開口形成後に凹部の斜面に金属膜を蒸着し、光を遮断する。あるいは蒸着は基板の裏面に行ってもよい。なお、最終的な開口寸法はこの蒸着膜厚に応じて絞られる。この製作法の利点は、シリ

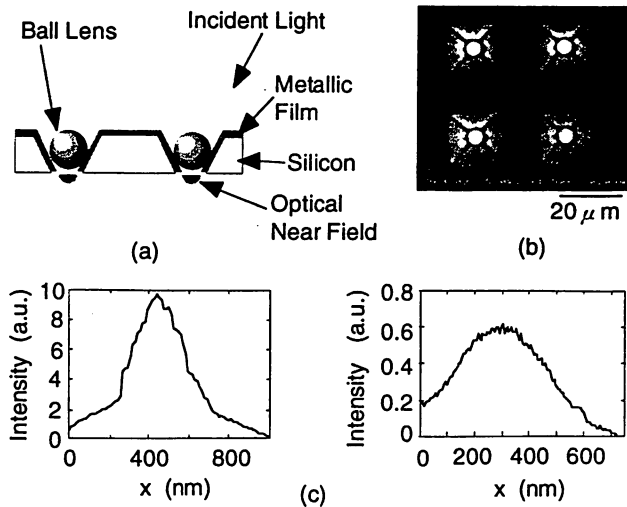


図6 ガラス球レンズ付きプローブアレーとその性能。(a)ガラス球レンズを凹部に設置したプローブアレーの断面形状。(b)作成したガラス球レンズ付きプローブアレーの走査電子顕微鏡写真。(c)プローブアレー中の開口の一つに発生した近接場光のパワーの空間分布の測定結果。これは開口面中心を通る断面分布を表す。左図はガラス球レンズ付き、右図はなしの場合。

Fig.6 A probe array with ball lenses and its near field optical performance. (a) Cross sectional profile of the probe array with ball lenses. (b) Scanning electron micrograph of a fabricated probe array with ball lenses. (c) Measured cross-sectional spatial profile of an optical near field intensity generated on an aperture with (left) and without (right) the ball lens.

コンのプレーナ技術で確立されたリソグラフィ技術のみを用いて、リソグラフィの解像限界を超えた微細な開口構造を均一かつ効率良く一括形成できることである。トラッキングの回避のためには各開口間の距離 L の精度が高いことが望まれるが、 L は入射光波長以上の値であり、従来のホトマスク技術で実現可能と考えられる。開口寸法のばらつきは各記録ビットからの散乱光強度のばらつきをもたらし、受信感度に影響を及ぼす。しかしこれは信号の時系列化の後に弁別しきい値を設けることにより回避できる。

実際に製作したプローブアレーの走査電子顕微鏡写真の例を図5に示す[24]。この場合、最適な製作条件を見出す目的も兼ね、 $L=30\mu\text{m}$ として製作した。また、蒸着する金薄膜の粒径により開口が埋まる場合もあるので、ここでは1辺が約300nmの開口となるように設計した。開口形成後は厚さ約50nmの金薄膜を蒸着した。1辺が約200nmの正形状の開口が再現性良く形成されたが、1辺の最小値として80nmのプローブアレーも実現している。

得られたプローブアレーの近接場光発生状況を調べ

るため、筆者らの開発した近接場光学顕微鏡で開口面上の近接場光パワーの空間分布を測定したところ、単峰性分布が得られ、良好な近接場光発生が確認された。また発生した近接場光の単位断面積当りのパワー密度はプローブアレー上面、すなわち光の入射面におけるパワー密度とほぼ等しいことが確認された。このことから筆者らの開発したプローブ[21]と同等以上の近接場光発生効率を実現していることがわかった。更に発生効率を増加させるため図6(a)に示すようにガラス球レンズを凹部に設置し、開口部に入射光が集中する構造を有するプローブアレーを製作した[18],[25]。図6(b)にその走査電子顕微鏡写真を示す。更に上記と同じ方法で近接場光パワーの空間分布を測定した結果を図6(c)に示す。この図においてガラス球レンズを使用しない場合の結果の縦軸との比較からガラス球レンズ使用によりパワーが16倍増加しており、このことから高効率の記録、高データ伝送速度の再生が期待されることがわかった。

4. むすび

本論文ではナノ加工の新技术としての近接場光学の原理と実験の現状について概説した。この応用の一つとして1Tbit/inch²の記録密度が期待される高密度光メモリの問題点を列挙した。更に筆者らの試みとして、平面プローブアレーによる記録再生、特に再生の場合のトラッキング回避の可能性について紹介し、プローブアレーの開発状況について報告した。

現在、国内外の研究成果をみると、近接場光学技術による光メモリの原理確認は既に終わったと言える。今後はいかに廉価で信頼性の高い技術を開発するかの段階に入ると考えられる。

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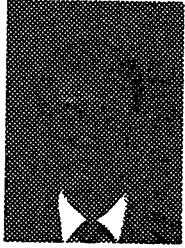


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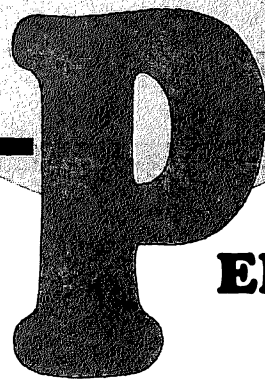


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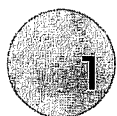
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小さな光でナノ、 原子を測る、加工する、操る

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1 なにを行うか？

21世紀は光の時代といわれて久しい。それは光を使えば通信、情報処理、加工、などいろいろなことが可能で、高度情報化、高度福祉社会のニーズに応え得ると期待されているからである。このような広い応用可能性は光の持つ性質、すなわち「真っ直ぐに速く進む波であること」に起因する。しかし、この性質のために光が不得意なことがある。それは小さい寸法の空間に光のエネルギーを集中させようとしても、光の波長（光の波が繰り返して振動するときの1単位分の長さ）よりも小さくならないことである。光の波長は約 $0.6\mu\text{m}$ （赤色光の場合： $1\mu\text{m}$ は $1/1000\text{mm}$ ）なので、これより小さい寸法の空間中には光のエネルギーは集中しないことになる。これは光の持っている回折という性質（すなわち狭いところに閉じ込めようとするとかえって広がってしまう性質）に起因する。

この回折の性質のために、光のエネルギーを使って約 $0.6\mu\text{m}$ 以下の寸法の微細加工をする事は不可能である。これに対し、電子ビー

ムなどを使って $0.1\mu\text{m}$ 以下、さらには原子一個のレベルの微細加工ができるようになりつつある現在、先端的と考えられてきた光利用技術には微細加工が不得意という大きな問題があった。しかし、21世紀を目前にした現在、光を使ったクリーンでソフトな微細加工は産業界をはじめとし、多くの人々の願いである。神奈川科学技術アカデミーの「フォトン制御」プロジェクトはこの願いをかなえることをめざしている。すなわち光（これを「フォトン」と呼ぶ場合がある）の状態を制御し、光の波長よりも小さい寸法にエネルギーが集中する光を作り、それを応用することを研究課題としている。



2 どのように作るか？

図1 (a) に示すように半径 a （これは光の波長より小さいとする。）の球に光を照射すると、殆どの光はそのまま通過してしまう。しかし光の性質を注意深く調べてみるとそのとき同時に球の表面にエネルギーが集中し、遠くへは飛ばずに表面にまつわりついた光

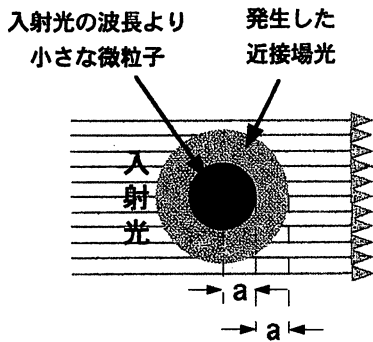


図 1 (a)
半径 a の微粒子の表面に発生する近接場光。

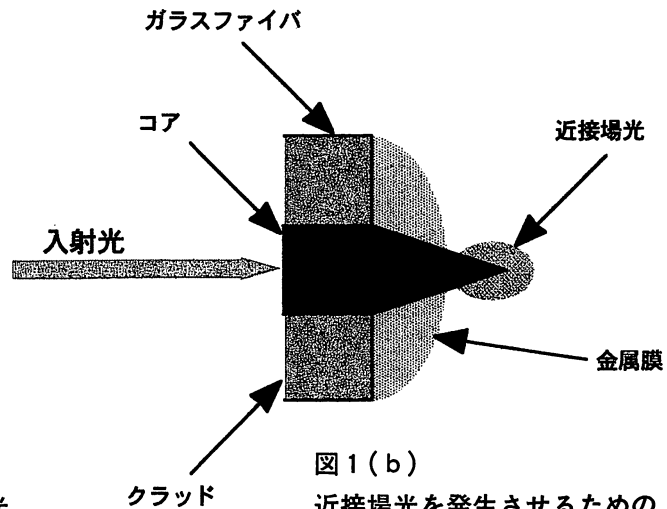


図 1 (b)
近接場光を発生させるための
ガラスファイバ製のプロブ。

(これは近接場光と呼ばれる。)が発生することがわかった。この光が球表面にまつわりつく厚みは球の半径 a 程度である。この厚みは光の波長よりずっと小さいので、この近接場光こそが我々の探していた小さな光である。ただしこのような光が存在することは今までだれも気がつかなかつたし、気がついたとしてもこの光は小さいが故にエネルギーが低く、さらにこのような小さな光を発生させるための小さな球を作ることが難しいので、だれもこれを利用することはできなかった。

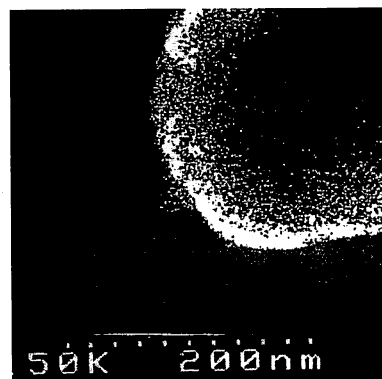
しかし、我々は前節にもあるような時代の

要請に応えるため、まず近接場光を発生させる道具を作ることを試みた。その道具は実際には図 1 (a) のような球ではなく、図 1 (b) に示すガラスファイバの針 (これはプロブと呼ばれる) である。すなわち、ファイバの後端から光を入れ、針の先端に近接場光を発生させる。近接場光の寸法は針先端の曲率半径程度である。我々はプロブを再現よく作るために特別な化学溶解法を発明し、図 2 (a) に示すように先端曲率半径 2 nm 以下、尖り角 20 度程度のプロブを再現性よく作ることに成功した。実際にはガラスファイバを



図 2 (a)
先鋭化ファイバの電子顕微鏡像。クラッドの外径は $8 \mu\text{m}$ 。

図 2 (b)
金属膜を蒸着し、先鋭化コア先端のみが突出しているプロブの電子顕微鏡。突出しているコアの根元の直径は 30 nm。



尖らせた後、図2 (b) に示すように根元に金属膜を塗布して先端の尖った部分のみに近接場光を発生させる。

図2のプローブは世界で最小の先端寸法を持っており、最も小さな測定、加工などを行うのに適している。我々はさらに進んだ化学溶解法を工夫して、図2のプローブに比べ100倍程度大きなエネルギーの近接場光を発生するものも製作することができた。以上のように近接場光を発生させる道具としてのプローブを他に先駆けて作ることができたのは、我が国の光エレクトロニクス産業が世界に誇るガラスファイバ製作技術をうまく利用したことも一因である。最近では尖ったプローブの先に特殊な色素分子などを乗せることも可能になっている。



3 何ができるか？

新しい道具としての小さな光が発生すると今まで原理的に不可能とされてきたことが可能になる。それは「見る」、「分析する」、「加工する」、「操る」などである。これらについて順次説明しよう。

3-1 見る

プローブの先端に小さな近接場光が発生すれば、そのプローブは微小な光源として使える。この光源で微小な物体を照明すると、その物体の像が見える。ここで近接場光の寸法は近接場光を発生させるのに使った光源からの光の波長よりずっと小さいので、波長以下の寸法をもつ微小物体の像が見えることになる。レンズを組み合わせた従来の光源顕微鏡では回折の影響により波長以下の寸法の物体像は見ることができない。これに対し近接場光学顕微鏡は従来の光学顕微鏡では不可能であった微小物体像の観測を実現した。これは近接場光学顕微鏡と呼ばれている。実際には物体を載せた試料台表面に添ってプローブを走査しながら像観察を行う。

顕微鏡が頻繁に利用される分野の一つは生物学であるが、その分野への応用例として図3 (a) にはサルモネラ菌の1本の鞭毛の端部を観察した結果を示す。この直径は30 nm (1 nm は $1/1000\mu\text{m}$) であり、光源の光の波長の約 $1/30$ なので、従来の光学顕微鏡では見えなかったものである。なお、この測定は空気中で行った。一方図3 (b) は水中に

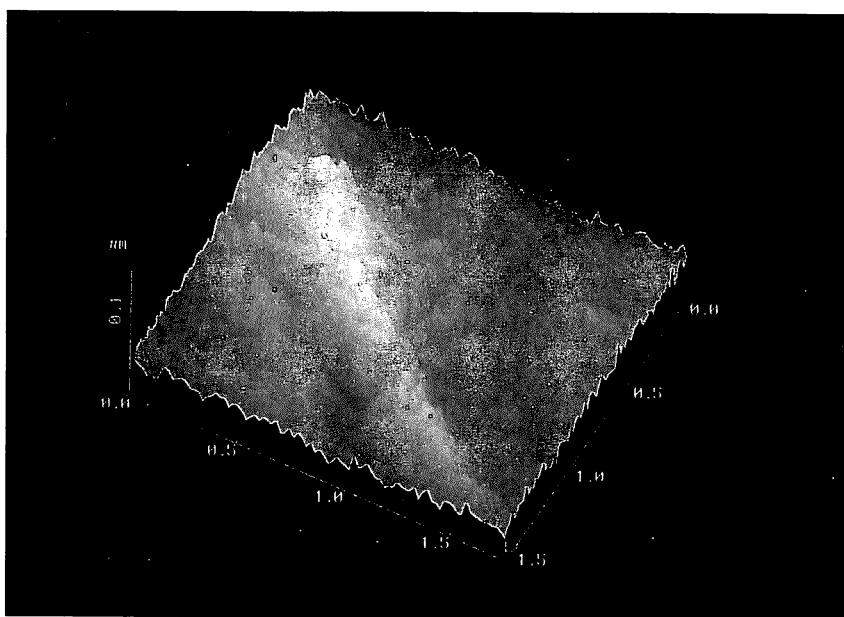


図3 (a)
サルモネラ菌の1本の鞭毛を空気中で観察した結果。鞭毛直径は30 nm

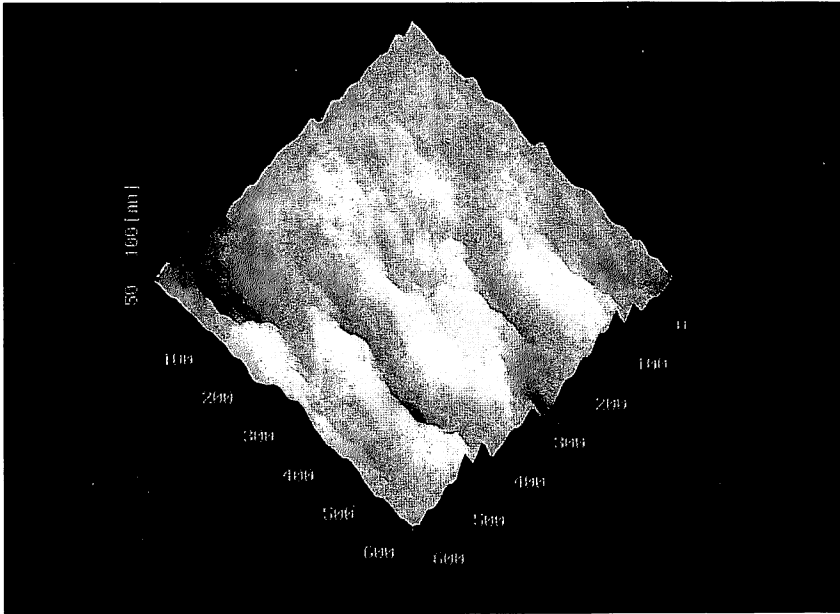


図3(b)

互いに隣接する5本の鞭毛を水中で観察した結果。鞭毛直径は30 nm。

5本の鞭毛が互いに隣接している状態での観察像であり、その直径は図3(a)の場合とほぼ同等である。このように近接場光学顕微鏡は空気中や水中など、特殊な環境で微小な物体像が観測できる特長を持っている。この他、豚の脳神経の端部にある軸管(細い管)中にあるマイクロチューブリンと呼ばれる細い糸状の物体(直径25 nm)の束を軸管の外側から観察したり、細胞中の色素分子からの発光を観察したりすることが可能になっており、生物学分野で活躍する高倍率顕微鏡としての期待が高まっている。

なお、近接場光学顕微鏡に関する研究開発は最近世界的に急激に活発化しており、米国では市販品も現れている。これらと比較して、我々の近接場光学顕微鏡では観察できる最小寸法は0.8 nmに達している。これは光源の光の波長の1/1000に達し、世界のトップデータであるが、このような優れた性能が得られたのも第2節で示したプローブの性能が優れているからである。

3-2 分析する

近接場光学顕微鏡は単に物体の形を見るだけの装置ではない。むしろ本節以降に概説される使用法が他の顕微鏡にはない優れた点である。その第一は物体の構造を分析することである。その一例として我々は化合物半導体の結晶成長の最先端技術を駆使して作製した半導体量子ドットと呼ばれる微小物体から発生する微弱な光を検出し、量子ドット内の電子の様子などを分析した。量子ドットは将来の半導体レーザー用の材料として現在最も注目を集めている物体である。ここではInGaAsの量子ドットを分析した。それは直径30 nm、高さ15 nmの円筒状をしており、これが結晶基板上の面積 $1\mu\text{m}^2$ あたり200個成長している。更にこの上を厚さ180 nmの保護用半導体層が覆っている。微弱な光を発生させるにはプローブ先端の赤色の近接場光を保護層の上からこの量子ドットに照射する。すると近赤外線の微弱な光(この光はフォトルミネッセンスと呼ばれている。)が発生するので、これを再びプローブ先端からファイバに導き、そのエネルギーおよび色を測定する。

従来は赤色の光を量子ドットに照射すると



図4
InGaAs量子ドットから発光するフォトルミネッセンスの強度分布の測定結果。
画像の一边は3 μm 。発光波長は928nm。測定温度は5 K。

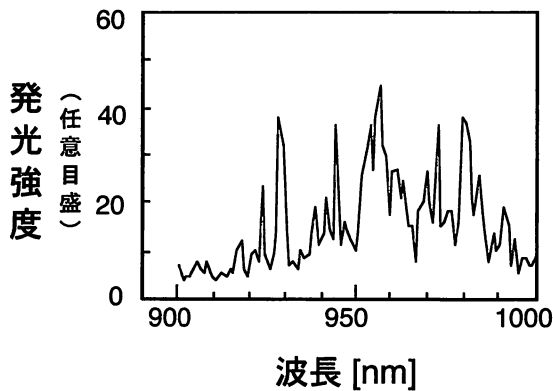


図5
図4に相当する量子ドットから発生するフォトルミネッセンスのスペクトルの測定結果。

き、凸レンズによって光ビームを絞って照射していたが、一度に約100個の量子ドットが照明されてしまい、一個の量子ドットから発生する微弱なフォトルミネッセンスを測定することは不可能であった。しかし、プローブを使えばそれが可能になる。実際の測定では量

子ドットを絶対0度付近の温度まで冷却して測定するので、今までの顕微鏡装置にはない、精密な技術的問題を解決する必要がある。これらを解決した後、各量子ドットから発する微弱なフォトルミネッセンスの分布を測定した結果を図4に示す。また図5の測定結果は10個の量子ドットから発するフォトルミネッセンスの波長がそれぞれ異なり、そのスペクトルが10本のピークに分かれていることを意味している。これらの2つの図は、各量子ドットからフォトルミネッセンスが明瞭に測定されたことを示している。さらに、発光させるための赤色光のエネルギーを変化させたときの発生したフォトルミネッセンスのエネルギーの変化のようすから各量子ドット中の電子エネルギーの状態などを分析できるようになった。

以上のフォトルミネッセンス測定による分析の他、微小物体のラマン分光分析も可能になった。たとえば半導体産業の中心をなすシリコン結晶基板、ニンジンに含まれる β カロチン、などのラマン分光スペクトルが得られている。さらに、半導体デバイスの代表であるCMOS中の電子の拡散のようす、集積回路表面のアルミ配線中の微小な空気の泡の存在、などが分析されている。これらの分析の一部は神奈川科学技術アカデミーの公開利用制度を通じて県内およびその近隣の企業と共同で行った。

3-3 加工する

プローブ先端に発生する微小な近接場光のパワーは小さい。しかし、寸法が小さいので単位断面積あたりのパワー密度は $100\text{W}/\text{cm}^2$ またはそれ以上の大きな値に達する。従ってこの光を物体表面に照射すると物体表面に穴が空いたり、その構造が変化する可能性がある。すなわち微小な加工ができる。加工精度はプローブ先端の寸法で決まり、その値は光源の光の波長よりずっと小さいので、光を

使った微細加工が実現する。

微細加工の応用として我々は超高密度光メモリの記録と再生を行った。フォトクロミック材料の薄膜を作製し、それにプローブ先端からの青色の高パワー密度の近接場光を照射すると、フォトクロミック材料の構造がトランス異性体からシス異性体へと変わった。この構造変化が記録に相当する。シス異性体は透明になるので、次にプローブ先端に低パワー密度の近接場光を発生させ、3-1節と同様の近接場光学顕微鏡により透明部分を測定すると、これにより記録を読み出す（再生する）ことができたことになる。

図6(a)はこのように再生した円形の記録形状の一つを示す。この直径は約50 nmである。この円が1ビットの情報を持つので、この円の面積が小さいほど記録密度が高い。この円の場合、従来の光メモリの記録密度の約100倍に相当する。すなわち1平方インチあたり約50 Gビットの記録密度となる。なおこの円の寸法はプローブの先端曲率半径より大きい。それはまだ十分に精密な装置が準備されていないからである。しかし、それが完成

すれば原理的には円の寸法はプローブの先端曲率半径と同等になり、そのときの記録密度は1平方インチあたり約1 Tビット (=1000 Gビット) に達することが試算されている。これは未だかつて光メモリや磁気メモリが実現しなかった超高密度であり、最近はこの新技術に対する社会的な興味が急激に高まっている。これを実現するために筑波研究学園都市にある通産省工業技術院産業技術融合領域研究所では次世代光基盤研究プロジェクトが発足し、産官学の共同により超高密度光メモリの開発が始まった。

なお、プローブを直線的に走査すると線が記録できる。図6(b)はこのようにして記録した線を、図6(a)と同じ方法で再生した結果である。この場合も図6(a)と同様、プローブを走査することにより任意のパターンを記録することができ、従来の光では不可能であった微細加工の可能性を示唆している。

最近はこの方法をもとに、フォトレジストの露光など、いくつかの微細加工が行われるようになった。

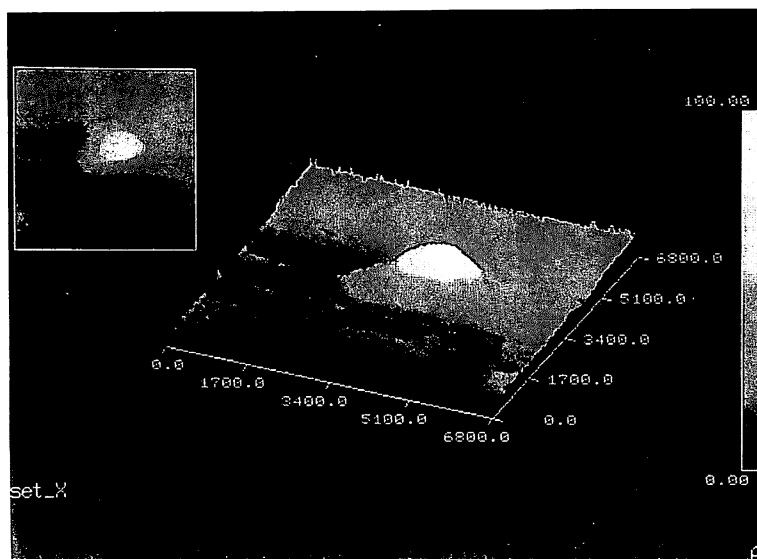


図6(a)

フォトクロミック薄膜に記録された円形の観察結果。直径50 nm。

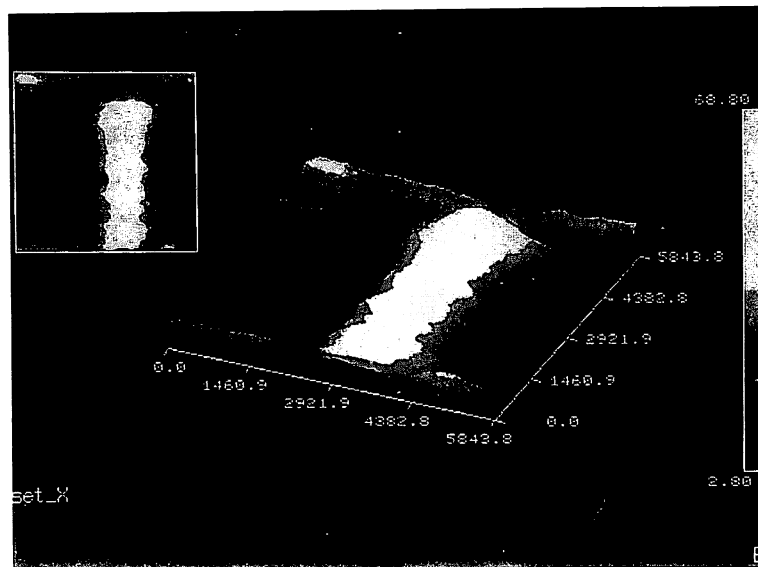


図 6 (b)
同薄膜に記録された線形の観察結果。幅50 nm。

3-4 操る

光はエネルギーを持つと同時に、物体に及ぼす圧力を持っている。微小な寸法の近接場光の場合、そのエネルギーと同様（ただし、3-3節で示したようにエネルギー密度は大きい。）圧力は小さいが微小な物体に対してはこの圧力の効果は無視できない。たとえば物体の基本的構成要素である原子が真空中を飛行する場合、近接場光の圧力でこの原子の飛行方向を曲げるなど、原子の運動を操ることができる。これができるれば、原子を結晶基板の上に導いて、人口的な微小物質を形成することができる。

以上の構想をもとに、我々は先にプローブ先端にしみ出す近接場光の圧力を利用して原子の飛行方向を変えたり、近接場光中に原子をつかまえたりすることを提案した。これまでに予備実験として、プローブではなく、図 7 (a) に示すような中空ファイバの内壁に円筒状の近接場光の薄膜を発生させ、真空中を飛行する原子が中空ファイバ内に飛び込んできたとき、近接場光の薄膜のもつ圧力で原

子を跳ね返しながら原子を中空ファイバ終端まで導くことを試みた。この場合、何種類かの中空ファイバを製作したが、その内径の最小値は300 nmである。このような小さな内径が実現したのは2節でも述べたように我が国のガラスファイバ製作技術の高さを物語っている。なお、実験に用いたファイバの長さは3 cmであるが、これは300 nmの内径と比較すると非常に長い。例えば仮に両者を一万倍すれば、内径3 mm、長さ300mになる。このような細くて長い管の中に微粒子を飛び込ませ、出口まで誘導することは容易ではないことが理解いただけるであろう。

しかし近接場光の薄膜を使うとこれが世界で初めて実現し、図 7 (b) に示すように光のパワーの増加とともに中空ファイバを誘導されて出てきた原子数の増加が観測された。なお、ここで使われた原子はルビジウム (Rb) である。この実験の応用として、中空ファイバを通り抜けた原子を結晶基板に吹き付けて新しい微小な人工物質を形成したり、さらに図 2 に示すようなプローブ先端にしみ出す近接場光に入射させ、その近接場光で原子をピ

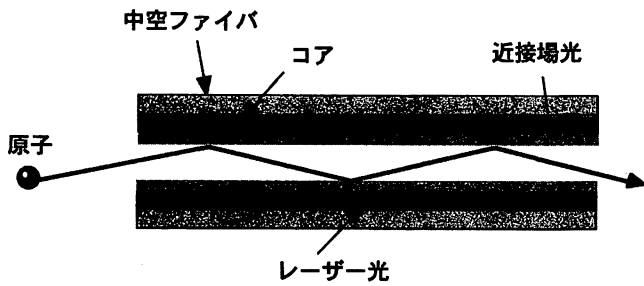


図7(a) 中空ファイバ内壁にしみ出した近接場光による原子の誘導の方法。

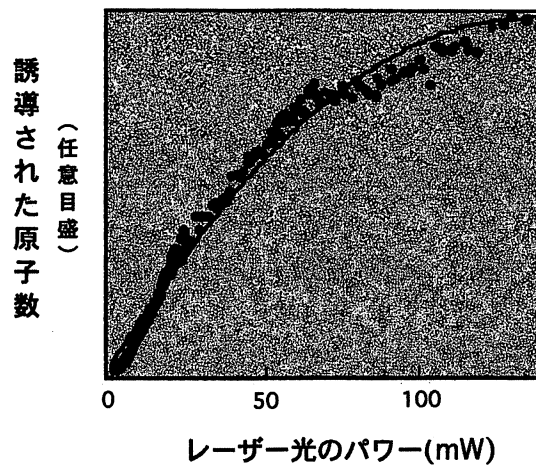


図7(b)
中空ファイバのコアを通るレーザー光パワーと誘導された原子数との関係の測定結果。

ンセットのようにつかんだ後、結晶基板の上に運んで人工物質を形成することが提案されている。一方ではこの中空ファイバによる誘導が原子の種類を選別性を持つことを利用して同位体分離の実験にも成功している。これらの実験は新しい材料科学・工学の幕開けを示唆している。この他にも原子の誘導に関する一連の実験結果には量子力学的な現象も含まれており、現代物理学にインパクトも与えている。

このように原子を操ることは基礎科学から産業的応用まで、未来を開拓する可能性を示している。

4 どこへ進むか？

以上で概説した近接場光の発生とその応用はナノメートル寸法の物質、さらには原子程度の寸法の物質を作りだし、さらにこれらの物質に新しい機能を持たせる可能性を含んでいる。このように新しい光としての近接場光を利用した工学を筆者は各々ナノ・フォトニクス、アトム・フォトニクスと呼んでいるが、さらにこれらを融合するとナノ/アトム・フォトニクスとでも呼ぶべき新しい工学分野、すなわち微小な物質と微小な光を使った通信・情報処理用の光集積回路などが実現するかもしれない。これができれば21世紀は本当の意味で光の時代といえるだろう。

光近接場を用いた原子の誘導

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Atom Guidance with Optical Near Field

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Optical near field realizes the precise control of atomic motion. As the first step, guiding of atoms through micron-sized hollow optical fiber is demonstrated. The two-step laser-photoionization spectroscopy shows the frequency dispersion properties of dipole interaction between atoms and optical near field. The species- and state-selective guidance is applied to in-line spatial separation of two stable Rb isotopes. In addition, the cavity QED (Quantum Electrodynamics) effect inside a dielectric cylinder is observed for the first time. The atom fiber has the faculty of carrying atoms to an arbitrary point over a long distance and then is useful as a novel scheme of optically controlled atomic deposition. The feasibility of creating dot-shaped nano-scale structures is illustrated through measurement of spatial distribution of the guided atom flux. Finally, a method of manipulating atoms beyond the diffraction limit of light waves is presented including sharpened optical fibers and an atomic funnel with optical near field.

Key Words: Optical near field, Atom guidance, Hollow fiber, Dipole force

1. はじめに

これまでの伝搬光を主体とした光技術の原理的限界を乗り越える新しい光科学の創造を目指して、光近接場¹⁾とその応用に関する研究が活発に行われている^{2,3)}。伝搬光の不具合は、顕微技術やナノ領域科学への応用において顕著となる。なぜなら、光波の回折効果によって半波長以下の分解能を得ることが基本的に困難だからである。一方、近接場光は回折の影響を受けず、ナノ寸法の範囲に局在させることが可能である。近年、著しい進歩を遂げたレーザー冷却をはじめとする光を用いた気体原子の熱運動の制御技術⁴⁾をナノ構造作製に応用しようとする研究が衆目を集めている^{5,6)}。光近接場はその局在性から、微小構造物の作製に対しても勝れて威力を発揮するものと期待される。

光近接場を用いた高精度な原子制御技術の開発は、量子力学ないしは量子光学に関わる基礎的物理研究への新しいアプローチ^{3,7-9)}を提供するのみならず、上に述べた回折限界を越えて任意形状のナノ構造を作るといった先端技

術につながる^{2,3,10)}などの点からも、重要な研究課題となっている。本稿では、その端緒として行った中空光ファイバを用いた原子の誘導と基礎物理学や工学への応用の幾つかについて紹介する。また、回折限界を越えて原子を操作するための構想についても簡単に触れる。

2. 中空光ファイバを用いた原子の誘導

2.1 光ポテンシャル

共鳴周波数近傍において、原子は空間的に強度変化する光の場から双極子力¹¹⁾を受ける。その強さは光電場強度の勾配に比例するため、表面から離れるに従って指数関数的に減衰する光近接場は強い双極子力を原子に及ぼす。一方、力の向きは共鳴周波数の前後で反転する。すなわち、光周波数が原子の共鳴周波数よりも低いとき(負離調)は光電場強度の強くなる向きに、高いとき(正離調)は弱くなる向きに作用する。光近接場で原子を制御しようとするとき、このような周波数分散特性を持った双極子力の使用が前提となる。

正離調の場合、光近接場は原子を反射する鏡のように振る舞う^{9,12,13}。原子の共鳴周波数 ω_0 よりも高い周波数 ω を持つ波数 k のレーザー光で強度 $P(r)$ の光近接場を誘起した場合、双極子力のつくる光ポテンシャル $U_{op}(r)$ は距離 r の関数として次のように表される^{9,11,12}。

$$U_{op}(r) = \frac{1}{2} \hbar \Delta \ln \left[1 + \frac{P(r)}{P_0} \frac{\Gamma^2}{4\Delta^2 + \Gamma^2} \right] \quad (1)$$

ここで、 P_0 と Γ はそれぞれ原子の光学遷移の飽和強度と自然幅を表す。また、 $\Delta = \omega - \omega_0 - k_{\parallel} v_z$ は周波数離調で表面に沿う方向の原子の運動に起因するドップラーシフト $k_{\parallel} v_z$ を含む。光ポテンシャルが原子の表面に垂直な方向の運動エネルギーに勝るとき、原子の反射が起こる。

2.2 原子誘導ファイバ

中空光ファイバ¹⁴の内部に光近接場(エバネッセント光)を誘起することによって、原子を運ぶ誘導路をつくることができる^{10,15}。現在までに、中空径 $20\mu\text{m} \sim 0.3\mu\text{m}$ の数種の中空ファイバを用いた実験が行われている。一例として、Fig.1に中空径 $7\mu\text{m}$ の中空ファイバの断面写真を示す。中空領域の周囲には、厚さ $4\mu\text{m}$ の同心円筒形の光伝搬コアがある。このコアにレーザー光を結合させて伝搬モードを励起すると、光近接場が中空領域にしみだす。正離調の場合、内壁から半波長程度の範囲に斥力の光ポテンシャルが生じ、近づいてくる原子を反射しながら誘導する光トンネルを形成する。

Figure 1に示した形状を持つ中空ファイバの光伝搬は、弱導波近似のもと LP_{m1} モードとして表される¹⁵。原子の誘導は、内壁円周に沿って節のない基本モードである LP_{01} モードの励起下で行うことができる。この場合の光近接場の強度 $P(r)$ は、0次の第1種変形ベッセル関数 $I_0(vr)$ を用い中空部の中心からの距離 r の関数として次のように表される¹⁵。

$$P(r) = C \frac{\beta}{\omega} I_0(vr)^2 \quad (2)$$

ここで、 β は伝搬定数を、 $v = (\beta^2 - k^2)^{1/2}$ は減衰定数を表す。また、 C は境界条件と伝搬光パワーの保存則から決まる係数である。式(1)および(2)から $k_{\parallel} = \beta$ として原子誘導の光ポテンシャルを見積もることができるが、後述するように微小中空ファイバ内では共振器QED効果¹⁶による引力の

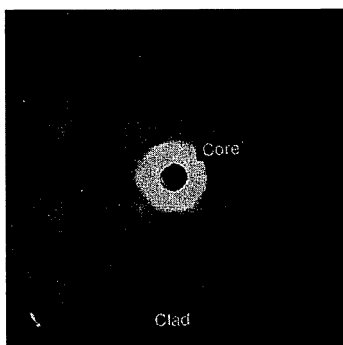


Fig.1 CCD image of the cross section of a 7- μm -hollow fiber with the LP_{01} mode.

共振器ポテンシャルが加わる。

2.3 2段階光イオン化分光実験

正離調光近接場による誘導実験はルビジウム(Rb)原子を用いて行われている^{17,18}。Rb原子の誘導には波長780nmの D_2 線($5S_{1/2} \leftrightarrow 5P_{3/2}$, $P_0 = 1.6\text{mW/cm}^2$, $\Gamma = 2\pi \times 6.1\text{MHz}$)が利用される。

レーザー光イオン化法によって誘導効果を検出する実験配置をFig.2に示す¹⁸。真空中(10^{-6}Pa 以下)でコリメートしたRb原子ビームを同軸上に置いた中空ファイバに導入する。また、チタンサファイアレーザーからの誘導光を中空ファイバに結合し、光近接場を誘起する。この際、中空ファイバの後端面をCCDカメラで観察しながら誘導に適する LP_{01} モードを選択的に励起する。中空ファイバからの透過Rb原子の検出は、 D_2 線に同調した半導体レーザー(LD)と波長476.5nmのアルゴンイオンレーザーを用いた2段階光イオン化を介して行う¹⁸。イオン化された原子は-3kVの負バイアスを印加したチャネル電子増倍管(CEM)に取り込まれ、単位時間あたりのイオン計数として処理される。このような2段階光イオン化では、半導体レーザーの同調周波数によって原子種および量子状態の選択が可能であり、誘導原子の分光を行うことができる。

Figure 3に、中空径 $7\mu\text{m}$ の中空ファイバ内を距離3cmに渡って誘導された基底上準位($5S_{1/2}$, $F = 3$)の ^{85}Rb 原子のドップラー・フリー光イオン化スペクトルを、誘導光の周波数離調の関数として示す。ここで、ドップラー項 βv_z を含まない周波数離調 $\delta = \omega - \omega_0$ は ^{85}Rb 原子の基底上準位($5S_{1/2}$, $F = 3$)を基準にして測られている。双極子力の周波

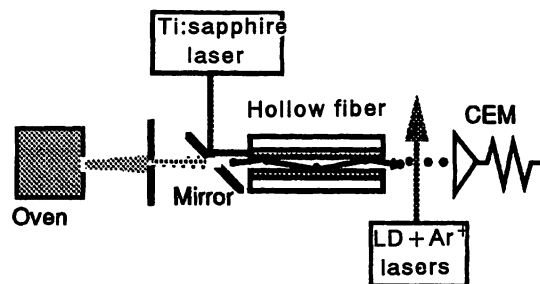


Fig.2 Experimental setup of two-step photoionization spectroscopy with a Rb atomic beam.

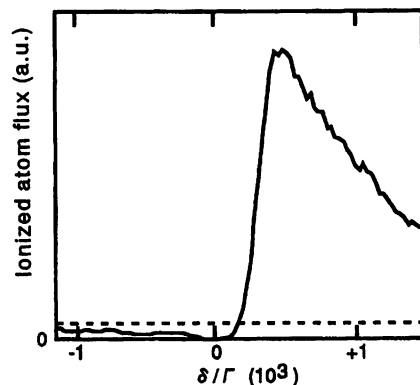


Fig.3 Doppler-free photoionization spectrum on the guided ^{85}Rb atoms in the $5S_{1/2}$, $F = 3$ upper ground state.

数分散性から期待されるように、正離調領域で透過原子束の顕著な増加が見られる。破線は誘導光がない場合の背景信号レベルで、中空領域を直進して通過してくる原子束に対応している。他方、負離調領域では背景信号レベルからの減少が見られるが、これは光近接場からの引力によって原子が内壁表面に引きつけられガラス表面に吸着されたためと推察される。

3. 原子誘導ファイバの応用

3.1 同位体分離

原子誘導ファイバは共鳴的な双極子相互作用を基にしているため、誘導光の周波数によって誘導原子の種類および量子状態を選別することが可能である。これを利用して、原子の同位体分離を行うことができる^{18,19)}。また、このような選択性は、後述する微小構造作製への応用にとっても優れた利点であり、純度の高い結晶を作れることを示唆する。

中空径7 μm の中空ファイバによって2種の安定な同位体⁸⁵Rbと⁸⁷Rbの分離を行った結果をFig.4に示す。ここでは、半導体レーザーの周波数を掃引することによって共鳴周波数の関数として光イオン化信号を得ている。Figure 4の上側の曲線は両同位体に対して正離調となるようにした場合で、両同位体の誘導が起きている。他方、下側の曲線は⁸⁷Rbに対しては正離調、⁸⁵Rbに対しては負離調となるように誘導光の周波数を設定した場合で、⁸⁷Rbだけが誘導されているのが分かる。

3.2 円筒形誘電体における共振器QED効果

光近接場と原子との相互作用はガラス表面のごく近傍で起きるため、ファン・デル・ワールス力の影響が無視できない場合がある²⁰⁾。微小な中空径を持つ中空ファイバ内に置かれた原子の場合には、このような現象は共振器QED効果として現れる²¹⁾。共振器QED効果は引力の共振器ポテンシャルを誘起し、表面近傍では原子誘導のポテンシャル障壁を著しく減じてしまう。

光近接場強度が弱くて光ポテンシャルが高くない場合には、共振器ポテンシャルによる相殺が生じ原子の誘導が

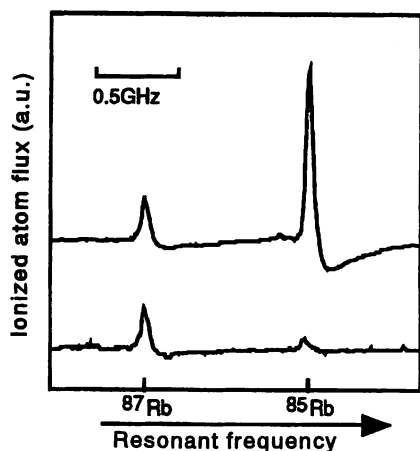


Fig.4 In-line spatial separation between ⁸⁵Rb and ⁸⁷Rb.

起こらなくなる。Figure 5に、誘導光パワーの関数として表した基底状態上準位(5S_{1/2}, F=2)の⁸⁷Rb原子束の光イオン化信号を示す。原点近傍を拡大した内挿図から見て取れるように、誘導の閾値が存在する。スケーリング形状因子を取り入れた近似解析によると、平行板形状に対する値¹⁶⁾より3~4倍ほど共振器ポテンシャルが大きくなっている。円筒形誘電体内部での共振器ポテンシャルの理論式が最近得られており²²⁾、実験結果との比較の高精度化が期待される。

3.3 光制御原子堆積に向けて

原子光学²³⁾の手法を用いた微小構造の作製に関しては、これまでに線状パターンの形成⁵⁾が行えるようになっており、次の課題として任意パターンの形成が進められている。原子誘導ファイバでは直接にドット形状を作ることが可能である²⁴⁾。それを示すために、透過原子束の空間分布を調べたものがFig.6である。ここでは、中空径7 μm の中空ファイバによって誘導されたRb原子束を、ファイバ後方12mmに位置する直径10 μm の白金の熱ワイヤを用いた表面イオン化によって検出している。Figure 6の空間分布曲線の半値全幅(20 μm)および誘導原子束の値から、中空ファイバ後方1mmの位置に基板を置いた場合には、17min/nmの

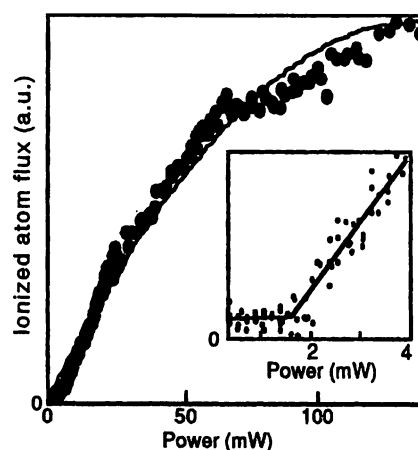


Fig.5 Guided ⁸⁷Rb atom flux in the 5S_{1/2}, F=2 upper ground state plotted as a function of coupled guide-light power.

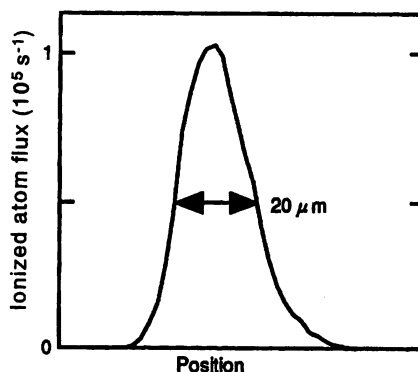


Fig.6 Measurement of the spatial distribution of Rb atom flux guided through a 7- μm -hollow fiber with a Pt hot wire.

堆積レート(の逆数)が見積もられる²⁴⁾。これは、ナノスケールのドット構造を作るのに適した値となっている。

原子誘導ファイバを用いた原子堆積のもう一つの際だった利点は、誘導光の周波数離調により堆積レートを精度よく制御できることである²⁴⁾。実際、原子ビームのオープン温度を低くして入射原子束を抑えた場合の光イオン化スペクトルからは、 $50\text{s}^{-1}/\text{GHz}$ という誘導原子束の周波数変化率を得ている。レーザー光の周波数ゆらぎを 1MHz 以下に抑えることは容易であるから、この結果は原子レベルで結晶成長が行えることを示す。ただし、誘導原子数の熱ゆらぎを抑えるためにはレーザー冷却した原子源を用いる必要がある。

4. 回折限界を越えた原子の操作

光近接場は針のように尖った先端にも生じる。このような自由度のない状況では、光近接場の拡がり具合は曲率半径程度の範囲であって、使用した光の波長によって制限されない。したがって、波長よりも十分小さい寸法に尖らせた針先に誘起した光近接場を用いれば、光波の回折限界を越えて原子を操作することが可能となる^{2,3,10)}。ナノ領域に局在した光近接場は、例えば先鋭化光ファイバを用いてつくることができる。Figure 7は、化学エッチング法によって作製した先鋭化光ファイバ^{2,25)}のSEM像である。先端の曲率半径は 2nm と見積もられる。このような先鋭化ファイバを用いた単一原子操作が提案されている^{2,3,26)}。

先鋭化ファイバで原子を操作するためには、原子の熱運動があらかじめ抑制されていることが望ましい。レーザー冷却技術の進歩により、等価温度 $1\mu\text{K}$ を切る原子集団を得ることが可能となっている。通常こうした超低温の原子集団はトラップ中に形成される。したがって、トラップから取り出した冷却原子を原子誘導ファイバを用いて先鋭化ファイバの針先に運ぶことが考えられる²⁷⁾。現在、原子誘導ファイバへの導入に適する超低温原子ビームの形成のために、光近接場を用いた原子ファネル²⁸⁾の開発が進められている。

Figure 8に、中空の円錐プリズム内壁面上にドーナツ形レーザー光を用いて誘起した原子反射ミラーで構成される原子ファネルの断面を示す。平均温度 $10\mu\text{K}$ のマクスウェル・ボルツマン分布に従う磁気光学トラップ(MOT)⁴⁾から落下する ^{85}Rb 原子の軌跡がモンテカルロ・シミュレーションによって描かれている。重力加速度が大きいため、

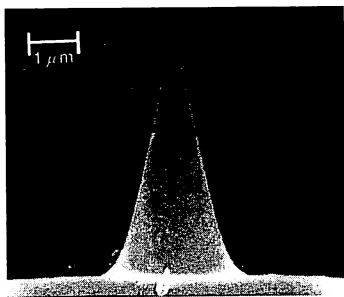


Fig.7 SEM micrograph of a sharpened optical fiber fabricated with chemical etching method.

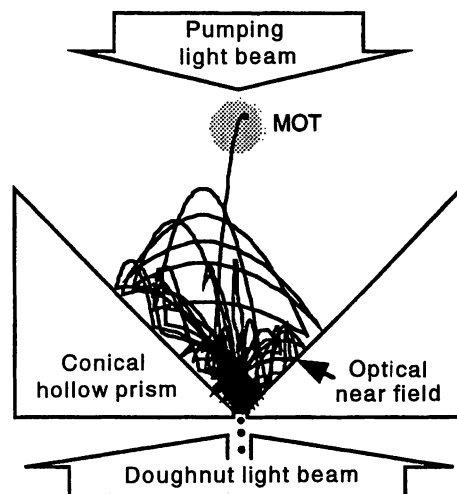


Fig.8 Atomic funnel composed of an optical near field mirror induced over a conical hollow prism. A trajectory of a ^{85}Rb atom released from a MOT is drawn based on the Monte-Carlo simulation.

数mmの落下で原子の運動エネルギーは通常のレーザーで誘起しうる原子反射ポテンシャルを優に越えてしまうことに注意しなければならない。これは、弱いポンピング光印加によるファネル内部での加速原子の再冷却²⁹⁾によって解決される。このような近接場光を用いたレーザー冷却は原子誘導ファイバ内でも行うことができ、実験が進行している。

5. むすび

光近接場を用いた回折限界を遥かに凌ぐ走査光学顕微鏡^{1,2,30)}の開発が進んでおり、生体試料観測³¹⁾や半導体物性³²⁾の研究など多方面に渡る利用が図られている。量子デバイスの開発においては、原子層エピタキシーなどの原子レベルでの結晶成長に対する関心が益々高まっている。原子誘導ファイバをはじめとする光近接場を用いた原子堆積が実現されれば、原子波レーザー³³⁾などとともに究極の結晶成長技術としての応用が期待されよう。

近接場光子は周期系でバンド構造をとる光子³⁴⁾と同じように“重い光子”として振る舞うことが指摘されている^{2,3,26)}。近接場光子を媒介とするナノ領域における近距離電磁相互作用の研究は、共振器QED効果も含めて、近接場光学顕微鏡の原理などとも密接に関わる表面近傍の局所的光学応答を理解する上で重要である^{2,35)}。原子誘導ファイバや先鋭化ファイバを利用した高精度な原子操作技術はそのような研究にとっても有用な手法を提供するであろう。

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レーザーワード

光近接場(optical near field)

日本語では近接場光とも呼ばれる。3次元形状をもつ物体に光を照射したとき、物体表面で散乱される光とともに同時に発生し、物体表面にしみだして局在する非伝搬光である。物体表面が平面である場合、その表面にしみだす光近接場はエバネッセント光と呼ばれ、光学の教科書に掲載されている。エバネッセント光のしみだしの厚みは光源の波長程度であるが、これは3次元形状の物体表面の光近接

場の特別な場合である。通常の物体は、3次元形状をもち、特に最近はその物体の寸法が光源の波長以下の場合をよく扱うが、その場合のしみだしの厚みは物体の寸法程度であり、従って光源の波長以下の厚みをもつことが分かっている。このような光近接場には回折限界がないため、微細構造の観察や加工また大容量光メモリなどへ応用することが考えられている。(大津 元一)

光CT(optical computed tomography)

ヘモグロビン、ミオグロビンなどの生体中の酸素キャリアは、近赤外域において酸素を含む状態と失った状態で吸収スペクトルが変化することが知られており、その波長特性から酸素濃度を知ることができる。このような分光学的手法と、X線CTなどに見られる計算機による画像処理技術を組み合わせて、生体物質やその状態の空間分布(断層画像)を得ることができれば、医療診断、予防に大変役立つものと考えられる。しかし、X線の場合とは異なり、生体は、

近赤外光に対して強い散乱体であり、光情報の画像化のためには、散乱の問題を解決する必要がある。現在、光CTの実現のために次の2つのアプローチがなされている。①画像再構成のためにX線CTのアルゴリズムを利用する。そのために、散乱媒質透過光の中から直線光路近くを伝播してきた成分のみを抽出する測定法を開発する。②生体内での多重散乱による複雑な伝播路の推定を行い、それを考慮した新しいアルゴリズムの開発を行う。(堀中 博道)

「近接場の光学」？ それとも 「近接場光の学」？

大津 元一

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近接場光とその応用に関する研究の最近の進歩はめざましく、その研究者数も急増している。原理的な提案は70年前にさかのぼるが、実現は1980年代に入ってからであり、回折限界を超えて波長より小さな領域を扱うこの分野は光学におけるルネッサンスなどと称されている。なお、この実現にはわが国の光エレクトロニクス、とくに光ファイバー技術がブレークスルーを与えていることは誇るべき事実である。

近接場光は非伝搬の光なので検出のためにはプローブにより散乱させる必要がある。このとき物質とプローブとは近接場光の多重散乱によって光学的に結合された状態になり、したがって物質表面に誘起された分極の分布もプローブの位置、構造の影響を受ける。このことは物質の屈折率がもはや基本的かつ不変の量でないことを意味する。

以上に注意すると近接場光について知るにはナノ寸法の物質表面に固有の光・物質相互作用を調べる必要があり、このためには屈折率などを基本量として展開する従来の光学の枠組みでの議論（すなわち「近接場の光学」）よりもむしろ新しい概念、理論モデル、すなわちナノ寸法の表面・界面物性を取り入れた「近接場光の学」が必要である。実験面でもレンズなどとは全く異なる光学素子、ナノ寸法材料の開発が不可欠である。

最近では近接場光の応用として、形を見るための光計測に使うのではなく、むしろ上記の光学的結合を利用してナノ寸法の物質を分析し、加工するなど、従来の顕微鏡などの装置にはない新機能に向かって進歩している。すなわち、従来の光学にはない新しい体系が形成されつつある。

ちなみにレーザーを扱う学問である量子エレクトロニクス・量子光学は光学から発したが、それらの持っている多様性、多機能性のためにわが国では光学分野からしだいに距離が遠ざかりつつあるようだ。これに対し、微小領域での光と物質との相互作用を扱う近接場光の研究開発がわが国では今後「近接場の光学」として従来の光学分野の中で進歩するのであろうか。それとも「近接場光の学」として光学から分離独立するのであろうか。

近接場光学顕微鏡による半導体微細・量子構造の分光評価

齋木 敏治*・大津 元一**

半導体量子構造や新材料の光学的評価をおこなう目的のために、高い空間分解能をもつ顕微分光技術の必要性が急速に高まりつつある。従来のレンズ光学系による手法はその利便さも手伝い、現在広く普及しているが、その分解能力に関しては必ずしも満足のいくものとはいえない。ごく最近、「光の波長に制限されない分解能」を謳い文句に登場した近接場光学顕微鏡はこの要求に応える道具立てとして、その活躍が大いに期待されている。近接場分光はその原理的な問題点として、高分解能の代償である感度の低さが指摘されているものの、レーザーや検出器など周辺技術の充実の後押しされ、数々のグループにより果敢な取り組みがなされている。

これまでの応用例をざっと振り返ってみると、近接場分光の用途は大きく以下の2つに分類される。1つは、試料表面におけるドーパント、欠陥などの分布、あるいはキャリアのダイナミクスを探る空間分解分光である¹⁻⁴⁾。この中には光・電子デバイスの基本型であるp-n接合の評価⁵⁻⁸⁾やレーザーダイオードなどのデバイス自体の診断⁹⁻¹¹⁾も含まれる。もう1つは、量子井戸のアイランド、量子細線・ドットなどサイズ不均一性をともなう粒子群から1つを選び出し、その本質的な特性を抽出する個別粒子分光である¹²⁻¹⁶⁾。前者の場合、概して高い分解能を必要とすることが多く、原子間力顕微鏡動作を併用して、表面形状との精密な対応づけがおこなわれることが特徴である。一方、後者に関しては、粒子

密度の許す限り、分解能に対する厳しい要求は緩められる。むしろ、観察対象のサイズが小さいこと、詳細な物性観測に立ち入る場合に弱励起測定が必要となることなどから、十分な検出感度を確保することが重要となる。

近接場分光においては、上で述べたように対象に応じて、トレードオフの関係にある分解能と感度のいずれを優先させるかが決定される。実際にこの兼ね合いはプローブ先端の金属開口径の制御により、自由に調整が可能である。このような融通性は、近接場光学顕微鏡を走査型プローブ顕微鏡ファミリーの中においてみたとき、他にはみられない特長といえる。本稿ではまず、従来の顕微分光技術と比較して近接場手法がもつ優れた特性について述べる。続いて空間分解分光、個別粒子分光の応用例としてp-n接合の多角的評価、単一量子ドットの低温における発光観察をそれぞれ紹介する。

1. 近接場分光の基本技術

1.1 原理と測定方法

近接場分光をおこなう際、使用するプローブ形状、光の照射、検出方法は観察試料に応じてさまざまである。ここでは対象を以下のように限定する。(1) プローブとしては微小開口型を用いる、(2) 試料は不透明基板上に成長させた半導体試料である(基板が透明の場合、測定上の困難は緩和される。具体的には、単一色素分子観測の成功例にならえばよい)、(3) 測定は、最も頻繁におこなわれるフォトルミネセンス検出を想定する。図1(a)に測定原理を示す。先鋭化した光ファイバーの先端部分に遮光のための金属膜をほどこし、光の波長よりも小さな開口を作製する。ファイバー中に導入され、微小開口を通過した直後の光(近接場光)は、2種類の

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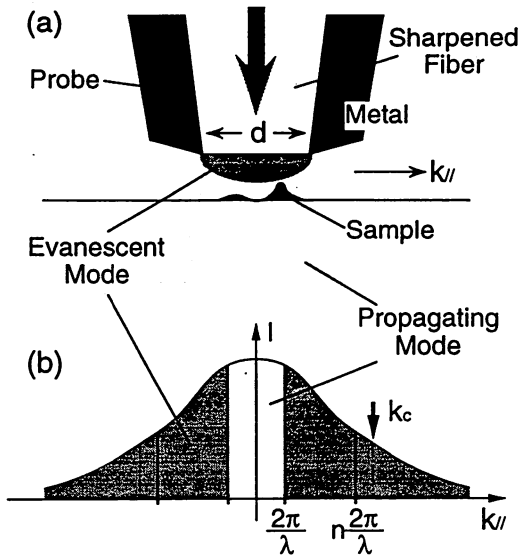


図1 (a) 近接場光学顕微鏡の原理, (b) 微小開口における回折波強度のスペクトル。

成分から構成されている。1つは伝搬光成分であり、開口直下では開口径程度のサイズに絞り込まれている。もう1つはエバネセント（非伝搬）成分であり、開口近傍に染み出ている。これらの成分は、開口面内の波数 ($k_{||}$) による回折波強度スペクトル上では図1(b)のように分布する（ここでスペクトルのひろがり（カットオフ k_c ）は、開口径 d によって決定され、 $k_c \sim \pi/d$ がおおまかな目安であることが実験的に見積もられている^{6,8)}）。つまり、 $k_{||} < 2\pi/\lambda$ の成分は空気中への伝搬が許され、それ以外はエバネセント成分としてふるまう。このような近接場光によって試料を照射することにより、局所的な励起が可能となる。信号としてのフォトルミネセンス検出は、不透明基板試料を用いているため、プローブと同じ側の外部集光、あるいは再び微小開口を通じた集光によっておこなわれることになる。以下では、半導体顕微鏡観察においてしばしば見られる問題点に関して、近接場技術を用いることによりそれがどのように克服可能であるかを詳述する。

1.2 エバネセント成分の発生

観察対象となるドーパントや粒子が表面近傍にのみ分布する表面試料とそれらが深さ方向にも一様に分布するバルク試料では、異なる光の照射方法が必要である。バルク試料の場合、高分解能を達成するためには、横方向だけでなく、深さ方向に対しても局所的な励起が重要となる。通常のレンズ系を含め、伝搬光を用いる限りそれは不可能であり（あとで述べるように吸収が強い場合は例外）、エバネセント成分を用いることが本質的な意味

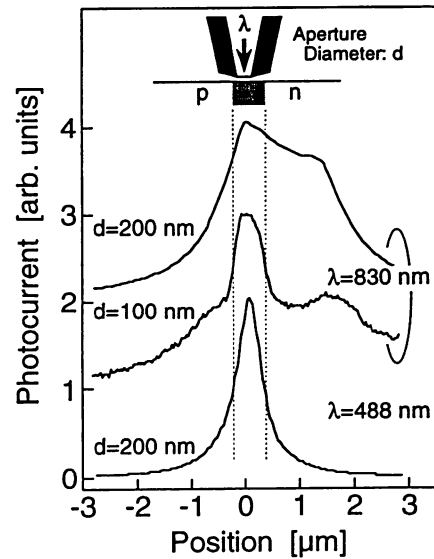


図2 近接場光電流信号の開口径、および励起波長に対する依存性。

をもつ。ただし、概して半導体試料は大きな屈折率 ($n = 3 \sim 4$) をもつため、試料中においても有意なエバネセント成分を発生させるには、開口径に厳しい条件が課せられる。この事情は以下のように説明される。図1において、実際に開口が屈折率 n の試料に対して近接場領域に近づいたとき、大気中とは異なり、 $k_{||} < n(2\pi/\lambda)$ の成分が試料中への伝搬を許される。そのため、試料内部に存在しうるエバネセント成分は残りわずかとなってしまふ。この相対的な寄与を増すためには、回折波スペクトルのひろがりが大きくなる必要があり、試料の屈折率に応じて、小さな開口を準備しなければならない。

バルク試料において、エバネセント成分の果たす役割を実験的に確認した結果を簡単に紹介する。手法としては p-n 接合における光電流検出を通して、試料内部での光のふるまいを探る方法を用いる^{5,6,8)}。p-n 界面（活性領域）近傍でプローブを走査することにより得られた光電流信号変化の様子を図2に示す。 $\lambda = 830 \text{ nm}$ のとき、開口径によって分解能が大きく異なっていることがわかる。開口径 $d = 100 \text{ nm}$ の場合、試料中においてもエバネセント成分が発生しているため、活性領域の真上にプローブがきたときに特に強い信号が検出され、高い分解能が達成されている。それに対し、 $d = 200 \text{ nm}$ のときには伝搬光成分の寄与が大半を占める。そのためプローブが真上からはずれたときでも、試料内部を光が進み、活性領域まで到達してしまい、分解能は大幅に低下する。この場合、分解能は結晶中への光の侵入距離で決定される。逆に、励起光として吸収の強い波長域のもの

を用い、この侵入距離を短くすることにより、高分解能を得ることも可能である。 $\lambda=488\text{ nm}$ の場合（侵入距離は約 80 nm ）の結果はそれを明確に示している。

1.3 微小開口による信号の集光

ここまでは、いかに局所的に光励起をおこなうかということを中心に話を進めてきた。ここで、半導体試料測定上の新たな問題点として、キャリアの拡散による分解能の低下について考える。頻繁に起こるこのような状況下では、信号の集光方法にも工夫が必要となる。図3(a)はそのようすを模式的に描いたものである。通常光励起された電子、ホールはフォトルミネセンスを放出するまでの間、それぞれの移動度にしたがい、空間的に拡散する。その距離は試料に強く依存するが、ミクロンオーダーにおよぶ場合が多い。そのため、いくら局所的にキャリアを生成しても、信号光を外部のレンズ系で集光する限り、その分解能は拡散距離、または回折限界によって制限されてしまう。これに対する解決策として、微小開口を通した信号の集光が分解能の向上をもたらすと期待されている^{7,15)}。ただし、そこで得られる分解能に関しては、単純に開口径では決まらず、プローブ先端形状などさまざまな要素が関わりあっていると考えられ、いまだ不明な点が多い。しかし、拡散距離、回折限界を超えた分解能を達成するには十分有効であることを、3.において具体的な成果として示す。

このように、開口を通した局所的な信号検出による分解能の向上が、これまでの顕微分光と比較した場合の注目すべき特長である。それにもかかわらず、実際の近接場分光の中でその適用例がほとんどみられないのは、開口による集光効率の低さに理由がある。この効率向上が近接場分光に携わる研究者の間での共通の課題となっている。一般に近接場プローブは、開口近傍において金属

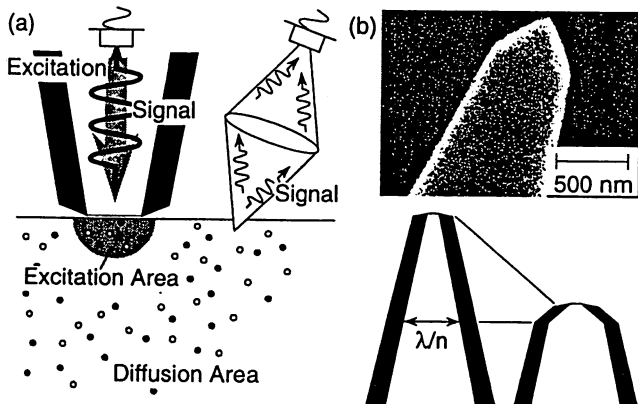


図3 (a) 局所検出による分解能の向上, (b) 二重テーパ形状による伝達効率の向上。

クラッド光導波路とみなすことができる。コア径が波長以下になるあたりから、カットオフの出現、金属クラッドによる吸収の増大などの影響が顕著になり、伝搬特性は大きく劣化する。そこで筆者らは、先端形状を図3(b)のように改良することにより、光の損失領域を短くし、高い伝搬効率を得ている¹⁷⁾（以下で紹介する測定例では、このようなプローブを用いている）。また最近では、カットオフをもたない表面プラズモンモードの利用も提案されており^{18,19)}、今後の進展が期待される。

2. 横方向 p-n 接合の多角的評価

横方向 p-n 接合は図4に示すように段差加工された GaAs 基板の上に1種類のドーパントを用いるだけで作製される新奇な構造をもつ²⁰⁾。横方向という形状の特徴を生かし、将来面発光素子としての機能が期待されている。ここでは、伝導型の変化やキャリアのダイナミクスを探る空間分解分光の例として、この p-n 接合の遷移、活性領域の観察結果を紹介する。

まず、発光スペクトルの空間依存性によって p 型から n 型への遷移領域を決定した結果を図4に示す⁷⁾。開口を通して励起光を照射し、局所的な発光を外部レンズで集光することにより、スペクトル測定をおこなう。発光のピーク波長はドーパントの伝導型、濃度を強く反映するため、その変化のようすから、遷移領域の正確な評価が可能である。特に着目すべき結果としては、上接合と下接合でその領域の幅が大きく異なっていることがあ

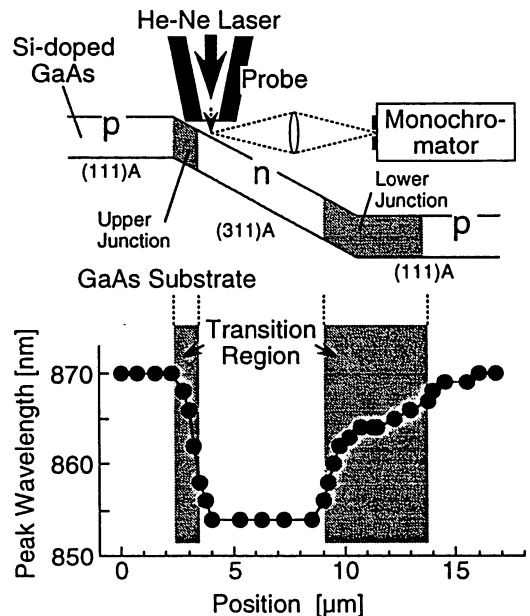


図4 横方向 p-n 接合の構造と発光スペクトルの測定方法、ならびに発光ピーク波長の位置依存性。

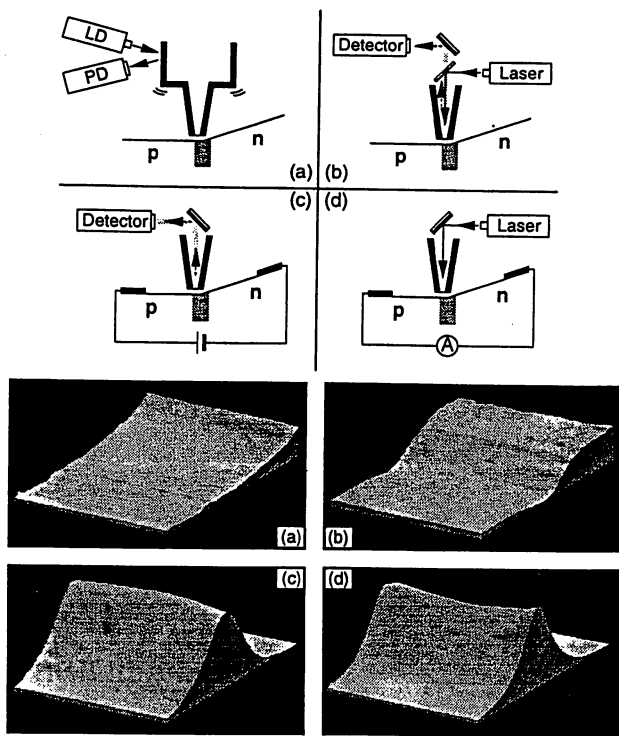


図5 p-n下接合における (a) シアフォース像, (b) フォトルミネセンス像, (c) 電流注入発光像, (d) 光電流像観察のための測定配置とその結果。

げられる。これは表面形状像（プローブ先端と試料表面間のシアフォース検出による凹凸像）で確認した，上と下での面方位の移り変わりの鋭さの違いによって説明される。

続いて，同様の試料に対して電極を設け，フォトルミネセンスだけでなく，電流注入発光，光電流も検出し，活性領域でのキャリアのふるまいを多角的に評価した結果について述べる。図5にそれらの測定配置と得られた画像（走査範囲は $4\mu\text{m}\times 6\mu\text{m}$ ）をそれぞれ示す。(b)~(d)の一連の測定は，装置間の接続の切り替えだけでおこなうことができるため，試料の同一箇所に対して，連続的な走査をおこなっている。下接合の活性領域において強い電流注入発光(c)と光電流(d)が観測されている。この領域でフォトルミネセンス強度(b)が逆に弱くなっているのは，光生成されたキャリアが内部電場によってそれぞれ発光に寄与しない方向にドリフトするためである⁷⁾。また，詳細は省略するが，これらの測定を通じ，電子とホールの拡散距離の実空間上での評価やp-n界面の内部での傾きの見積りなども定量的におこなわれている⁸⁾。

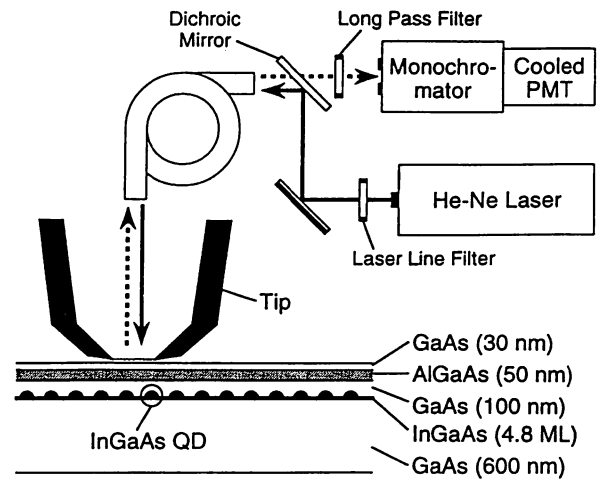


図6 量子ドット試料の構造とその発光観察の測定配置。

3. 低温における単一量子ドットの発光観測

半導体量子ドットは，離散的準位構造と強い光学遷移をもち，その特徴は発光スペクトルに強く反映される。またこのスペクトルの幅は，電子，ホール（励起子）の緩和のダイナミクスの情報を提供するため，その正確な評価が基礎，応用の両面から強く望まれている。従来の分光法においては，その分解能の低さのために，サイズゆらぎのある多数のドットを同時に観測してしまい，単一のドットに関する重要な知見が不均一性の中に埋もれてしまっていた。ここでは，近接場顕微鏡の分解能を生かした個別粒子分光の一例として，単一量子ドットの発光スペクトル，発光イメージング測定をおこなった結果を紹介する。

試料は図6に示すようにGaAs基板上に成長させた自己形成InGaAs量子ドットを用いる²¹⁾。ドットのサイズは直径30 nm，高さ15 nm程度であり，成長密度は $2\times 10^{10}\text{cm}^{-2}$ と非常に高い。試料，プローブ，スキャナーなどヘッド部をクライオスタット中に組み込むことにより，ヘリウム温度での測定が可能となっている。開口径500 nm，先端角の大きな二重テーパ型プローブを用いて，He-Neレーザー光を試料に照射する。キャリアはGaAs層においても生成され，1~2 μm 拡散した後，大部分はドットの閉じ込め準位に緩和する。本実験では，観測ドット数をさらに制限するため，ドットからの発光もまた，開口を通じて集光する。

図7に発光スペクトルを示す。(a)はプローブを1 μm 程度試料から離すことにより得られたファーストフィールド領域でのスペクトルである。多数のドットを同時に観測しているため，スペクトル形状はなめらかであり，

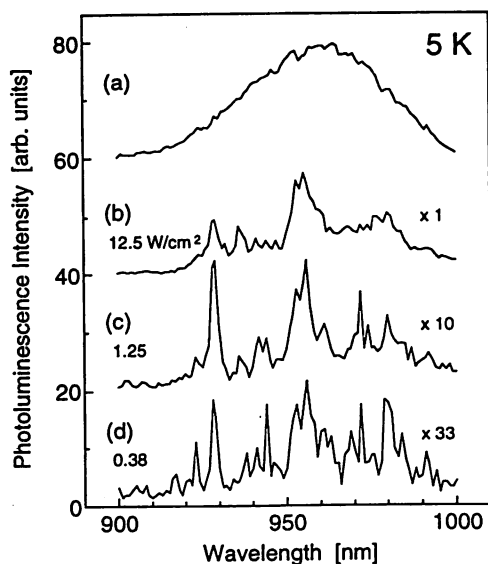


図7 (a) ファーフィールド, (b)~(d) ニアフィールドにおける量子ドットからの発光スペクトル。

サイズの不均一性の大きさを反映した情報だけを与えている。一方(b)~(d)は、プローブをニアフィールド領域まで接近させて測定したスペクトルであり、細かなピーク構造がみられる。この結果は、高分解能により観測ドット数が10個以下に制限され、個々のドットからの発光が分離して観測されていることを示している。(c)、(d)の励起強度領域では、スペクトル形状が変化せず、発光強度は線形に増加していることから最低励起状態からの発光のみを検出していることがわかる。それに対し、(b)の励起強度に達すると、最低励起状態の飽和が始まっていることがわかる。本測定では、最も鋭い発光線幅として波長分解能程度の0.5 meVが確認されている。

次に、発光の検出波長を923, 928, 968 nmに固定し、プローブを走査することにより得られた画像を図8(a)~(c)に示す。走査範囲は、 $3\ \mu\text{m} \times 3\ \mu\text{m}$ である。それぞれの波長に対し、全く異なった発光画像が得られている。1つ1つのスポットが同じ発光波長をもつ単一ドットの分布のようすを示している。スポット密度の違いは、図7(a)の不均一ひろがりを実空間上で再現している。図8(d)には比較的小さなスポットについて、信号強度の断面のようすを示す。ドットの実際のサイズが30 nmに対して、画像上でのスポットサイズが160 nm程度($\lambda/6$)となっている。励起されたキャリアはGaAs層で拡散しているため、この分解能は集光領域によって決定されていると解釈される。またこの大きさは開口径よりも十分小さく、開口を用いた集光が非常に効

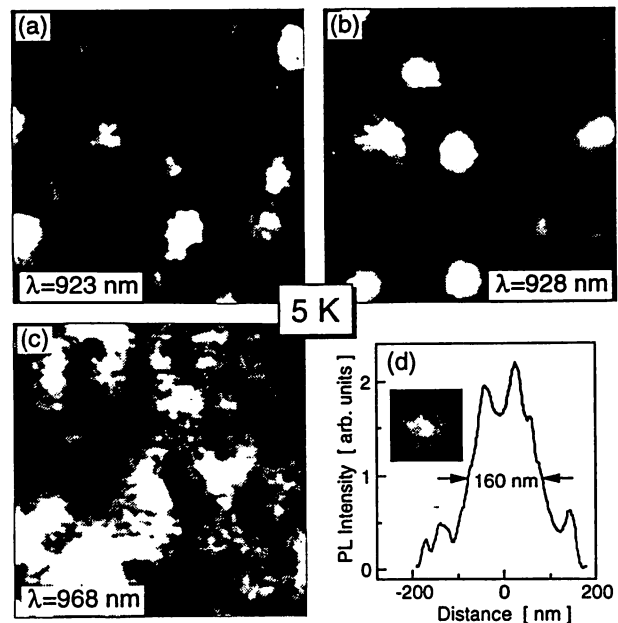


図8 (a)~(c) 検出波長の固定により得られた単一量子ドットからの発光画像, (d) 発光画像の断面。

果的であることを示している。装置の安定性、分解能の再現性についても良好であり、単一ドットの高次準位、あるいは励起子分子の発光も観測されるに至っている。

本稿ではまず、これまでの顕微分光技術と比較し、近接場光学顕微鏡がもつ際立った特長について詳述したが、それらは以下の2点にまとめられる。(1) エバネセント成分を利用した励起により、深さ方向にも局所的な励起が可能である、(2) 開口を通した集光により、キャリア拡散やバックグラウンド信号などの影響を大幅に除去できる。一方、感度に関してはいまだ大きな課題が残されている。近接場分光のもつ「高分解能」が従来の顕微分光の「簡便さ+高感度」という魅力に勝り、便利な道具として抵抗なく利用されるようになるためには、この問題の克服が必須である。3.で紹介した大きな開口をもつ改良型プローブによる高感度かつ高分解能測定はその取り組みの第一歩である。今後は高い分解能が本質的な重要性をもつ、反射、吸収、非線形信号などの測定が不自由なくおこなえるよう、システム性能の向上を進めていく必要がある。

ここでは特に、近接場顕微鏡による「観測」としての側面のみを述べた。しかしプローブを用いるという観点からは「制御」という潜在能力を引き出すことが可能である。プローブ先端の誘電体部あるいは金属部の試料への接近は、局所的な環境の変化をもたらす。その結果、たとえば発光物質に対しては発光寿命の変化や励起エネ

ルギーの移動を誘発する。また鋭い共鳴構造をもつ物質に対してはその共鳴シフトを生じさせ、新たな光機能を発現させるであろう。このように今後取り組むべき課題は山のようにある。波長にとらわれない自由な光操作という夢の技術を手にするための出発点として、近接場分光が担う役割は十分大きいと考えられる。

本稿をまとめるにあたり、横方向 p-n 接合に関しては、NHK 放送技研・斎藤信雄氏、量子ドット測定においては、NEC 光エレ研・西研一氏、東工大・横山吉隆氏（現 NEC）にそれぞれご協力いただきました。またプローブ作製については、神奈川科学技術アカデミー・物部秀二氏にお手伝いいただきました。これらの方々に深く感謝いたします。

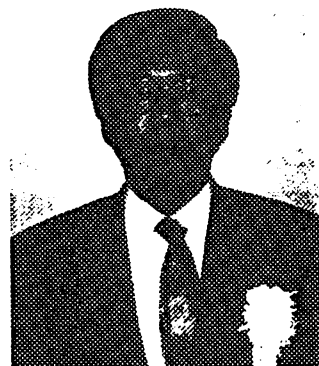
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光で見えないものを光で見る、 あやつる、加工する

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東京工業大学の天津でございます。どうぞよろしくお願ひいたします。

伊賀先生のお話は、非常に豊かな発想に基づきまして着実なご努力をなされた結果、産業界の大きな分野を開かれた業績のご紹介でしたが、これからの私の話は、まだ技術の成長段階でいいますと、幼稚園から小学校1年生ぐらいのところになったかなという話ですので、まだ不確かなところもありますが、将来もあるだろうということで、気楽にお聞きになってくだされば結構です。

「光で見えないものを光で見る、あやつる、加工する」というタイトルは、実は伊賀先生の先ほどのお話とあわせてご説明しますと、広がったり集まろうとする性質を持つ光を使ったのでは見えないような非常に小さいものを、別の種類の光を使って見たり、あやつったり、加工することはできないかというお話です。

話の筋としては、まず導入のお話と、それから簡単な原理的なお話、それから新しいタイプの光を発生させるためのプローブ——というのはガラスの針のことですが、そんなものを作る話、それからこういうものが出来た暁に、実際に見たり、分析する話、それから、話の都合上、タイトルの順序とは逆にさせていただきますけれども、その小さな光を使って物質を加工するという話です。一例として従来の光ディスクに比べて、1,000倍ぐらいの多くの情報を記録できるような、光メモリを加工するという話です。それから次は小さい光を使って物質の基本的な

構成要素である原子をあやつる話をしてまとめさせていただきますと思います。

申し遅れましたけれども、私は東京工業大学で教えておりますが、同時にこういった研究は、財団法人の神奈川科学技術アカデミーで、ここ何年間かやらせていただいております。それも含めたご紹介ということをしていただければと思っております。

まず、それでは原理的なお話をします。

最初は、伊賀先生がお話しになったことの復習になります。まず、いままでのお話で出てきた光というのは、要するに広がるものです。光は電波の親戚ですけれども、たとえばレーザーから出てきた光は、広がって遠くまで飛んで行きます。それはものすごいスピードで飛んで行くわけです。それですから、光の通信とか、光の情報処理とかいうことができるわけです。

この光を集めるには、ある直径を持つ凸レンズを持ってきて、その中に光を通します。並行な光は凸レンズを通すと、焦点のところに点状に集まります。点光源の場合にもある程度集まるわけですが、しかし、波は広がろうという性質を持ちますので、レンズで集めようと思っても、実際には点にはならず、若干ぼけが生ずるわけです。

ぼけの大きさがどのくらいかというと、空間的に振動しながら飛んで行くときの1周期の長さ、——これは光の波長といっているわけですが、その光の波長程度です。本当は点になると思ったのが、もともと光が広がろうとする性質があるために、レンズで集めて

も、光の波長の大きさ程度の直径までしか小さくならないということです。

そういうことが起こると、往々にして悪いことがあるんですけれども、たとえば図1に示すように、ここに隣接して2つの光源があったとします。この距離は非常に近い。そこから出てくる光をレンズで集めて、たとえばここに仮に点が出来れば、この光源の間隔ぐらいのところだけ離れた点が生じますね。だから、この点が2つあるということを見て、ここには光源が2つあるということがわかります。しかし、もとの1個の光源から出てくる光が、ここで光の波長程度のぼけを生じますと、特にこの2つの光源の間隔が光の波長以下ですと、光の波長程度広がった点が、1つあるのか2つあるのかわからなくなります。ということは、逆に、ここに光源がもともと1つあるのか、2つあるのかわからないということになります。

これは何を言っているかという、こういうレンズで小さいものを見ようとする、ここに小さいものが光の波長程度、またそれ以下の間隔離れて置かれていたときに、その2つの物体を区別することができないということです。言い換えてみますと、たとえばこれを顕微鏡として考えますと、顕微鏡で見ることのできる最小の寸法は、光の波長程度であるということになります。

この原因はどこにあるかという、もともと光というのは遠くまで伝搬していくとともに、広がろうとする性質を持つ。ですから、いくらレンズで集めようとしても広がろうとする性質が残って、その大きさが光の波長程度だということです。

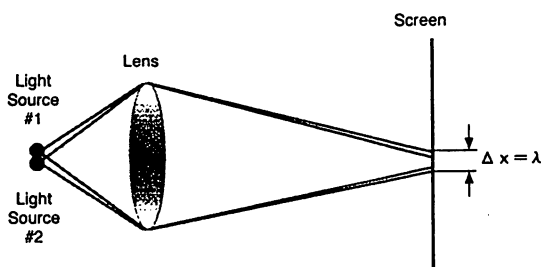


図1 レンズで集光したときの像のぼけ Δx λ は光源からの光の波長。

よく、「壁に耳あり障子に目あり」というようなことわざがありますけれども、この話は、光だけではなくて、一般に波について言えることです。音は音波で、光とはちょっと違いますけど、それは波に違いないですね。ですから、壁に耳ありというのは、壁に必ずしも耳をつけなくても、壁の向う側の人の話し声はどこかに隙間があれば、その音が回って、隙間を出たあとと広がろうとするので、仮に後ろにいても聞こえるということを行っていますので、音でも広がろうとする性質を持つということから来ることわざ、というふうに言うことができます。もちろん壁に耳をつければ、その壁を通じて音が聞こえるわけですが、必ずしも壁に耳をつけなくても、壁の後ろ側にちょっと隠れていても、音は壁を回って広がろうとする性質を持つので、聞こえるということです。

翻って光に関して考えてみますと、光が広がろうとする性質を持ちますと、あまり小さいものを見たり、加工することができなくなります。たとえばレーザーの光をレンズで集めて、半導体の板を持ってきて、これを熱すると穴があきます。穴があくといっても、その穴の大きさは光の波長程度ですので、光の波長程度より小さい穴をあけることはできないということです。

一方、レーザーというのは光を閉じ込める箱でありまして、その中に光の波を閉じ込めなくてはいけません。あまり小さい箱を用意して閉じ込めようとする、光は広がろうとする性質がありますので、反発して広がってしまっ、小さい箱の中にはちっとも光が閉じ込められないことになります。つまり、あまりレーザーの寸法を波長よりも小さくすると、レーザーの装置はできなくなるということです。ということなので、レーザーをたくさん並べた光の集積回路などの最小寸法は、やはり光の波長程度以下になりません。

皆さんご存じのように、電子の動きを使ったトランジスタ、それを集積化した集積回路というものがありますね。LSIといってい

まして、最近はその上にUというのをつけて、ウルトラLSIとっていますが、あれはものすごく小さくなってきてまして、原子数個分または数十個分ぐらいの寸法の構造の加工によって作ったようなデバイスになっているわけです。最近では、原子を1個1個つかむとかいうような加工技術も出てきておりまして、物質は非常に小さいんですけども、それに対して光というのは、今いいましたように、波長程度以下の寸法のところには閉じ込めることができないほど大きいということです。

ということで、世の中は高度情報化社会、それから高度福祉社会になってきておりますので、光を使った通信などで多量で多様な情報を送ろうとすると、光の波長程度の大きさのデバイスでは追いつかなくなる時代が、わりと早くやってくるかもしれません。すなわち光の波長以下には光は小さく閉じ込められないので、早晚、光技術の限界がやってきます。

しかし、本日のこれからのお話は、実際には必ずしもそういうデッドロックにはのり上げないかもしれないという内容です。

それにはどういうことをすればいいかということですけども、それは広がろうとする光以外の光を使えばよろしい。いろいろ探してみますと、そういう光が実はあります。

それは近接場の光といいます。図2に示しますように光の波長に比べてずいぶん小さい直径を持つ微粒子があったとします。これに光を当てます。この光はこの微粒子が小さ

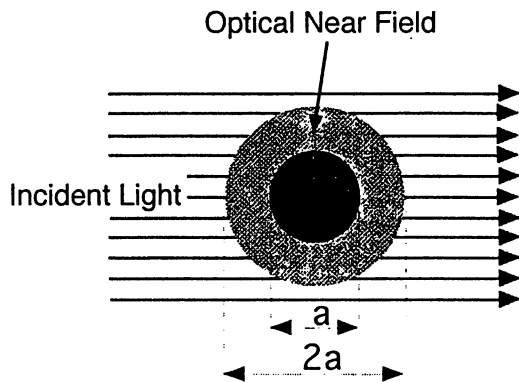


図2 直径aの微粒子の表面に発生する近接場の光の寸法

いために、ほとんど気がつかないで、通り抜けて行ってしまいます。しかしこの微粒子の表面を注意深く見てみますと、光がじわじわと染み出していまして、その光の染み出しの厚みが、この微粒子の直径程度です。それからこの光は、微粒子の周りをぐるぐる回っている表面波のようなもので、遠くへは飛んで行きません。これが物質の表面に近接したところにある光の場なので、近接場の光といっております。

これは先ほど言いましたように、遠くまで飛んで行かないで、この物質の周りに染み出していますので、広がる光、集まる光ではなく、染み出しているだけで飛んで行かない光です。

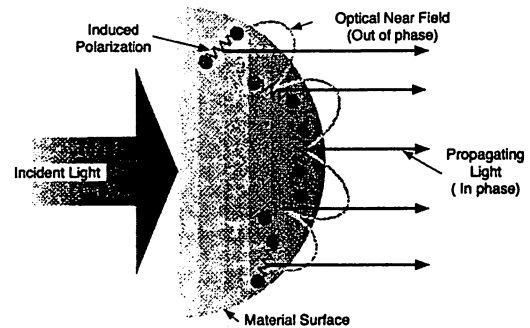


図3 近接場の光の発生の機構の説明

何でこんな光が出るのかというのを簡単に説明しますと、図3のようになります。左から光が入ってきまして、微粒子に当たります。微粒子というのは、電子とか原子がいっぱい詰まったものです。ここに光が入ると、この中の原子の周りを回っている電子が振動を始めまして、非常に小さなアンテナが出来るようになります。言ってみれば、これは原子のアンテナです。原子のある場所によって出来るアンテナの方向がいろいろ違いますので、そのアンテナから発する電波、すなわち光の出方が原子ごとに少しずつ違います。光は波ですので、原子のアンテナから出てくる光の振動は位相が少しずつずれています。そのうちの位相の合った成分は、互いに足し合わさって遠くまで飛んで行きます。これが

入ってきた光があたかも通り抜けて出てきたことになっているわけで、これが実は従来の光です。

それ以外に、互いに隣同士位相がずれていると、出てくる光が弱め合ったりするような干渉という効果により、すぐ消えていってしまうわけです。言ってみれば、この物質の表面だけに先ほどの干渉の縞々みたいなものが出来る。いわゆるアンテナ同士が互いに干渉し合って、やがて消えてしまうような、ささやかな光がここに出てきます。これが先ほどの、染み出して遠くに飛んで行かない光、つまり近接場の光です。

これが発生する光近接場の実態と考えられるわけですが、実はいままでの説明の中に、光の波長ということばは全然出てきませんでした。光の波長がどうであろうと、こういう光の膜といいますか、近接場の光が存在するわけです。だから、この微粒子の直径を小さくして、その周りにこんな非常にささやかな光を出させ、この光をペンライトのように使うことができれば、その物質が仮に光の波長に比べて小さい寸法を持っていたとしても、このペンライトの光を物質に当てまして、照明すれば、それを見ることができます。それからこの小さな光の膜を半導体の板に持って行って、そこを加熱すると、そこに光の波長よりずっと小さい穴があきます。しかし実際には問題があります。この光は遠くまで飛んで行きませんので、見えないわけです。見えなければ利用ができません。まず、見るためにはどうしたらいいか。

そのためには、たとえばこの微粒子に光を当てたときに光近接場が出来たとしますと、その光を測定するために、図4(a)に示すようにもう1つの微粒子を持ってきます。その微粒子で先ほどの光の膜を散乱させるわけです。要するに微粒子を光の膜の中に突っ込み、その微粒子で先ほどの光の膜を散らせるわけです。散ったあとは、もうこれは普通の遠くまで飛んで行く光になりますので、遠くに目を置いていても見えます。このようにもう1

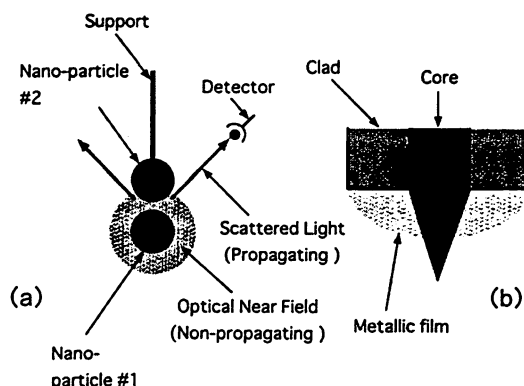


図4 プローブによる近接場の光の検出
(a) 原理図
(b) コア部を先鋭化し、根元を金属膜で覆ったガラスファイバによるプローブの断面図

個の微粒子が必要だということです。この微粒子を、プローブとっています。

では、どのくらいの大きさのプローブが必要かということ、一番よく光を散乱させるためには、この微粒子と同じ大きさの微粒子を持つてくるのが一番いいということがわかっています。たとえば1 nm (ナノメートル)、すなわち波長の1,000分の1ぐらいの大きさの微粒子の周りに光の膜を発生させ、それを観測したり利用するには、1 nmの大きさのもうひとつの微粒子を最初の微粒子の表面近く、1 nm程度まで近づけないといけないということです。

ただ、1 nmの微粒子を作るとするのは、必ずしも容易ではありません。1 nmというと非常に小さくて、原子が約100個からなる物質です。ですから、このような光を利用するには、非常に小さな微粒子を加工して作るというような、材料工学の助けが必要になってきます。

実は、ここまで込み入ったようなメカニズムに関してのお話ではありませんが、関連する素朴な提案が1928年に出ているんですね。イギリスのシンゲという人が物理学の雑誌に書いておりますが、その論文を見ていると非常に面白くて、英語そのものが日本でいうと古文みたいな感じで、われわれが高校で習ったような英語とはちょっと違う文法に基づいて書いてあります。よく読んでみると、「私

はこのような論文を書く気はしなかった。しかしながら、ボスがこういうものを書いて、特許を書いて発表しておく、のちのちにいいことがあるかもしれない、というので書きました」ということを書いてあります。確かにその頃は、小さいものを作るということではできなかったものですから、こういうものを提案しても荒唐無稽だったと思います。

けれども、科学の長い歴史の中では、そういうものを実現するような技術の研究も進んでいまして、最近になってそういった研究を実現するための周辺技術がかたまってきましたので、1980年代の中頃に、ほぼ4つの研究グループで初めて実験が成功しました。それはIBMのスイスのチューリッヒの研究所、アメリカのコーネル大学、それからアメリカのオークリッジ国立研究所、それからわれわれの東京工業大学です。

それからもう1つ面白いのは、有名なインシュタインが友達に宛てた手紙の中に、シンゲと同様のアイデアも書いているということです。

さて、実際にはどのようにして加工するかということですが、微粒子を、宙に浮かすということもできませんので、何らかの支えが必要で、したがって、現代の宝石といわれる非常に純粋度の高いガラスのファイバーを加工して作ります。

ガラスのファイバーというのは、中心部にゲルマニウムなどの原子が入って、屈折率が高い部分を持ち、これをコアといいます。周りにはクラッドといい、コアを支えるものがあります。光はこのコアの中を伝搬してきます。

これを図4(b)に示すように針のように尖らせまして、根元に不透明な金属膜を塗ります。そうすると先端だけが透明で飛び出します。この部分を微粒子として使うわけです。この非常に小さい先端で近接場の光を散乱させるわけです。散乱させたらファイバーの中を通して、ファイバーの後端にある光検出器で測定します。

なぜ、1980年の中頃になって、実験がう

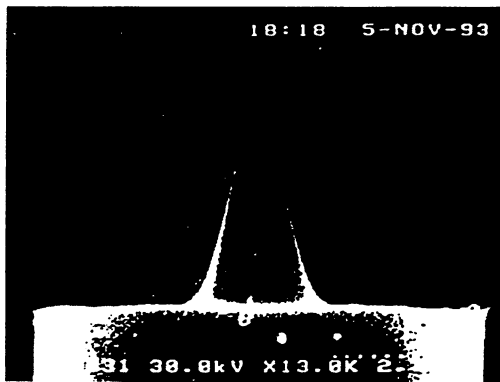
まくいくようになったかということ、特に日本のファイバーメーカーの技術力の高さによって、ガラスファイバーが、非常に性能よく出来るようになりまして、かつこれを熱で溶かしたり、それから最近は酸で溶かして尖らすという技術が非常に進みまして、先端の曲率半径が非常に小さくなるようなものが出るようになりました。

それ以外に、IBMのチューリッヒ研究所がその当時ノーベル賞を取った、電子顕微鏡の非常に倍率の高い走査型トンネルプローブ顕微鏡というものがあるんですが、そういうような技術を使えるようになりました。それまでの約60年間は、ほとんど空白の時代でした。

図5はわれわれのところであまくいった、ガラスファイバーを尖らした結果の写真です。ガラスファイバーの周りのクラッドの中心にコアが尖っています。これはフッ酸で溶かし出したものです。この先端部分が少し暗いですけれども、拡大すると先端曲率直径、先ほどの微粒子に対応するものの直径ですけれども、それは30Å未満。原子が20~30個ぐらいしかくっついていないような状態になっています。次に金属膜を根元に塗りまして、円錐状のものを飛び出させます。円錐状のもの寸法が30nmくらいですから、光の波長の20分の1ぐらいの大きさになっています。もちろん、この先端部分は光の波長の1,000分の1程度の大きさになっています。こういうものを使うと、先ほど言ったような光の膜を使って見る、加工する、あやつるということが出来ます。

最近ではガラスファイバーを、ただ尖らして鋭くするだけではなくて、出入りする光の量が多くなるように、尖っている部分の長さを短くしたり、それから先端に色素や半導体の微粒子などをつけて、違う色の光を出したり、いろいろな機能をするプローブが出来るようになってきています。

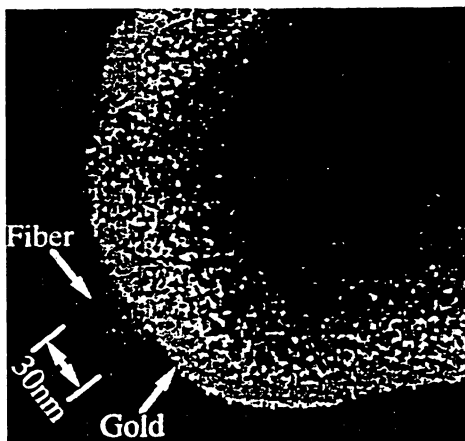
それからもう1つ申し上げたほうがいいと思われるのは、図4(a)はあくまでも原理的な説明でして、両者の微粒子の役割を逆にす



先鋭化ファイバー



先端部の拡大図



根本に金属膜を蒸着後

図5 ファイバークローブの電子顕微鏡写真

することもできます。すなわちガラスのファイバーの後端から光を入れますと、先端部分に非常に小さな近接場の光が染み出します。それをもともとは測定対象である微粒子で散乱させるということもできます。ですから、こちらの下の微粒子の周りの光をプローブで散乱させるか、ファイバーから出てきた近接場の光を微粒子で散乱させるか、どちらかを使

います。どちらの方法を使うかは、応用の内容によるわけです。

それでは、見る、分析するというお話から始めさせていただきます。

まず、見るということですが、先ほどの2通りのやり方があるんですが、1つはガラスファイバーを近接場の光のある位置に固定します。そして散乱される光のパワーを測定します。その測定値をグラフ用紙の縦軸のところに書いておくわけです。横軸はこの針の位置です。次に、この針をちょっと動かして、やはり次のパワーの値を測定します。そして、グラフ用紙の横軸の隣の点に、この測定値を書きます。これを繰り返して針をずっと動かしながら、針の各位置のところで光のパワーの測定値をグラフで書いていきます。実際には針を一次元上に動かすわけではなく、奥行き方向にも動かして、二次元の地図の上に光のパワーの測定値を書いていくわけです。それは何を表しているかということ、近接場の光の各場所でのパワーの大きさを表していることにほかなりません。ということは、この微粒子の寸法、形状を測定していることになります。

これは微粒子の形を見るための装置、いわゆる顕微鏡を作っていることにほかなりません。これを近接場光学顕微鏡といっているわけです。

具体的にそんなようなやり方でものの形を見た例を、これからいくつかご覧に入れます。

その装置では、ガラスのファイバーをもちろん手で動かすわけにもいきませんし、それから装置をコンパクトに作りませんと、周りの温度が1度変わると、たとえばアルミニウムの板などは1 μ mぐらい簡単に伸び縮みしてしまいますから、きれいな像が見えません。ですから、お茶筒のような容器の中に収めて、ひっくり返しても壊れないような装置を注意して作るわけです。

たとえば図6は、サファイアの非常に平坦な板を見たものです。平坦な板でも、ところどころ原子の数でいうと、1層とか2層ぐ

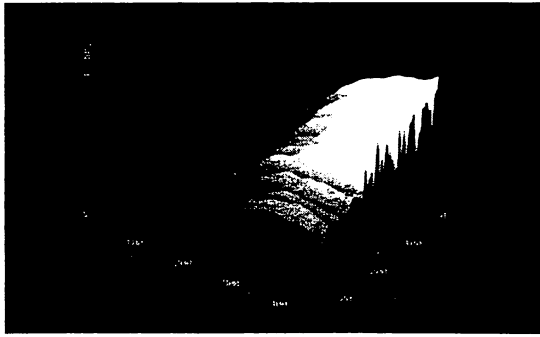


図6 超平坦サファイア基板上の原子層ステップの測定結果

らのステップがあります。サファイアというのは透明ですから、後ろから光を当てて、先ほどの近接場の光をサファイアの板の上に発生させます。そこにプローブを近づけていって、サファイアの板の面に沿わせて動かしながら、測定される光のパワーを図示したものです。縦軸のこの高さが10Åぐらいです。ですから、原子でいうと2層分ぐらいの段差があるということで、光の波長の1,000分の1ぐらいの厚みがはかれるように出来ているということです。

それ以外に、誘電体のリチウムナイオベートと称する結晶を、特別な方法で先ほどのサファイアの板の面の上に一辺20nmぐらいの微粒子として作ります。それに光を当て、出てくる近接場の光をファイバーで拾い出すと、図7のように小さい結晶の像が見えます。

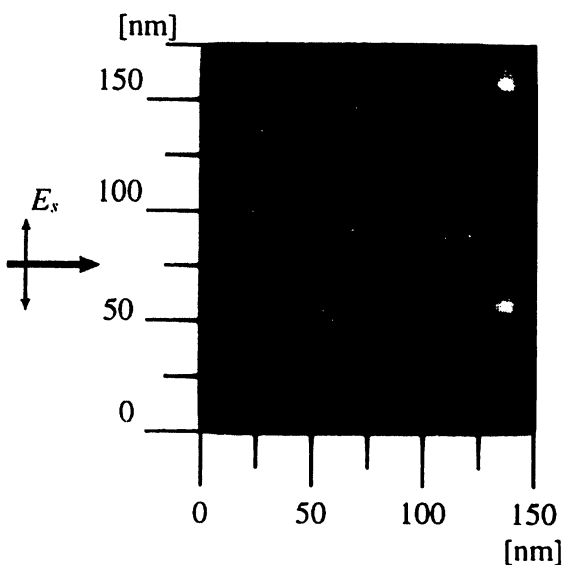


図7 リチウムナイオベートの微粒子の測定結果

この図では光の波長の4分の1×4分の1ぐらいの視野を見てるわけです。普通の顕微鏡ですと、先ほど凸レンズで焦点面上に光を集めたときの光のぼけのために、このような小さい視野の中には像があるのかないかわかりません。しかし、このようなプローブをうまく使いますと、こういうふうには結晶の微粒子が見えます。

これは要するに顕微鏡なわけですが、顕微鏡の大きなマーケットは、生物とか医学とか、そちらのほうの生体試料を見るような分野です。図8は直径がだいたい20から30nmぐらいのバクテリアの鞭毛と称する小さなひげを、ガラスの基板の上に固定しまして、空気中で見たものです。この変な黄色いものがその鞭毛の末端の部分で、直径はだいたい30nmですから、光の波長の20分の1から30分の1ぐらいの値です。



図8 バクテリアの鞭毛の空気中での測定結果

これはもちろん、従来は光では見えませんでした。どうしていたかという、電子顕微鏡を使って見ていたわけですね。すなわちガラスなどの基板の上にこのひげを置き、金属膜を塗り、それを真空装置の中に入れて、電子のビームを当てて見たということです。ですから、電子顕微鏡ですと、生体試料そのものを殺して見ているということになります。しかし、光の場合には空気中で、この程度の大きさまで、この顕微鏡で見えるということです。

ただ、実際には生物試料というのは、溶液中で生きていますので、溶液中で見る必要があります。電子顕微鏡では溶液中の試料とい

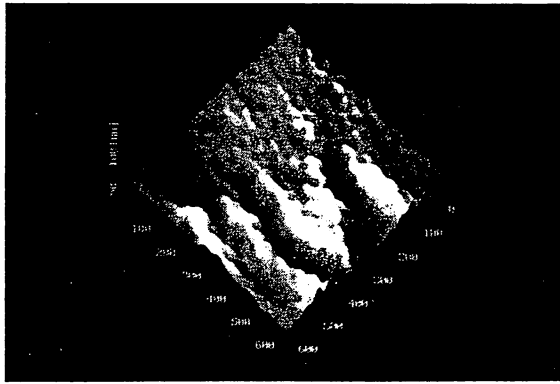


図9 バクテリアの鞭毛の水中での測定結果

うのは見えません。ここでは先ほどの鞭毛をガラスの上に載せて、それを水の中に入れます。水の中にガラスのファイバーを浸けて、この鞭毛の下から光を当てたときに発生する近接場の光を見ています。図9は互いに近接する5本のひげが見えた例です。これもだいたい、30から太いところで50nmぐらいの寸法のものが見えます。

見るということに関してはきりがありません。たとえば神経細胞の本体から出ている軸索と称するパイプの中にマイクロチューブリンと称する細いひものような束がいっぱい詰まっています。このマイクロチューブリンと称するひもの直径は、電子顕微鏡で見たときは25nmぐらいといわれています。これを電子顕微鏡で見るためには、このパイプの軸索を切り、中からマイクロチューブリンの束を出し、そのうち1本を板の上に載せて、金属膜を塗って、真空の中に入れなくてはなりません。

しかし、光の場合には、実はこの軸索が半透明ですので、プローブをこの軸索の外側に這わせると、中の模様が見えます。ですから軸索を切って中身を出さずに見ることができる、というのがプローブを使った方法の特長です。

実際に見た1本のマイクロチューブリンの直径が25~26nmぐらいです。つまり電子顕微鏡で見たのとほとんど同じぐらいのものが、軸索のパイプを切ることなく見えるということです。

いまの段階でどのぐらいの小さいものまで見えるかといいますと、代表的になっていきますわれわれの例ですと、8 Åぐらいのものまででしたら見えます。ですから光の波長の1,000分の1ぐらいのものが見えるようになってきているということです。

実は、光を使ってこんな小さいものを見ようとするときには、もちろん強いライバルとして電子顕微鏡などがあります。しかし、光には電子顕微鏡ではできない機能がありまして、それは分析するという事です。最近の動きは、見るだけではなくて、非常に小さいものの構造を分析ということが加わっておりますので、この後は、先ほどの近接場の光を使った大きな応用として、分析する話をご紹介させていただきたいと思います。

たとえば、当てる光の波長を変えていきます。ここで、どの波長の光を当てたときに、相手の微粒子はその光をどのくらい強く吸収するか、またはどの波長の光を当てたときに、その微粒子はどんな色の光を発生するかということ調べますと、微粒子の内部構造がわかるということです。内部には電子とか原子が詰まっていますので、たとえば原子や電子がどんな振る舞いをしているかということがわかります。

代表的な例を1つだけご覧に入れることにしますが、最近はやってきています半導体の量子ドットというものの構造の分析です。

半導体の量子ドットというのは、半導体の小さな粒とだけ思えば結構です。たとえば半導体の板の上に、結晶成長の最先端技術を使って非常に小さな半導体の微粒子をたくさん並べます。この微粒子の寸法は中に入っている電子の動きを束縛してしまうほど小さいので、電子はもはや自由に動き回れません。すると電子はどんなエネルギーも好き勝手に取り得るのではなくて、特別の値しか取り得なくなり、電子のエネルギーが離散化します。

これに赤い光を当てると、この電子が活性化しまして、この中に閉じ込められながら、

近赤外の光を出します。ですから、出てきた近赤外の光を使うと、このような小さい微粒子をいっぱい集めて、効率の高いレーザーが出来るといわれています。

各々のドットからは、非常に単色性の高い強い光が出てくるといわれています。しかし現在の技術は、1個1個のドットを規則的に同じ大きさで、同じ構造で作ることが難しく、やはりばらついています。このばらつきをなくして、より高い精度で、ばらつきの少ないドットを作っていくにはどうしたらいいか、という設計指針を得るためには、まず、このドット1個1個から出てくる光のスペクトルの特性を正確にはかって、そのばらつきの原因がどこにあるかというのを知って、最適な材料の製作を設計することが重要です。

しかし、従来は多数のドットに、レンズを使って赤い光を当てて、それで近赤外の光を出して、それをレンズで集めていました。この場合、赤い光の像のぼけが1 μm ぐらいあります。そうしますと、その1 μm ぐらいのぼけの中に、ドットはだいたい100個とか200個ぐらい入ってしまいます。ですから、1個のドットだけからの近赤外の光を測定しようとしても、それは無理なわけですね。ですから、多数のドットのスペクトルの包絡線が測定されるにすぎません。しかし、この1個のドットからのスペクトルだけをはかりたい。そのためには、1個のドットに光を選択的に与えられればよろしい。そのためにはプローブの後端から赤い光を入れて、先端の鋭いところに近接場の光を染み出させて、それを1個のドットに近づけて、1個のドットだけを活性化して、そこから出てくる近赤外の光を測定することが必要でしょう。

もちろん、電子顕微鏡は、こういう目的には使えませんが、近接場の光を使ったようなやり方ではうまくいきます。

最近、半導体レーザーの材料のインジウム・ガリウム・砒素という化合物半導体の、直径30nm、高さが15nmぐらいの量子ドットが作れるようになってきました。それはガリ

ウム砒素の基板の上の、だいたい1 μm ×1 μm の面積内に200個ぐらいの密度で作られています。実際には、並べられたドットの上にガリウム砒素や、アルミニウム・ガリウム砒素などの保護層を置いています。ですから、このドットというのは地下深く埋蔵されたものなわけですね。ですから上からは平らに見えて、このドットがあるかどうかはわかりません。しかし図10に示すように、このプローブから出てくる近接場光でこのドットを基板を通して照明すると、電子の活性化によって生ずる近赤外の光が出てきます。当てる光は赤い光で波長が0.6 μm 、出てくる光の波長は0.8~0.9 μm です。そのスペクトルを見ようというものです。

もう1つの問題は、普通はこのドット1個からの近赤外の光は弱いので、こういった測定は試料を液体ヘリウム温度、すなわち絶対5~10度程度の極低温にまで冷却して近赤外の光の発生効率を増加させる必要があります。それから実際には、プローブを通して光を当てても、この保護層のところで電子が活性化されて、その電子がいろいろ動き回ります。そうすると、たとえば保護層のある位置で活性化された電子が、ずっと遠くまで行って、遠方のドットを活性化したりすることもあるわけです。それなので、必ずしもある位置で光を局所的に照射しても、逆に遠くの方から近赤外の光が出てくる場合があります。

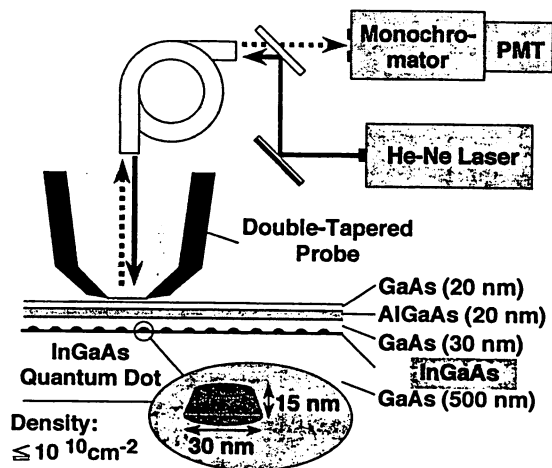


図10 InGaAs半導体の量子ドットおよび試料の構造とフォトルミネッセンス測定装置の説明

その光を全部集めてしまうと、1個1個のドットからの光を受けることにはなりません。そこで近赤外の光もこのファイバーの針を通して受けて検出します。すなわちプローブの先端から出てきた赤い光でドットを活性化して、ドットから出てきた近赤外の光を、もう1回プローブで集めて検出するというような、そういった効率の悪いことをせざるを得ません。けれども、プローブが適当な形をしていますと、首尾よく検出できます。

実際に極低温で測定してみますと、従来ですと図11の上部の曲線のような、数多くのドットからの発光スペクトルの包絡線しか見えなかったのですが、ここでは下部の曲線のように1本1本のスペクトルが見えるようになりました。

ドットは半導体ですので、電子が活性化され伝導帯と称するエネルギー準位に上がりますと、価電子帯と称するエネルギーの準位の中に、正孔と称する正の電荷を持つような擬似粒子が発生します。あるときに負の電荷を持つ電子が正の電荷を持つ正孔とくっつきますと、エネルギーを失って、そのかわりそのエネルギーが光となって出ます。実際にわれ

われは、この光を近赤外の光として見ているわけです。この電子と正孔のペアのことを励起子（エキシトン）という言い方をしていますが、この励起子が発光するのが、先ほどの近赤外の光です。

それから、同時に電子と正孔が2組ペアになる場合があります。これは励起子分子とよばれます。これは水素の原子核と電子が組み合わせあって出来る水素原子が2つ結合して水素分子を作るようなものです。

こういった水素分子状の励起子分子も光を出します。2つの励起子がペアをつくることによる結合エネルギー分だけ低いエネルギーの光が出ますので、実際には1個の励起子から出てくる光よりも、波長が少し長い光を出します。これをバイエキシトンとっているんですが、バイエキシトンのこのような励起子分子のスペクトルも見ることができると、ドットの中に電子と正孔のペアが1組あるものが発光しているのか、水素分子状の2組あるものが発光しているのか、という構造分析ができます。

さらに付け加えると、励起子でも、特に電子が一番最低のエネルギーの値をとるのが一番安定なんですが、このエネルギー準位をとり得る電子が中にいっぱいになりますと、次の電子は上のエネルギーの値をとります。この上のエネルギーにいる電子が正孔と結合して出す光は、高いエネルギーを持っており、したがって、短い波長の光になります。ですから、こういう波長の短いスペクトルが出たら、この中には高いエネルギーの電子があることがわかります。

実際には、広い波長範囲にわたってスペクトルをはかります。その結果が図12です。それから、この図12のパラメータは、ドットに当てる赤い光のパワー密度の大きさです。このパワー密度を上げていくと、最初のエネルギーの低い電子の発光が弱くなったり、逆にそのあとエネルギーの高い電子からの発光が増えたりというようなことがあって、それぞれ当てる光のパワー密度に対して一番エネ

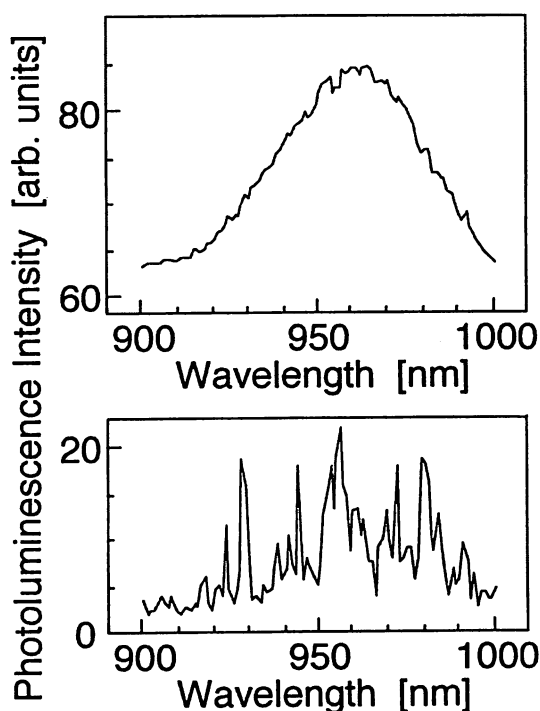


図11 量子ドットのフォトルミネッセンスのスペクトルの測定結果（上図：従来方法による下図：近接場の光による）

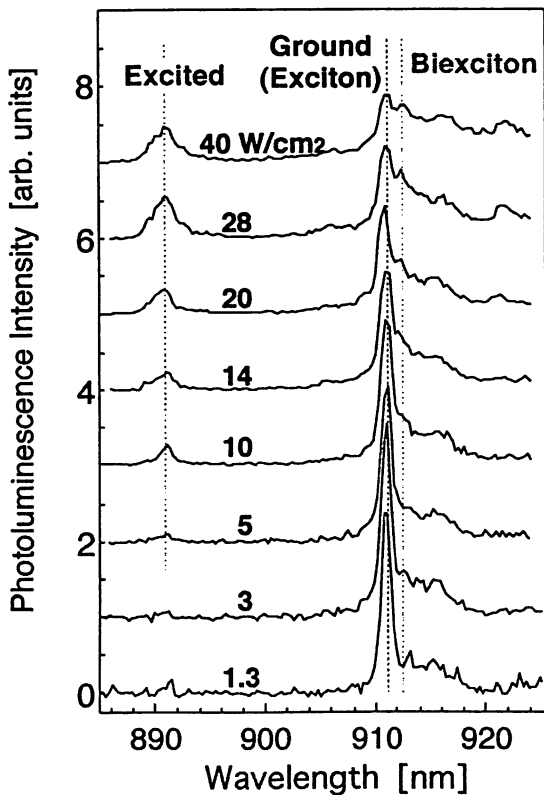


図12 照射する光のパワー密度をパラメータとした量子ドットのフォトルミネッセンスのスペクトルの測定結果

ギーの小さい電子が発光する強度、それから励起子分子が発光する強度、それから高いエネルギー準位の電子が発光する強度の振る舞いというのが違ってきます。このようなものを注意深く見ると、どのドットにどんなエネルギー状態の電子が含まれているかがわかります。

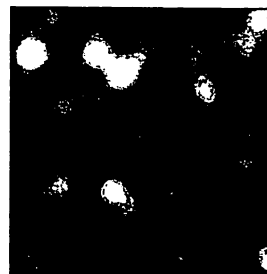
たとえばいろんなドットから出てくる発光の強度を、当てる光のパワー密度をだんだん増やしながら調べますと、最終的には図13のような分類ができて、励起子分子の発光、最低次のエネルギーを持つ励起子の発光、それからエネルギーの高い準位を持つ励起子の発光というように、すべてのドットに対してどんなエネルギー状態の電子が含まれているかがわかるようになります。

次は、加工するという話に入ります。いままでは、見たり、分析したりする話でしたので、物質の構造を変えたりするような積極的なことはしませんでした。しかし、次のよう

なやり方で加工することができます。加工の例としては、光メモリをご覧に入れます。

原理は簡単です。ガラスファイバースコープの後端から光を当てますと、近接場の光がプローブの先に発生します。この近接場の光のパワーは、1 pW (ピコワット) ~ 1 μW (マイクロワット) ぐらいです。ですから、伊賀先生のmW (ミリワット) などのレーザーの光からすると、3桁から9桁ぐらい小さい値です。しかし、このプローブの寸法も非常に小さく、数ナノメートルですので、単位断面積当たりのパワーの大きさ、いわゆるパワー密度は、1平方cm当たりに換算すると、100W ~ 1 kWになります。そうしますと、ファイバースコープの先に強いエネルギー密度の光が出ますので、この光を適当な膜、材料、結晶の板に当てますと、そこだけが集中的に加熱される場合があります。場合によってはそこだけが光化学反応を起こしまして、構造が変わります。それを利用して、この光のエネルギーで板に穴をあけるか、または構造を変えて、この光の直径の大きさ程度の寸法のメモリ、ビットを表す点1個と記録することができます。

この点を書いたことが、光メモリの記録に対応します。レコード針による録音というこ



3 μm x 3 μm

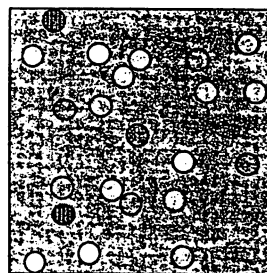


図13 各量子ドットから発光するフォトルミネッセンス像とその原因の帰属

とになります。再生は、もう少し弱い光を使って、この穴のあいたところや構造の変わったところを、顕微鏡として測定します。ここに穴や点があったということを知ると、これは記録の再生になるわけです。場合によっては、別の波長の光を入れますと、この穴を修復することができます。メモリの消去ということになります。これを繰り返しますと、消去可能なメモリが出来ます。

このような原理の実験は、われわれのところですでに、ずいぶん前にうまくいってしまっていて、たとえば有機材料の膜に、紫外線の光をファイバーを通して当てますと、図14のように直径が50nmぐらいの穴があきます。場合によっては、ファイバークローブを一直線上に走らせれば、線が描けたりする場合もあるわけです。

このような実験がきっかけになりまして、実は去年ぐらいから日本の国全体としても、こういった方法を使って、超高密度の光メモリを作る大型プロジェクトが進みつつあります。

なぜこんなことが採用されたかというのと、広がる光、集まる光を使って、従来の光メモリを作っていると、だいたい1平方インチ当たり1ギガビット程度の記録密度になります。しかし、光メモリの強大なライバルとして、磁気メモリ、ハードディスクメモリがありまして、その性能がどんどん上がってきますし、それを追い越そうとして、広がる光、集まる光を使っても、レンズで集められるスポットの大きさというのは、光の波長程度しか

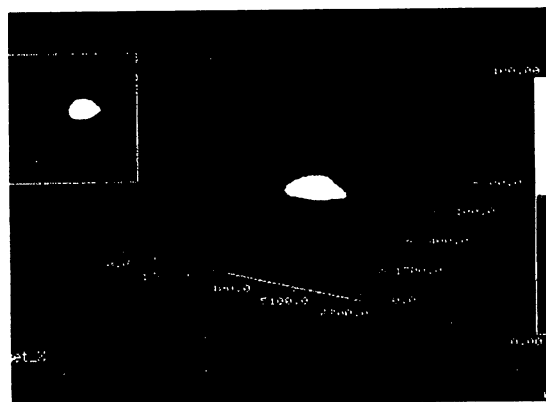


図14 有機薄膜に記録された直径50nmの円形スポットの像

ならないわけです。ですから、それよりも小さいものを加工してメモリを作ろうとしても、それは無理です。

けれども、われわれの場合には、原理的にはガラスのファイバーのプローブ程度の寸法まで小さな穴があきます。それは記録密度でいうと1平方インチ当たり1テラビットということになります。テラビットというのは1,000ギガビットということで、原理的には従来の光メモリの1,000倍の記録密度が得られるということです。

これはたとえば、動画像を1つのディスクの中に記録するとか、気象情報、医療情報、図書館1つ分ぐらいの文献情報を記録するとか、その程度の大きな容量を持っています。

そういうことで、将来の超高密度の光メモリの担い手の1つと言われて、図全体が応援をしてくださっているわけですが、ただし、1テラビットという値は大き過ぎることが問題です。したがってプローブで書くというような原理実験から、皆さんが日常的に使えるようになるためには、越えなければならない大きな技術的問題があります。

たとえばどんな情報を提供するかによって、書き換え可能な必要があるのか、記録だけでいいのかという問題がありますし、それからプローブのかわりにどんなデバイスを作って記録したり、再生する必要があるのかとか、プレイヤーそのものの装置はどういうふうにしたらいいのか、記録される材料としてはいままでの材料でいいのかどうか、などが問題になってきます。

さらに、たとえば1nmのように小さいところに光を当てたときに、そこで熱が発生します。普通は熱というのは周りに拡散していくわけですが、その拡散というのが、そんな小さいところでは、従来の大きなものに熱を起こしたときの拡散の状態と同じなのかどうか、という非常に物理の細かいところにかかわるような問題も、解決されなければいけません。

とはいっても、実際にはこれらを1つ1つ

解決していく必要がありまして、そのうちのわかりやすいような例を1つだけ挙げますと、いま開発が進み始めているのは、再生スピードを上げるために、プローブを何本も用意しまして、同時に何本かのプローブで書かせたり、読み出させることです。

けれども、活け花の剣山を逆さにしたようなプローブの束を作ったとしても、プローブの高さにばらつきがあって、このばらつきを10nm以下ぐらいにすることは、現在のマイクロプロセスと称する半導体の加工技術では無理だろうと言われていています。ですから言ってみれば、ここではわれわれが開発したような、1個1個の細いプローブを束ねて使うことはやめるということです。

そのかわりシリコンの板に適当なエッチングで小さな穴をあけまして、穴の上から光を当てて、穴のところに出てくる近接場の光を使って記録したり再生するほうがよからうということが言われ始めています。

この穴はばらつきなく作れますし、その穴をたとえば100個×100個、アレイ状に作ることもできますし、それから裏の面が平らですので、記録材料の上に非常に薄い潤滑剤を数ナノメートルぐらいの膜厚で塗りまして、その上にこれを滑らせると、精度よく適当なスピードで滑らせることができます。

図15は穴が2次元的にあいているシリコ

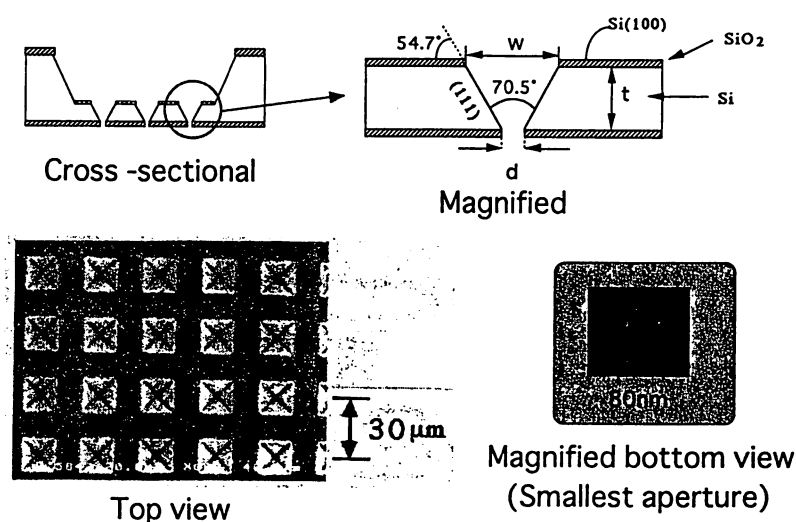


図15 シリコン結晶板に2次元的に穴の空いたプローブの構造と電子顕微鏡写真

ンの板で、その穴の1個を拡大すると、最小で80nmぐらいの四角い穴があいているものが出来ていることを表しています。

それから最近はさらに凝ったことができるようになってきていまして、上から光を当てて、穴のところに発生する近接場の光のパワーを上げるために、凸部に1 μm程度の直径をもつボールレンズを1個1個埋め込みまして、上から入ってくる光がこの穴のところで焦点を結ぶようなものもできるようになってきています。

さて、最後に、あやつるという話をさせていただきたいと思います。

いままでの話は、見る、分析する、それから加工するということだったのですが、その寸法は、もちろん光の波長よりははるかに小さく、だいたいナノメートルぐらいの寸法の物質を扱っていました。しかし、ナノの下はオングストロームで、物質を構成する基本的な構成要素としての原子の寸法となるわけです。近接場の光というのは小さいですし、パワーも弱いんです。だけど、相手が原子ぐらいまで小さくなってくると、その原子をかなりいろいろな自由度であやつることができるかもしれないという夢があるわけです。

ということで、いままでの話は、ナノの寸法のものを扱うというような光エレクトロニクスでしたので、ナノフォトニクスとよぶことにしますと、そのナノフォトニクスというのは画像計測だったり、分析だったり、メモリを作ったりするような加工だったりしたわけです。

もう1つ先に、アトムフォトニクスというのがあるのではないかと、ということです。それはたとえば真空中に浮いたり、飛行している原子を、近接場の光を使ってあやつるということです。

その原理は、図16のようになっています。すなわち近

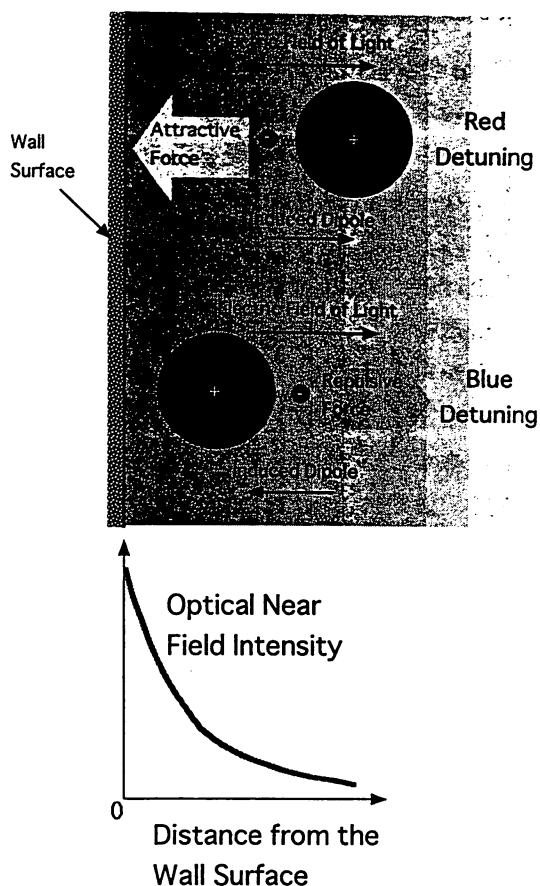


図16 近接場の光により発生する原子に対する双極子力の説明

接場の光を発生する物質の表面がここにあります。この図ではわかりやすいように表面が平面の場合について描いています。近接場の光のパワーの分布は、物質表面の法線方向に対して、急激に減衰します。その中に原子が入ったとします。近接場の光はもちろん電波の一種ですから、近接場の光が電磁波として持っている電界の振動方向に原子核と電子が変位しまして、同じ方向の分極が出来る場合と、それから場合によっては、同じ原子でも反対方向の分極が出来る場合と、両方あります。仮に、近接場の光の電界によって、電界が振動するのと同じ方向、言ってみれば同じ位相で振動するような分極が出来ると、出来た分極と電界との間の相互作用で、この原子は光のパワーの強い方向に引っ張られます。逆の方向の振動の仕方をするると、原子は光の弱い方向に引っ張られます。この力を双極子力といっていますが、こういうものを使うと、

真空中に浮いている原子が近接場の光の中に飛び込みますと、原子はこの物質の表面にくっつくか、はねかえされるか、ということになるわけです。

この実験は超高真空中で行います。くっつくか、はね返されるかは、使っている近接場の光の色によります。色というのは、実は言い方がおかしくて、色を区別するのは電磁波としての光の周波数です。毎秒何回、光の電磁波が振動しているかということです。

原子にはいろんな構造がありまして、その構造によって決まる原子に特別に共鳴する周波数がありまして、その周波数に対して光の色を表す周波数が若干高ければ、先ほどの図16のように、原子は物質表面からはね返されるような力を受けます。原子の特別の周波数に対して光の周波数がちょっと低ければ、原子は物質表面にくっつきます。

それを利用して、われわれは、ガラスのファイバースコープから近接場の光を出させて、この近接場の光の周波数を適当に調節しておく、場合によっては原子はくっついたり、はね返されるかもしれない、ということを考えています。しかし、これはなかなか難しく、一朝一夕には実現しませんので、その1つ前の方法を考えています。

そのためには、プローブの先の近接場の光ではなくて、図17に示すようにちくわ型のファイバーを用意します。ちくわですから中身が抜けているわけですが、内壁の周りを、断面がドーナツ状のコアが取り巻いているようなファイバーです。その外側はクラッドでコアを支えています。ドーナツ状のコアのと

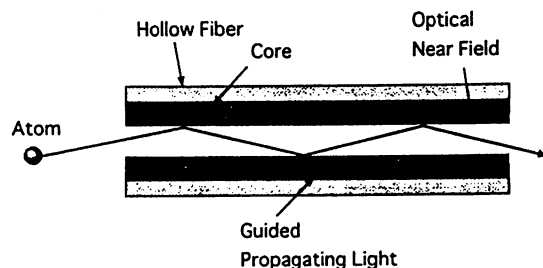


図17 中空ファイバ内壁面に発生する近接場の光による真空中の原子の誘導の説明

ころにドーナツ状の光を通しますと、この内壁のところに近接場の光が膜状に出てきます。もともと、ドーナツ状の光の周波数を、原子の構造によって決まる周波数よりも少し高くしておきます。そうすると、原子は内壁からはね返されます。すなわち、原子がちくわの中に飛び込んできたときに、この近接場の光の力を使って、ちくわの中を誘導することができるかもしれません。

原子がちくわの中を誘導されれば、誘導された原子をファイバプロブの先の近接場の光に当てて、そこでプロブの先でつかまえない、という希望もっています。

ということで、いくつかの実験を行ってみましたので、それらをご紹介します。日本のファイバメーカーの技術力はものすごく高く、このようなちくわのファイバが実際に出来ます。その内径の一番小さいのは、だいたい300nmぐらいですから、使う光の3分の1ぐらいの直径です。

いろんな原子で実験できるのですが、ここでご紹介するのは、ルビジウムという原子です。これはナトリウムの親戚です。ルビジウムは室温では水銀のような状態になっています。これを温めると蒸発し、ガス状に原子が吹き出します。それを穴のあいた板を通して、ある程度平行に真っ直ぐに飛ばし、その先にさきほどのちくわのファイバを置いておきます。同時にドーナツ状の光を真空の外から、窓を通して入れます。そうすると内壁に近接場の光の膜が、ドーナツ状にしみ出てきます。この中を原子が通り抜けるはずで、図18は、その実験結果の例で、光のパワーの増加とともに通り抜ける原子の数が増えることを示しています。ちなみに、使ったガラスファイバの内径は300nm程度、長さは3cmです。これは原子から見るとほぼ無限に大きい長さです。ちなみに、300nmの直径で3cmの長さというものを、両者1万倍していただくと、内径3mm、長さ300mとなりますので、このように細くて長い管の中を微粒子が通るといふことになります。

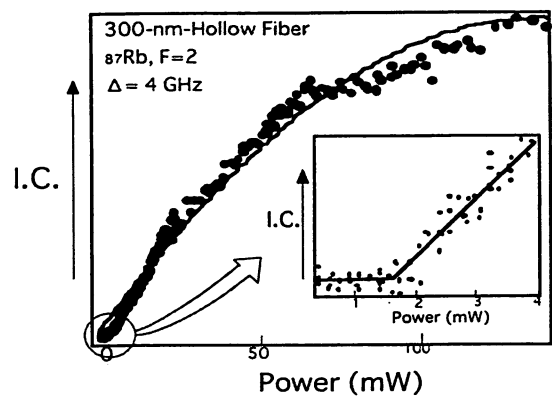


図18 内径300nmの中空ファイバを誘導された原子の数の測定結果
横軸はファイバに入射した光のパワー。縦軸は原子の数。内挿図は原点付近の拡大図。

応用には科学から技術までいろんなものがあります。たとえば、科学的には量子力学的な効果を検証する、技術的には飛び出した原子を結晶の板の表面に吹きつけて新しい物質を作っていく。それから化学の人はご存じかもしれませんが、同位体を分離するというようなものです。

それ以外にも、いくつか量子力学的な効果もあるんですが、時間が残り少なくなったので省略します。将来はちくわ型のファイバの中を通り抜けた原子をファイバプロブの先に出てきた近接場の光でつかまえ、さらにはつかまえた原子を基板の上にくっつけて、新しい物質を作る提案がなされていて、理論的な考察や、予備的な実験がなされています。

この技術というのは、実はルビジウムだけにしか対応できないということではなくて、目的とする原子が決まれば、原子の構造による特有の周波数に対応する光を使うことにより、いろいろな原子に対して使えます。たとえば紫外線を使いますと、シリコンをあやつることができます。言ってみれば、適当な色の光を出すレーザーの光源さえ用意できれば数多くの原子が扱えます。

今後、原子1個1個を小さな近接場の光で扱う技術が進むのではないかと期待しております。

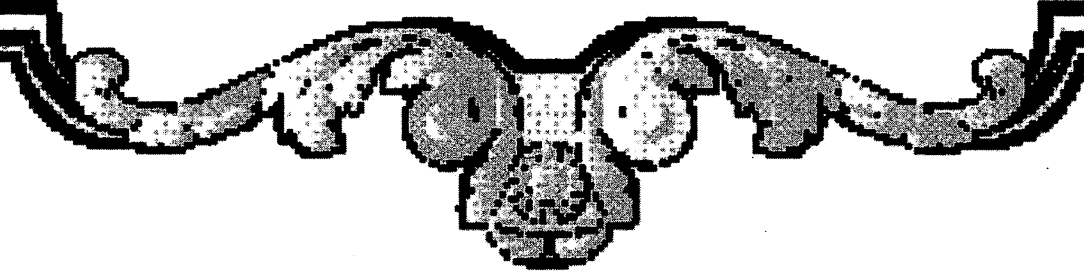
最後に今後の展望について述べます。過去には計測ということに主体が置かれて、近接

場の光を使って、分解能としては20nmぐらいというのがチャンピオンデータといわれていた時代があります。これが1980年代の中頃から90年代に入る頃です。最近はプローブ技術の開発が進んで、計測の場合の分解能が0.8nmぐらいになってきています。

今後は単なる計測、分析ではなくて、加工、それから1個の量子ドットにスイッチのような機能を持たせ、非常に小さな光ICのようなものが出来るかもしれません。それから究極の技術として、原子操作ができると期待され、近接場光の光の本質や、物質との総合性の本質を調べながら、先ほど申しましたナノフォトニクスとかアトムフォトニクスというのができるかもしれません。ただ、これらの技術やその基礎となる科学は、先ほど申しましたように、ナノ技術に支えられたものです。小さいものを加工できる人が、こういった方向に向かって進むことができますので、光だけではなくて、物質を、それから材料を絶えず加工する努力が、どうしても必要です。

ただ、非常に幸いなことに、この分野は非常に日本の強い分野で、総合力としては欧米各国をしのいでいるといわれています。そのようなことで、ナノフォトニクスとかアトムフォトニクスが、ある程度並行してできると、それを融合して、新しいタイプの光エレクトロニクス、フォトニクスの分野ができるのではないかと考えております。

ご静聴どうも有難うございました。



[II] PAPERS IN OTHER FIELDS

Modulation Characteristic of Waveguide-Type Optical Frequency Comb Generator

Takanori Saitoh, Shigenori Mattori, Shigeru Kinugawa, Koichiro Miyagi, *Member, IEEE*, Akira Taniguchi, *Member, IEEE, Member, OSA*, Motonobu Kourogi, and Motoichi Ohtsu, *Senior Member, IEEE, Member, OSA*

Abstract—We report the modulation characteristics of a waveguide-type optical frequency comb generator (WG-OFCG) with the advantages of compactness, high modulation index and low driving power. The characteristics of the optical frequency comb (OFC) generated when the modulation index exceeds 2π are discussed. The power of the modulation sidebands was measured by the optical heterodyne method with an external-cavity laser as a local oscillator. The dependence of the modulation index of the WG-OFCG on the modulation frequency up to 40 GHz was measured. The generation span of the OFC at the modulation frequency detuned from an integer multiple of the free spectral range (FSR) is discussed.

Index Terms—Frequency control, frequency domain synthesis, frequency measurement, optical pulses, optical waveguides, phase modulation, wavelength division multiplexing.

I. INTRODUCTION

AMONG all physical quantities, frequency can be observed with the highest precision. For example, the frequency of microwaves can be measured with a precision of better than 10^{-10} . However, the frequency of light cannot be measured directly because its value, which is as large as several hundred THz, is far beyond the bandwidth of conventional photo-detectors. Consequently, the frequency of light is estimated from its wavelength and the velocity of light. The wavelength is measured either by an optical spectrum analyzer (OSA) utilizing the wavelength selectivity of a diffraction grating, or by an optical wavelength meter (OWM) which counts the number of interference fringes. However, the accuracies obtained by an OSA and an OWM are limited to 10 GHz and 100 MHz, respectively. The wavelength division multiplexed (WDM) system is a candidate for high-capacity optical networking [1]. Each channel of the WDM system requires a light source with an accuracy of 1 GHz [2]. Although a laser diode (LD) with the frequency controlled by an OWM meets this requirement, it is not practical to use optical wavelength meters for all channels of the WDM system.

On the other hand, a low-cost and high-precision method has been used to control the LD frequency by using the spectrum of an atom or molecule with a known absorption frequency as a frequency discriminator [3], [4]. By using this method, the LD frequency accuracy reaches 100 MHz. However, the system requires the same number of frequency references as LD's, and the LD frequencies cannot be controlled without the absorption frequency of the atom or molecule. To overcome these difficulties, an optical frequency comb generator (OFCG) has been developed to generate many modulation sidebands from a frequency-reference laser [5]–[13]. The principle of the OFCG is based on a Fabry–Perot electrooptic modulator [14]. The OFCG is composed of an optical phase modulator mounted in an optical resonator. The laser light propagating in the OFCG is deeply phase modulated by traveling back and forth in the phase modulator; the output has many modulation sidebands separated from each other by the phase modulation frequency. Each modulation sideband has the same frequency stability and linewidth as those of the frequency-reference laser. The first OFCG was composed of a microwave cavity and an optical phase modulator in an optical resonator composed of two discrete mirrors [5]–[8]. However, to increase the generation efficiency, a bulk-type OFCG (B-OFCG) with an optical resonator fabricated by coating high-reflection films on the two end facets of a monolithic phase-modulation crystal [9], and a multiple-cavity-type OFCG with three or four mirrors, were developed [10].

A waveguide-type optical frequency comb generator (WG-OFCG) [15]–[20] with the advantages of compactness, high modulation index, and low driving power has been developed. This paper reports the characteristics of the OFC when the modulation index exceeds 2π , as well as the dependence of the modulation index on modulation frequencies up to 40 GHz. The generation span of the OFC at the modulation frequency detuned from an integer multiple of the free spectral range (FSR) is discussed.

II. STRUCTURE OF WAVEGUIDE-TYPE OPTICAL FREQUENCY COMB GENERATOR

Fig. 1 shows the structure of the WG-OFCG. This was fabricated originally for the optical-waveguide-type phase modulator operating at a wavelength of $1.5 \mu\text{m}$ (Anritsu SCB05A), and the optical resonator was formed by coating the two end facets of the waveguide with high-reflection films. The reflectivities of the high-reflection films are estimated to be

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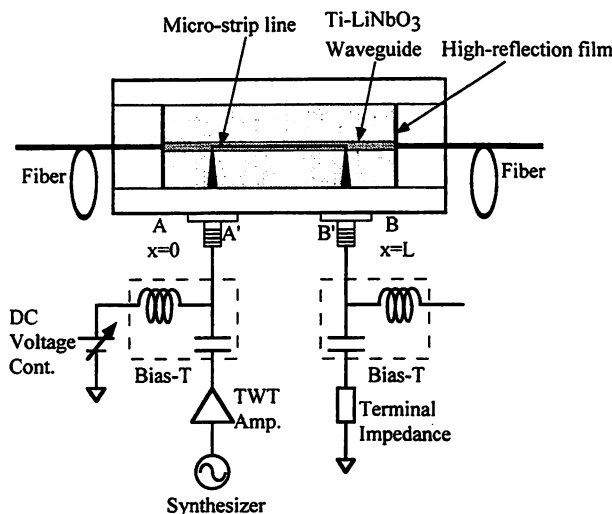


Fig. 1. Schematic illustration of WG-OFCG.

TABLE I
PERFORMANCE OF WG-OFCG AND B-OFCG

	WG-OFCG	B-OFCG
Modulation index	$>2\pi$	0.2π
Driving microwave power	1 W	10 W
Finesse	30	300
Sideband power	$\propto \exp\left(-\frac{2 k \pi}{mF}\right)$	$\propto \exp\left(-\frac{ k \pi}{mF}\right)$

96%. Two polarization-maintaining fibers were attached to the two end facets. The waveguide was formed by diffusing titanium into a lithium-niobate (LiNbO₃) substrate. A microstrip line was formed on the top surface of the waveguide. The velocity of the microwave propagating in the microstrip line was matched to the velocity of the light propagating in the waveguide by controlling the thickness of the microstrip line. Since this phase modulator is a traveling-wave type, only the light copropagating with the microwave is modulated. This phase modulator can carry out phase shifting of π radian by applying a voltage of 5.5 V-dc. The maximum phase modulation index resulting from applying microwaves with a frequency of 10 GHz is about 2π . The waveguide length was 38 mm. The measured FSR and finesse of the optical resonator were 1.905 GHz and 30, respectively. The light transmittance of the resonator at the resonance frequency was about 5%. Table I shows the performance of the WG-OFCG and a B-OFCG. The WG-OFCG is characterized by a high phase modulation index and low finesse compared to the B-OFCG.

The microwave output from a synthesizer with a bandwidth and maximum power of 1–40 GHz and 10 dBm, respectively,

is amplified by a traveling wave tube (TWT) amplifier with a bandwidth and maximum power of 4–18 GHz and 43 dBm, respectively, transmitted to a bias-T on the input port (A' in Fig. 1) and input to the microstrip line of the WG-OFCG. There is another bias-T on the output port (B' in Fig. 1) of the microstrip line with a 50- Ω terminal impedance. To adjust the resonance frequency of the optical resonator, a dc voltage is applied to the microstrip line via the bias-T on the input port.

III. MODULATION INDEX DEPENDENCE OF OPTICAL FREQUENCY COMB

As shown in the Appendix, the WG-OFCG resonance frequency is modulated with a frequency of f_m and an amplitude of $m_0 \text{FSR}/(2\pi)$ where m_0 and FSR are the modulation index and free spectral range of the optical resonator, respectively. Therefore, the WG-OFCG can be regarded as a shutter that allows light transmission when the resonance frequency matches the injected light frequency ν_0 .

A. Shallow Modulation ($m_0 < 2\pi$)

When m_0 is smaller than 2π , the light output from the WG-OFCG is composed of a superposition of two pulse trains with a repetition frequency of f_m . Each waveform of the pulse is similar to the resonance spectral profile of the WG-OFCG, and its half-height peak width is $1/(m_0 \cos \theta F f_m)$, where $\theta = \sin^{-1}\{2\pi(\nu_0/\text{FSR} - M)/m_0\}$ and M and F are the maximum integer in $<\nu_0/\text{FSR}$ and the finesse of the optical resonator, respectively. Consequently, from approximation of (A5) in the Appendix, the electric field of the light output from the WG-OFCG $E(t)$ can be described as shown in (1) at the bottom of the page where E_{in} is the electric field amplitude of the injected light. By Fourier transformation of (1), the electric field of the k th sideband $E_k(t)$ can be expressed as

$$\begin{aligned}
 E_k(t) &= f_m \int_{-1/(2f_m)}^{1/(2f_m)} E(t) \exp(-2\pi f_m kt) dt \\
 &= \frac{\pi E_{\text{in}} A}{m_0 \cos \theta F} \exp\left(-\frac{|k|\pi}{m_0 \cos \theta F}\right) \\
 &\quad \times \exp\{j2\pi(\nu_0 + kf_m)t\} \\
 A &= \begin{cases} 1 & k \geq 0 \\ e^{j\pi k} & k < 0 \end{cases} \quad (2)
 \end{aligned}$$

This equation shows that the sideband power decreases exponentially with increasing $|k|$. To generate sidebands over a wide frequency range, the product of $m \cdot F$ must be enlarged. The sidebands have maximum power when $\theta = 0$, that is, when the injected light frequency is equal to the unmodulated resonance frequency of the resonator. As shown in Table I, at the same value of $m \cdot F$, the sideband powers generated by the WG-OFCG and the B-OFCG are different. In the WG-OFCG,

$$E(t) \cong \sum_{l=-\infty}^{\infty} \left\{ \frac{1}{1 + j2m_0 \cos \theta F f_m \left(t - \frac{2\pi l + \theta}{2\pi f_m}\right)} + \frac{1}{1 - j2m_0 \cos \theta F f_m \left(t - \frac{2\pi l - \theta + \pi}{2\pi f_m}\right)} \right\} \times E_{\text{in}} e^{j2\pi\nu_0 t} \quad (1)$$

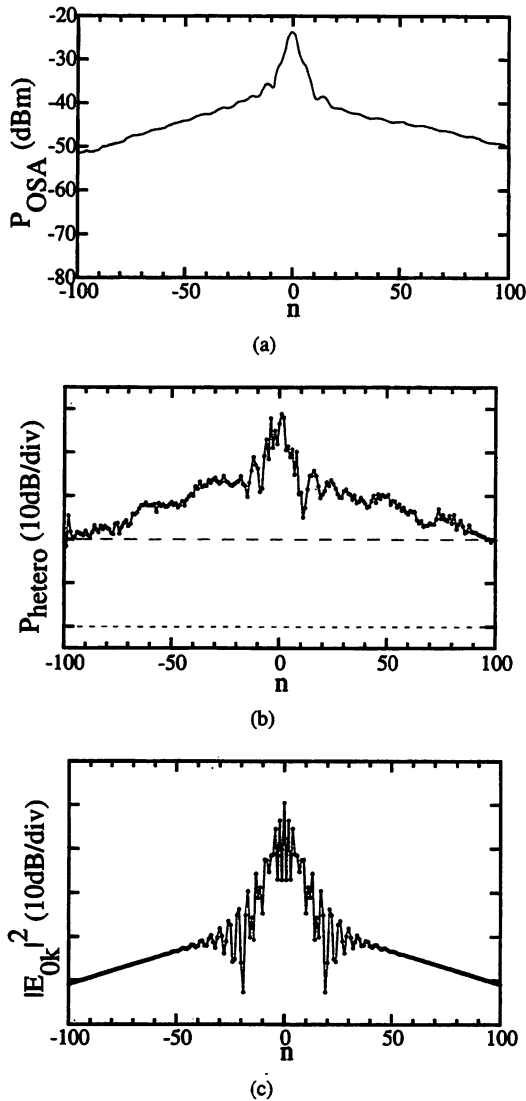


Fig. 2. OFC's at $\theta = 0$: (a) OSC measured by OSA with resolution of 0.1 nm (n = order of modulation sideband), (b) powers of heterodyne signals between n th modulation sideband and external-cavity laser, and (c) OSC calculated from (5).

only the light copropagating along the traveling microwave, is modulated. In the B-OFCG, the propagating light is modulated as it travels back and forth along the phase modulator by the standing microwave.

B. Deep Modulation ($m_0 > 2\pi$)

In the B-OFCG, it is difficult to apply a large electric potential gradient to the LiNbO_3 crystal used as the phase modulator, because the crystal thickness is in the order of several mm. Consequently, the modulation index m_0 is limited to 0.1–0.2 π even when a microwave power as high as 10 W is applied to the crystal. However, in the WG-OFCG, a large electric potential can be applied to the waveguide because it is as thin as several μm . Therefore, WG-OFCG phase modulation can exceed 2π . The modulation index is proportional to the square root of the microwave power, and is described as

$$m_0 = 2.2\pi \sqrt{P(W)}. \quad (3)$$

The microwave power is restricted by the heat resistance of the WG-OFCG and is about 1 W. However, the WG-OFCG can achieve a phase modulation index as large as 2.2π at a microwave power of 1 W. At $m_0 < 2\pi$, the condition of $\theta < m_0$ is required for generation of the OFC [8]. However, at $m_0 > 2\pi$, the OFC can be generated irrespective of θ because it is always smaller than m_0 . Figs. 2(a) and 3(a) show OFC's generated when $\theta = 0$ (when the unmodulated resonance frequency of the WG-OFCG [ν_{R0}] is tuned to the injected light frequency (ν_{LD}]) and $\theta = \pi$ (when ν_{R0} is maximum detuned from ν_{LD}) measured by an OSA at a resolution bandwidth of 0.1 nm. The modulation index and the modulation frequency are 2.2π and 3.81 GHz, respectively. For ν_{R0} tuning, a dc voltage was applied to the microstrip line via the bias-T. As shown in these figures, both OFC's have similar spans, but the OFC at $\theta = \pi$ has large ripple in its envelope. To examine the cause of the ripple, the powers of the modulation sidebands were calculated as shown below. The light output from the WG-OFCG at $m_0 > 2\pi$ is composed of a superposition of four or more pulse trains as shown (A5). The electric field of the light output from the WG-OFCG $E_0(t)$ and the k th modulation sideband $E_{0k}(t)$ at $\theta = 0$ are described as

$$E_0(t) \cong \sum_{l=-\infty}^{\infty} \left\{ \begin{aligned} & \frac{1}{1 + j2m_0 F f_m \left(t - \frac{l}{f_m} \right)} \\ & + \frac{1}{1 - j2m_0 F f_m \left(t - \frac{2\pi l + \pi}{2\pi f_m} \right)} \\ & - \frac{1}{1 + j2m_0 \cos \varphi_0 F f_m \left(t - \frac{2\pi l + \varphi_0}{2\pi f_m} \right)} \\ & + \frac{1}{1 - j2m_0 \cos \varphi_0 F f_m \left(t - \frac{2\pi l + \varphi_0 + \pi}{2\pi f_m} \right)} \\ & - \frac{1}{1 + j2m_0 \cos \varphi_0 F f_m \left(t - \frac{2\pi l - \varphi_0}{2\pi f_m} \right)} \\ & + \frac{1}{1 - j2m_0 \cos \varphi_0 F f_m \left(t - \frac{2\pi l - \varphi_0 + \pi}{2\pi f_m} \right)} \end{aligned} \right\} \times E_{in} e^{j2\pi\nu_0 t}, \quad \text{where } \varphi_0 = \sin^{-1} \left(\frac{2\pi}{m_0} \right) \quad (4)$$

and

$$E_{0k}(t) = \left\{ \begin{aligned} & \frac{\pi}{m_0 F} \exp \left(-\frac{\pi|k|}{m_0 F} \right) + \frac{2\pi}{\sqrt{m_0^2 - 4\pi^2 F}} \\ & \times \exp \left(-\frac{\pi|k|}{\sqrt{m_0^2 - 4\pi^2 F}} \right) \cos \varphi_0 k \end{aligned} \right\} \times E_{in} A e^{j2\pi(\nu_0 + k f_m)t}. \quad (5)$$

Similarly, their values at $\theta = \pi$, $E_\pi(t)$, and $E_{\pi\kappa}(t)$ are described as

$$E_\pi(t) \cong \sum_{l=-\infty}^{\infty} \left\{ \begin{aligned} & \frac{1}{1 + j2m_0 \cos \varphi_\pi F f_m \left(t - \frac{2\pi l + \varphi_\pi}{2\pi f_m} \right)} \\ & + \frac{-1}{1 - j2m_0 \cos \varphi_\pi F f_m \left(t - \frac{2\pi l + \varphi_\pi + \pi}{2\pi f_m} \right)} \\ & + \frac{-1}{1 + j2m_0 \cos \varphi_\pi F f_m \left(t - \frac{2\pi l - \varphi_\pi}{2\pi f_m} \right)} \\ & + \frac{1}{1 - j2m_0 \cos \varphi_\pi F f_m \left(t - \frac{2\pi l - \varphi_\pi + \pi}{2\pi f_m} \right)} \end{aligned} \right\} \times E_{in} e^{j2\pi\nu_0 t} \quad \text{where } \varphi_\pi = \sin^{-1} \left(\frac{\pi}{m_0} \right) \quad (6)$$

and

$$E_{\pi k} = \left\{ \frac{2\pi}{\sqrt{m_0^2 - \pi^2 F}} \exp \left(-\frac{\pi|k|}{\sqrt{m_0^2 - \pi^2 F}} \right) \sin \varphi_\pi k \right\} \times E_{in} A e^{j2\pi(\nu_0 + k f_m)t} \quad (7)$$

From (5) and (7), the slopes of the envelopes at $m_0 = 2.2\pi$ are 0.985 and 0.991 [dB/order of modulation sideband], respectively. Since these values are similar, the OFS's at $\theta = 0$ and $\theta = \pi$ have similar widths. The envelope of the OFC at $\theta = \pi$ has a ripple with a period of about 7 [= $\pi/(2\varphi_\pi)$] sidebands. The calculated results agree with the measured results as shown in Figs. 2(a) and 3(a). From (6) and (7), the ripple is due to interference between the OFC's caused by pulse trains with different phases.

To estimate the span width of an OFC that can be used as a frequency reference, the heterodyne signal between a local oscillator and n th modulation sideband in the range of $-100 \leq n \leq 100$, was measured. To perform measurement, laser light emitted from a DFB laser with a linewidth of 10 MHz and a power of 10 mW was injected into the WG-OF CG. An external-cavity laser with a linewidth of 100 kHz and a power of 2 mW was used as a local oscillator. Fig. 4 shows the heterodyne signal between the external-cavity laser light and the -40 th modulation sideband measured by an RF spectrum analyzer at a resolution bandwidth of 3 MHz. Figs. 2(b) and 3(b) show the powers of the heterodyne signals at $\theta = 0$ and $\theta = \pi$, respectively. The dotted line is the noise level detected by the RF spectrum analyzer.

If frequency offset locking is performed using heterodyne signal feedback, heterodyne detection requires a bandwidth of about 300 MHz [20]. Consequently, the noise level at frequency offset locking is 20 dB larger than at 3 MHz bandwidth detection as shown in Fig. 4. The broken lines in Figs. 2(b) and 3(b) are the noise levels when the detection bandwidth is 300 MHz. From this estimation, it is found

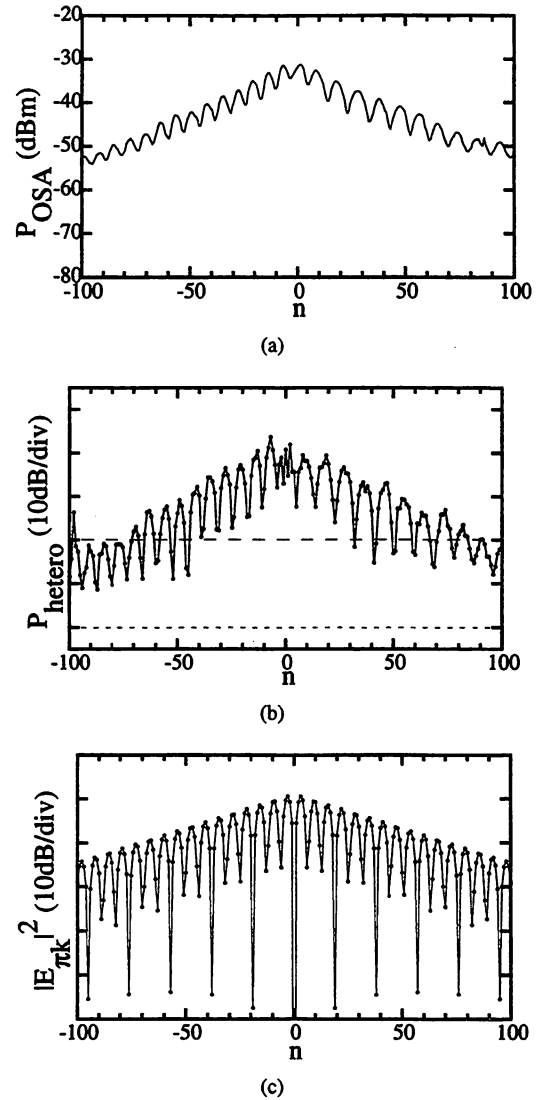


Fig. 3. OFC's at $\theta = \pi$: (a) measured by OSA, (b) powers of heterodyne signals, and (c) OSC calculated from (7).

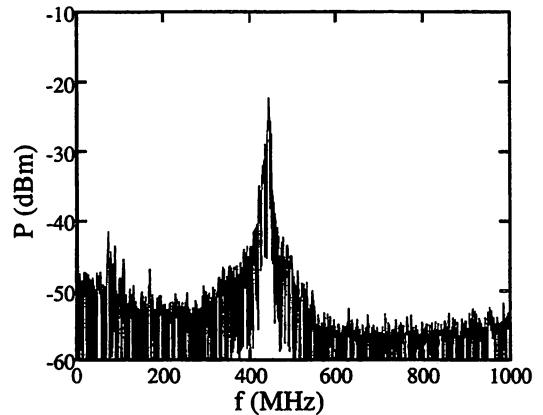


Fig. 4. Heterodyne signal between external-cavity laser and -40 th modulation sideband.

that the modulation sidebands of $|k| < 100$ can be used for frequency offset locking. Equations (5) and (7) show that the modulation sideband power depends on θ although the span of the OFC is held. Fig. 5 shows this dependence of the powers

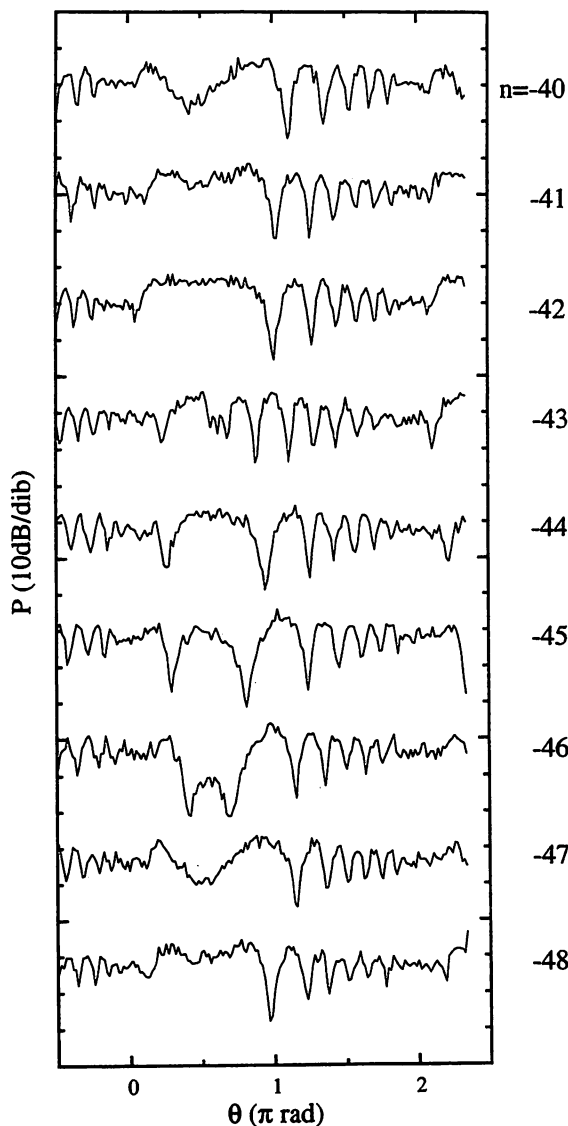


Fig. 5. Dependence of power of heterodyne signal between n th modulation sideband and the external-cavity laser on θ .

of the heterodyne signal between the external-cavity laser light and the modulation sidebands with orders from -40 th to -48 th. The value of θ was adjusted by controlling the dc voltage applied to the microstrip line. As shown in the figure, the power of the heterodyne signal decreases by about 20 dB when θ is varied. The condition of $|\theta| < 0.1\pi$ is required to stabilize the power of the heterodyne signal.

IV. MODULATION FREQUENCY DEPENDENCE OF OPTICAL FREQUENCY COMB

When $f_m = N \cdot \text{FSR}$ (N : integer), modulation sidebands can be generated with high efficiency. Fig. 6 shows OFC's measured by an OSA with a resolution of 0.1 nm at $N = 2-20$. The power of the injected laser light was 10 dBm. The solid curves in this figure are the results when a microwave with a power of 10 dBm output from the synthesizer was applied directly to the WG-OFCG. The dotted curves are the results when a microwave with a power of 30 dBm amplified by the TWT amplifier was applied to the WG-OFCG. Since the

TWT amplifier has a bandwidth of 4–18 GHz, a microwave with a power of 30 dBm was applied to the WG-OFCG only when $N = 2-10$. When $N \geq 8$, separate sidebands are observed because the modulation frequencies are higher than the resolution bandwidth of the OSA.

Fig. 7 shows the modulation indices estimated from Fig. 6 and (2) and the microwave transmission coefficient of the microstrip line. The open and filled circles are the estimated results for microwave powers of 10 and 30 dBm, respectively. This figure shows that the modulation index does not depend on f_m when $f_m \leq 10$ GHz. The solid curve shows the microwave transmission coefficient measured by a network analyzer. When $f_m \leq 10$ GHz, the microwave transmission coefficient does not depend on f_m . However, when $f_m > 10$ GHz, the microwave transmission coefficient decreases monotonically. The modulation index is in proportion to square root of the applied microwave power as shown in (3). Consequently, the modulation index decreases monotonically when $f_m > 10$ GHz.

The modulation index decreases with increasing modulation frequency, but the span of the OFC increases due to expansion of the space of the modulation sidebands. Fig. 8 shows the modulation frequency dependence of the span of the OFC when the powers of the modulation sidebands exceed -70 dBm. The open and filled circles are the span of the OFC's for microwave powers of 10 and 30 dBm, respectively. From this figure, it is found that the span of the OFC increases when the modulation frequency increases from 0 to 10 GHz, reaching the maximum at a modulation frequency of about 10 GHz.

V. ADJUSTING SPACE OF SIDEBANDS

If the WG-OFCG is used in the WDM system as a frequency reference, it would be convenient to space sidebands at $1/\text{integer}$ of the grid space required by the WDM system. For example, when the grid space is set to 25 GHz, the modulation frequency required by the WDM system is 25, 12.5, 6.25, ... GHz. If the modulation frequency is set to 12.5 GHz, it is advantageous to set FSR of the WG-OFCG to 1.786 GHz ($=12.5/7$). Consequently, the length of the waveguide must be 38.45 mm because the group index of the waveguide is 2.183. In general, since the accuracy of the waveguide length determining by polishing is about 0.05 mm, the optimum modulation frequency for the fabricated WG-OFCG (integer multiple of FSR of optical resonator) is 12.5 ± 0.01 GHz, and the allowable difference from the modulation frequency required by the WDM system may be 0.01 GHz. If the modulation frequency deviates from the optimum modulation frequency, the OFCG cannot perform high-efficiency phase modulation. Fig. 9 shows the relationship between the detuning frequency from the optimum modulation frequency $\delta f_m (= |f_m - N \cdot \text{FSR}|)$ and the span of the envelope of the modulation sidebands with powers exceeding -70 dBm in the OFC. The filled circles are the measured results for a WG-OFCG with a finesse, modulation index, FSR and modulation frequency of 30, 2π , 1.905, and 11.43 GHz, respectively. The open circles are the measured results for a WG-OFCG with a finesse, modulation index, FSR and modulation frequency of

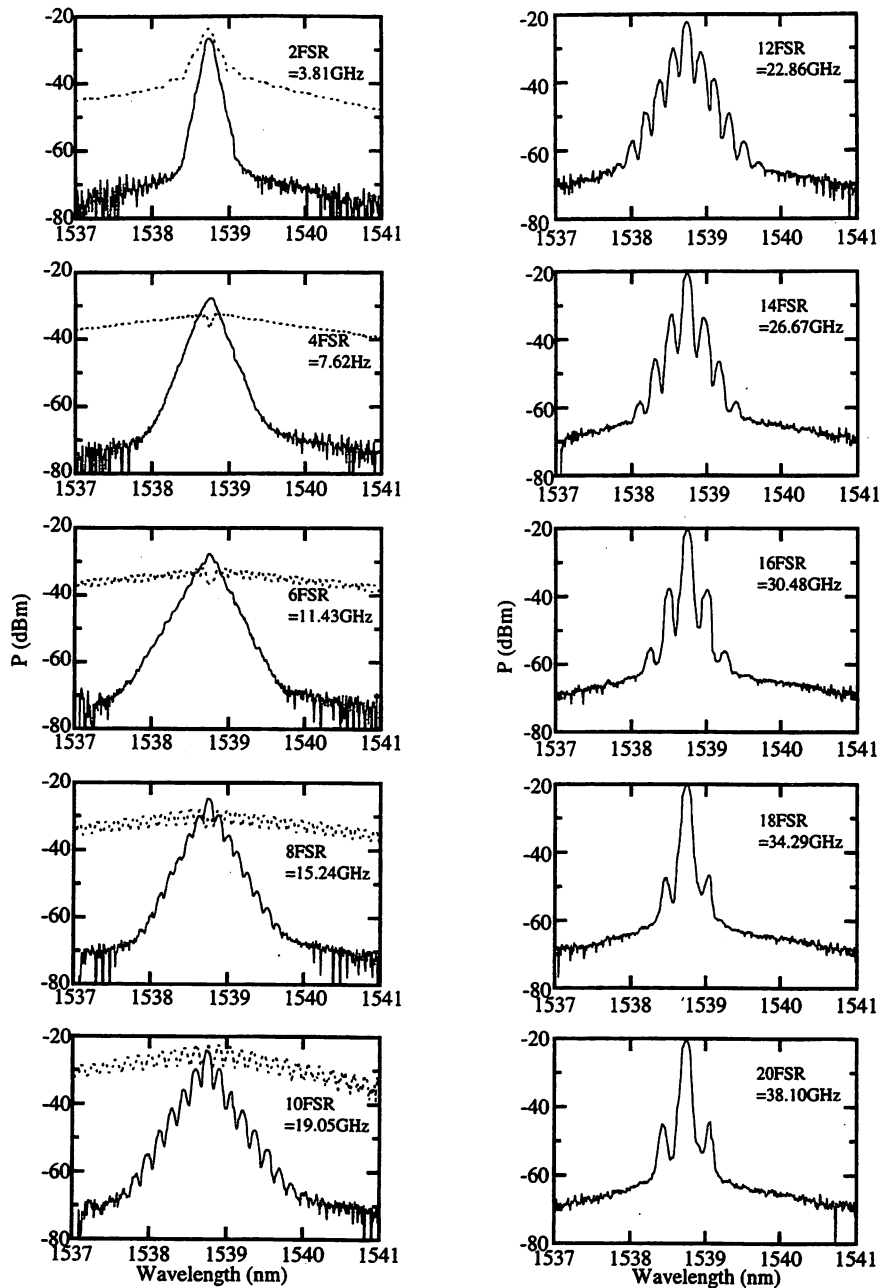


Fig. 6. OFC's measured by OSA with resolution of 0.1 nm for various modulation frequencies. The solid and dotted curves are the measured results for microwave powers of 10 and 30 dBm, respectively.

10, 2π , 1.692, and 10.152 GHz, respectively. The WG-OFCG with a finesse of 10 was made by degrading the reflectivities of the facets to about 70%. The dotted and solid lines are calculated by the numerical Fourier transformation of (A5) for finesses of 10 and 30, respectively. As shown in this figure, the span of the OFC is independent of the detuning frequency for $\delta f_m < \delta f_{m-c}$, and decreases monotonically for $\delta f_m > \delta f_{m-c}$, which is represented by using the cut-off detuning frequency δf_{m-c} . At the maximum detuning frequency ($\delta f_m = 0.5\text{FSR}$), the span of the OFC is minimum. The detuning frequency has no effect on the performance of the WG-OFCG, even if the optimum modulation frequency deviates 10 MHz ($< \delta f_{m-c}$) below the modulation frequency required by the WDM system, due to polishing inaccuracy.

The effect of detuning becomes larger with increasing finesse because finesse corresponds to the number of laser light round trips in the resonator. Fig. 10 shows the calculated span of the OFC versus the detuning frequency for a range of finesses from 10 to 60. The cut-off detuning frequency decreases with increasing finesse. At $\delta f_m = 0$, the span of the OFC increases with increasing finesse. However, when $\delta f_m > 0$, the span of the OFC has the smallest upper limit, represented by the dotted line. Fig. 11 shows the relationship between the finesse and the span of the OFC at $\delta f_m = 10$ MHz. The span of the OFC increases when the finesse increases from 0 to 30, but when the finesse exceeds 30, the span is nearly constant. This confirms that when the finesse of the OFCG is 30, the OFC is not affected by detuning of the modulation

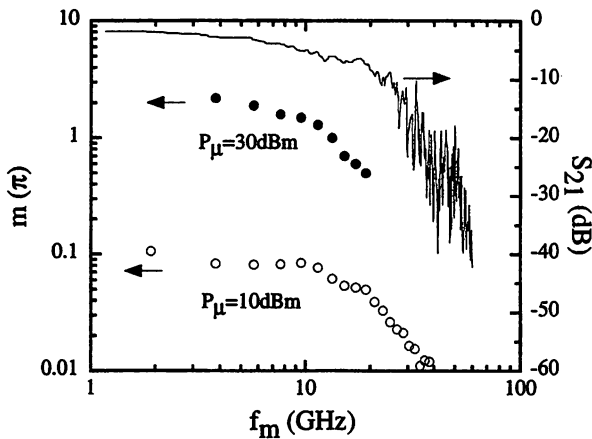


Fig. 7. Modulation index m estimated from Fig. 9 and (2) and microwave transmission coefficient S_{21} of microstrip line of WG-OFCG. The open and filled circles are the estimated results for microwave powers of 10 and 30 dBm, respectively. The solid curve is the S_{21} of the microstrip line measured by a network analyzer.

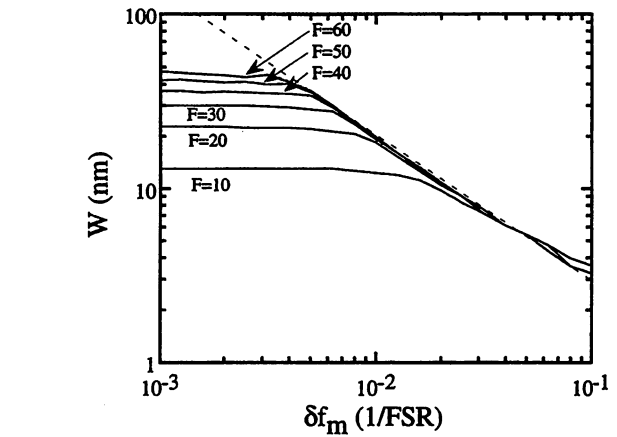


Fig. 10. Span of OFC W calculated from numerical Fourier transformation of (A5) versus detuning frequency δf_m for finesses range from 10-60.

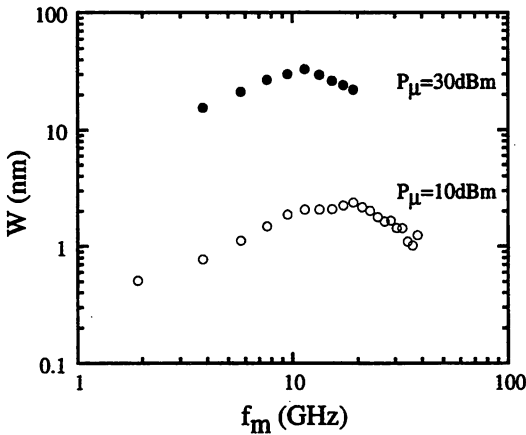


Fig. 8. Relationship between modulation frequency f_m and span W of envelope of modulation sidebands with powers exceeding -70 dBm. The open and filled circles are the measured results for microwave powers of 10 and 30 dBm, respectively.

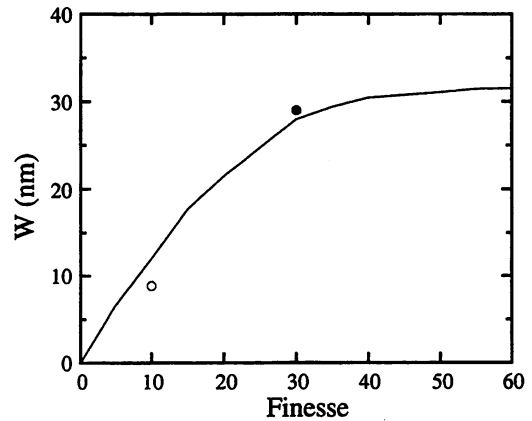


Fig. 11. Relationship between finesse and span of OFC W at $\delta f_m = 10$ MHz. The solid curve is the span of the OFC calculated from numerical Fourier transformation of (A5). The open and filled circles are the measured results for finesses of 10 and 30, respectively.

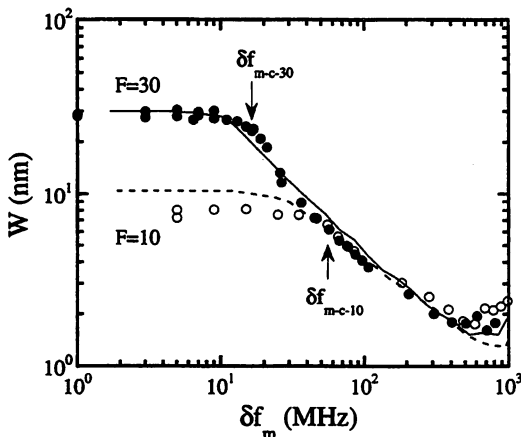


Fig. 9. Relationship between detuning frequency δf_m and span W of envelope of modulation sidebands with powers exceeding -70 dBm. The open and filled circles are the measured results for finesses of 10 and 30, respectively. The dotted and solid curves are the calculated results from numerical Fourier transformation of (A5) for finesses of 10 and 30, respectively.

frequency, even if the detuning modulation frequency due to polishing inaccuracy of the waveguide length is about 10 MHz. Therefore, a WG-OFCG, with a modulation index exceeding 2π and a finesse of 30 is a promising candidate for the frequency reference for the WDM system.

VI. SUMMARY

We have discussed a WG-OFCG with the advantages of compactness, high modulation index and low power. The OFC generated when the modulation index exceeds 2π is discussed. The powers of the modulation sidebands were measured by heterodyne detection. The dependence of the modulation index on the modulation frequency up to 40 GHz was measured. The generation span of the OFC at the modulation frequency detuned from an integer multiple of the FSR, is discussed. It is found that the span of the OFC is not affected by detuning of 10 MHz when the finesse of the OFCG is 30. The WG-OFCG is useful for adjusting the space of the modulation sidebands due to its low finesse and high modulation index.

APPENDIX

In Fig. 1, the microwave injected from A' travels in the microstrip line and is output from B' . Since the velocity of the

$$\begin{aligned}
m_k &= \begin{cases} m_0 \sin \left\{ 2\pi f_m \left(t - \frac{nL}{c} \right) \right\} & (k = 0) \\ \sum_{q=0}^k m_0 \sin \left\{ 2\pi f_m \left[t + \frac{(2q - 2k - 1)nL}{c} \right] \right\} - \sum_{q=0}^{k-1} \frac{c}{2\pi f_m nL} m_0 \\ \quad \times \sin \left\{ 2\pi f_m \left[t + \frac{2(q - k)nL}{c} \right] \right\} \sin \left(\frac{2\pi f_m nL}{c} \right) & (k > 0) \end{cases} \\
&= m_0 \sin \left\{ 2\pi f_m \left[t - \frac{(k+1)nL}{c} \right] \right\} \left\{ \frac{\sin \left[\frac{2\pi f_m (k+1)nL}{c} \right]}{\sin \left(\frac{2\pi f_m nL}{c} \right)} - \frac{c}{2\pi f_m nL} \sin \left(\frac{2\pi f_m k nL}{c} \right) \right\}. \quad (A3)
\end{aligned}$$

$$\begin{aligned}
E(t) &= E_{\text{in}} (1 - R) \sum_{k=0}^{\infty} R^k \exp \left(j \left\{ 2\pi \nu_0 \left[t - \frac{(2k+1)nL}{c} \right] + m_k \right\} \right) \\
&= E_{\text{in}} \frac{(1 - R)}{\exp \left\{ -j \left[-\frac{\pi \nu_0}{\text{FSR}} + m_0 \sin(2\pi f_m t) \right] \right\} - R \exp \left(-j \frac{\pi \nu_0}{\text{FSR}} \right)} e^{j2\pi \nu_0 t} \quad (A5)
\end{aligned}$$

microwave is matched to the velocity of the light propagating in the waveguide, the electric potential $V(x, t)$ is described as

$$V(x, t) = V_0 \sin \left\{ \frac{2\pi f_m n}{c} \left(\frac{c}{n} t - x \right) \right\} \quad (A1)$$

where V_0 , f_m , n , and c are the amplitude and frequency of the microwave, the index of the waveguide, and the velocity of the light in the vacuum, respectively. Propagating light starting from A ($x = 0$) at $t = t_1$ accepts a constant electric potential $V = V_0 \sin(2\pi f_m t_1)$ until its arrival at B ($x = L$). After the light is reflected at B at $t = t_2$, it propagates to A and accepts an electric potential described as $V = V_0 \sin\{2\pi f_m(2t - t_2 - nL/c)\}$. If the phase modulation index is m_0 , corresponding to the microwave of (A1), the phase modulation accepted when the light propagates from A to B and from B to A in the waveguide is described as

$$m = \begin{cases} \frac{m_0 \sin(2\pi f_m t_1)}{c} & (A \rightarrow B) \\ \frac{2\pi f_m nL}{c} m_0 \sin(2\pi f_m t_2) \\ \quad \times \sin \left(\frac{2\pi f_m nL}{c} \right) & (B \rightarrow A). \end{cases} \quad (A2)$$

From (A2), the light injected from A at $t - (2k+1)nL/c$, propagates $2k+1$ times in the waveguide, is output from B at t , and is phase modulated as shown in (A3) at the top of the page. Equation (A3) shows that the effect of the phase modulation is integrated by the light traveling back and forth in the waveguide when $f_m N \cdot 2nL/c = N \cdot \text{FSR}$ (N : integer). However, when $f_m \neq N \cdot \text{FSR}$, the light cannot be modulated with high efficiency because phase of the microwave does not conform between each propagation of the light. The phase modulation when $f_m = N \cdot \text{FSR}$ can be described as

$$m_k = (-1)^N (k+1) m_0 \sin(2\pi f_m t). \quad (A4)$$

Consequently, the electric field of the light output from the WG-OF CG can be derived and is expressed in (A5) shown at the top of the page from the sum of the propagating light using (A4) where E_{in} , ν_0 , R , and FSR are the electric field amplitude, the frequency of the injected light, the reflectivity of the reflection film, and the FSR of the optical resonator, respectively. Consequently, the power of the light output from the WG-OF CG is described as

$$P(t) = \frac{|E_{\text{in}}|^2}{1 + \frac{4R}{(1-R)^2} \sin^2 \left\{ -\frac{\pi \nu_0}{\text{FSR}} + \frac{m_0}{2} \sin(2\pi f_m t) \right\}}. \quad (A6)$$

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Tunable UV CW generation at 276 nm wavelength by frequency conversion of laser diodes

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Abstract

We developed a tunable ultraviolet continuous-wave light source by frequency up-conversion of laser diodes. The ultraviolet light was generated in a BBO crystal placed in an external resonant enhancement cavity by sum-frequency mixing between the 778 nm output of a laser diode and the second harmonic of the 857.2 nm output of the laser diode which was generated in a KNbO₃ crystal placed in an external resonant enhancement cavity. The ultraviolet power of 35.7 nW was obtained at 276.4 nm wavelength with input fundamental power of 50 mW at 778 nm and 200 mW at 857.2 nm. The tuning range of the ultraviolet generation was from 275.5 nm to 276.4 nm. © 1998 Elsevier Science B.V.

A laser diode (LD) provides a compact, efficient and reliable source of coherent radiation. The use of LD is limited only by their wavelength range which is typically restricted to the red and infrared part of the spectrum. However, radiation of ultraviolet (UV) frequency can be generated by nonlinear optical frequency conversion of a LD output.

There have been several reports on the generation of blue light by frequency doubling of the LD output using a KNbO₃ (KN) crystal in a resonant enhancement cavity [1,2]. It has been reported that the tunable UV continuous-wave (CW) radiation was generated by frequency quadrupling of a high power semiconductor laser which combines a low power single mode master oscillator with a high power amplifier, i.e. the KN in a resonant enhancement cavity generated the second harmonic generation (SHG), which was further frequency-doubled by β -barium borate (BBO) in a resonant enhancement cavity [3]. In related works, tunable UV pulsed radiation was produced by frequency quadrupling of a mode-locked external cavity compound semiconductor laser containing a tapered GaAlAs amplifier with single pass SHG in KN following BBO [4]. It was suggested that tunable UV CW radiation can be generated by frequency up-conversion of LD.

In this paper we report on a tunable UV CW radiation

near 276 nm generated by frequency up-conversion of the LD output. We have reported the generation of a tunable UV CW radiation near 252 nm by frequency tripling of a tunable CW Ti:sapphire laser which can be replaced by a LD [5]. This wavelength is appropriate for laser-induced fluorescence detection of silicon (Si). The allowed transition, ³P₀–³P₁ (transition wavelength 252.487 nm), can be used for laser cooling of Si atoms [6]. Recently, laser spectroscopy has been also reported on rubidium (Rb) atoms which were guided by an evanescent wave in a small hollow-core optical fiber [7,8]. These results show that our UV light source is applicable to cooling and guidance of Si.

A schematic of the experimental setup is shown in Fig. 1. The fundamental lights (LD1 and LD2) are provided by single-stripe, index-guided GaAlAs diode lasers, with maximum output power specified to be 200 mW at 855 nm and 50 mW at 778 nm at room temperature, respectively. Tunable UV CW generation was conducted by two successive stages. The first stage was frequency doubling of the LD1 output in a KN crystal placed in an external resonant enhancement cavity. The second stage was sum-frequency generation between the LD2 output and the second harmonic (SH) of the LD1 output in a BBO crystal placed in an external resonant enhancement cavity. The LD1 output

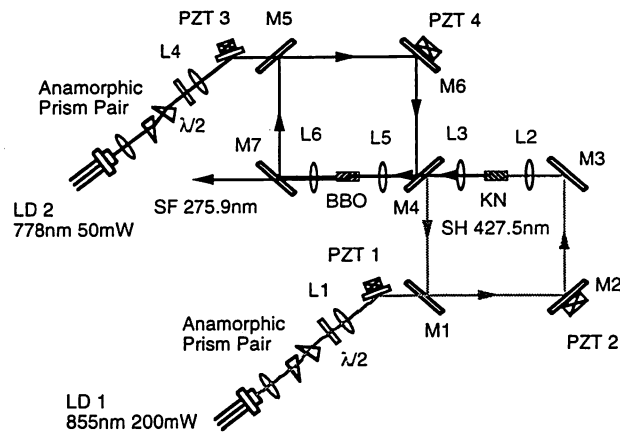


Fig. 1. Schematic of the experimental setup for frequency up-conversion of LD output. PZT, piezo-electric transducer; $\lambda/2$, half-wave plate; $\lambda/4$, quarter-wave plate.

was collimated by lenses and its ellipticity was compensated for by an anamorphic prism pair. The output beam was mode-matched with an external resonant enhancement cavity by using a plano-convex lens L1 ($f=400$ mm) followed by a half-wave plate ($\lambda/2$). The cavity for SH consists of four flat dielectric mirrors (M1–M4) and two biconvex lenses (L2 and L3; focal length $f=40$ mm). The reflectivities of mirrors M2–M4 are higher than 99.6% at 855 nm. The transmission of lenses L2 and L3 are higher than 99.9% at 855 nm and 99% at 427.5 nm. Total round-trip loss in the cavity was estimated to be about 5%; therefore, the input coupling mirror M1 for the fundamental wave had reflection of 95% at 855 nm. We have used the relation which is effective for optimized input mirror transmission of a cavity [9]. It showed that the input coupling mirror M1 for the fundamental wave had reflection of 95% at 855 nm. The input coupling mirror was selected to impedance match the resonant enhancement cavity.

The stabilization of the LD1 frequency to the cavity resonance frequency is achieved by optical feedback from the counterpropagating fundamental mode that is excited in the ring cavity [10]. The distance from the LD1 to the cavity and the length of the cavity can be controlled by piezoelectric transducers (PZT1 and PZT2), and the phase of feedback light from the cavity, as well as the resonance frequency of the cavity, can be scanned. We chose a noncritically phase-matched a-cut KN crystal. An optimum phase matching is achieved by temperature-tuning of the crystal [11]. The temperature of the KN crystal is controlled by a Peltier element within 0.01°C . Both crystal facets were antireflection (AR)-coated for the fundamental wave at 860 nm and for the SHG at 430 nm. The crystal length was 5 mm. The crystal was installed at the center between the two biconvex lenses L2 and L3, where the beam waist of the cavity was located. Under this condition, the beam waist was estimated to be $25\ \mu\text{m}$. This suggested that the second harmonic conversion coefficient should be

about $3.6 \times 10^{-3}\ \text{W}^{-1}$. The generated SH in the crystal passed through the biconvex lens L3 and the flat mirror M4.

The resonant enhancement factor (P_C/P_I) of the cavity was estimated to be about 20, where P_C and P_I are the intracavity and the incident power, respectively. The SH power was estimated to be 6.0 mW for the fundamental power of 200 mW at 857.2 nm. In the experiment, the intracavity fundamental power P_C was measured to be 1.3 W; therefore the observed spatial mode-matching factor was 33%. The highest SH power obtained was 3.2 mW, which was smaller than the estimated value because of the surface reflection loss of the output mirror M4 and the prism and the transmission loss of the UV-pass filter being used. The output mirror M4 allowed the power transmission for the SHG to be larger than 72% with AR-coated at 428.6 nm wavelength. The tuning curve of the SHG is shown in Fig. 2. We tuned the SH wavelength by tuning the fundamental wavelength while adjusting the KN crystal temperature in the external build-up cavity. The SH was tuned from 426.5 nm to 428.6 nm, which was limited by the capability of the temperature controller for the crystal, and by dewing of the crystal surface. The SH power increased with increasing wavelength because of the wavelength-dependence of the reflectivities of the cavity mirrors. The linewidth of the SH light was estimated to be smaller than 2 MHz by the free spectral range and finesse of the build-up cavity. The frequency of the SH could be scanned by scanning of the LD1 current, the cavity and the phase mirror.

The UV light was generated by sum-frequency mixing between the LD2 output and the SH of the LD1 output. The LD2 output beam was collimated by lenses and its ellipticity was compensated for by an anamorphic prism pair. The output beam was mode-matched with an external resonant enhancement cavity by using a plano-convex lens L4 ($f=200$ mm) followed by a half-wave plate ($\lambda/2$). We employed a BBO crystal that was cut at an angle $\phi=0^\circ$, $\theta=43.4^\circ$ with respect to the optical axis, that permitted type I phase matching of 430 nm and 780 nm wavelength fundamental beams [12,13]. The crystal length

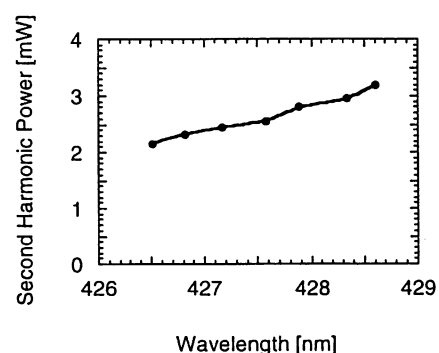


Fig. 2. The tuning curve of the SH obtained by frequency doubling of the LD1 output in a KN crystal in the external resonant enhancement cavity.

was 7 mm. Usually, both BBO crystal facets were coated to protect the crystal against hydropical damage. Since this protection coating reduced the transmission for the UV light, we applied AR-coatings for the fundamental wavelength at 780 nm, the SH wavelength at 430 nm and the sum-frequency (SF) wavelength at 277 nm to both surfaces of the crystal, which appeared also to be effective to protect the surface of the crystal against the hydropical damage. The BBO crystal was placed in a resonant enhancement cavity similar to the first resonant enhancement cavity. Two biconvex lenses (L5 and L6) have focal length $f = 50$ mm. The reflectivities of four flat dielectric mirrors (M4–M7) are higher than 99.9% at 778 nm. The transmission of lenses L4 and L5 are higher than 99.9% at 778 nm, 99% at 427.5 nm and 87.7% at 275.9 nm. Total round-trip loss in the cavity was estimated to be about 2%; therefore, the input coupling mirror M5 for the fundamental wave had reflection of 98% at 778 nm. The input coupling mirror M5 for the fundamental wave was selected to had reflection of 98% at 778 nm in the same way the input coupling mirror M1 was. The stabilization of the LD2 frequency to the cavity resonance frequency is achieved by the same way as for the LD1. The distance from the LD2 to the cavity and the length of the cavity can be controlled by PZT3 and PZT4. The crystal was installed at the center between the two biconvex lenses L5 and L6, where the beam waist of the cavity was located. The generated SF in the crystal passed through the biconvex lens L6 and the flat mirror M7. This beam was detected by a solar blind calibrated photomultiplier (Hamamatsu C1053) with cut off wavelength at 320 nm.

The conversion coefficient of sum-frequency mixing between the fundamental wave at 778 nm and the SH at 428.6 nm in the BBO crystal was calculated to be about $5.2 \times 10^{-5} \text{ W}^{-1}$. With the fundamental power of 50 mW at 778 nm wavelength and the SH power of 2.5 mW, the SF power at 276.4 nm wavelength was estimated to be about 117 nW. The highest SF power obtained was 35.7 nW, which was smaller than the estimated value because of the surface reflection loss of the lens L6 and the output mirror M7. Part of the SF power was dissipated by the surface reflection loss and transmission loss in the fused silica prism. The mirror M7 allowed the power transmission for the SF to be larger than 85% with AR-coated at 276.4 nm wavelength. The tuning curve of the SF is shown in Fig. 3. The SF was tuned from 275.5 nm to 276.4 nm which was limited mainly by the tuning capability of the SHG. The UV power increased with increasing wavelength because of the tuning characteristic of the SHG shown in Fig. 2. We estimated that an UV light power of several mW would be required for application to cooling, nanometer-scale deposition and atomic wave guidance of Si. When the LD1 and LD2 in our experimental setup could be replaced by a high power LD which will be made in the near future, the spatial mode-matching factor of the cavity will increase, the UV light power will be expected to reach several mW. Therefore, our UV light source is applicable

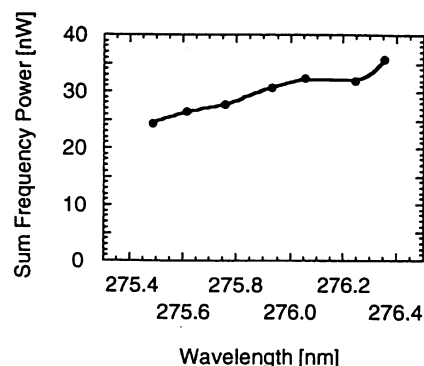


Fig. 3. The tuning curve of the SF obtained by sum-frequency mixing between the LD2 output and the SH of the LD1 output in a BBO crystal.

to cooling, nanometer-scale deposition and atomic wave guidance of Si.

In summary, tunable CW UV generation by LD was demonstrated. It was accomplished by frequency doubling of the 857.2 nm output of the LD in the KN crystal placed in an external enhancement cavity and sum-frequency mixing between the 778 nm output of the LD and the SH of the 857.2 nm output of the LD in the BBO crystal. As a result, a 35.7 nW tunable UV CW light source was obtained at 276.4 nm wavelength and the tunable range of 275.5–276.4 nm with 30 nW of average power.

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30-THz Span Optical Frequency Comb Generation by Self-Phase Modulation in an Optical Fiber

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Abstract—The width of an optical frequency comb (OFC) was increased to 30 THz by using self-phase modulation (SPM) in an optical fiber. This value is 2.7 times larger than the maximum OFC span obtained by the OFC generator alone. We compare the resulting spectrum to numerical simulations to confirm that the SPM and the higher order dispersion of the fiber contribute to broaden the spectral profile.

Index Terms—Electrooptic modulation, optical frequency conversion, optical propagation in nonlinear media, optical pulse compression, optical pulse generation.

I. INTRODUCTION

TUNABLE coherent light sources whose absolute frequencies are stabilized, e.g., to the Cs microwave standard, are required for various applications such as high-resolution spectroscopy, the measurement of fundamental constants [1], [2], metrology, and optical communications. Although absolute optical frequencies have been measured with the help of harmonic frequency chains [3], this procedure is complicated as it needs a variety of different oscillators. In contrast to conventional frequency chains, difference frequency synthesis systems have been proposed [4]–[6]. In these systems, the frequency gap of some laser at a frequency f and its second harmonic $2f$ is measured. This frequency gap is equal to the optical frequency f . In this way, absolute optical frequency measurements might be simplified. If optical interval divider stages [4] are used that recursively divide arbitrary large frequency gaps by a factor of two, the number of stages required is determined by the limited bandwidth of the photodetector.

An optical frequency comb generator (OFCG) can be used to measure the difference frequency of two lasers that might be as high as several terahertz [7]–[9]. The OFCG is based on a Fabry–Perot (FP) electrooptic modulator (EOM) [10], [11]. It generates a broad comb of equally spaced optical frequencies separated by the modulation frequency. The accuracy of the frequency intervals between the sidebands is given by the accuracy of the microwave oscillator that drives the OFCG. The optical frequency comb (OFC) can significantly reduce the number of required optical interval divider stages of a difference frequency synthesis system [12]. If the OFC is broad enough to contain the second harmonic of the lower frequency modes f it can measure the absolute frequency of these modes without any interval divider stage. After

second harmonic generation of the lower frequency mode, the frequency gap between f and $2f$ is determined with the OFCG. Because the absolute frequencies of all the sidebands are also determined at the same time, an optical frequency counter is realized by using the OFC. For this application, an OFC with a spectral width that equals the carrier frequency has to be used.

Although the width of currently available OFCG's has been expanded to several terahertz by improving the modulation index and the finesse of the FP cavity [13], [14], it is still limited to about 5% of the carrier frequency. To further increase the span, one is facing problems such as limited mirror reflection bandwidth, incident microwave power limitation (to prevent damage to the EOM), and chromatic dispersion of the crystal.

In order to overcome these difficulties, we have used the spectral broadening provided by an optical fiber to coherently add modes to the OFC generated by a conventional OFCG. For this purpose we employ an optical fiber amplifier to gain enough power in the pulses provided by the OFCG (see Fig. 5). The intensity-dependent refractive index of the fiber induces self-phase modulation (SPM) of these pulses. As SPM is a $\chi^{(3)}$ process, it is able to generate new modes by four-wave mixing. The additional modes of the OFC emerge from the sum frequency of two modes from which a third mode is subtracted. In this way, a new frequency ω_1 will be generated by mixing three existing modes with the frequencies ω_2 , ω_3 , and ω_4 such that they obey the relationship $\omega_1 + \omega_2 = \omega_3 + \omega_4$ which states conservation of energy. From this, it is clear that the uniformity of the extended OFC will be maintained if the spacing between the initial frequencies ω_2 , ω_3 , and ω_4 is uniform.

So far a maximum width of 1600 THz of a such a super continuum (SC) was produced by a 2-TW Q -switched laser together with a rare gas as SPM medium [15]. In this way, neither a high repetition rate nor a narrow linewidth of each mode is obtained as required in our intended application. Optical fibers are low-loss and low-dispersion media for far-infrared wavelengths because the material dispersion of the fiber can be compensated by the structural dispersion. The modulation index which depends on the power density is readily increased by the use of a small fiber core. SC at the 1.55- μm region up to 200 nm was generated in a dispersion-shifted fiber by a 6.3-GHz mode-locked fiber ring laser which outputs 1.7-W 3.3-ps pulse train [16]. It indicates the possibility to broaden the output spectrum of an OFCG operated in a pulsed mode.

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In this paper, we report on a 30-THz span OFC produced by a dispersion-flattened optical fiber. We analyze the evolution of the spectrum of the generated side bands numerically and compare it with the experimental results.

II. THEORETICAL ANALYSIS

For our calculations, we used the output characteristics of an OFCG at 1.55 μm and a dispersion-flattened fiber (DFF) with an anomalous dispersion of 0.12 ps/km/nm, a mode field diameter of 6.1 μm , and a transmission loss of 0.23 dB/km [17]. The DFF's material dispersion is compensated by a double cladding layer. Its absolute dispersion is less than 1 ps/km/nm over a wide range of wavelengths extending from 1.5 to 1.6 μm . Since SPM under the anomalous dispersion supports the formation of a soliton, we can expect efficient spectral broadening by the pulse compression of the higher order solitons [18].

Evolution of a pulse in the DFF can be described by a nonlinear propagation equation including higher order dispersion terms [19]

$$\frac{\partial A}{\partial z} = -\frac{i}{2}\beta_2 \frac{\partial^2 A}{\partial T^2} + \frac{1}{6}\beta_3 \frac{\partial^3 A}{\partial T^3} + \frac{i}{24}\beta_4 \frac{\partial^4 A}{\partial T^4} - \frac{\alpha}{2} + i\gamma|A|^2 A. \quad (1)$$

Here A is the slowly varying envelope function, T is the time defined in a frame which moves with the pulse at the group velocity $v_g = 1/\beta_1$, α ($= 0.23$ dB/km) is the fiber loss coefficient, and γ ($= 4.6$ W $^{-1}$ ·km $^{-1}$ for the DFF in the 1.55- μm region) is the nonlinear coefficient which depends on the nonlinear refractive index and the mode field diameter. The wavenumber β is expanded in a Taylor series about the carrier frequency ω_0 using the coefficients $\beta_n = [d^n \beta / d\omega^n]_{\omega=\omega_0}$. Terms up to the fourth order are included ($\beta_2 = -1.5 \times 10^{-28}$ s 2 /m, $\beta_3 = 2.2 \times 10^{-43}$ s 3 /m, $\beta_4 = 5.6 \times 10^{-55}$ s 4 /m). The term proportional to β_2 represents the group velocity dispersion (GVD). The terms proportional to β_3 and β_4 represent the higher order dispersion. Under the presence of the higher order dispersion, the anomalous dispersion range is limited within the wavelength range from 1.52 to 1.58 μm which corresponds to the frequency span of 7.5 THz. Because of the flatness of the GVD profile around 1.55 μm , β_4 is dominant in the higher order dispersion and the dispersion characteristic is symmetric about the carrier frequency.

We assumed that the initial pulse envelope is that of a typical output from an OFCG. The output of the OFCG is a pulse train whose repetition rate is twice the modulation frequency, if the modulation index is less than π rad. We can consider the effect of EOM as a sinusoidal modulation of the length of the FP cavity. Sweeping across the OFCG resonance by varying the cavity length yields two resonance lines per one sweeping period. For example, one resonance line is obtained when the cavity expands and the other line when the cavity contracts. Therefore the output consists of two pulses for each period of modulation. When the FP cavity of OFCG is exactly resonant with the input light frequency, pulses are generated at equal intervals with the narrowest width. Although both of the pulse intensities are Lorentzian line shapes, they have inverted

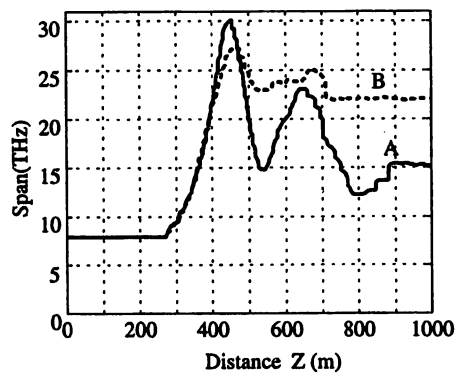


Fig. 1. Theoretical results showing the variations of the OFC span after propagating the distance Z in the dispersion-flattened fiber: curve A in the absence and curve B in the presence of higher order dispersion.

complex amplitudes to each other, because the directions of the cavity sweep for the pulses are opposite. Therefore, each of them forms exponential decaying sidebands on the positive or negative side of the input frequency. The maximum sideband order is limited by GVD of EOM used in OFCG. As a result, the output of OFCG has both side exponential decaying sidebands with stepwise discontinuities on both sides of the spectrum. GVD causes the distortion of the pulse shape, still they have inverted complex amplitudes to each other. The FWHM and the peak power of each pulse are 1 ps and 2 W, respectively, which agree with the experimental condition.

To solve (1), the split-step Fourier method was used [19]. We first calculated the variation of the OFC span as a function of the propagated distance. Curve A of Fig. 1 shows a variation of the OFC span along the propagated distance Z under the absence of the higher order dispersion. The full width of the comb is defined by the region in which the mode power exceeds 1 nW because this is the typical power required for beat frequency measurement. After the pulse is launched into the fiber, the spectral width stays at the initial value for $Z \leq 300$ m because of the stepwise discontinuities of the initial spectrum. Then the span increases rapidly by the effect of SPM and anomalous GVD to the maximum value of 30 THz at $Z = 450$ m. Then the span width changes periodically along Z , while the peak value of each period gradually decreases. As the pulse duration oscillates for higher order solitons as a function of the distance traveled, we expect at the same time an oscillation of the OFC span. Without the higher order dispersion, the maximum span at the optimum fiber length is limited by the lowest order GVD and the input power. The optimum fiber length is 450 m for this theoretical calculation. Unless we use the optimum length fiber, the initial spectrum is not effectively broadened. Curve B of Fig. 1 shows a variation of the OFC span under the presence of the higher order dispersion. Since the effect of the higher order dispersion is negligible while the OFC span is narrow, the two curves are identical up to a distance of 400 m which corresponds to a spectral width of 20 THz. The maximum width that is reached is higher in the absence of higher order dispersion. Compared with curve A, the variation of the spectral width along Z is strongly damped in curve B. However, the span at $Z = 1000$ m is larger than that of curve A.

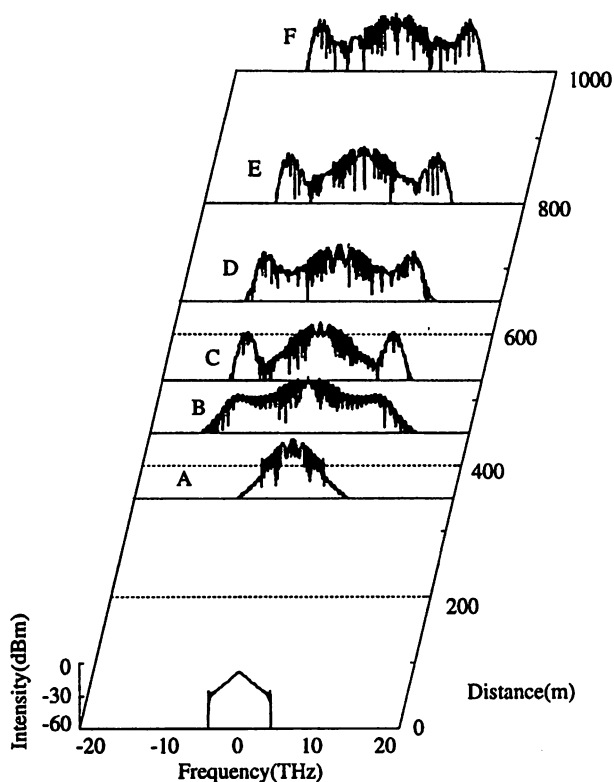


Fig. 2. Spectral evolution in the presence of the higher order dispersion.

Fig. 2 shows the evolution of the spectral profile under the presence of the higher order dispersion. As the OFC span is broadened, the peaks of both sides of the spectrum are generated. They are not generated without the higher order dispersion. Both the peaks are generated in the normal dispersion range and the difference frequencies between the peaks and the carrier are about 9 THz. Without the higher order dispersion, the width of the broadened spectrum at the optimum fiber length is rapidly decreased at larger distances. The peaks under the presence of the higher order dispersion are stable in spite of the periodic evolution of the other components. Therefore, the OFC span remains large at the larger distances.

Fig. 3 is the temporal evolution of the initial pulse along Z under the presence of the higher order dispersion. In Fig. 3, curves A–F show pulse shapes of the spectra in A–F of Fig. 2. Only one pulse is shown in Fig. 3. Another pulse has almost the same waveform except for the inverted complex amplitude. This is due to the dispersion characteristics of the DFF. As the pulse is compressed, the oscillatory structure appears as shown in the inset of Fig. 3, where the oscillation period is about 110 fs. At larger distances, the pulse is split into two or three distinct pulses similarly to the evolution of a fifth-order soliton. The generation of the oscillatory structure is due to the optical wave breaking [20] by the spectral broadening under the presence of the higher order dispersion. Because of the frequency chirp induced by SPM, red-shifted light is generated in the leading edge and blue-shifted light is generated in the trailing edge of the pulse. When the spectral range exceeds the normal-dispersion regime, the red-shifted light near the leading

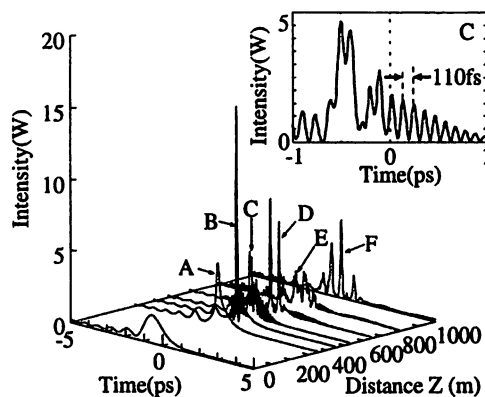


Fig. 3. Temporal evolution under the presence of higher order dispersion. The inset shows the oscillatory structure at the distance that corresponds to curve C.

edge travels faster and the blue-shifted light near the trailing edge travels slower than the unshifted light in the normal-dispersion regime. The red-shifted light overtakes the unshifted light so that interference fringe appears at the leading edge. The blue-shifted light is overtaken by the unshifted light and it also generates an interference fringe on the trailing edge. The fringes induce index changes proportional to the intensity due to the SPM and modulate the pulse at the period of the fringe. Therefore, the difference frequencies between the peaks and carrier are about 9 THz which corresponds to a reciprocal of the period of the oscillation. Since the components around the side peaks are separated from the pulse evolution, these components remain unperturbed. The pulse distortion induced by the higher order dispersion causes the stable three-peak structure.

The difference frequency between the peaks and the carrier can be explained by the degenerate four-wave mixing (FWM) in which two photons at carrier frequency ω_1 are annihilated and two photons at ω_2 and ω_3 , are created where $\omega_2 < \omega_1 < \omega_3$. A phase-matching requirement under the presence of the higher order dispersion demands $\omega_3 - \omega_1 = 9.2$ THz. The frequency components that satisfy this condition build up most rapidly to form the three-peak structure.

The spectral envelope in the simulation included large intensity discontinuities between odd sidebands and even sidebands indicated in Fig. 2. These discontinuities appear because the typical output of an OFCG consists of two pulses per modulation period. As indicated in [11] and [21], any beat note between two adjacent modes is exactly matched by one of opposite phase from the corresponding part of the mode spectrum on the opposite side of the input frequency. However, their spectra overlap after the broadening action of the fiber. According to the calculation, the modulation sidebands are added constructively or destructively depending on their order. For this reason, the intensity of the modulation sidebands after the fiber action are discontinuous between odd and even sidebands. If we calculate the resulting spectrum starting from one side of the initial spectrum only, the large variation of the intensity between odd and even sidebands disappears.

Uncertainty of sideband frequency is of great importance for our intended application. The uncertainty is mainly caused

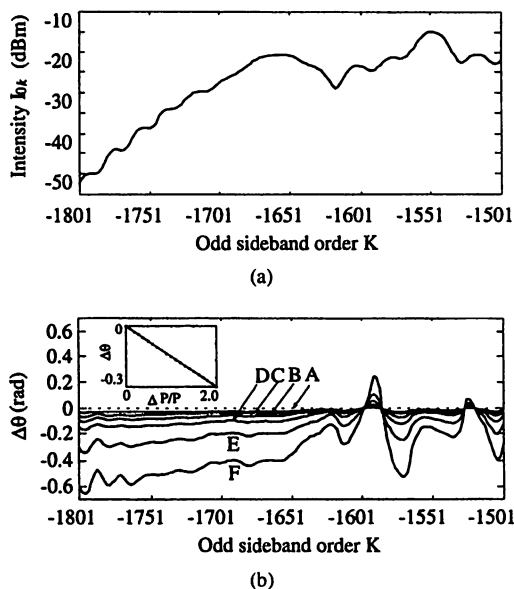


Fig. 4. Theoretical results showing the intensity and the phase variations as a function of odd-order mode number k . (a) The intensity of the k th sideband I_{0k} with a total input power P . (b) The phase variations $\Delta\theta$ when P increased to $P + \Delta P$. The inset shows $\Delta\theta$ as a function of $\Delta P/P$ for $k = -1651$. Curves A–F show the phase variations for $\Delta P/P = 0.1\%$, 0.2% , 0.3% , 0.5% , 1% , and 2% , respectively.

by power drift of input pulses and temperature drift of the fiber. Small fluctuations are imposed on the output power of the OFCG because of the laser power fluctuation and imperfect control of a coupling cavity (mirror M3). Because the power drift in the optical fiber induces the variation of the pulse evolution, the phase of the sidebands are disturbed by the output power fluctuations. We estimated the phase variations of the OFC generated by SPM due to the input power variations. We show the result of the odd-order sidebands only, as the even-order sidebands behave similarly. Fig. 4(a) shows the intensity of the k th-order sideband I_{0k} in the range between -10.8 and -9 THz from the carrier that is created at $Z = 1000$ m (see Fig. 2). Since we selected the odd sidebands from the result of the calculation, we obtain a smooth spectral profile. Fig. 4(b) shows the phase variations $\Delta\theta$ of the odd-order k th sidebands after the total output power P of the OFCG increased by ΔP . Curves A–F display the phase variations when $\Delta P/P$ is 0.1% , 0.2% , 0.3% , 0.5% , 1% , and 2% , respectively. The inset shows $\Delta\theta$ as a function of $\Delta P/P$ for $k = -1651$. Even though $\Delta\theta$ depends on k , it is proportional to ΔP as indicated by the inset. The maximum phase variations are less than 0.4 rad per 1% of average input power variation. If the power variation is maintained lower than 1% for a measurement time of 1 h, the uncertainty of the sideband frequency induced by the power drift is less than 20 μHz .

The uncertainty induced by the temperature drift is increased due to the temperature dependence of the refractive index of optical fiber. Temperature sensor application of optical fiber has been demonstrated based on this effect [22]. From the sensitivity, the variation of the effective optical fiber length can be estimated about 1 cm per temperature shift of 1 K for 1 -km-long unjacketed single-mode fiber. If temperature

drifts 1 K per hour, uncertainty of the carrier frequency is about 2 Hz. Since the uncertainty is much larger than that induced by power drift, it is required that temperature shift is controlled at least less than 1 mK. Although the phase shift has small dependence on the wavelength, the phase shifts of all sidebands can be compensated by detecting the phase shift of carrier. If we split off part of the laser output and pass it through a frequency shifter, we can detect the phase shift by observing the beat signal between the part of the laser output and the carrier of the DFF output.

III. EXPERIMENTS AND DISCUSSIONS

A. Experimental Setup

The experimental setup is shown in Fig. 5. We used a coupled cavity OFCG (C-C-OFCG) [23]–[26] to compress input optical energy into pulses and make the peak power of the pulses higher than the average power of input light. The C-C-OFCG transformed the CW laser diode (LD) [27] output ($\lambda = 1.54$ μm , 2 mW) into a pulse train with a repetition rate that is twice the modulation frequency. The C-C-OFCG consists of a monolithic FP cavity in a resonant microwave cavity and a mirror [28]. The monolithic cavity is a lithium niobate crystal ($1 \times 1.2 \times 23.4$ mm^3) with high-reflection-coated mirrors M1 (radius of curvature ∞ , reflectance ($R = 99.6\%$) and M2 (radius of curvature 50 mm, $R = 99.6\%$) on both facets. The finesse and the peak transmission of this cavity were 600 and 30% when the cavity was not modulated. The resonance frequency f_m of the microwave cavity was adjusted to 6.06 GHz to match twice the free spectral range (FSR) of 3.03 GHz. In this way, the generated sidebands are resonantly enhanced. A modulation index of 0.75 rad was measured at the resonance of the microwave cavity. To improve the input coupling efficiency to the monolithic cavity, a third mirror was used which was servo-controlled for the minimum back reflection (M3: radius of curvature 42 mm, $R = 99.3\%$). For this purpose, the mirror M3 was mounted on a dithered piezoelectric transducer. The reflected light was demodulated in order to lock the cavity resonance frequency to the zero crossing of the demodulated signal, i.e., the resonance frequency of the coupling cavity. This coupling mirror was placed at a distance of 5 mm from the cavity input mirror M1. The finesse of the cavity formed by the mirrors M3 and M1 was 600 when the monolithic cavity was not modulated and decreased to 400 when it was modulated.

To stabilize the monolithic OFCG with respect to the input laser we split off a small part of the output from the OFCG and directed it to a fast photodiode PD1 (Fig. 5). The output from the photodiode was mixed (demodulated) with the modulation frequency f_m using the double balanced mixer DBM1. Because the repetition frequency in the pulsed mode is $2f_m$ [10] the signal from the mixer vanishes at this point providing the error signal to lock the OFCG to the input laser. To remove the offset fluctuation of the error signal from DBM1, we used an acousto-optic modulator (AOM) as a chopper before the photodiode PD1. The signal from DBM1 was demodulated at the chopping frequency by DBM2. The

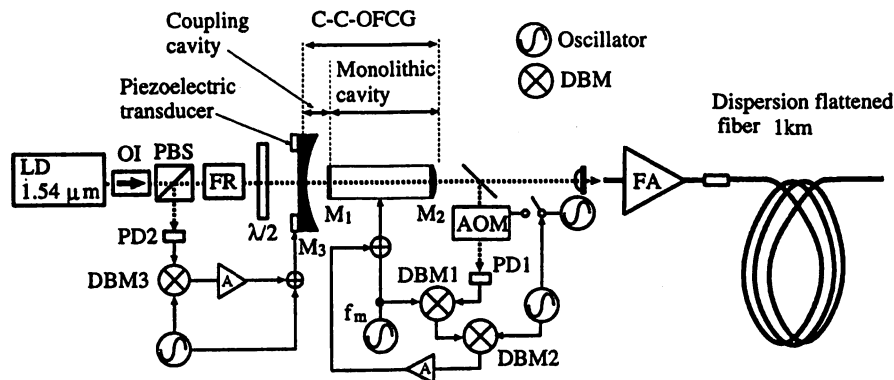


Fig. 5. Experimental setup. LD: extended-cavity laser diode; OI: optical isolator; PBS: polarizing beam splitter; FR: Faraday rotator; $\lambda/2$: half-wave plate; A: amplifier; FA: optical fiber amplifier; AOM: acousto-optic modulator; PD: photodiode; DBM: double-balanced mixer.

cavity was locked to the zero crossing of the demodulated signal by adding dc voltage on the crystal as a feedback signal.

When both the coupling cavity and the monolithic cavity were locked, the efficiency, i.e., the total output power divided by the input power, of the cavity was 25%. This is an improvement of 100 as compared to the monolithic OFCG without the coupling cavity. Since the output power of the C-C-OFCG was still too low, we amplified it with an erbium–ytterbium co-doped fiber amplifier before introducing it into a 1-km-long DFF.

B. Experimental Results

Curves A–D of Fig. 6 show the spectral envelopes observed with the help of a monochromator with a resolution of 1 nm. Curve A shows the spectral envelope of the output from the C-C-OFCG when the monolithic cavity was locked to the laser frequency. In this case, the spectral width is limited by material dispersion of the EOM [13] to about 7.7 THz. When the monolithic cavity was locked to the point slightly detuned from the laser frequency [13], the OFC span increased to 10.8 THz as shown in curve B in which case it is still limited by the material dispersion. The shortest pulses are obtained when the monolithic cavity is exactly locked to the laser frequency (curve A), even though curve B has a larger spectral width. Because a short pulse and a high peak power are important for efficient spectral broadening by SPM in an optical fiber we employed the output of the C-C-OFCG as shown in curve A. To use the maximum OFC span was not mandatory, as both the spectral envelopes of curves A and B exceeded the width of the gain profile of the fiber amplifier.

Curve C shows the spectral envelope after the amplification. The average power of $220 \mu\text{W}$ was increased to 40 mW. Because the span of the OFC was more than twice the bandwidth of the amplifier, the gain profile of the fiber amplifier affected the OFC profile. We monitored the output from the amplifier with an autocorrelator, whose time resolution was 100 fs. We determined the full width at half maximum (FWHM) of the autocorrelation signal to be 2.0 ps, as is shown in Fig. 7. Since the FWHM of the autocorrelation signal is predicted to be twice that of the output of the OFCG assuming a Lorentzian pulse shape and that the amplifier maintains this shape, the FWHM and the peak power of the output from the amplifier

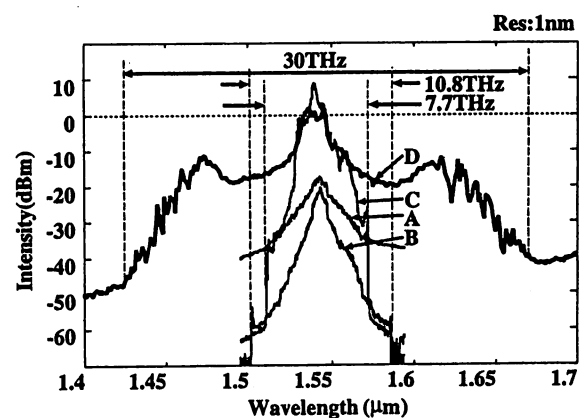


Fig. 6. Spectral envelopes of the OFC. Curve A shows the C-C-OFCG output. Curve B displays the output of the C-C-OFCG locked to the monolithic cavity to a point slightly detuned from resonance. Curve C shows the spectral profile obtained by introducing the spectrum of curve A into the fiber amplifier. Curve D is obtained from C by the self-phase-modulating action of the DFF.

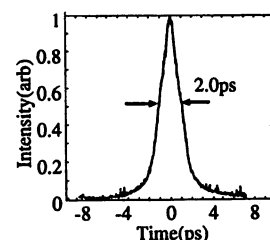


Fig. 7. Autocorrelator trace of the pulses that leave the fiber amplifier.

are predicted to be 1.0 ps and 2 W, respectively. Although the FWHM of the output of an OFCG indicated in [10] predicts that the pulse duration is 200 fs for the present experimental condition, the dispersion of the EOM distorts the pulse shape and broadens the pulse.

Curve D shows the spectral envelope of the output of the DFF. The spectrum is broadened to 30 THz symmetrically with a three-peak structure as predicted by the numerical simulations. The side peaks are formed around 80 nm (9 THz) away from carrier. The power of modulation sidebands around the carrier frequency of curve C, which shows the spectrum after the fiber amplifier, is transferred to both wings of curve D. The large intensity variation between odd and even sidebands

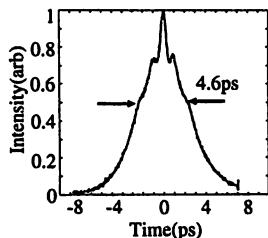


Fig. 8. Autocorrelator trace of the pulses that leave the dispersion flattened fiber.

is not resolved. The spectral width after the DFF is 2.7 times larger than the widest span of an OFC obtained without a DFF. The resulting OFC includes more than 5000 modulation sidebands. The measurable difference frequency reached 15% of the carrier frequency. Since we did not observe effects that makes the spectrum asymmetric, such as stimulated Raman scattering, we confirmed that the spectral broadening is caused by SPM and the effect of the higher order dispersion in the fiber. Fig. 8 shows the autocorrelation signal of the output from the DFF. Besides the broadening to 4.6 ps, we observed the generation of two side lobes which shows that the pulse has split into several short pulses as predicted by the numerical simulations.

Spectral broadening of the OFC was not observed when the output of the amplifier was launched to the opposite direction of the fiber. This indicates that the dispersion characteristic at the other end of the fiber was significantly different. Also, the width of the OFC obtained in the experiment was larger than that of the theoretical result, though the three-peak structure agreed with the numerical simulations. These discrepancies might be due to the fact that we ignored variations of the dispersion characteristics along the fiber which are caused by the core diameter variations around the designed value. The GVD characteristic of DFF is varied by the core diameter variations through the structural dispersion. Compared to the result of the simulations, the intensity of the side peaks in the three-peak structure is lower and the width of each peak is broader. The broadening is thought to be caused by dispersion characteristic variations of the fiber because the exact position of the side lobes depends on the dispersion.

The amplifier's gain profile that we ignored in the theoretical simulation limited the peak power and the pulsewidth. Since we used the amplifier to amplify both sides of the OFC, the amplified power and the bandwidth were distributed to both the pulses. If one side of the OFC, which consists of one pulse per modulation period only, was amplified, achievable peak power would be twice as large. Since the efficiency of the pulse compression and the spectral broadening by SPM under the anomalous dispersion is roughly proportional to the square root of the peak power [18], we expect to obtain an OFC of more than 40 THz.

The dominant noise source in our experiments was the amplified spontaneous emission (ASE) added in the amplifier. In the DFF, its behavior is not simply additive because the system is nonlinear. When a noise field is added to the input pulse, part of the noise is incorporated into the pulse. It is well known that, in the presence of SPM and GVD, the amplifier

noise causes random timing jitter problems [29]. Although the timing jitter affects the uniformity of mode spacings in the short term, the repetition rate is maintained in the long-term average. Therefore, the timing jitters caused by the ASE do not contribute to the uncertainty of the long-term frequency measurements. Furthermore we can avoid the noise effect of the amplifier by using a high-power laser. The average power of 200 mW is enough to induce SPM of the DFF because of the high efficiency of the C-C-OFCG.

Another noise source is spontaneous emission in the optical fiber. In the fiber, the incident pulse amplifies new modulation sidebands as well as the spontaneous emission through the nonlinearity. The noise level of the output depends therefore on the evolution of the incident pulse. We have not investigated the noise generated in the fiber theoretically. We will find the effect experimentally by measuring the beat signal between the OFC and another laser.

IV. SUMMARY

The spectral width of an optical frequency comb (OFC) was increased to 30 THz by self-phase modulation (SPM) in an optical fiber. The OFC includes more than 5000 sidebands separated by 6.06 GHz generated with external modulation from a single-mode diode laser. A high-repetition-rate pulse train originated from a cavity-coupled OFC generator, whose maximum spectral width was 10.8 THz, was amplified by a fiber amplifier to yield a peak power of 2 W and a pulse duration of 1 ps. Because of higher order dispersion in the optical fiber used for SPM, a three-peak structure appeared in the spectrum. Numerical simulations agreed with the observations. The phase variations of the OFC was investigated theoretically confirming that it can be maintained low enough for beat frequency measurements if the power fluctuation of the pulses launched into the fiber is less than 1%.

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M. Kourogi, photograph and biography not available at the time of publication.

M. Ohtsu (M'88-SM'90), photograph and biography not available at the time of publication.

Phase change of a probe due to oscillation of cold atoms in an optical standing wave

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We abruptly subject laser-cooled ⁸⁷Rb atoms to the one-dimensional, blue-detuned, optical standing-wave potential generated by two crossed probe laser beams and detect the phase change of the probe beam due to cold-atom dynamics. Damped oscillations of the observed phase signal are attributed to a temporal change of the root-mean-square position of the cold atoms relative to the minima of the periodic optical potential. The damping rate is in good agreement with numerical results, which take into account the potential anharmonicity but neglect spontaneous emission. The agreement indicates that the decay is mainly caused by dephasing due to trap anharmonicity. For sufficient low initial temperature, our calculations show quantum revivals of the wave-packet oscillation. [S1050-2947(98)51001-3]

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Laser-cooling techniques make it possible to prepare very cold atomic samples whose center-of-mass motion can be controlled by optical potentials. When such cold atoms are confined by a periodic optical potential, atomic energies are quantized and discrete band structures appear. The discrete vibrational energy spectrum has been observed using stimulated Raman transitions [1,2] and resonance fluorescence [3]. More recently, we have observed the oscillation of the atomic position in the time domain (i.e., pendulum motion of the center of the atomic density distribution) via the transient response [4,5] of recoil-induced resonance [6].

From a quantum-mechanical point of view, such an oscillation is the result of coherence between the n th and $(n+1)$ th vibrational states. The damping rate of the transient signal is related to the spectral widths of the sidebands for absorption and emission [1–3], since the sidebands are associated with the autocorrelation of the atomic oscillation. It was recently realized that the observed signal decay rate was much smaller than the inelastic optical scattering rate predicted by Lamb-Dicke theory [7]. Consequently, it was suggested that for a perfectly harmonic potential, the signal damping rate be determined by the cooling rate, which is much smaller than the inelastic scattering rate.

In our previous work [4,5] the oscillation of the mean atomic position was observed by the change in intensity of the probe beam. Alternatively, one can detect the change of phase of the probe beam. In this case, in contrast to intensity detection, one observes the temporal change of the root-mean-square (rms) position spread of cold atoms in the potential wells. Thus, the phase of the probe light allows one to measure periodic contraction and expansion of the atomic density distribution caused by coherence between the n th

and $(n+2)$ th vibrational states. By detecting both intensity and phase signals, one can obtain complementary information on the atomic dynamics in the potential. It also opens up the possibility of active control of atomic motion by temporal change of the optical potential.

In this paper, we describe an observation of the phase change of the transmitted probe beam caused by the oscillation of the rms position of trapped atoms. The rms position of cold atoms has also been measured in optical lattice experiments using Bragg scattering [8–11] and fluorescence [12]. The method presented here has the advantage that the atomic dynamics is not perturbed by an additional probe laser beam.

In our experiment, a cold cloud of ⁸⁷Rb atoms is prepared by a magneto-optical trap (MOT) [13] and polarization gradient cooling (PGC) [14] in a vapor cell. The $5S_{1/2}F=2 \rightarrow 5P_{3/2}F=3$ cyclic transition of the ⁸⁷Rb D_2 line was used for cooling. Figure 1 shows our experimental setup to observe recoil-induced resonance and its transient response. Inside the cold sample, two probe beams [with wave vectors \vec{k}_1, \vec{k}_2 ($\vec{q} = \vec{k}_2 - \vec{k}_1$), angular frequencies ω_1, ω_2 ($\delta \equiv \omega_2 - \omega_1$) detuned from the cooling transition by Δ ($\Delta \gg \delta$), and parallel linear polarizations] are crossed at an angle θ . These two probe beams are generated from the same extended cavity diode laser, each one having passed through a 20- μ m pinhole to obtain a uniform wave front. Under these conditions, an optical potential proportional to $\cos(qr_q - \delta t)$ is generated in the beam-crossing area. The velocity of the optical potential in the laboratory frame, $v_0 = \delta/q$ along \vec{q} ($q \equiv |\vec{q}|$), is controlled by changing δ . Here, r_q is the projection of the position vector on \vec{q} , and the spatial period of the potential is $2\pi/q$. One of the transmitted probes is divided by a beam splitter (BS), one part being detected by a photodetector PD1 for intensity measurement, and the other one being mixed by a 1-mm-thick étalon plate (ET) and then detected by PD2 for phase measurement. Reflectivities of the front and rear faces of the ET are 38% and 100%, respec-

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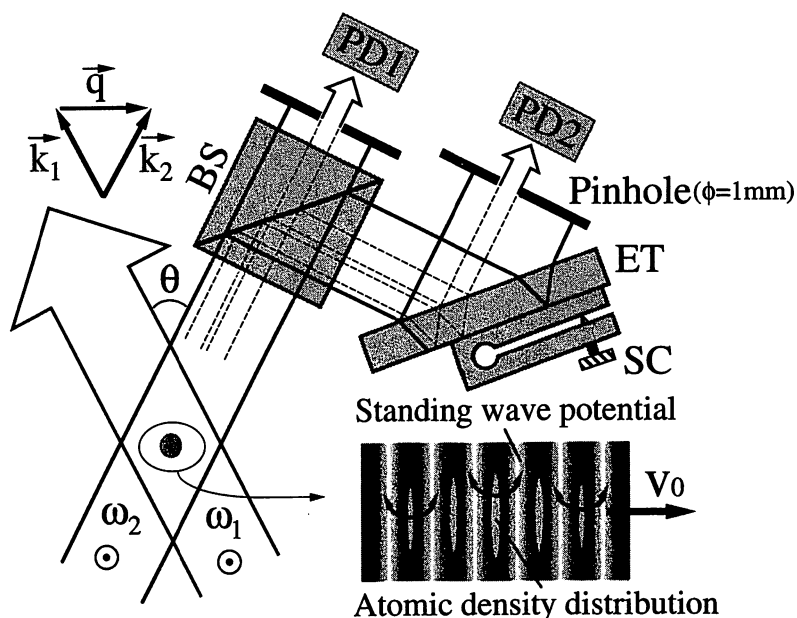


FIG. 1. Experimental scheme to observe recoil-induced resonance and its transient response.

tively. Because the diameter of the probe beams (5 mm) is bigger than the size of the atomic cloud (< 1 mm), only the central part of the probe interacts with the cold atoms. The central and the outer part of the probe beam are overlapped by the ET, and thus the output signal of PD2 is a convolution of both the intensity and phase changes of the transmitted probe beam. The reflection angle of the ET is adjusted by a fine screw (SC) in such a way that phase sensitivity is maximized.

First the temperature of the cold atoms is measured by using recoil-induced resonance [4,6]. We record the output of PD1 versus the frequency difference δ , and estimate the temperature to be as low as $4.7 \mu\text{K}$. Here, the intensity of the probe beams is 4.2 mW/cm^2 , $\theta = 12.5^\circ$, the frequency difference δ is scanned at a rate $d(\delta/2\pi)/dt = 54 \text{ kHz/ms}$, and $\Delta/2\pi = +920 \text{ MHz}$. The large detuning ensures that the optical potential is shallow enough that the atomic momentum distribution is not perturbed.

Next, we observe the change in both the intensity and phase of the transmitted probe due to the transient response of recoil-induced resonance. Temporal changes of PD1 and PD2 outputs are monitored for fixed δ , i.e., the cold atoms are exposed to one-dimensional standing waves with a constant, δ -dependent drift speed. Figure 2 shows typical photodetector signals for three frequency differences and for the intensity and crossing angle stated above. The detuning is $\Delta/2\pi = +90 \text{ MHz}$, and the frequency difference $\delta/2\pi$ is set smaller than 6 kHz . Under these conditions, the atomic kinetic energy in the frame of the moving optical potential, $(m\delta/q)^2/2m \ll (3.7\hbar k)^2/2m$, is much smaller than the optical potential depth $(8.9\hbar k)^2/2m$, and thus localized atomic motion is achieved. Atoms preferably populate the $F=2, m=0$ ground state due to optical pumping. Therefore, our optical potential depth refers to the transition $F=2, m=0 \rightarrow F'=3, m=0$.

When $\delta/2\pi = 6 \text{ kHz}$ [Fig. 2(a)], damped oscillation of the output from PD1 is observed, which reflects the oscillation of the mean atomic position relative to the slowly moving

optical potential [5]. Such an oscillation of the mean atomic position cannot be induced when atoms are exposed to a stationary standing wave because of the symmetry of the system. We actually observed that the damped oscillation of PD1 output vanished at $\delta/2\pi = -0.4 \text{ kHz}$ [Fig. 2(b)]. We interpret this as follows: Due to an intensity imbalance between the MOT and PGC counterpropagating cooling laser beams, the atoms were cooled to a nonzero average velocity. Therefore, a small δ (i.e., small drifting speed of the potential) was needed to compensate that motion. Even though the oscillation of the output of PD1 vanished, we still observed a damped oscillation of the PD2 signal [Fig. 2(c)]. This implies that the residual atomic dynamics caused an oscillatory phase change of the probe beam but no intensity modulation. The oscillation frequencies of Figs. 2(a) and 2(c) are $5.38 \pm 0.05 \text{ kHz}$ and $9.0 \pm 0.1 \text{ kHz}$, respectively.

We now present the physical mechanism responsible for

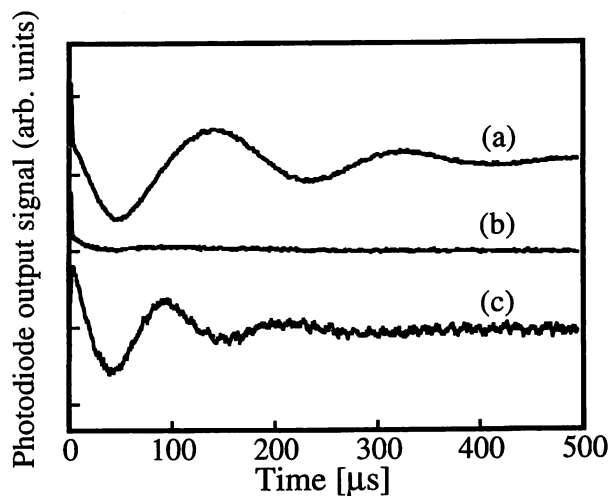


FIG. 2. The output signals of two photodetectors [(a), (b)] PD1 and (c) PD2. The frequency differences for each signal were (a) $\delta/2\pi = 6 \text{ kHz}$ and [(b), (c)] $\delta/2\pi = -0.4 \text{ kHz}$, respectively.

the observed oscillatory phase change. The intensity and phase change of the probe arise from a recoil-induced resonance and can be represented by the imaginary and real part of the induced polarization P , respectively, approximately given by

$$P \cong -\frac{2\mu\Omega}{\Delta} \int dr_q \exp[i(qr_q - \delta t)] \overline{\psi^*(r_q, t)\psi(r_q, t)}. \quad (1)$$

Here Ω and μ are the Rabi frequency and the transition dipole moment, respectively. $\psi(r_q, t)$ is the atomic center-of-mass wave function, and $\overline{\psi^*(r_q, t)\psi(r_q, t)}$ represents the ensemble average of the density distribution for all of the atoms (atomic density grating), since they all contribute to the signal. From Eq. (1), we can understand that an intensity change of the transmitted probe beam originates in a modulation of the atomic density distribution proportional to $\sin(qr_q - \delta t)$. Because the optical potential has a periodicity of $\cos(qr_q - \delta t)$, the time evolution of the average atomic position relative to the minima of the periodic potential leads to the intensity change of the transmitted probe beam. On the other hand, a change of the rms position spread of the atoms relative to the potential minima affects only the real part of the polarization, implying that a temporal change of the rms position spread of the atoms manifests itself as a phase change of the probe beam. In Fig. 2, even when the intensity change vanished [Fig. 2(b)], a phase change was still observed [Fig. 2(c)]. Thus the damped oscillation of the observed phase change corresponds to an oscillation of the width of the position distribution.

In order to investigate the characteristics of signal decay, we simulated the atomic dynamics when cold atoms are nonadiabatically exposed to the stationary standing wave. We neglected spontaneous emission and calculated the time evolution of the atomic density operator, assuming that its initial value was given by a Maxwell-Boltzmann distribution with temperature T . The time evolution of the density operator after switching on the probe light abruptly was determined by the Hamiltonian, which consists of the atomic kinetic energy and the optical potential,

$$i\hbar \dot{\rho}_{p'p''} = \frac{1}{2m} (p'^2 - p''^2) \rho_{p'p''} - \frac{U_0}{4} (\rho_{p' - \hbar q, p''} + \rho_{p' + \hbar q, p''} - \rho_{p'p'' + \hbar q} - \rho_{p'p'' - \hbar q}), \quad (2)$$

where the density operator is given in momentum representation, and U_0 is the potential depth.

In Fig. 3, solid lines show the time evolution of the rms position spread of the atoms calculated by Eq. (2). Dashed lines are results calculated with Newton's equation in the same periodic potential. The crossing angle, detuning, and intensity of each probe beam were the same as in Fig. 2. The initial rms momentum Δp_q of the atoms along \vec{q} was set to (a) $\Delta p_q/\hbar q = 0$, (b) $\Delta p_q/\hbar q = 1$, and (c) $\Delta p_q/\hbar q = 17$, respectively. Here, Fig. 3(c) corresponds to our experimental condition of $T = 4.7 \mu\text{K}$. In Fig. 3, the time is given in units of $2\pi/\omega$, where ω is the Bohr frequency between the lowest and first excited vibrational levels. Both the classical and quantum-mechanical results show that the oscillation of the

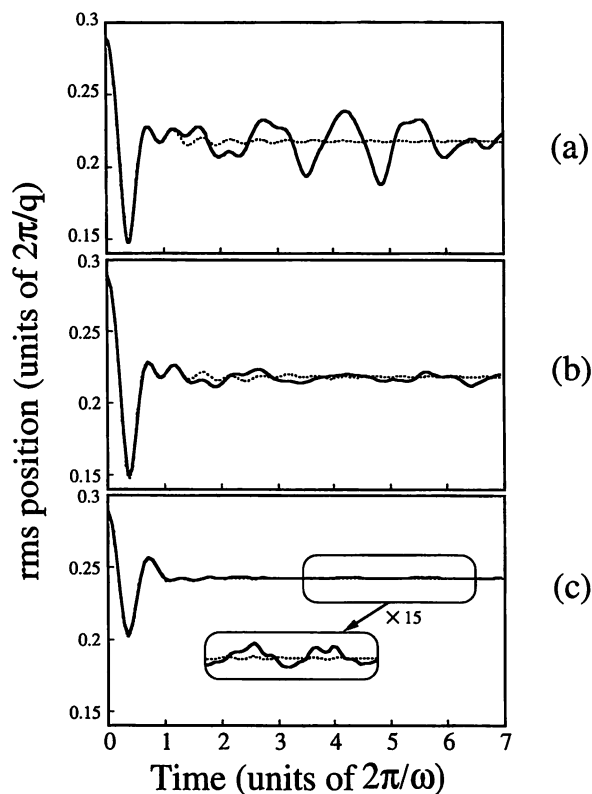


FIG. 3. Numerical results for the time evolution of the root-mean-square (rms) position of the atomic density distribution relative to the minima of the standing-wave potential. Solid and dotted lines are quantum and classical results, respectively. The rms position spreads were estimated inside one spatial period of the potential, $2\pi/q$. The initial rms momentum Δp_q of the cold atoms along \vec{q} are (a) $\Delta p_q/\hbar q = 0$, (b) $\Delta p_q/\hbar q = 1$, and (c) $\Delta p_q/\hbar q = 17$, respectively.

rms position of the atoms damps rapidly due to the dephasing effect caused by the trap anharmonicity. When $\Delta p_q/\hbar q = 0$, a remarkable difference between classical and quantum simulations appears, which is a quantum revival of the atomic oscillation [solid curve in Fig. 3(a)]. [Our calculation shows that our optical potential contains approximately 50 discrete bound states. Under conditions such as in Fig. 3(a), the sudden turn-on of the potential excites the atoms to a distribution of vibrational levels whose mean excitation quantum number is 23. Here, the effect of the periodicity of the lattice and tunneling from one well to another is taken into account.] For large initial rms momentum of the atoms, such as in Fig. 3(b), and in particular in Fig. 3(c), the quantum revival, visible in Fig. 3(a), diminishes [solid curves in Figs. 3(b) and 3(c)]. Nevertheless, although the quantum calculation approaches the classical one for the calculation of Fig. 3(c), the calculation shows that a slight revival still exists [see inset of Fig. 3(c)].

Figure 4 shows both (a) experimental [same as Fig. 2(c)] and (b) numerical results of the phase change signal of the transmitted probe, where Fig. 4(b) was calculated using Eqs. (1) and (2) for the conditions of Fig. 3(c). Figure 4(a) is in good agreement with Fig. 4(b), which indicates that the decay rate of the phase signal was mainly determined by dephasing due to trap anharmonicity. While a slight quantum

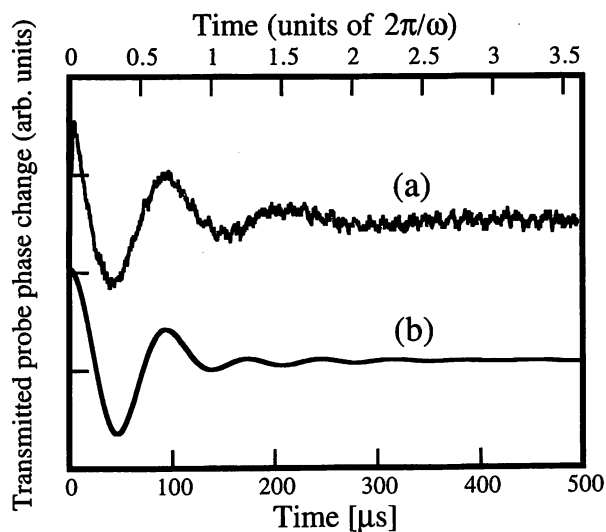


FIG. 4. (a) Experimental and (b) numerical results for the phase change signal of the transmitted probe. Note that (a) is identical to Fig. 2(c).

revival appears in Fig. 3(c), we did not observe such a revival in our experiment. We believe that there are two reasons why we could not observe the revival. First, our signal-to-noise ratio was insufficient. Second, the population of sublevels other than the $F=2$, $m=0$ state was not zero. At-

oms in these sublevels experience optical potential depths shallower than that for the $F=2$, $m=0$ state due to the smaller relative transition strength, which causes an additional spread of frequencies and increases the damping rate of the atomic density grating oscillation. However, using a large crossing angle and a subrecoil cooling technique [15,16], the value of $\Delta p_q/\hbar q$ could be decreased such that the quantum revival of the atomic oscillation could be observed.

The calculated oscillation frequencies of the rms width (measured by PD2) and mean position (measured by PD1) are $(1.49 \pm 0.08)\omega$ and $(0.88 \pm 0.02)\omega$, which are smaller than 2ω and ω due to the potential anharmonicity. The corresponding experimentally observed frequencies of 1.2ω (9.0 kHz) and 0.73ω (5.4 kHz) are 17% smaller than the numerical values. We believe this is because atoms in sublevels other than the $F=2$, $m=0$ state experience a shallower potential depth. The experimental value of the ratio of these frequencies is 1.7, which agrees perfectly with our theoretical result.

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Kozuma *et al.* Reply: The Comment [1] on our recent Letter [2] gives a new insight into the decay mechanism of the recoil-induced resonance signal [2–4] or the atomic motion of optically trapped atoms [5,6]. They suggested that the anharmonicity and the cooling rate, not the optical inelastic scattering (IS) rate, are important for the signal decay. In our Letter, the signal decay was attributed to two effects, i.e., IS and the anharmonicity. We also mentioned that the effect of IS can be suppressed by Lamb-Dicke effect [7,8]. However, we recently found out the signal decay rate was mainly determined by the anharmonicity for our experimental conditions. We also found that, as the Comment says, the calculated IS rate appeared to be larger than the experimental signal decay rate even though one takes into account the Lamb-Dicke effect. The Comment suggests a concise explanation of the signal decay mechanism. In this Reply, we reexamine the IS rate and the dephasing rate due to anharmonicity in order to check the validity of the new idea.

In our Letter, we used the optical saturation power of 1.6 mW/cm^2 which is for the transition between $F = 2, m = 2$ and $F' = 3, m = 3$. Considering the relative transition strengths between Zeeman sublevels, the $F = 2, m = 0$ ground state is estimated to be mostly populated. Thus, our two-level approximation can be applied to this transition. The relative transition strengths between $F = 2, m = 0$ and $F' = 3, m = 0$ states is 0.6 of that between $m = 2$ and $m = 3$. Thus, the optical scattering rate R becomes smaller than that in the Comment, i.e., $\epsilon = 0.4$ and $R = 3 \times 10^5 \text{ s}^{-1}$. Nevertheless, the IS rate of $n = 0$ state is still about 10^5 s^{-1} which is larger than the experimental decay rate $2.5 \times 10^4 \text{ s}^{-1}$. Moreover, it should be noted that, in our experiment, the cold atomic cloud was nonadiabatically exposed to a standing wave optical potential. Therefore, atoms are represented as a superposition of many vibrational states. The calculation performed in terms of energy bands revealed that there are 52 discrete vibrational levels and the mean quantum number is 23 under our experimental conditions. These results indicate that the IS rate $R(2n + 1)\epsilon$ is much larger than the experimental decay rate. It means that conventional Lamb-Dicke theory fails to explain our experimental results.

On the other hand, calculations of the decay rate of probe signal due to anharmonicity without the IS yield the rate of $4 \times 10^3 \text{ s}^{-1}$. In spite of this value being smaller than the experimental value ($2.5 \times 10^4 \text{ s}^{-1}$), it might be explained considering the contribution of ground state sublevels. The population of the sublevels other than $F = 2, m = 0$ state are not zero. Atoms in these sublevels feel different optical potential depths due to the difference of the relative transition strength, which causes an additional spread of frequencies and increases the damping rate of the atomic density grating oscillation.

Here, one question still arises. If this new idea is reasonable, the atomic coherence time becomes much longer than that predicted by the Lamb-Dicke theory. Therefore the coherence time is long enough for the revival of oscillations of the atomic density grating to be observed. However, we did not observe such a revival effect. Under our experimental conditions, the initial de Broglie wavelength was much smaller than the spatial period of the potential and many vibrational levels were populated due to the nonadiabatic optical potential switching. Although the atomic motion approaches the classical one for such a condition, our calculation showed that a small quantum revival exists. The calculation was performed with parameters of the actual experiment using atomic density operator in momentum space, where IS effect was neglected. In the real experiment, due to the existence of Zeeman sublevels, the oscillation damps faster than expected from a two-level calculation. We presume this is the reason why we did not observe the revival effect. The optical lattice, where many atoms are cooled down to the lowest vibrational level, is the one of the candidates to observe such a quantum mechanical revival phenomenon.

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CThY4

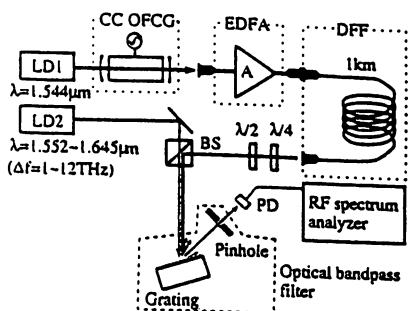
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Beat signal measurement of 12-THz separated lasers by using self-phase-modulation in optical fiber

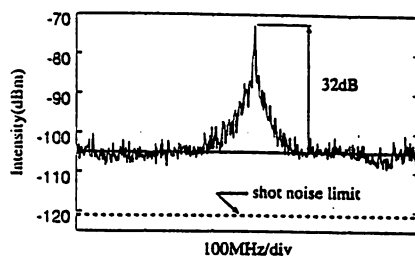
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An optical frequency comb generator¹ (OFCG) is based on a Fabry-Perot electro-optic modulator² and generates >1000 sidebands that are equally spaced by the modulation frequency. When the frequency of one of the sideband components is compared with that of another laser, the difference frequency between the two lasers can be measured with an accuracy as high as that of the microwave oscillator.

To increase the OFC span, we applied self-phase-modulation (SPM) in optical fiber to the output of the OFCG. Since the comb generated by OFCG forms a short pulse train whose repetition rate is determined by the modulation frequency, the spectral broaden-



CTHy4 Fig. 1. Experimental setup. CC OFCG: coupled-cavity monolithic OFC generator. EDFA: erbium-ytterbium-doped fiber amplifier. DFF: dispersion-flattened optical fiber. $\lambda/4$: quarter-wave plate. $\lambda/2$: half-wave plate. BS: beam splitter. PD: photodetector.



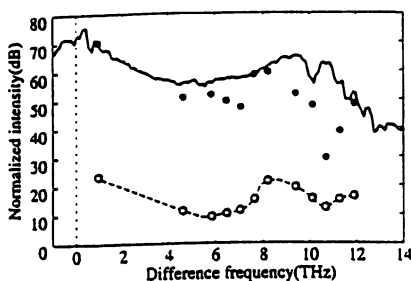
CTHy4 Fig. 2. Measured beat spectrum when the difference frequency of two lasers was 12 THz. Span and resolution bandwidth are 500 and 1 MHz, respectively. Dashed line shows shot-noise limit.

ing of the pulse train is equivalent to additional generation of equally spaced sidebands. We have already realized the spectral broadening of the output of the OFCG up to 30 THz³ by using an optical fiber amplifier and a 1-km-long dispersion flattened optical fiber (DFF).⁴

To verify the applicability of the sidebands and examine the effect of the spontaneous emission noise added in the amplifier, we measured beat signals between the additionally generated sidebands and another laser. As shown in Fig. 1, the laser beam from a 1.54- μm semiconductor laser (LD1) was introduced into a coupled-cavity monolithic OFCG,⁵ where the modulation frequency and modulation index were 6 GHz and 0.75 rad, respectively. After amplification up to a peak power of 2 W with 1 ps pulse width, the OFC span was increased to 30 THz in a DFF (anomalous dispersion $0.12 \text{ ps} \cdot \text{km}^{-1} \cdot \text{nm}^{-1}$ at 1.54 μm). The output of the DFF was mixed with the laser beam from a 1.6- μm semiconductor laser (LD2) that is tunable from 1.55 to 1.64 μm . Because a 2-GHz-bandwidth photodiode was used for the mixed beam detection, only the lowest frequency beat signal was measured with a radiofrequency spectrum analyzer.

The measured beat spectrum is shown in Fig. 2 when the difference frequency of the two lasers was 12 THz. The signal-to-noise ratio was 32 dB when the resolution bandwidth was 1 MHz. The measured noise intensity was 16 dB larger than the shot-noise limit, and it was determined by beat noise between the output of LD2 and noise in the DFF output.

Dependencies of beat signal and noise in-



CTHy4 Fig. 3. Dependencies of the beat signal and noise intensities normalized to shot-noise limit. Closed circles are signal intensity. Open circles are noise intensity. Solid curve is beat signal intensity estimated from measured spectrum.

tensities on the difference frequency are shown in Fig. 3. The beat signal and noise intensities are normalized to the shot-noise limit. The solid curve is beat signal intensity estimated from the measured spectrum. The measured beat signal intensities (closed circles) depend on the spectral profile. Since spontaneous emission spectrum added in the amplifier is broadened by the signal pulse intensity, dependency of noise intensity (open circles) has a similar profile to that of the measured spectrum. Because of these noise characteristics, the beat signal-to-noise ratios were almost constant at any difference frequencies.

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Super wide span Optical Frequency Comb generation by nonlinear self-phase-modulation in an optical fiber

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An optical frequency comb (OFC) generator², which is based on an Electro-Optic modulator enclosed in a high finesse optical cavity⁶ driven by microwave with a constant frequency, generates more than one thousand of equally spaced sidebands on large frequency span as wide as several THz from a cw laser light. Those sidebands are called as an OFC. When frequency of one of the sideband-components in the OFC is compared with the other laser by using photodetectors, the difference frequency between the laser and the carrier frequency of the OFC can be determined with an accuracy as high as that of the microwave frequency, and frequency and phase of lasers can be controlled very precisely within the span of the OFC. The OFC generator is a suitable device for frequency measurement of widely wavelength-tunable lasers such as semiconductor lasers used in optical communication networks. An optical frequency atlas of semiconductor lasers locked to 1.5 μ m bands acetylene lines have been made by using an OFC generator⁷. Therefore, there has been a heightened interest in expanding OFC generation.

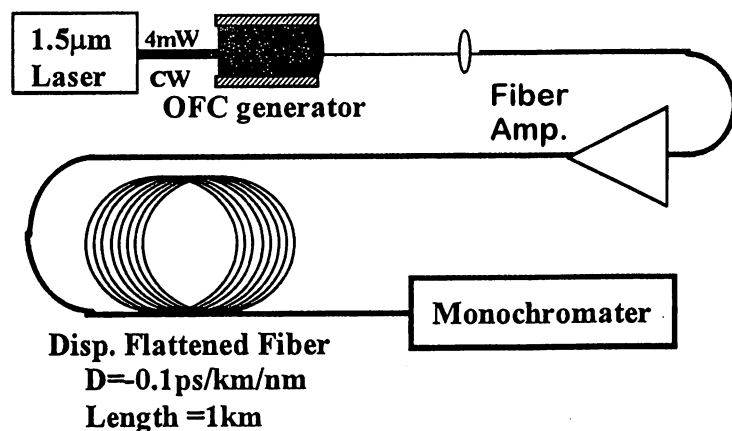


Fig. 1 Experimental setup.

In order to expand the frequency span of an OFC much more, we applied nonlinear self-phase-modulation (SPM) technique⁸ to output of an OFC generator. Since, additionally generated sidebands by SPM is also equally spaced, SPM can be applied for expansion of OFC span.

The experimental configuration is shown in Fig.1. The laser beam (4mW) from an 1.54 μ m semiconductor laser was introduced into a coupled-cavity monolithic OFC generator². Then the output of the OFC generator was introduced into a dispersion-flattened-fiber⁹ (DFF) via an optical fiber amplifier. The modulation frequency of the

OFC generator and the efficiency of OFC generation were 6GHz and 25%, respectively. The output of the OFC forms a train of short optical pulses. The width of optical pulse have been measured as 1ps. The dispersion of DFF was -0.12ps/km/nm at 1.54 μ m wavelength. The optical fiber amplifier, connected before the DFF, amplified the average OFC power to 40mW which corresponds to peak power of pulses as high as 2W.

The spectral envelopes of OFC's are shown in Fig.2. Where curves a, b and c are the spectral envelopes of the original OFC, the amplified OFC, and the OFC after passing through the DFF, respectively. Though the gain curve of the fiber amplifier affected the shape of the OFC envelope as shown in curve b, the curve c shows that the envelope was extended to a span as large as 47THz with power larger than 1nW. This value corresponds to 8000 sidebands generation, and is about 7 times larger than the span of OFC obtained without DFF (curve a).

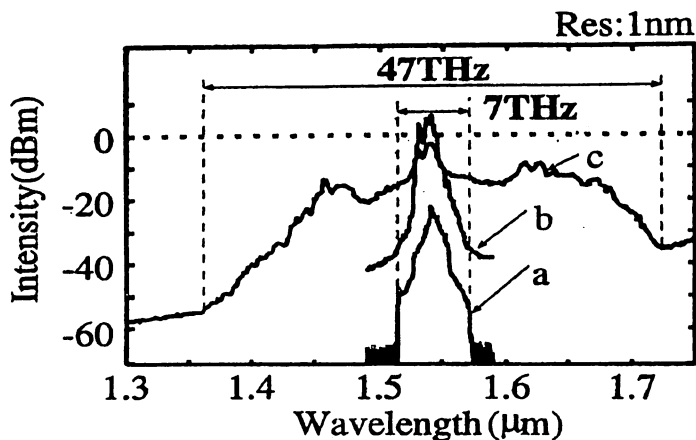
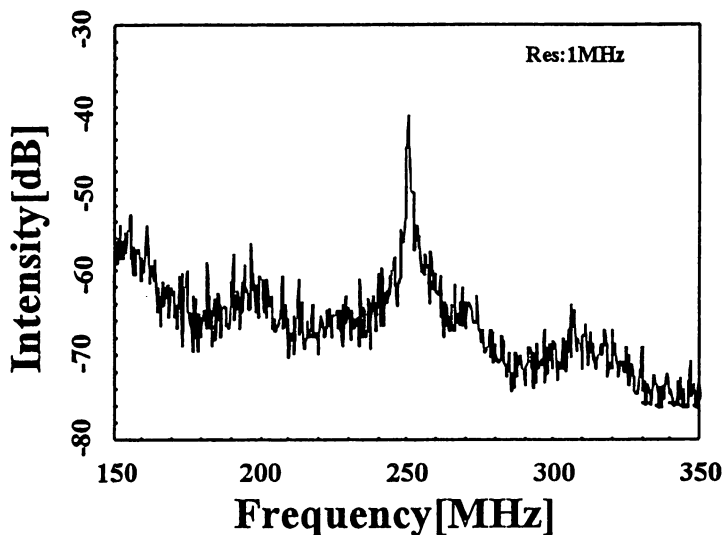
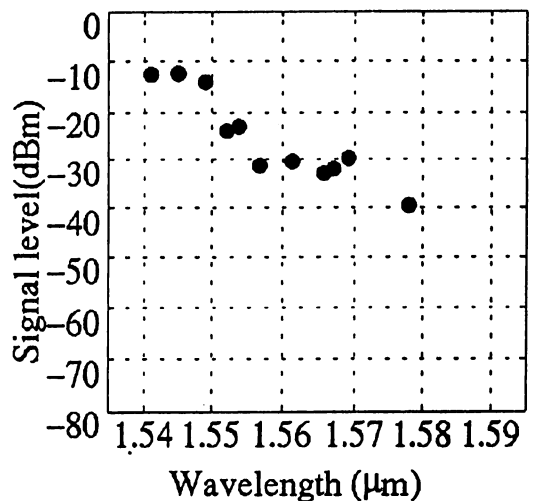


Fig.2 Spectral envelopes of OFC.
a:original OFC.
b:amplified OFC.
c:OFC after passing through the DFF.



(a)



(b)

Fig.3 Beat signal. (a) Spectrum of the beat signal. (b) Signal level of the beat signal as a function of the wavelength of the reference laser.

The authors would like to thank Mr.M.Onishi, Sumitomo Electric Industries for the helpful discussions about optical fibers.

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広スパン光周波数コムの発生と応用

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広い周波数スパンをもつ光コムを発生する方法として、電気光的手法に加え、光による自己位相変調の効果を用いる方法を中心に紹介した。これにより、得られた光コムのスパンは 50 THz に達した。さらにこの応用の一例として、搬送波周波数から 12 THz 離れた光コムの一成分と他のレーザー光の間のビート信号検出について記した。

Keywords : optical frequency comb, electro-optic modulator, laser frequency measurement, laser frequency stabilization, self phase modulation, optical soliton, super continuum

1. ま え が き

電磁波はその振幅、周波数、位相を制御することにより、通信、計測など多様な用途に用いられる。このような制御はマイクロ波周波数帯では高精度に実現される。これは超高精度な周波数標準の存在と、異なるマイクロ波間の周波数や位相を比較し制御できる電子回路技術の成果である。一方、光の周波数や位相の比較制御は容易ではない。なぜなら、電子回路で取り扱える周波数が高々 100 GHz (10^{11} Hz) であるのに対し、光の周波数は 1 THz (10^{12} Hz) ~ 1 PHz (10^{15} Hz) に達するからである。しかし、今後の光エレクトロニクス発展のためには光をマイクロ波と同等以上に制御する必要が増大すると考えられる。この要請に応えるために近年光の周波数や位相を測定するための光周波数コム(以下光コムと略記)の研究が盛んになっている¹⁻¹⁵⁾。

これに対しわれわれは、まずファブリ・ペロー形電気光学変調器(FP/EO変調器)^{16,17)}を光コムの発生に用い¹⁻⁹⁾、発生周波数範囲(以下スパンと略記) 10 THzを実現した。同様な原理かつ光導波路構造のコンパクトな光コム発生器も最近実現されている^{8,9)}。これらにより発生した光コムは周波数多重光通信のためのレーザー周波数制御や、広範囲に分布する吸収線の周波数測定などに使用されている。例えば、波長 1.5 μm 帯に広く分布するアセチレン分子の数十本ある吸収線の周波数測定に利用された¹³⁾。さらに、この方法によるレーザー光の差周波数測定の精度は 10^{-14} に達し¹⁴⁾、より高い精度の測定を要求する物理定数の決定にも有効である。実際、光コムは水素原子の 1S-2S 状態の紫外遷移周波数測定装置に組み込まれ、リュードベルグ定数や重陽子構造半径の測定に用いられた^{18,19)}。

光コムのスパンのさらなる拡大のために近年われわれは

光ファイバー中での光の自己位相変調を利用し、従来の 5 倍のスパンを実現した¹⁵⁾。以下ではこれらについて概説する。

2. 光コムとは何か

マイクロ波周波数帯では、広い周波数範囲で周波数変換を行う装置としてコムジェネレーターがある。これは図 1 (a) に示すように、入力周波数の逆数に等しいくり返し時間間隔で幅の狭い電圧パルスが発生する装置である。このパルスは周波数軸上で図 1 (b) に示すように入力周波数の整数倍のみ成分をもつ櫛(コム)形のスペクトルとなり、その信号はコムとよばれる。コムの各高調波は周波数軸上のマーカーとして機能し、受信器、位相同期システムの基準周波数、局部発振器に利用されている。周波数軸上

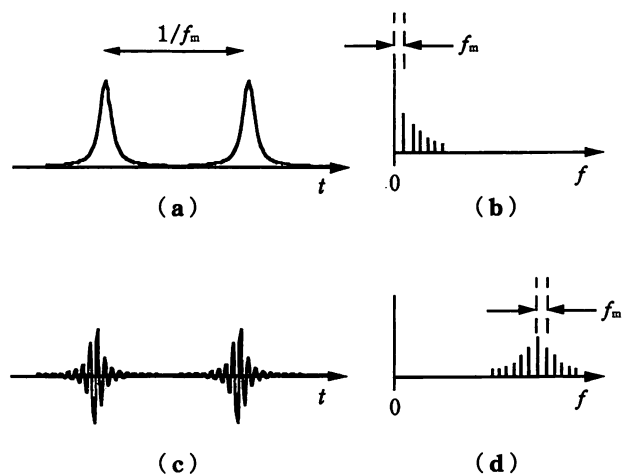


図 1 マイクロ波周波数と光の場合のコムの時間軸上の波形とスペクトル形状。(a)マイクロ波の場合の波形。(b)スペクトル形状。(c)光の場合の波形。(d)スペクトル形状。

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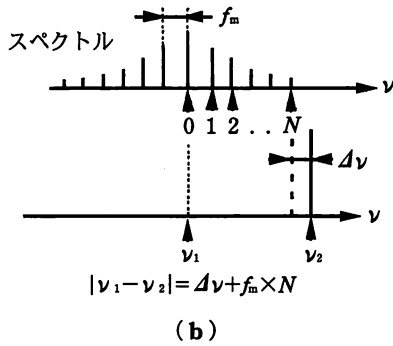
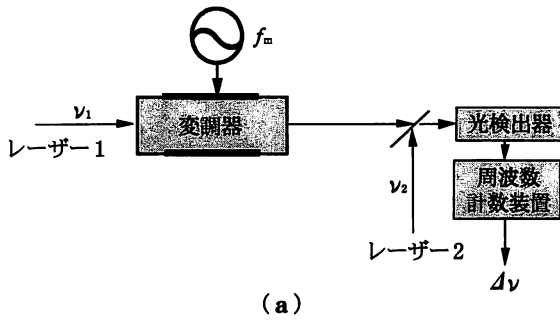


図2 (a)光コムによる差周波数測定の方法。(b)光コムとレーザーの周波数の関係。周波数 f_m で変調された周波数 ν_1 の光と周波数 ν_2 の光を重ね合わせ光検出器に入射し、光周波数コムとして発生した第 N 番目の変調サイドバンドとの間のビートの周波数 $\Delta\nu$ を測定することによって差周波数が決定される。

でのコムのスパンはパルス幅の逆数程度なので、これを広げることはパルス幅を狭めることに等価である。

一方、光では図1(c)に示すような包絡線パルスの発生が可能であり、パルス幅の最小値は数 fs に達する。これは、図1(d)のような光コムのスパンとして 1 PHz に達する値の実現可能性を意味する。しかし光パルスは包絡線パルスであるため搬送波をもち、搬送波の周波数が安定でなければ、安定な光コムとはならない。これを解決する一つの方法は、シングルモードレーザーの外部変調による変調サイドバンドを光コムに用いることである。この場合パルス変調、位相変調どちらでもよい。この時、搬送波の周波数は被変調レーザーに等しいため、周波数安定化レーザーを用いれば安定な光コムが得られる。

安定な光コムが発生すれば、それを基準に他のレーザーの周波数を決定することができる。すなわち、図2のように周波数 f_m で変調された周波数既知のレーザー1 (周波数 ν_1) の光と周波数未知のレーザー2 (周波数 ν_2) の光を重ね、光コムの第 N 番目の変調サイドバンドとの間のビート周波数 $\Delta\nu$ を測定すれば、 $|\nu_1 - \nu_2|$ が決定され、また ν_2 が決定される。

3. 電気光学効果によるコムの発生

現在最も広いスパンの光コムを発生できる変調器は、図3のファブリ・ペロー (FP) 共振器に EO 変調器を挿入

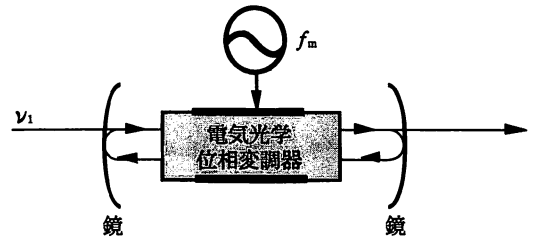


図3 FP/EO 変調器の構成。2枚の鏡で構成されたFP共振器にEO変調器(位相変調器)を挿入している。

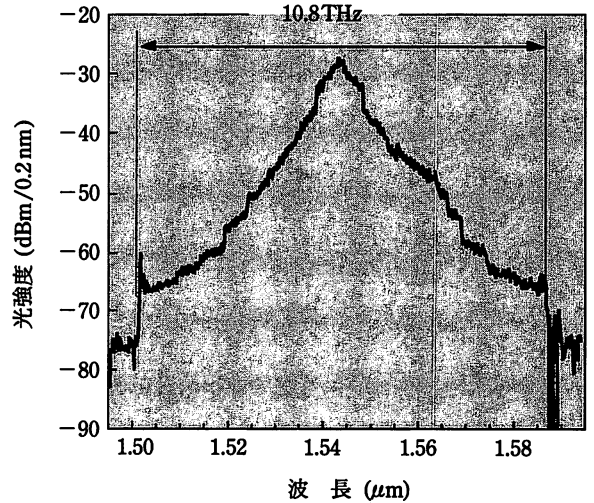


図4 FP/EO 変調器を用いて得られた、光コムの包絡線の形状。

した FP/EO 変調器である。これ自体を光周波数コム発生器とよぶこともある。この変調器では FP 共振器に共鳴した光が多重に EO 変調器を往復することにより深い変調が得られ、高次のサイドバンドを発生できる。このとき変調周波数は FP 光共振器の自由スペクトル域の整数倍に一致させる必要がある。時間軸上での出力の波形は変調周波数の 2 倍の周期のローレンツ関数型の光パルス列となる。このときパルス幅 τ は、

$$\tau = \frac{1}{2Ff_m\beta} \quad (1)$$

となる。ここで F は FP 共振器のフィネス、 β は EO 変調器の片道の変調指数である。フィネスが大きいほどパルス幅は小さく、したがってその逆数に対応するスパンは広くなる。

そこで、より高いフィネスを得るためにわれわれは、EO 変調器用の結晶端面に高反射膜を蒸着することによって、結晶自体を FP 光共振器にしたモノリシック構造の FP/EO 変調器を開発した⁴⁻⁷⁾。これによりフィネスは 400 から 600 を得た。これを用いて得られた光コムのスペクトルを図4に示す。ここで使用した半導体レーザーの波長は 1.5 μm である。 β は 0.7 rad であった。使用した分光器が変調周波数 6 GHz に等しい間隔をもつ光コムの一つ一つの各

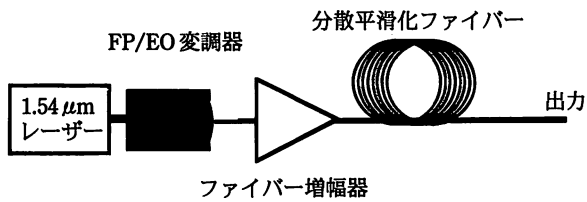


図 5 光コムのスパン拡大のために光ファイバー中の自己位相変調効果を用いた実験系。

サイドバンド成分を分離測定するほど分解能が高くなかったために、光コムの包絡線のみが得られている。この図よりスパンは約 10 THz に達することがわかる。

このような光コムの差周波数測定への応用が可能であることを確認するために、そのサイドバンドの一つと他のレーザーとの間のビート信号を観測したところ、コムの搬送波とそのサイドバンドとの間の差周波数から 5 THz 離れたサイドバンドまでビート信号の観測が可能であった。しかしながら、図 4 を見てわかるようにスペクトルの両端で急峻に光コムのパワーが減少している。また、この場合の(1)式で推定されるパルス幅は 200 fs ($1 \text{ fs} = 10^{-15} \text{ s}$) であるが、実測値は 1 ps であった。これらは、EO 結晶の分散によって FP 共振器の自由スペクトル域がもはや一定ではなく波長の関数となったことに起因する。

EO 変調器による光コムのスパンの分散限界 $f_{d\text{-limit}}$ は

$$f_{d\text{-limit}} = \sqrt{\frac{8c\beta}{D\pi\lambda^2}} \quad (2)$$

式で表される^{5,20)}。ここで D は群速度分散である。実験で得られたスパンの値は、ほぼこの分散限界に等しいことが確認された。

4. 自己位相変調によるスパンの拡大

先に述べたようにコムのスパンの限界は EO 結晶の分散で決まるので、分散補償を行えばスパン拡大ができるはずである。しかしながら分散補償を FP 共振器中で行うと、フィネスが低減すると考えられる。この問題を回避するためにわれわれは光ファイバー中の光の自己位相変調²¹⁾によるスペクトル拡大を利用した。先に述べたように、コム自身は時間軸上では高速な光パルスであるので、光ファイバー材質のシリカのように光の強度に応じて屈折率の変化する非線形光学媒質中では光パルスの強度変化により自己位相変調ができる。パルス周期は自己位相変調の後も変わらないので、自己位相変調で新たに発生したサイドバンドの間隔も変調周波数に等しくなり、光コムとして使用できる。自己位相変調によるスペクトル拡大はスーパーコンティニウム^{22,23)}の発生にも用いられる方法であり、原理的には光パワーが大きければ 1 PTHz のコムの発生が期待できる²³⁾。

図 5 に実験系を示す。FP/EO 変調器 に入力する半導体

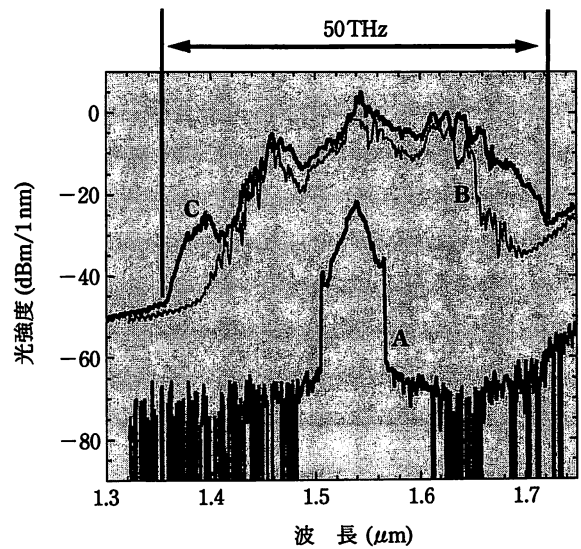


図 6 FP/EO 変調器と光ファイバー内の自己位相変調効果を用いて得られた、光コムの包絡線の形状。A: 光ファイバーを通過する前。B, C: ファイバー増幅器と光ファイバーを通過した後。B: 平均パワー 100 mW。C: 平均パワー 250 mW。

レーザーの波長は $1.544 \mu\text{m}$ である。先に述べた FP/EO 変調器でこの光を変調し、この平均パワーを光ファイバー増幅器で 100~250 mW 程度まで増幅した。このときの光コムのパルス幅は 1 ps と測定され、これからピークパワー 10~25 W と推定される。この増幅された光を、1 km の分散平滑化ファイバー²⁴⁾に入射させた。分散平滑化ファイバーとは群速度分散と、群速度分散の傾きを補償した光ファイバーである。群速度分散は $1.544 \mu\text{m}$ の波長においてわずかに (-0.1 ps/nm/km) 異常分散となるように設定されている。これにより、光ファイバーを通過する間に光は高次のソリトンの振舞により短パルス化し、狭い時間幅に光強度が集中し、さらに大きな自己位相変調を引き起こす。

図 6 は光ファイバーを通過する前の光コム(A)と、通過した後の光コム(B: 平均パワー 100 mW, C: 平均パワー 250 mW)のスペクトルを示している。特に曲線 C により、この方法によって発生した光コムのスパンが 50 THz 程度まで拡大したことがわかる。この値は、より高次の光ファイバーの分散により制限されていると考えられている。さらに波長 $1.552 \mu\text{m}$ から $1.645 \mu\text{m}$ まで波長可変な半導体レーザーを用意し、曲線 B の光コムの一つとサイドバンドとの間のビート信号を観測した。図 7 に搬送波からの周波数差に対するビート信号(A)と雑音強度(B)の依存性を示している。ここで 0 dB はショット雑音で決まる測定限界である。この図からビート信号と雑音強度の変化がともに光コムの包絡線のスペクトル形状変化に相当しており、12 THz までほぼ 30 dB 以上の SN 比を維持していることがわかる。A と B の差である SN 比の値は約 30 dB に達し、周波数測定への応用には十分である。雑音レベルは光ファ

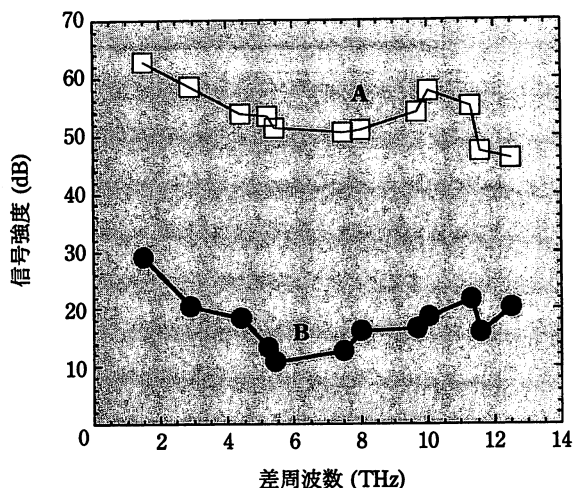


図7 コムのサイドバンドと他のレーザーとの間のビート信号と雑音強度のレベル。横軸はコムのキャリアとそのサイドバンドとの間の差周波数。ここで雑音測定時の帯域幅は1 MHzに設定された。0 dBはショット雑音レベルを表す。
A: 信号。 B: 雑音。

イバー増幅器から発生する自然放出光の影響によって発生したものと考えられる。

今後はより広範囲に波長を掃引できるレーザーを用いれば、12 THzを超える範囲でビート信号の観測が可能になると期待される。また、光ファイバーのより高度な分散制御を行えばスパンのさらなる拡大も可能であると考えられる。

5. まとめと展望

本稿では、広い周波数スパンをもつ光コムの発生方法として、電気光学的手法に加え、光の自己位相変調の効果を用いる方法を用いることで光コムのスパン50 THzが得られることを示した。さらに、この応用として搬送波周波数から12 THz離れた光コムのサイドバンドの成分と他のレーザー光の間のビート信号の観測結果を紹介した。

現在の光コムの周波数安定度は、搬送波であるレーザーで決定される。これはマイクロ波周波数帯におけるコムとの大きな違いである。しかしながら、スパンがさらに拡大すれば、搬送波の周波数をも周波数標準に同期できるようになる。これによりマイクロ波周波数帯と同等の安定度をもつ光コムを実現できる。

例えば、何らかの周波数をもつ基本波と、その第二次高調波との間をカバーできるような光コムを考えてみる。このような光コムがあれば、図2と同様に、基本波と第二次高調波の間の差周波数が測定される。ところが、この測定された差周波数はまさに基本波の周波数そのものである。すなわち、相対的な光の周波数の測定を行う光コムでも、絶対的な光の周波数の測定ができる。また、同時に光コムの搬送波の周波数も決定でき、それを用いてコムの搬送波の周波数を周波数標準に同期できる。

この例が示唆するように近い将来マイクロ波帯と同等に安定な光コムが近い将来実現し、広い分野に応用されると期待できる。

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