Nutation in energy transfer of dressed photons between nano-particles

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Abstract

Experimental results on the temporal behavior of dressed photon (DP) energy transfer are presented. Nanometer-sized particles (NPs) of cubic CuCl crystals grown in a NaCl crystal were used as the sample for the experiments. By measuring the temporal variations of the photoluminescence (PL) intensities emitted from these NPs, unique characteristics of DP energy transfer were found. Namely, the PL intensities exhibited nutation that originated from the bidirectional DP energy transfer between the small and large NPs. The period of the nutation was 50 ps. The periodic variation of the PL intensity emitted from the small NP had a phase lag of $\pi/3$ behind that from the large NP. The duration of the forward DP energy transfer was longer than that of the backward transfer. The difference between these times resulted in temporal modulation of the PL intensities. The temporal variations exhibited additional pulsatory variations whose period was one-*fourth* the nutation period.

1. Introduction

A dressed photon (DP) is an off-shell quantum field created by the interactions among photons, electrons, and phonons in a nanometer-sized space [1-4]. During DP energy transfer between nanometer-sized particles (NPs), the energy is dissipated by transformation to the energies of propagating free photons. These energy transfer and dissipation processes have been used to detect the DP, and have been applied to realize innovative nanometer-sized optical devices, nanometer-resolution fabrication technologies, energy conversion devices, and so on [5,6].

The novel phenomena of size-dependent resonance and autonomy in the DP energy transfer have been experimentally observed [7]. Further experimental studies have identified that the spatio-temporal properties of these phenomena corresponded to the quantum walk (QW) process [8]. Based on this identification, novel studies on quantum probability theory have commenced recently [9].

This paper presents the results of experiments on the temporal behavior of DP energy transfer carried out in order to promote these studies, especially to establish theoretical models of the QW.

2. Experimental methods

This paper presents experimental results that correspond to the QW system in Fig. 1. Two semiconductor NPs played the roles of nodes 1 and 2. The DP, transferred between these NPs bidirectionally, played the role of a link. The light incident into and emitted from the NPs played the role of the input and output signals, respectively. The sample used for the experiments consisted of cubic NPs of CuCl crystals (volume density of $2\times10^{17}/\mathrm{cm}^3$) that were grown in a NaCl crystal (Fig. 2(a)). The average center-to-center distance between the adjacent NPs was 12 nm, which was estimated by assuming that the random spatial distribution of the grown NPs was a Weibull distribution.

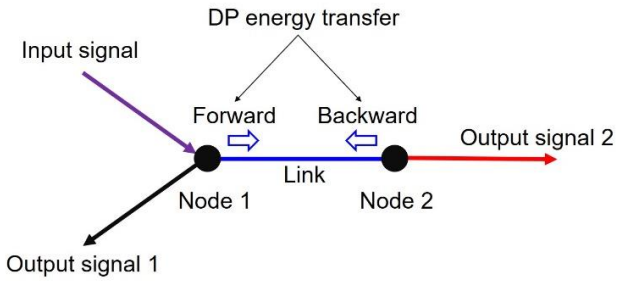

Fig. 1 The QW system to be experimentally realized in the present paper.

Two of the NPs in the NaCl crystal were used as nodes 1 and 2. As is schematically explained by Fig. 2(b), they were small and large cubic CuCl-NPs (NP_S and NP_L, respectively). Pulsed light (10 ps pulse width and 381 nm wavelength) was used as the input signal. For the output signals 1 and 2, photoluminescences (PLs) emitted from the excitons in NP_S and NP_L, respectively, were used. Quantum states of these excitons were identified by a set of three quantum numbers (1,1,1). The wavelengths of these PLs were 383 nm and 385 nm, respectively. The sample temperature was maintained at 80 K.

Two strategies, **(S1)** and **(S2),** were planned to experimentally realize the system of Fig. 1 by using the sample in Fig. 2(a):

(S1) For selective application of the input signal only to node 1:

As is schematically explained by Fig. 2(b), a pair of NPs, consisting of NP_S and NP_L, with $a_{S,eff}$: $a_{L,eff} = 1:\sqrt{2}$, was used. Here, $a_{S,eff}$ and $a_{L,eff}$ are the effective side lengths, representing the spatial extent of the quantum field of the exciton. The required length ratio was achieved by selectively using NP_S and NP_L of 3.9 nm ($= a_{S,eff}$) and 5.5 nm($= a_{L,eff}$), respectively[1)]. Their center-to-center distance was 12 nm, estimated as described above.

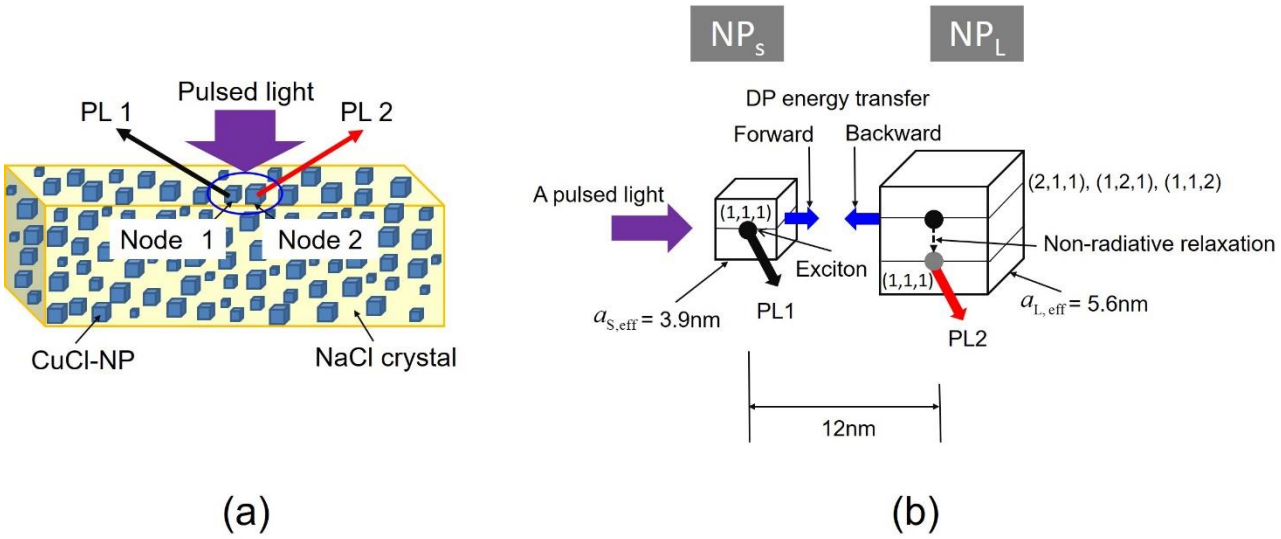

Fig. 2 The sample used for the experiments.

(a) Cubic NPs of CuCl crystals grown in a NaCl crystal. (b) Small and large cubic CuCl-NPs (NP_S and NP_L, respectively), used as nodes 1 and 2 in Fig. 1.

By irradiating the sample with pulsed light, an exciton was excited to the (1,1,1) state of NP_S. It should be noted that, because of the length ratio above, the energies of the triply degenerate states (2,1,1), (1,2,1), and (1,1,2) in NP_L were equal to that of the (1,1,1) state of NP_S. However, even under such resonant condition, the exciton could not be exited to these states because they are electric dipole-forbidden states with respect to propagating light. This indicated that the incident light was effective to excite the exciton only in NP_S, realizing selective application of the input signal to node 1.

1) Their geometrical side lengths were 4.6 nm ($= a_S$) and 6.3 nm ($= a_L$), respectively.

(S2) For distinguishing between output signals 1 and 2:

[Output signal 1]

The PL emitted from the exciton in the (1,1,1) state of NP_S was used as output signal 1. It should be noted that this exciton also creates a DP.

[Output signal 2]

{Creation} The energy of the DP, created by the exciton of the (1,1,1) state in NP_S, transferred to NP_L and excited the exciton to the triply degenerate states (2,1,1), (1,2,1), and (1,1,2). This excitation was possible because these states were electric-dipole allowed states with respect to the nanometer-sized DP. In other words, the long-wavelength approximation, popularly employed in conventional optical science, was violated by the DP. The exciton excited by the DP above emitted the PL. It also created the DP, and its energy transferred back to NP_S, resulting in bidirectional DP energy transfer between NP_S and NP_L. This transfer phenomenon has been called nutation.

{Measurement} It might be possible to use the PL emitted from NP_L as output signal 2. However, its wavelength was equal to that of output signal 1 due to the resonance condition above. Therefore, output signal 2 could not be distinguished from output signal 1, and thus, it could not be selectively measured. For selective measurement, a rapid non-radiative relaxation, specific to the CuCl-NPs, was used by de-exciting the exciton from the triply degenerate states to the state (1,1,1), allowing subsequent emission of the PL. Its wavelength (385 nm) was longer than that (383 nm) of output signal 1 due to the energy dissipation by the non-radiative radiation. Although the magnitude of this dissipation was as low as 1/100 times the photon energies of output signal 1, it was sufficiently large to allow output signals 1 and 2 to be distinguished. Thus, this PL could be used as output signal 2.

As a result of the planned strategies **(S1)** and **(S2)** above, the realized system of Fig. 2(b) exhibited the following differences **(D1)** – **(D3)** from the system of Fig.1. They were:

(D1) Node 2 was split into three because of the triple degeneracy of the exciton state in NP_L.

(D2) A part of the DP energy dissipated at node 2 due to the non-radiative relaxation of the exciton in NP_L .

(D3) Since the effective side lengths ($a_{S,\mathrm{eff}}$ and $a_{L,\mathrm{eff}}$) of NP_S and NP_L were different from each other, the magnitudes of the created DP energies and their spatial extents, represented by a Yukawa-type function $V(r)$ ($V(r) \propto \exp(-r/a)/r$, where a is the size of the NP), were different. Thus, their DP energy transfer times, being inversely proportional to $V(r)$, were different. That is, since $a_{S,\mathrm{eff}} < a_{L,\mathrm{eff}}$, the transfer time of the forward path of the link (from NP_S to NP_L) was longer than that of the backward path (from NP_L to NP_S).

By noting the differences **(D1) - (D3)**, Fig. 1 was revised to Fig. 3(a). However, for a basic discussion of the QW process, it should be possible to approximate Fig. 3(a) by the simpler system of Fig. 3(b) because of the sufficiently low magnitude of the energy dissipation in **(D2)**.

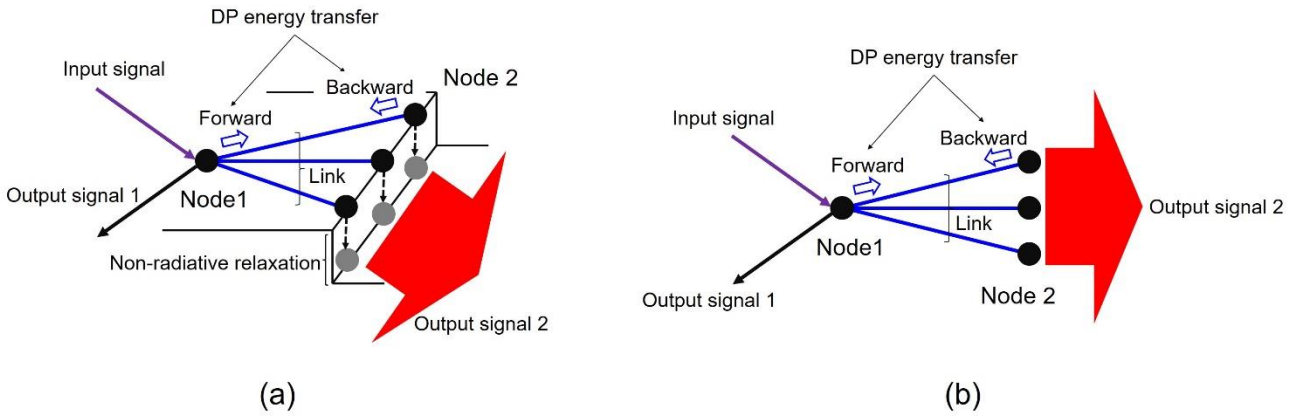

Fig. 3 The revised QW system.

3. Experimental results and discussions

The black and red curves in Fig. 4 represent the measured temporal variations of the outputs 1 and 2, respectively. They were the PL intensities emitted from the (1, 1, 1) states of the excitons in NP_S and NP_L. They exhibited monotonic decreases with time, which originated from the conventional radiative relaxation. In the time span 0–500 ps immediately after applying the pulsed input signal, the temporal decrease was attributed to the QW process and was fitted by $\exp(-t)$ [9]. The subsequent decrease in the time span from 500 ps to 4 ns was slower and was fitted by $\exp\left(-\sqrt{t}\right)$, which was attributed to the random walk process. Since the present paper focuses on the phenomena relevant to the QW process, especially on the nutation of the DP energy transfer, experimental results acquired in the time span 0–500 ps were analyzed. For these analyses, Fig. 5 was acquired by expanding the horizontal axis of Fig. 4.

The black and red curves in Fig. 5 indicate the temporal variations of the DP energy transfer of the forward and backward paths, respectively. Their pulsatory variations represent nutation with a period of 50 ps. This value of the period was compatible with the period (40 ps) that was estimated from the relation between the center-to-center distance (10 nm) and the transferred DP energy (1×10^{-4} eV) [10].

Figure 5 indicates the following unique characteristics **(C1)**–**(C3)** of DP energy transfer:

(C1) The phases of the pulsatory variations of the two curves are different from each other: In order to estimate the magnitude of this difference, the Fourier components of the 50 ps-period were extracted from these curves and are shown in Fig. 6. The black and red curves in this figure represent nutation, and the sinusoidal variation of the black curve had a phase lag of $\pi/3$ behind that of the red curve. This lag originated from the triply degenerate states of the exciton in NP_L (**(D1)** in Section 2). That is, the *triple* degeneracy caused a lag of one-*third* of π [2]. In other words,

within one-*third* of the DP energy transfer time of the backward path, the (1,1,1) state in NP_S was promptly occupied by the exciton that was initially created in the triply degenerate states in NP_L.

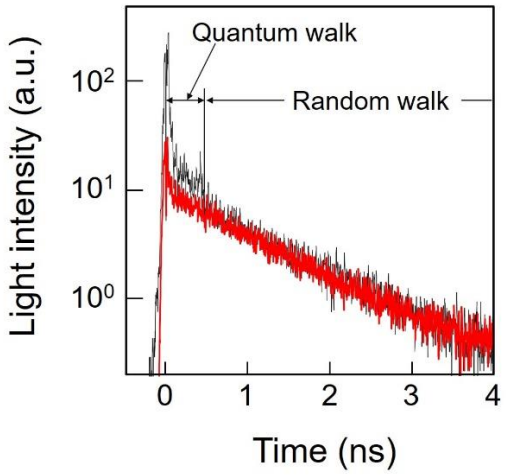

Fig. 4 The measured temporal variations of the two PLs.
The black and red curves represent outputs 1 and 2, respectively.

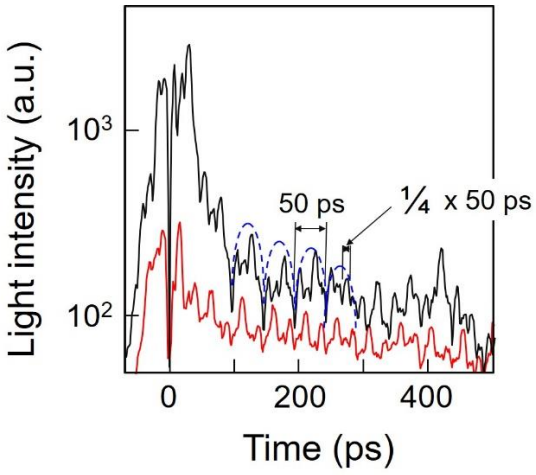

Fig. 5 The measured temporal variations of the two outputs.
They are the experimental results in the time span as short as 0–500 ps in Fig. 4

Since the DP energy was bidirectionally transferred between NP_S and NP_L, the profiles of the two curves in Fig. 6 should be anti-correlated, and thus, their phase difference should be as large as π. However, the phase lag was found to be $\pi/3$. The reason for this discrepancy was considered to be as follows: Since a large number of CuCl-NPs were buried in the NaCl crystal, the DP energies could transfer not only between NP_S and NP_L but also between multiple NP_Ls (or between multiple NP_Ss). Furthermore, it was confirmed by analyzing the measured absorption spectral profile of the sample that the number of NP_Ls was larger than the number of NP_Ss. This

indicated that the DP energy transfer between neighboring NP_Ls could contribute to the phase difference between the two curves. However, since the phase of the DP energy transfer between these NP_Ls is random, the anti-correlation characteristics did not clearly appear, and thus, the phase difference was maintained as small as $\pi/3$.

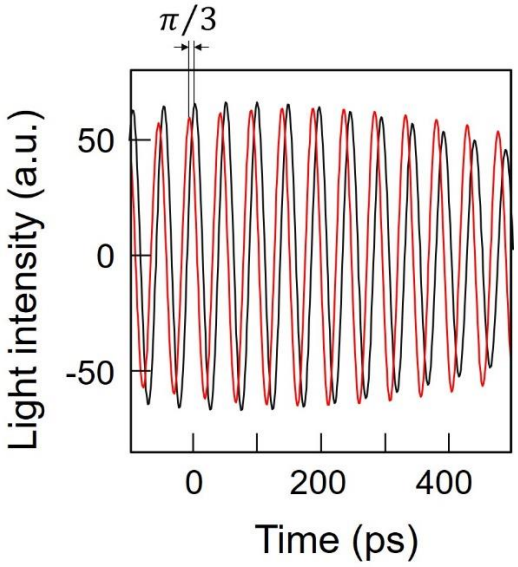

Fig. 6 The measured temporal variations of the two outputs.
They are the Fourier components of the 50 ps-period, extracted from the two curves in Fig. 5.

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2) This kind of phase lag has never been observed when a large number of NPs is used as the macroscopic material system for inducing conventional optical phenomena. This is because the NPs in this system have been approximated as a coupled quantum state having a singlet state of the exciton.

(C2) The pulsatory variations of the PL intensities were modulated (blue broken curve in Fig. 1): This was attributed to the fact that the transfer time along the forward path was longer than that along the backward path (**(D3)** in Section 2).

(C3) Additional pulsatory variations existed whose period was one-*fourth* that of the nutation: Such a short period was attributed to the fact that, among the *four* energy levels (the (1,1,1) state level in NP_S and the triply degenerate states in NP_L) relevant to the bidirectional DP energy transfer, only the (1,1,1) state in NP_S was initially occupied by an exciton at the commencement of the nutation.

Figure 7 shows the Fourier spectral profiles of the two curves in Fig. 5. The spectral peak (A) at 20 GHz corresponded to the nutation period of 50 ps. Because the profiles of the measured pulsatory variations deviated from sinusoidal curves, the spectral peak (B) of the second-order higher harmonic can be seen. The absence of the third-order spectral peak was attributed to the modulation in the PL intensity mentioned in **(C2)**. In other words, the odd-order higher harmonics

were missing due to the difference between the forward and backward transfer times. The spectral peak (C) was attributed to the superposition of the fourth-order higher harmonic and the additional pulsatory variations whose period was one-*fourth*, as mentioned in **(C3)**.

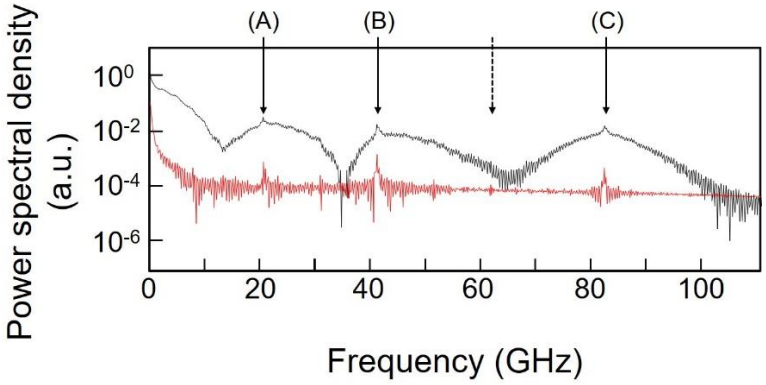

Fig. 7 The Fourier spectral profiles of the two curves in Fig. 5.

4. Summary

This paper presented experimental results on the temporal behavior of DP energy transfer in order to promote studies on the QW process. Cubic NPs of CuCl crystals grown in a NaCl crystal were used as the sample for the experiments. By measuring the temporal variations of the PL intensities emitted from these NPs, unique characteristics of DP energy transfer were found. They were:

[1] The PL intensities exhibited nutation that originated from the bidirectional DP energy transfer between the small and large NPs. The period of the nutation was 50 ps.

[2] The periodic variation of PL intensity emitted from the small NP had a phase lag of $\pi/3$ behind that from the large NP. This lag originated from the triple degenerate states of the exciton in the large NP.

[3] The duration of the forward DP energy transfer was longer than that of the backward transfer. This was attributed to the difference in the magnitudes of the created DP energies localized at the small and large NPs. This difference temporally modulated the PL intensities.

[4] The spectral profile exhibited additional pulsatory variations whose period was one-*fourth* the nutation period because the (1,1,1) state in the small NP was initially occupied by an exciton at the commencement of the nutation.

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