Non-adiabatic relaxation process that lies behind the adiabatic relaxation of dressed-photon–phonon transfer

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Abstract

This paper reports the results of numerical calculations based on a quantum walk (QW) model in order to analyze the temporal behavior of the dressed-photon–phonon (DPP) transfer. Since a random walk (RW) process lies behind the QW process, the time evolution operator of the interpolation model is used by introducing a probability p. The results reveal that: The crossover time t_x decreases with increasing p and χ / J (the ratio between the DP hopping energy J and the DP-phonon coupling energy χ). In contrast, th slope S_{RW} of the curve for the RW process increases. The slope S_{QW} of the curve for the QW process increases with increasing χ / J, which indicates that a part of the adiabatic relaxion energy is apt to be spontaneously converted to the non-adiabatic relaxation energy. In the case of $\chi / J \ll 1$, the nutation cycle is long and the slopes are small, which is advantageous for operating novel nanometer-sized devices.

1 Introduction

A dressed photon (DP) is created by a photon-exciton (or electron) interaction in a nanometer-sized particle (NP). Furthermore, the DP creates a dressed-photon–phonon (DPP) when it couples with a phonon. A previous article reviewed experimental results on the temporal behavior of DPP transfers among NPs [1]. Notable features of this behavior were: When two semiconductor NPs (NP_1 and NP_2) are illuminated by an optical pulse, a DPP is created and transferred bi-directionally between the two NPs (Fig. 1(a) in [1]).

As a result, these NPs emitted photoluminescence components (PL_1 and PL_2). The temporal behaviors of the light intensities PL_1 and PL_2 were as follows:

[1] In the early short time-span, the emitted light intensities pulsated due to the DPP nutation between the two NPs and decreased with a short time constant τ_{QW}. This decrease was due to the adiabatic energy relaxation, which was fitted by the function $y_{QR} = \exp(-t / \tau_{QW})$ of a quantum walk (QW) model.

[2] Subsequently, intensities decreased with a long time constant τ_{RW}. This was due to non-adiabatic energy relaxation, which was fitted by the function $y_{RW} = \exp(-\sqrt{t/\tau_{RW}})$ of a random walk (RW) model [2]. Finally, the nutation in **[1]** was buried in the non-adiabatic energy relaxation. Measured values of these time constants were τ_{QW} = 150 – 600 ps and τ_{RW} =0.7 – 15 ns. The nutation cycle was τ_r =50 – 155 ps [3].

This paper reports the results of numerical calculations carried out in order to analyze the experimentally found features **[1]** and **[2]** described above.

2 Methods and results of numerical calculation

Recent theoretical studies have found that a RW process lies behind the QW process, and the time evolution operator $\mathcal{L}$ for the interpolation model is represented by the sum of those of the QW and RW models ($\mathcal{L}^{\mathrm{QW}}$ and $\mathcal{L}^{\mathrm{RW}}$, respectively):

$$\mathcal{L} = (1-p)\mathcal{L}^{\mathrm{QW}} + p\mathcal{L}^{\mathrm{RW}}, \qquad (1)$$

where p $(0 \le p \le 1.0)$ is a probability [4,5].

This section presents the numerically calculated results of the temporal behaviors of the emitted light intensity in the early short time-span and in the subsequent long time-span based on eq. (1). For calculation, $\mathcal{L}^{\mathrm{QW}}$ is chosen with probability $1-p$, while $\mathcal{L}^{\mathrm{RW}}$ is chosen with probability p at each time step, independently. As a result, one can evaluate the temporal behavior of the emitted light intensity. The ratio χ/J between the DP hopping energy J and DP-phonon coupling energy χ was used as the physical parameter. It should be noted that the phonon is a quantum of the crystal lattice vibration that can induce non-adiabatic energy relaxation.

Figure 1 shows the calculated dependence of the emitted light intensities on the probability p in the case of χ/J =1.0. The slopes S_{QW} and S_{RW} correspond to the rates of intensity decreases that are due to the adiabatic and non-adiabatic relaxations (**[1]** and **[2]** in Section 1), respectively. Figure 1(a) represents the result at p =0, that is, the light intensities due to the adiabatic relaxation. Pulsative variations of the curves represent the nutation of the DPP transfer. Figure 1(b) is the result at p =0.15. Here, the nutation due to the adiabatic relaxation is seen in the early time-span shorter than t_x. In the time-span longer than t_x, the light intensity due to the non-adiabatic relaxation surpasses that due to the adiabatic relaxation. This means that the nutation is

buried in the non-adiabatic energy relaxation, as was pointed out in **[2]** in Section 1. Thus, t_x is named the crossover time. Figure 1(c) shows the results at p=1.0, which are governed by the non-adiabatic relaxation. It is easily confirmed that t_x is shorter than that in Fig. 1(b). The nutation can be faintly seen in the early time-span shorter than t_x.

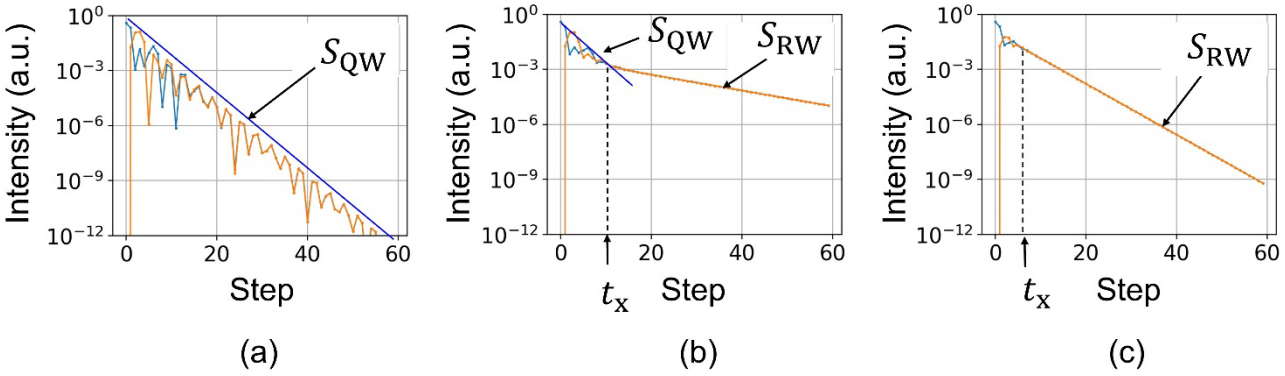

Fig. 1 Temporal variations of the emitted light intensities at χ / J =1.0.

Blue and red curves represent the intensities of PL_1 and PL_2, respectively. t_x is the crossover time.

(a) The results at p=0. Pulsation represents the nutation. S_{QW} is the slope due to the adiabatic relaxation. (b) The results at p=0.15. S_{RW} is the slope due to the non-adiabatic relaxation. (c) The results at p=1.0.

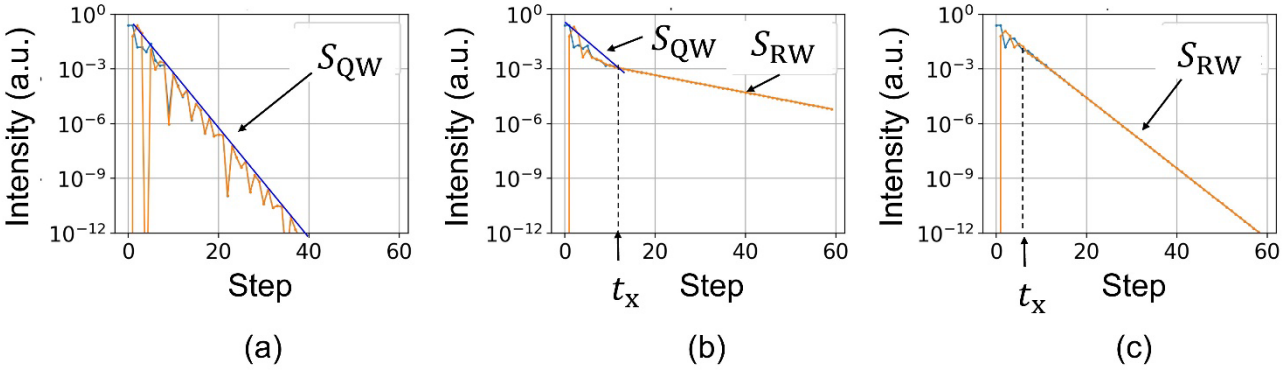

Fig. 2 Temporal variations of the emitted light intensities at χ / J =1.41.

(a) p=0. (b) p=0.15. (c) p=1.0.

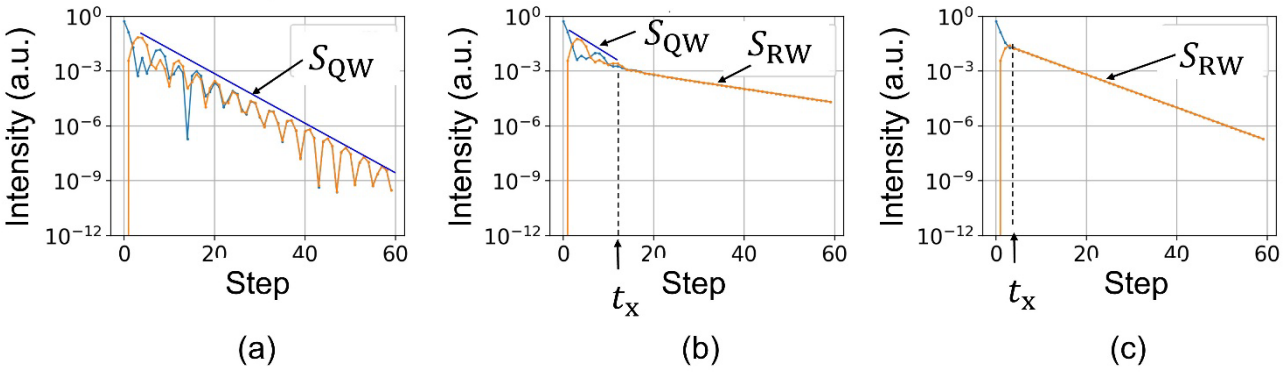

Fig. 3 Temporal variations of the emitted light intensities at χ / J =0.816.

(a) p =0. (b) p =0.15. (c) p =1.0.

Figures 2 and 3 represent the results at χ / J =1.41 and 0.816, respectively. Figure 4 presents the features of the curves in Figs. 1, 2, and 3. They are:

(1) (Fig. 4(a)) The crossover time t_x decreases with increasing p and χ / J. For practical applications, it should be pointed out that a longer crossover time t_x is advantageous for improving the performance in novel nanometer-sized devices such as a nano-optical condenser and an optical buffer memory [1].

(2) (Fig. 4(b)) The slope S_{RW} increases with increasing p and χ / J.

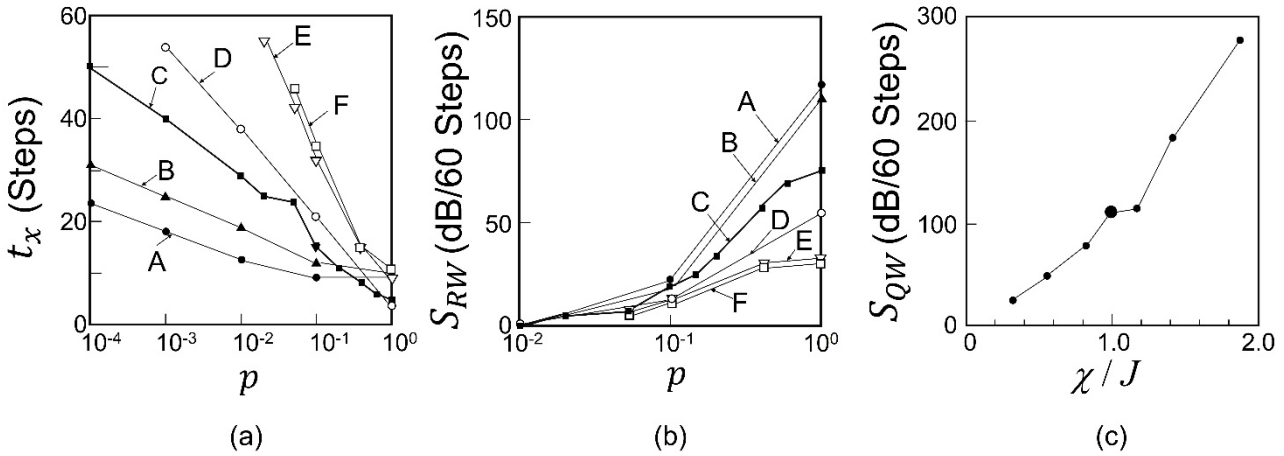

Fig. 4 Crossover time t_x, the slopes S_{RW} and S_{QW} of the curves.

χ / J =1.84 (A), 1.41 (B), 1.00 (C), 0.816 (D), 0.585 (E), and 0.329 (F).

(3) (Fig. 4(c)) The slope S_{QW} increases with increasing χ / J. This increase indicates that a part of the adiabatic relaxion energy is apt to be spontaneously converted to the non-adiabatic relaxation energy. The conversion rate is represented by ε in eqs. (4) and (5) of ref. [1].

Previous calculations using the QW model have confirmed that the DPP localized at the B atom-pairs in a Si crystal when $\chi / J \gg 1$ [6]. This localization has been used to invent a novel method of DPP-assisted annealing, resulting in the successful fabrication of revolutionary Si-LED and Si-laser devices [7]. To examine these revolutionary works, Fig. 5(a) represents the calculated results at $\chi / J = 10$. It is easily found that the crossover time t_x is shorter than that in Fig. 4(a), which means that the adiabatic relaxation is rapidly buried in the non-adiabatic relaxation. This is because the phonon, a possible source of the non-adiabatic energy relaxation, can efficiently couple with the DP when $\chi / J \gg 1$. Furthermore, it is found that the slope S_{QW} is larger than that in Fig. 4(c), which means that a part of the adiabatic relaxion energy is apt to be spontaneously converted to the non-adiabatic relaxation energy, as was presented in **(3)** above. In this situation, the DPP efficiently localizes at the position of the B atom-pair in the Si crystal for optimizing the spatial profiles of the B atom-pair by DPP-assisted annealing. As a result, highly efficient Si-LED and Si-laser devices were successfully fabricated [6].

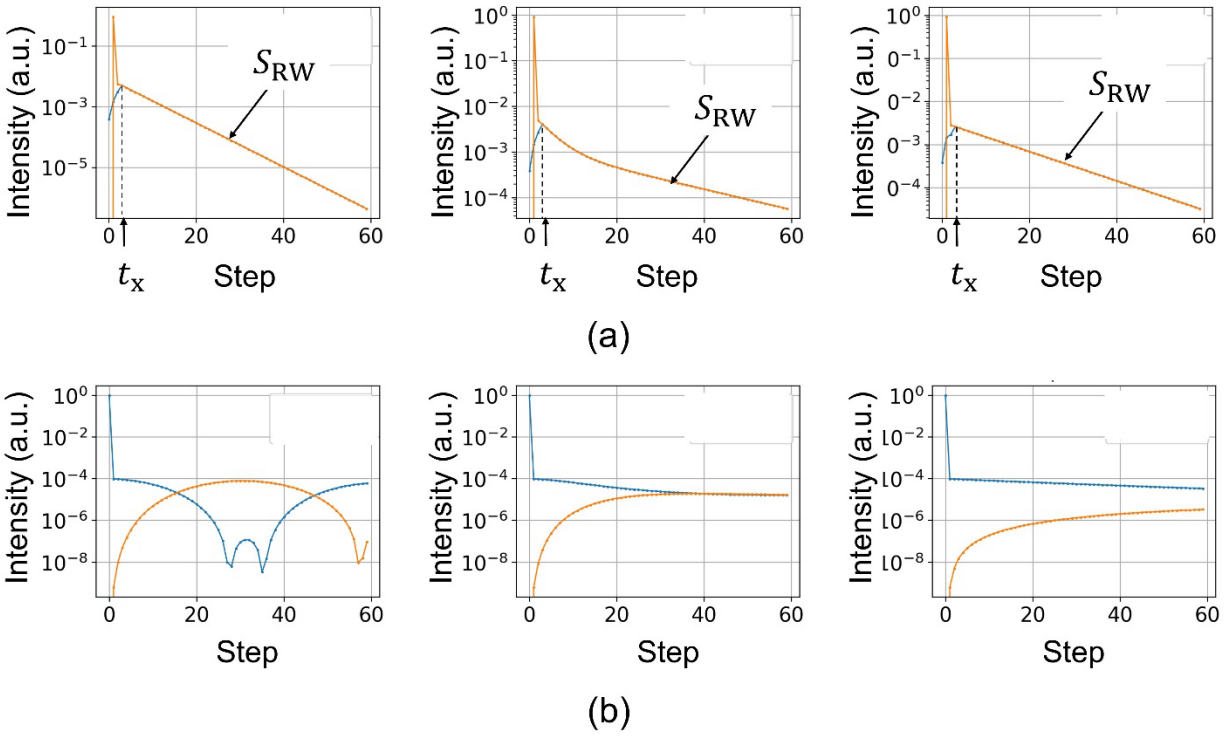

Fig. 5 Calculated results. (a) χ / J=10. (b) χ / J=0.1.
Left, center, and right figures are at p=0, 0.15, and 1.0, respectively.

Figure 5(b) represents the calculated results at $\chi / J = 0.1$. The profiles of the curves in this figure are conspicuously different from those in Fig. 5(a) and Figs. 1-3. The left figure shows that the nutation cycle is much longer than those in Figs. 1(a), 2(a), and 3(a). Furthermore, the slopes S_{RW} and S_{QW} are small. This is because the phonon, a possible source of the non-adiabatic relaxation, does not efficiently couple with the DP when $\chi / J \ll 1$. Thus, in this figure, the leading role in the energy transfer is played by the DP and not the DPP. The long nutation cycle and small slopes indicate that the condition $\chi / J \ll 1$ is advantageous for operating the novel nanometer-sized devices above.

5 Summary

In order to analyze the experimentally found features of the temporal behavior of the DPP transfer, this paper used the time evolution operator of the interpolation model because a RW process lay behind the QW process. The calculated results indicated the following unique features: The crossover time t_x decreased with increasing p and χ / J. In contrast, the slope S_{RW} increased. The slope S_{QW} increased with increasing χ / J, which indicated that a part of the adiabatic relaxation energy is apt to be spontaneously converted to the non-adiabatic relaxation energy. In the case of $\chi / J \ll 1$, the nutation cycle was long and the slopes were small, which are advantageous for operating novel nanometer-sized devices.

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