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Coherent-phonon-assisted excitation transfer via optical near fields in dilute magnetic semiconductor nanostructures

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Abstract Optical near-field (ONF) phenomena observed in nanoscale systems have been applied to nano-fabrication and nano-photoelectronic devices for the last decade. Excitation transfer (ET) is an elementary process caused by ONF interaction localized at the nanoscale in real space. Recently, we have observed spin-dependent ET controlled by an external field and discovered the sign of assistance of phonons. In this work, we elucidate that the assistance of coherent phonons in ET originates from two types of mechanism: energy resonance between exciton levels renormalized by phonon effects and enhancement by phase matching between coherent oscillation of populations and phonon oscillation. This study will stimulate the development of the application of ET via a ONF to nano-photoelectronic devices.

1 Introduction

Optical near-field (ONF) phenomena have been observed in nanoscale systems recently and ONF interaction is going to be applied to unique nano-fabrication and nano-photoelectronic devices [1–3]. Excitation transfer (ET) is an elementary process originating from ONF interaction localized at the nanoscale in real space, which is fundamentally different from the one via propagating electromagnetic fields in free space [4–8]. Recently, we observed spin-dependent ET in double quantum wells (DQWs) consisting of ZnCdSe of a non-magnetic-semiconductor quantum well (NMSQW) and ZnCdMnSe of a dilute-magnetic-semiconductor quantum well (DMSQW) as shown in Fig. 1 (a) [9]. The ET is controlled by an external magnetic field by utilizing the Zeeman effect by which an exciton energy level in the DMSQW splits into two levels dependently on spins in contrast to the NMSQW as shown in Fig. 1 (b). Moreover,

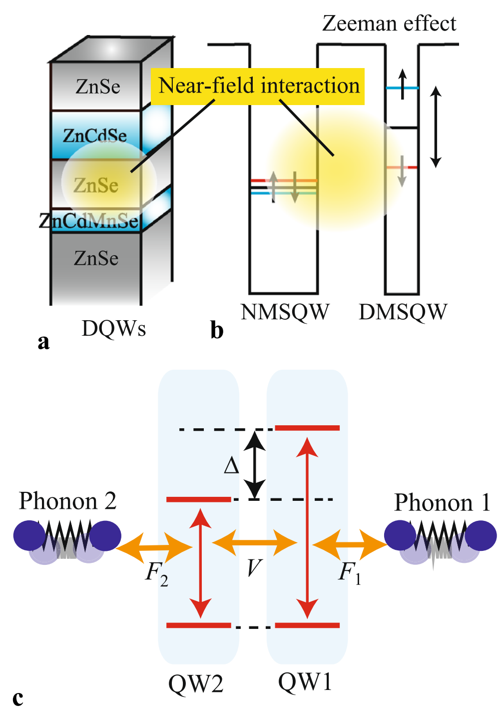

Fig. 1 Schematic drawing of an experimental system and a theoretical model. (**a**) Double quantum wells of ZnCdSe and ZnCdMnSe coupled by an optical near field. (**b**) Spin-dependent excitation transfer via an optical near field controlled by the Zeeman effect. (**c**) Theoretical model of a coupled system of two-level systems and phonons

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in this experiment, ET can occur when exciton levels in each QWs are not resonant, and the level spacing is nearly equal to the energy of LO phonons, which indicates that the ET is assisted by not only ONFs but also LO phonons.

In this work, we present a quantum-mechanical theory on ET via a ONF in excitonic systems coupled to LO phonons, and investigate the mechanism of an assistance of LO phonons in ET. In particular, effects of coherent phonons are considered and two types of mechanism of coherent-phonon-assisted ET are elucidated. The first one originates from the energy resonance between exciton energy levels renormalized by phonon effects in DQWs while the second one originates from the enhancement by phase matching between coherent oscillation of populations in DQWs and phonon oscillation. We derives the conditions to realize two types of coherent-phonon-assisted ET, where the condition of phase matching is more intriguing than that of the energy resonance for the assistance of phonons. This study will stimulate the development of the application of optical-near-field ET to nano-photoelectronic devices.

This paper is organized as follows. In Sect. 2, we present a quantum-mechanical theory on coherent-phonon-assisted ET in DQWs. In Sect. 3, we calculate ET dynamics and discuss two types of mechanism of the ET. We elucidate the conditions to realize the assistance of coherent phonons and their physical meaning. Finally, we summarize the study and give perspectives in Sect. 4.

2 Theoretical formulation

We consider a theoretical model as shown in Fig. 1 (c). Two-level systems (TLSs) represent excitons in DQWs. The exciton in QW1 (QW2) interacts with the LO phonon system in QW1 (QW2), and excitonic states in QW1 and QW2 are modified by LO phonons in QW1 and QW2, respectively. Moreover, excitons can transfer between QW1 and QW2 by ONF coupling, which is different from the electronic tunneling. However, LO phonons cannot transfer between DQWs by ONF coupling. The Hamiltonian of the total system is described as $H = \hbar(\omega_{\mathrm{ex}} + \Delta)a_1^\dagger a_1 + \hbar\omega_{\mathrm{ex}} a_2^\dagger a_2 + \hbar V(a_1^\dagger a_2 + a_2^\dagger a_1) + \hbar\omega_1^{\mathrm{LO}} b_1^\dagger b_1 + \hbar\omega_2^{\mathrm{LO}} b_2^\dagger b_2 + \hbar F_1 a_1^\dagger a_1 (b_1 + b_1^\dagger) + \hbar F_2 a_2^\dagger a_2 (b_2 + b_2^\dagger)$. The first and second terms represent the energies of excitons in QW1 and QW2, respectively, where $a_1^\dagger$ and a_1 ($a_2^\dagger$ and a_2) are creation and annihilation operators of excitons in QW1 (QW2). The level spacing between excitons in QW1 and QW2 is written as Δ. The third term represents the exciton transfer between QW1 and QW2 by the ONF coupling $\hbar V$. The fourth and fifth terms represent the energies of LO phonons in QW1 and QW2, respectively, where $b_1^\dagger$ and b_1 ($b_2^\dagger$ and b_2) are creation and annihilation operators of LO phonons and $\hbar\omega_1^{\mathrm{LO}}$ ($\hbar\omega_2^{\mathrm{LO}}$) is the energy of a LO phonon in QW1 (QW2). The sixth and seventh terms represent the interaction between excitons and LO phonons in QW1 and QW2 by their coupling constants F_1 and F_2, respectively.

The dynamics of the system is described by one-body density-matrix elements, which are population of excitons in QWl ($l = 1, 2$): $N_l(t) = \langle a_l^\dagger a_l\rangle(t)$, transfer between QW1 and QW2: $T(t) = \langle a_2^\dagger a_1\rangle(t)$, which imaginary part shows positive (negative) sign in case of transfer from QW1 (QW2) to QW2 (QW1), and mean field of coherent phonons in QWl ($l = 1, 2$): $B_l(t) = \langle b_l^\dagger\rangle(t)$. We obtain the closed-form equations of motion for such density-matrix elements by utilizing the second-order cluster-expansion truncation [10, 11] as follows:

$$\frac{d}{dt}N_1(t) = -2V\mathrm{Im}\big[T(t)\big], \tag{1}$$

$$\frac{d}{dt}N_2(t) = 2V\mathrm{Im}\big[T(t)\big], \tag{2}$$

$$\frac{d}{dt}T(t) = -i\tilde{\Delta}(t)T(t) + iV\big[N_1(t) - N_2(t)\big], \tag{3}$$

$$\frac{d}{dt}B_1(t) = -i\omega_1^{\mathrm{LO}}B_1(t) - iF_1N_1(t), \tag{4}$$

$$\frac{d}{dt}B_2(t) = -i\omega_2^{\mathrm{LO}}B_2(t) - iF_2N_2(t), \tag{5}$$

where $\tilde{\Delta}(t) = \Delta + 2\{F_1\mathrm{Re}[B_1(t)] - F_2\mathrm{Re}[B_2(t)]\}$ is the level spacing between exciton levels renormalized by coherent LO phonons in QW1 and QW2.

3 Results and discussion

3.1 Outline of numerical calculations

The equations of motion derived in Sect. 2 are numerically calculated. We assume that $\omega_1^{\mathrm{LO}} = \omega_2^{\mathrm{LO}} = \omega^{\mathrm{LO}}$ and $F_1 = F_2 = F$. The ONF coupling constant and exciton-LO phonon coupling constant are fixed to $V/\omega^{\mathrm{LO}} = 0.1$ and $F/\omega^{\mathrm{LO}} = 0.5$, respectively. Then we investigate the dependence of ET dynamics on Δ. In the initial state, the exciton populations in QW1 and QW2 are given as $N_1(0) = 1$ and $N_2(0) = 0.2$, respectively, and the other elements of density matrix are zero. We look for such conditions that coherent phonons prominently enhance ET, and elucidate the physical meaning of such conditions.

3.2 Two types of coherent-phonon-assisted excitation transfer

In case of no phonon assistance ($F = 0$), it has been already known that coherent oscillation of populations by ET occurs most strongly when exciton levels are resonant

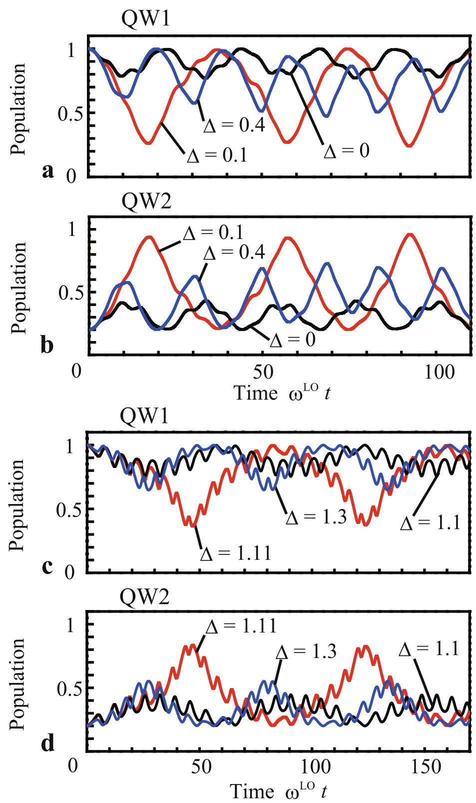

Fig. 2 Population dynamics by two types of coherent-phonon-assisted excitation transfer (ET). (**a**) and (**b**) show population dynamics by ET of type I in quantum well (QW) 1 and QW2, respectively, when $\Delta = 0$, 0.1, and 0.4. (**c**) and (**d**) show population dynamics by ET of type II in QW1 and QW2, respectively, when $\Delta = 1.1$, 1.11, and 1.3. Other parameters are fixed to $F = 0.5$ and $V = 0.1$. Δ, F, and V are normalized by ω^{LO}. Time is normalized by $1/\omega^{\mathrm{LO}}$

($\Delta = 0$), whose mechanism is often interpreted as the resonant dipole–dipole interaction. If level spacing Δ increases, the ET oscillation is damped and becomes rapid. The frequency is derived as $\Omega = (\Delta^2 + 4V^2)^{1/2}$, which is known as the Rabi frequency.

On the other hand, we calculate ET in case with the phonon assistance ($F = 0.5$). Figures 2 (a) (c) and (b) (d) show the population dynamics by ET in QW1 and QW2, respectively. In contrast to case of $F = 0$, Figs. 2 (a) and (b) reveal that the coherent oscillation of populations is enhanced strongly when $\Delta = 0.1$. This indicates that the coherent-phonon-assisted ET originates from a new type of mechanism besides the resonance between exciton levels $\Delta = 0$.

In addition, Figs. 2 (c) and (d) reveal that the ET also emerges strongly in case of $\Delta = 1.11$, also. Moreover, the time scale of the ET is much longer than the coherent oscillation of populations corresponding to the rapid oscillation. Summarizing the results of this subsection, we found two types of coherent-phonon-assisted ET; type I emerges in case of $\Delta = 0.1$ while type II does in $\Delta = 1.11$.

3.3 Mechanisms of two types of phonon assistance

In this subsection, we elucidate two types of mechanism of ET. First, we explain the mechanism of type I. The real exciton levels, which are $\hbar\omega_1^{\mathrm{ex}} = \hbar(\omega_{\mathrm{ex}} + \Delta)$ in QW1 and $\hbar\omega_2^{\mathrm{ex}} = \hbar\omega_{\mathrm{ex}}$ in QW2, are renormalized to the exciton levels, $\hbar\tilde{\omega}_1^{\mathrm{ex}}(t) = \hbar\omega_1^{\mathrm{ex}} + 2\hbar F\mathrm{Re}[B_1(t)]$ and $\hbar\tilde{\omega}_2^{\mathrm{ex}}(t) = \hbar\omega_2^{\mathrm{ex}} + 2\hbar F\mathrm{Re}[B_2(t)]$, respectively, by effects of coherent LO phonons. Therefore, the level spacing between excitons in QW1 and QW2, Δ, is also modified to the level spacing, $\tilde{\Delta}(t) = \Delta + 2F\{\mathrm{Re}[B_1(t)] - \mathrm{Re}[B_2(t)]\}$. The well-known resonance condition $\Delta = 0$ in case of no phonon effects is transformed to $\tilde{\Delta}(t) = 0$ in case with phonon effects. Such a condition for the phonon assistance of type I is drawn in Fig. 3 (a) schematically.

Second, we explain the mechanism of type II. The usual Rabi frequency of the coherent oscillation, $\Omega = (\Delta^2 + 4V^2)^{1/2}$, is modified to the effective Rabi frequency, $\tilde{\Omega}(t) = [\tilde{\Delta}(t)^2 + 4V^2]^{1/2}$, by phonon effects. Equations (1) and (4) ((2) and (5)) reveal that the exciton populations oscillate connecting with the mean field of coherent phonons. Therefore, when the condition of $\tilde{\Omega}(t) = \omega^{\mathrm{LO}}$ is satisfied, the phase matching between the coherent oscillation of exciton populations and the coherent-phonon oscillation is realized and the ET oscillation is enhanced strongly. We consider that this mechanism is more intriguing than type I. Such a condition for the phonon assistance of type II is drawn in Fig. 3 (b) schematically.

We show the evidence for two types of mechanism of the coherent-phonon-assisted ET in Fig. 4. Figures 4 (a), (b), and (c) show the population dynamics, time evolutions of $\tilde{\Delta}(t)$, and $\tilde{\Omega}(t)$ in case of type I. We found that $\tilde{\Delta}(t)$ oscillates around zero although $\tilde{\Omega}(t)$ does not change around ω^{LO}. These results prove that the ET of type I does not satisfy the phase-matching condition $\tilde{\Omega}(t) = \omega^{\mathrm{LO}}$, but satisfies the virtual resonance condition $\tilde{\Delta}(t) = 0$.

Figures 4 (d), (e), and (f) show the population dynamics, time evolutions of $\tilde{\Delta}(t)$, and $\tilde{\Omega}(t)$ in case of type II. We found that $\tilde{\Omega}(t)$ oscillates around ω^{LO} although $\tilde{\Delta}(t)$ does not oscillate around zero. These results prove that the ET of type II does not satisfy the virtual resonance condition $\tilde{\Delta}(t) = 0$, but satisfies the more interesting condition of phase-matching, $\tilde{\Omega}(t) = \omega^{\mathrm{LO}}$.

4 Conclusions and perspectives

We presented the quantum-mechanical theory on ET via an optical near field assisted by coherent phonons, and elucidated that the assistance of coherent phonons in ET originates from two types of mechanism; type 1 originates from the energy resonance between exciton levels renormalized by phonon effects in DQWs and type 2 originates from the

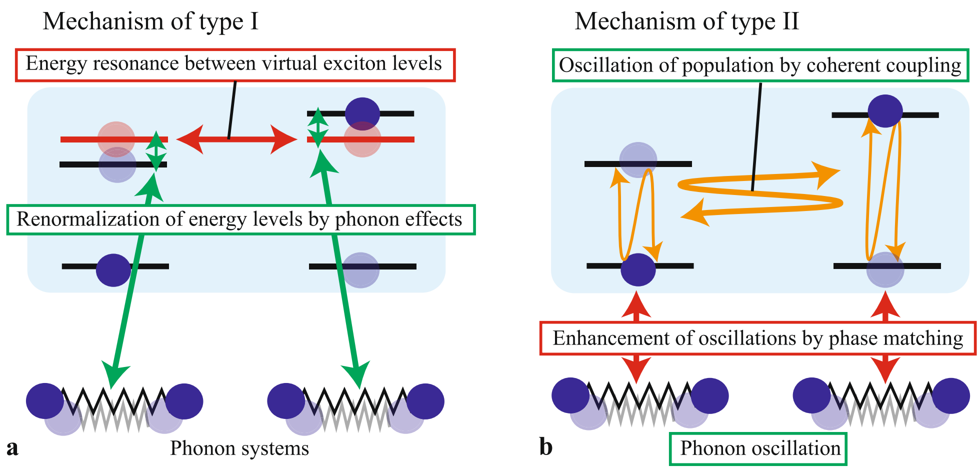

Fig. 3 Schematic drawings of two types of mechanism of coherent-phonon-assisted excitation transfer. (**a**) Mechanism of type I: energy resonance between virtual exciton levels renormalized by phonon effects in quantum well (QW) 1 and QW2. (**b**) Mechanism of type II: enhancement by phase matching between coherent oscillation of populations in double QWs and phonon oscillations

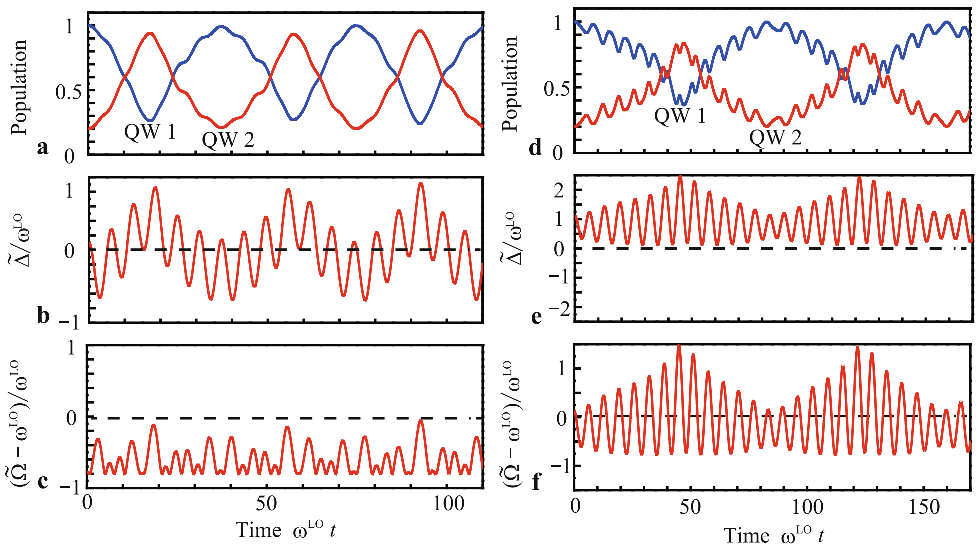

Fig. 4 Evidences for two types of mechanism of the coherent-phonon-assisted excitation transfer (ET). (**a–c**) Case of type I when $\Delta = 0.1$. (**d–f**) Case of type II when $\Delta = 1.11$. (**a**) and (**d**) show population dynamics in double quantum wells (DQWs) 1 and 2. (**b**) and (**e**) show time evolutions of effective level spacing between excitons renormalized by phonons in QW1 and QW2, $\tilde{\Delta}$. (**c**) and (**f**) show time evolutions of the effective Rabi frequencies of the coherent oscillation of populations in DQWs, $\tilde{\Omega}$. Horizontal broken lines in parts (**b**), (**c**), (**e**), and (**f**) represent zero. Other parameters are fixed to $F = 0.5$ and $V = 0.1$. Δ, F, and V are normalized by ω^{LO}. Time is normalized by $1/\omega^{LO}$

enhancement by phase matching between coherent oscillation of populations in DQWs and phonon oscillation. The conditions to realize the ET of types I and II are obtained as $\tilde{\Delta}(t) = 0$ and $\tilde{\Omega}(t) = \omega^{LO}$, respectively, where $\tilde{\Delta}(t)$ is the effective level spacing of excitons in QW1 and QW2 and $\tilde{\Omega}(t)$ is the effective Rabi frequency of the coherent-phonon-assisted ET. The phase matching in case of type II is more intriguing than the resonance condition in case of type I. In this paper, we introduced only the coherent element of phonons. However, incoherent phonons are expected to cause relaxation, dephasing, and energy dissipation, which are expected to affect ET. Therefore, in the forthcoming paper, we will investigate effects of incoherent phonons beyond the second-order cluster-expansion truncation. Moreover, we will clarify a microscopic origin of optical-near-field interaction given as a parameter V in this paper. In particular, such an expansion is necessary in order to introduce the quantum nature of optical near fields. This study will open up a new way to control of environment behavior and stimulate the development of the application of optical-near-field ET to nano-photoelectronic devices.

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