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Quantum Generation Dynamics of Coherent Phonons: Analysis of Transient Fano Resonance

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Abstract. We study the transient Fano resonance of a semiconductor Si observed in the early time region of coherent phonon generation induced by an ultrafast pump laser. We particularly examine effects of the detuning on the transient Fano resonance, where the detuning is defined by the difference between the central frequency of the pump laser and the band gap. It is clarified that asymmetric profiles of transient induced photoemission spectra, implying the Fano resonance, strongly depend on the detuning. This is attributed to energetically adjacent bosonic states, whose energy levels are strongly influenced by the detuning.

1. Introduction

Coherent phonon (CP) generation in semiconductors is one of the representative phenomena induced by irradiation of an ultrafast pulse laser [1]. The mechanism of the CP generation has been discussed with the phenomenological models such as the impulsive stimulated Raman scattering model [2] and the displacive excitation of CP model [3], where the initial phase of the CP oscillation is considered to be a key parameter. Although several theoretical works focusing on the initial phase have been carried out [4, 5, 6], the CP generation mechanism remains a point of controversy. Furthermore, recent time-resolved spectroscopy techniques unveil the early time dynamics of the CP, where a vestige of the Fano resonance (FR) [7] is transiently observed in a lightly n-doped Si [8, 9]. This FR is manifested only in about 100fs after the laser irradiation, and is different from the conventional FR in equilibrium, for example, observed in heavily-doped Si [10, 11]. In order to clarify the CP generation dynamics accompanying the transient FR, a microscopic theory based on a quantum mechanical model is required.

The present authors have presented a comprehensive quantum theory of the transient FR and the subsequent CP dynamics in semiconductors based on a polaronic-quasiparticle (PQ) picture, where the transient induced photoemission spectra have asymmetric profiles in non-polar crystals while symmetric in polar crystals [12]. The difference is mainly ascribed to a time-dependent phase factor of electron-phonon interactions; the phase factor is real in an optical-phonon deformation potential interaction, whereas pure-imaginary in a Frölich interaction. In

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the present paper, we examine effects of a detuning on the transient induced photoemission spectra, where the detuning is defined by the difference between the central frequency of the pump laser and the band gap of the semiconductor. We have found that the asymmetric profiles of the spectra strongly depend on the detuning, which is attributed to energetically adjacent bosonic states.

2. Theory

In this chapter, we briefly summarize our theory of Ref. [12]. The total Hamiltonian of the system is represented by $\hat{H} = \hat{H}_e + \hat{H}'(t) + \hat{H}_p + \hat{H}_{e-p}$, where $\hat{H}_e = \sum_{b\boldsymbol{k}} \varepsilon_{b\boldsymbol{k}} a^\dagger_{b\boldsymbol{k}} a_{b\boldsymbol{k}} + \frac{1}{2}\sum_{\boldsymbol{q}\neq 0} V^{(C)}_{\boldsymbol{q}} \sum_{bb'\boldsymbol{k}\boldsymbol{k}'} a^\dagger_{b\boldsymbol{k}+\boldsymbol{q}} a^\dagger_{b'\boldsymbol{k}'-\boldsymbol{q}} a_{b'\boldsymbol{k}'} a_{b\boldsymbol{k}}$ is the two-band Hamiltonian of electrons with the Coulomb term, and $\hat{H}'(t) = -\sum_{\boldsymbol{k}} [\Omega_{cv}(t)\, a^\dagger_{c\boldsymbol{k}} a_{v\boldsymbol{k}} + \Omega_{vc}(t)\, a^\dagger_{v\boldsymbol{k}} a_{c\boldsymbol{k}}]$ is the electron-light interaction. Here $a^\dagger_{b\boldsymbol{k}}$ ($a_{b\boldsymbol{k}}$) represents a creation (destruction) operator of an electron with the Bloch momentum $\boldsymbol{k}$ in the b (=c for conduction or v for valence)-band. The electric dipole moment between b and b'-bands is given by $d_{bb'}$ and $\Omega_{bb'}(t) = d_{bb'}F(t)$, where $F(t)$ is the electric field of a pump laser. The Hamiltonian of the LO-phonon is given by $\hat{H}_p = \sum_{\boldsymbol{q}} \omega^{(LO)}_{\boldsymbol{q}}\, c^\dagger_{\boldsymbol{q}} c_{\boldsymbol{q}}$ and the electron-phonon interaction is represented by $\hat{H}_{e-p} = \sum_{b,\boldsymbol{q},\boldsymbol{k}} (g_{b\boldsymbol{q}} c_{\boldsymbol{q}} a^\dagger_{b\boldsymbol{k}+\boldsymbol{q}} a_{b\boldsymbol{k}} + g^*_{b\boldsymbol{q}} c^\dagger_{\boldsymbol{q}} a^\dagger_{b\boldsymbol{k}} a_{b\boldsymbol{k}+\boldsymbol{q}})$, where $c^\dagger_{\boldsymbol{q}}(c_{\boldsymbol{q}})$ is a creation (destruction) operator of an LO-phonon. $g_{b\boldsymbol{q}}$ is a coupling constant of the b-band electron with the LO-phonon; the coupling corresponds to the deformation potential interaction in non-polar crystals and to the Frölich interaction in polar crystals, respectively. Here, it should be noted that we use the atomic units in this paper unless otherwise stated.

In our theory, we consider the equation of motion for a composite operator $A^\dagger_{\boldsymbol{q}}(\boldsymbol{k}bb') \equiv a^\dagger_{b\boldsymbol{k}+\boldsymbol{q}} a_{b'\boldsymbol{k}}$. We need to treat the commutator expressed by

$$[\hat{H}_e + \hat{H}'(t), \bar{A}^\dagger_{\boldsymbol{q}}(\boldsymbol{k}bb')] \approx \sum_{\tilde{\boldsymbol{k}}\tilde{b}\tilde{b}'} \bar{A}^\dagger_{\boldsymbol{q}}(\tilde{\boldsymbol{k}}\tilde{b}\tilde{b}') \bar{Z}_{\boldsymbol{q}}(\tilde{\boldsymbol{k}}\tilde{b}\tilde{b}', \boldsymbol{k}bb'), \tag{1}$$

where $\bar{A}^\dagger_{\boldsymbol{q}}$ represents that the rotational wave approximation is employed [12]. We diagonalize the non-hermitian matrix $\bar{Z}_{\boldsymbol{q}}$ as $U^{L\dagger}_{\boldsymbol{q}} \bar{Z}_{\boldsymbol{q}} = \mathcal{E}_{\boldsymbol{q}} U^{L\dagger}_{\boldsymbol{q}}$ and $\bar{Z}_{\boldsymbol{q}} U^R_{\boldsymbol{q}} = U^R_{\boldsymbol{q}} \mathcal{E}_{\boldsymbol{q}}$. Here $\mathcal{E}_{\boldsymbol{q}}$ is the eigenvalue diagonal-matrix and $\{U^L_{\boldsymbol{q}}, U^R_{\boldsymbol{q}}\}$ are the associated biorthogonal set of eigenvectors. Then we introduce an α-th adiabatic quasi-boson operator with time fixed as an adiabatic parameter defined as

$$B^\dagger_{\boldsymbol{q}\alpha} = \sum_{\boldsymbol{k}bb'} \bar{A}^\dagger_{\boldsymbol{q}}(\boldsymbol{k}bb') U^R_{\boldsymbol{q}\alpha}(\boldsymbol{k}bb'), \tag{2}$$

and finally obtain the adiabatic coupled equations represented by

$$-i\frac{dB^\dagger_{\boldsymbol{q}\alpha}}{dt} = B^\dagger_{\boldsymbol{q}\alpha}\mathcal{E}_{\boldsymbol{q}\alpha} + c^\dagger_{\boldsymbol{q}} M'^*_{\boldsymbol{q}\alpha} + i\sum_{\alpha'} B^\dagger_{\boldsymbol{q}\alpha'} W_{\boldsymbol{q}\alpha'\alpha} + M''_{-\boldsymbol{q}\alpha} c_{-\boldsymbol{q}} \tag{3}$$

and

$$-i\frac{dc^\dagger_{\boldsymbol{q}}}{dt} = \sum_{\alpha} B^\dagger_{\boldsymbol{q}\alpha} M_{\boldsymbol{q}\alpha} + c^\dagger_{\boldsymbol{q}} \omega^{(LO)}_{\boldsymbol{q}}, \tag{4}$$

where $W_{\boldsymbol{q}\alpha'\alpha}$ is a non-adiabatic coupling between the α-th quasi-boson and the α'-th one [12]. $M_{\boldsymbol{q}\alpha}$, $M''_{-\boldsymbol{q}\alpha}$ and $M'^*_{\boldsymbol{q}\alpha}$ are effective couplings between the α-th quasi-boson and the LO-phonon. Almost all of the quasi-boson states are electron-hole pair excitations, resulting in the quasi-boson continuum. Embedding the LO-phonon level $\omega^{(LO)}_{\boldsymbol{q}}$ in the continuum $\mathcal{E}_{\boldsymbol{q}\alpha}$ is likely to induce the FR [12].

The coupled equations of motion (3) and (4) are considered as a scattering problem [7], and a new PQ operator $F_{\boldsymbol{q}}^{\dagger} = [B_{\boldsymbol{q}}^{\dagger}, c_{\boldsymbol{q}}^{\dagger}]V_{\boldsymbol{q}}^{R}$ is introduced, where $V_{\boldsymbol{q}}^{R}$ is obtained by solving a matrix equation [13]. Then, the retarded Green's function of $F_{\boldsymbol{q}}^{\dagger}$ and relevant physical quantities including the dielectric function and transient induced photoemission spectrum $\bar{A}_{\boldsymbol{q}}(t_p;\omega)$ are also calculated in our theoretical framework [12].

3. Results and Discussion

In this work, we have carried out calculations for Si, and before showing obtained results we introduce a key parameter, the detuning

$$\Delta\omega = \omega_0 - E_g, \tag{5}$$

where ω_0 is the central frequency of the pump laser and E_g is the band gap of Si. In addition, it should be noted that we employ a squared wave for the pump laser and the pulse width is set to 15fs, which ranges from $t = -7.5$ fs to 7.5 fs [12]. Then Ω_{cv} is set to 29 meV.

Figure 1 shows calculated results of $\bar{A}_{\boldsymbol{q}}(t_p;\omega)$ at $t_p = 7.5$ fs for $\Delta\omega = -27$ meV and $\Delta\omega = 82$ meV, respectively. Although asymmetric spectral profiles characterizing the FR appear for both cases, positions of the spectral peaks and the dips are clearly different; for $\Delta\omega = -27$ eV the peak is in the higher energy side of the dip while for $\Delta\omega = 82$ meV the peak is in the lower energy side of the dip. This difference is found to result from the behavior of the effective coupling $M_{\boldsymbol{q}\alpha}$. Figure 2(a) shows $M_{\boldsymbol{q}\alpha}$ for $\Delta\omega = 82$ meV as a function of time. We can see that changes its sign from positive to negative at $t = 26.8$ fs and at $t = 71.3$ fs, and negative to positive at $t = 70.2$ fs, which implies that the phase factor $M_{\boldsymbol{q}\alpha}$ abruptly changes between 0 and π. At $t_p = 7.5$ fs, $M_{\boldsymbol{q}\alpha}$ for $\Delta\omega = 82$ meV is positive and thus the phase factor is 0. By contrast, the phase factor for $\Delta\omega = -27$ meV is equal to π, because $M_{\boldsymbol{q}\alpha}$ for $\Delta\omega = -27$ meV is negative for any t. As is already discussed in Ref. [12], the spectral profiles strongly depend on the phase factor of $M_{\boldsymbol{q}\alpha}$. Thus the spectral profile for $\Delta\omega = 82$ meV is different from that for $\Delta\omega = -27$ meV.

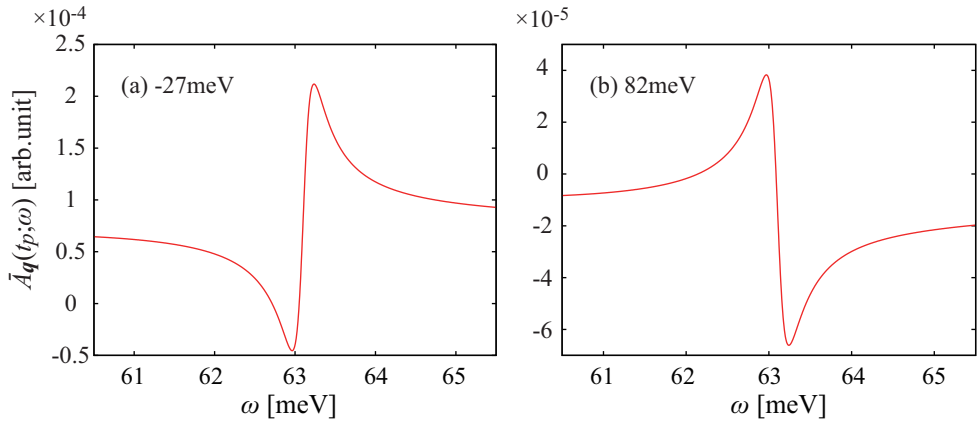


Figure 1. (a) Transient induced photoemission spectra $\bar{A}_{\boldsymbol{q}}(t_p;\omega)$ at $t_p = 7.5$ fs for $\Delta\omega = -27$ meV and (b) that for $\Delta\omega = 82$ meV, respectively.

Now, we focus on the the singular behavior of $M_{\boldsymbol{q}\alpha}$ for $\Delta\omega = 82$ meV; it seems to diverge at $t = 24.4$ fs and at 71.0 fs. In particular, the latter diverging behavior results in the change of the sign of $M_{\boldsymbol{q}\alpha}$ at $t = 70.2$ fs and at 71.3 fs. We have found that these singularities are attributed to the coupling between energetically adjacent adiabatic quasi-boson states. Figure 2 (b) shows

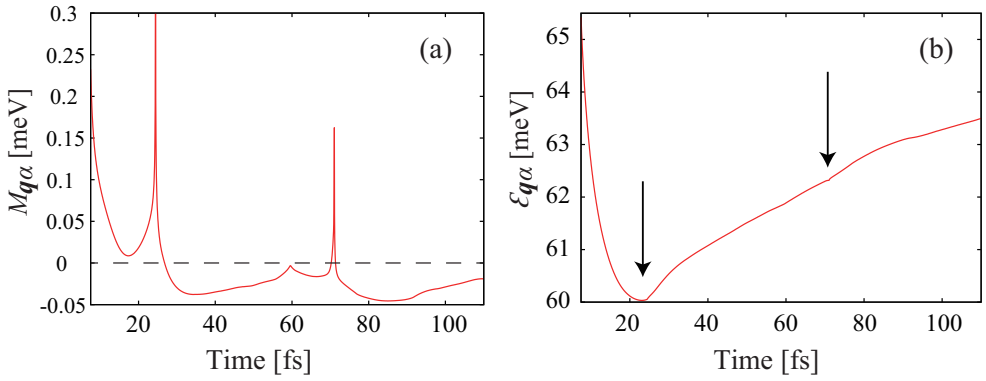


Figure 2. (a) The coupling constant $M_{q\alpha}$ and (b) associated adiabatic energy $\mathcal{E}_{q\alpha}$ for the detuning parameter $\Delta\omega = 82$ meV as a function of time.

the associated adiabatic energy $\mathcal{E}_{q\alpha}$ for $\Delta\omega = 82$ meV as a function of time. The abrupt energy changes occur at $t = 24.4$ fs and at 71.0 fs represented by arrows. At these points, $\mathcal{E}_{q\alpha}$ and the energy of the adjacent state labeled α' show the level crossing, resulting in the singularity of $M_{q\alpha}$. Here it should be noted that the level crossing shown here is considered to be closely related to the exceptional point (EP) [14, 15, 16], which is widely observed in non-hermitian problems in physics.

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