Ultrafast control of electron-phonon entangled systems in bulk solids

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Abstract: A simple theory is presented for the ultrafast coherent control of electron-phonon entangled systems in condensed matter which agrees well with the transient reflectivity measurement by the sub-femtosecond phase-locked dual pulse technique in bulk GaAs.
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1. Introduction

Irradiating solids by short optical pulses, both the electronic coherence and the phonon coherence may be created. Although a number of experimental works have been carried out to observe the coherent dynamics in electronic and phonon degrees of freedom, most of these works were done separately with different techniques such as the four-wave mixing and the transient reflectivity measurements. One of the reasons is the great difference in the time-scale of oscillation of signals which is a signature of quantum coherence. Recently, we succeeded in simultaneous observation of the quantum coherence of electronic states and phonon states in GaAs crystal at room temperature by the transient reflectivity measurement[1]. The crystal was excited by a pair of phase-locked short pulses (~20 fs: half width at half maximum) with delay time t_{12} controlled in sub-femtosecond accuracy. The induced reflectivity change ΔR was measured with delay t_{13} from the first pulse. As shown in Fig.1 (a), the amplitude $[\Delta R]$ of the oscillating component originating from the LO phonon clearly showed a fringe pattern as a function of t_{12} with frequency of the optical frequency of the pump-pulse $\Omega/2\pi = 370$THz, which is superimposed on the fringe pattern with frequency of the LO phonon $\omega/2\pi = 8.7$THz. This means that the electronic degree of freedom is entangled with that of phonon and is coherently controlled at least for a short delay (~80 fs). Here we propose a simple theory for describing the coherent control of the electron-phonon entangled system in solids, namely in dissipative environment. It is shown that the rapid phase relaxation for the electronic part results in the degradation of the visibility of the high frequency component in $[\Delta R]$ indicating the collapse of the entanglement.

2. Model and formulation

Consider a two-state model descried by the Hamiltonian

$$H = \varepsilon_g |g\rangle\langle g| + \varepsilon_e |e\rangle\langle e| + \hbar\omega b^+ b + \alpha\hbar\omega\left(b + b^+\right)|e\rangle\langle e|, \quad (1)$$

where $|g\rangle$ and $|e\rangle$ are the ground state and the optically excited state with energy ε_g and ε_e, respectively, b is the annihilation operator for the LO phonon, and α is the coupling constant. Since α is small, we can safely confine ourselves to the reduced three-level system, namely, the zero-phonon state in $|g\rangle$, the zero-phonon and the one-phonon state in $|e\rangle$,

$$H = \varepsilon_g |g,0\rangle\langle g,0| + \varepsilon_e |e,0\rangle\langle e,0| + \left(\varepsilon_e + \hbar\omega\right)|e,1\rangle\langle e,1|, \quad (2)$$

in which the phonon numbers are indicated in each state vectors. Within the rotating wave approximation, the interaction with the phase-locked dual pump-pulses is given by

$$H_I(t) = \mu\left\{E(t)e^{i\Omega t} + E(t-t_{12})e^{i\Omega(t-t_{12})}\right\}\left\{|e,0\rangle\langle g,0| + \alpha|e,1\rangle\langle g,0|\right\} + \text{H.c.}, \quad (3)$$

where μ is the transition dipole moment which is assumed to be a scalar quantity, $E(t)$ is the envelope of the optical pulse centered at $t = 0$. In order to take into account the phase relaxation phenomenologically, we adopt the stochastic model of Anderson[2], in which the excited state energy ε_e contains a randomly fluctuating part $f(t)$ which obeys the Markoffian-Gaussian stochastic process defined by the two-time correlation function $\langle f(t)f(t')\rangle = \hbar^2 D^2 \exp\left(-\gamma|t-t'|\right)$, where γ is the effective frequency of the random field and D is its intensity. The

phonon relaxation is neglected here. The Schrödinger equation for the state vector $|\psi(t)\rangle$ is solved to the first order in μ in the interaction representation with resonant condition $\hbar\Omega = \varepsilon_e - \varepsilon_g$ as

$$|\psi(t)\rangle = |g,0\rangle - \frac{i\mu}{\hbar}\int_{-\infty}^{t} d\tau\, e^{iF(\tau)}\left\{E(\tau)+E(\tau-t_{12})e^{i\Omega t_{12}}\right\}\left\{|e,0\rangle+\alpha\, e^{i\omega\tau}|e,1\rangle\right\}, \quad (4)$$

with $F(\tau)\equiv\hbar^{-1}\int_0^{\tau} f(\sigma)d\sigma$. The above form indicates that $|\psi(t)\rangle$ is an entangled state between the electronic states and the phonon states. The density matrix $\rho(t)$ is given by $\rho(t)=|\psi(t)\rangle\langle\psi(t)|$. The oscillation amplitude of the reflectivity for the probe pulse is proportional to the cross-term of the zero- and one-phonon states in the excited state subspace. By taking the average over stochastic process, we find

$$\left\langle\{\rho(t)\}_{e,0;e,1}\right\rangle = \frac{\alpha\mu^2}{\hbar^2}\int_{-\infty}^{t} d\tau \int_{-\infty}^{t} d\tau'\, \Phi(\tau-\tau')\left\{E(\tau)+E(\tau-t_{12})e^{i\Omega t_{12}}\right\}\left\{E(\tau')+E(\tau'-t_{12})e^{-i\Omega t_{12}}\right\}e^{-i\omega\tau'}. \quad (5)$$

Here, $\Phi(\tau-\tau')=\exp\left[\frac{D^2}{\gamma^2}\left\{1-\gamma|\tau-\tau'|-e^{-\gamma|\tau-\tau'|}\right\}\right]$ is the memory function[2]. The amplitude of the phonon signal for the probe pulse is given by

$$[\Delta R]\propto \lim_{t\to\infty}\mathrm{Re}\left\langle\{\rho(t)\}_{e,0;e,1}\right\rangle. \quad (6)$$

3. Nemerical results and discussion

If the pulse width is short enough, the integrals of Eq. (5) can be performed approximately and we find a much simpler expression for $[\Delta R]$

$$[\Delta R]\propto\left|1+\cos\omega t_{12}+\Phi(t_{12})\{\cos\Omega t_{12}+\cos(\Omega+\omega)t_{12}\}\right|.$$

In Fig. 1 (b) and (c), examples of the theoretical values for the delay-time dependence of the phonon amplitudes given by the above formula are plotted for $\gamma = 0.01\,\mathrm{fs}^{-1}$ with the phase decay constant D/γ as a parameter. The optical and the phonon frequencies are chosen from the experimental values for GaAs. The high frequency fringe with oscillation period 2.7fs in the short delay-time region originates from the electronic interference, while the long scale fringe with period of 114fs is due to the phonon coherence. The former is superimposed on the latter in the short delay-time region. This indicates the persistence of the entanglement which is created by the first pump-pulse. As the delay-time increases, the visibility of the electronic interference decays from 1 to 0 rapidly, while the visibility of the phonon interference survives rather long. Figure 1 clearly reveals the difference in the so-called coherent control in the electronic system and phonon systems.]

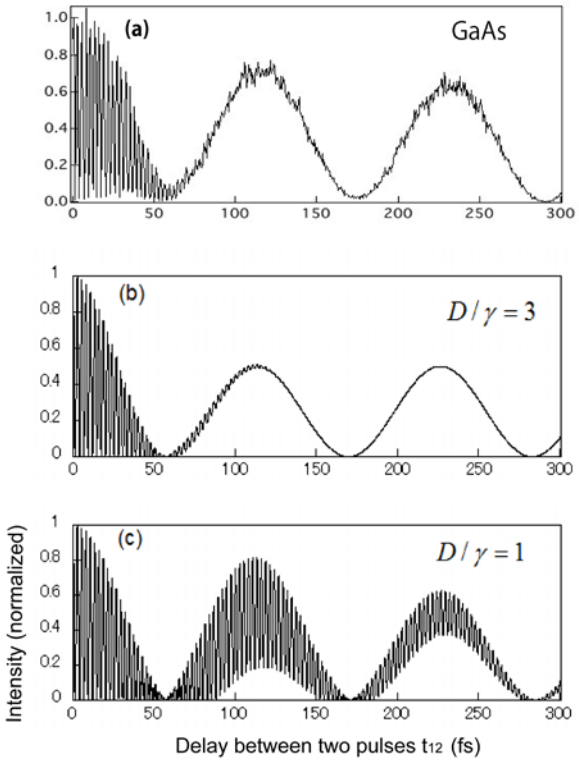

Figure 1. The interferogram for the LO phonon ; (a) experiment for GaAs at RT and (b), (c) theory.

The essential features of experimental data of the interferogram for GaAs at room temperature shown in Fig.1 (a) are well reproduced by the curve in Fig.1 (b) with $D/\gamma = 3$. This technique of pump-probe measurement by sub-femtosecond phase-locked dual pulses will open up the possibility to investigate the detailed mechanism of ultrafast phase relaxations in condensed matter under various conditions such as the temperature and the carrier density of the sample.

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