# Dual role of longitudinal optical phonons for generation of coherent oscillations in gallium arsenide under optical pumping

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We present a novel and simple picture of the generation dynamics of coherent longitudinal optical (LO) phonons and LO-phonon-plasmon-coupled (LOPC) modes by the ultrafast infrared pump-pulses in gallium arsenide (GaAs) employing the low-temperature approximation. LO phonons exhibit a pronounced coupling with plasmons formed by the optically excited electrons in the excited states of GaAs. This coupling results in the coherent oscillation of the LOPC modes in the excited states. The pump pulse also induces stimulated Raman scattering, which generates the coherent LO-phonon oscillation in the ground state. This picture is incorporated into a simplified model, and the time evolution of the density operator is calculated using the Lindblad-type quantum master equation. The theoretical results explain well the reported experimental results on the coherent oscillation of LO phonons and LOPC modes observed through transient reflection measurements. Above all, our model provides a natural reason for the simultaneous manifestation of the LO phonons and the LOPC modes.

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### I. INTRODUCTION

The interplay of quasiparticles in solids plays an essential role in a wide range of physical phenomena [1,2]. For instance, electron-phonon interactions hold an important position in condensed matter physics [3,4] and are also critical in discussions concerning ultrafast phenomena. In certain metallic and semiconducting materials, the plasmon appears as a quasiparticle associated with the collective motion of electrons [5,6]. In polar semiconductors such as GaAs, LOphonon-plasmon-coupled (LOPC) modes form [7-10], and have been observed by both Raman spectroscopy [11,12] and coherent phonon spectroscopy with transient reflectivity measurements [13–16]. The frequency of the plasmon is proportional to the square root of the conduction electron density. If the plasmon frequency is close to that of the LO phonon, new modes denoted  $L_+$  and  $L_-$  are generated via the LO-phonon-plasmon interaction, the eigenstates of which are obtained through the diagonalization of the interaction Hamiltonian [8,9].

Nevertheless, it is often reported that the bare LO phonon oscillation continues to manifest in transient reflection measurements along with the  $L_+$  and  $L_-$  modes [14–16] and in Raman spectroscopy [12]. This is strange because the LOPC modes are real eigenmodes in the excited states and the LO mode is simply its building block. The simultaneous coexistence of the LO phonon and LOPC modes has been discussed in connection with an inhomogeneous density distribution of electrons-for example, a surface depletion layer for a depth distribution of doped electrons [12,15] and an in-plane distribution of excitation densities produced by focusing the laser beam tightly on the sample [10,13]. In this model, the LOPC modes and the bare LO phonons are assumed to be spatially segregated depending on the electron density in the crystal.

Coherent optical phonons are generated by irradiating ultrashort optical pulses shorter than the phonon period [17, 18]. The coherent phonon oscillations are generated not only during the absorption process but also in the stimulated Raman process even under opaque conditions [19]. We have performed a study of the generation processes of coherent LO phonons in GaAs in pump-probe measurements employing the double pulse pumping technique using a pair of optical pulses with a relative phase controlled to within an attosecond accuracy [20,21]. This technique enabled us to clarify the ultrafast generation processes of the coherent phonon through quantum-path interference. An important result is that, with no contribution from the absorption process, the coherent LO phonons are generated through the Raman process.

In the present work, we propose a different and simple model to explain the spectral coexistence of the LO mode and the LOPC modes in coherent phonon generation within a GaAs crystal. The amplitude of the coherent phonons is calculated by solving the Lindblad-type quantum master equation while adopting a low-temperature approximation. The almost bare LO phonons are shown to be generated in the ground state through a stimulated Raman process, whereas the LOPC modes are generated in the excited states through the impulsive absorption process.

### **II. THEORETICAL MODEL**

We consider a composite system of electrons, optical phonons, and a plasmon system, and focus on coherent

phenomena. In this model, and within the low-temperature approximation, we assume that coherently photoexcited electrons interact with phonons and plasmons in which the doped electron density is negligibly small.

The wavelength of the optical pulse is much longer than the lattice constant, and hence the electron-hole pairs are uniformly distributed near the  $\Gamma$  point. Therefore, optical phonons need only be considered near the  $\Gamma$  point ( $q \approx 0$ ). We modify the standard Hamiltonian for an electron–phonon– plasmon system [8,9,22–24]. The Hamiltonian used in this paper is described as follows (details are given in Supplemental Material [25]);

$$H_{0} = \varepsilon_{g} |g\rangle \langle g| + \varepsilon_{e} |e\rangle \langle e| + \hbar \omega_{ph} b^{\dagger} b$$
  
+  $\alpha \hbar \omega_{ph} (b + b^{\dagger}) |e\rangle \langle e| + \sum_{\eta = g, e} \hbar \omega_{pl,\eta} c_{\eta}^{\dagger} c_{\eta} |\eta\rangle \langle \eta|$   
+  $\gamma \sum_{\eta = g, e} \hbar \sqrt{\omega_{ph} \omega_{pl,\eta}} (b + b^{\dagger}) (c_{\eta} + c_{\eta}^{\dagger}) |\eta\rangle \langle \eta|.$  (1)

Here,  $|g\rangle$  and  $|e\rangle$  are the electronic ground and excited states, and  $\epsilon_g$  and  $\epsilon_e$  are its energy, respectively. The ground state  $|g\rangle$  represents a state, in which the valence bands are fully occupied and a vacant conduction band, and the excited state  $|e\rangle$  represents a state, in which electrons are excited from the valence band to the conduction band. The parameters  $\omega_{\rm ph}$ ,  $\omega_{\rm pl}$ represent the LO phonon and plasmon frequencies, respectively. For n-doped semiconductors, the plasmon frequency in the conduction band depends on the donor induced and photoexcited electrons (n). The density of the thermally excited electrons from the dopant is approximately  $10^{17}$  cm<sup>-3</sup> for the Si-doping density of 10<sup>18</sup> cm<sup>-3</sup> at 90 K, and its plasmonphonon coupling maybe negligible. Moreover, in the valence band, the plasmon frequency is dependent on the donor induced electrons, which is negligible at low temperatures. We also set  $\omega_{\rm pl,e} = \sqrt{ne^2/m^*\epsilon_{\infty}\epsilon_0}$  and  $\omega_{\rm pl,g} = 0$ , where *n* is the electron density,  $m^*$  is the effective mass of an electron, eis the elementary charge, and  $\epsilon_0$  is the vacuum permittivity.  $\omega_{\rm pl,e}$  and  $\omega_{\rm pl,g}$  are plasmon frequencies at electronic excited and ground states, respectively. The parameter  $\alpha$  represents the dimensionless coupling constant of the electron-phonon system. In the bulk, the Huang-Rhys factor ( $\alpha^2$ ) is considered very small ( $\alpha^2 \ll 1$ ). The parameter  $\gamma$  denotes the coupling constant between phonon and plasmon and is defined by  $\gamma = \frac{1}{2}\sqrt{1 - \epsilon_{\infty}/\epsilon_s}$ , with high-frequency and static-dielectric constants:  $\epsilon_{\infty}$  and  $\epsilon_s$ . In the limit of large electron density *n*, the frequency of  $L_{-}$  is known to approach the frequency of the transversal optical phonon  $(\omega_{TO})$  [7–9]. The operators  $b^{\dagger}$  and b denote, respectively, the creation and annihilation operators of an optical phonon at the  $\Gamma$  points. Similarly, the operators  $c^{\dagger}$  and c denote the creation and annihilation operators of the plasmon.

Adopting the rotating-wave approximation and the dipole interaction, the light-electron interaction Hamiltonian  $H_I(t)$  is expressed as

$$H_I(t) = \mu E(t) e^{-i\Omega_0 t} |e\rangle \langle g| + \text{H.c.}, \qquad (2)$$

where  $\mu$  denotes the transition dipole moment,  $\Omega_0$  is the frequency of the optical pulse, and E(t) is the optical envelope of the pump pulse, represented by  $E(t) = E_0 f(t)$  for a single

pulse. The function f(t) represents a Gaussian envelope function with a width of  $\sigma$ , and  $E_0$  the amplitude of the electric field. The amplitude of the electric field is estimated as

$$E_0 = \sqrt{\frac{\sigma}{c\epsilon_0}} F \sqrt{\frac{\pi}{2}},\tag{3}$$

where *F* denotes the fluence  $[J/cm^2]$  of the pump pulse, *c* is the speed of light, and  $\epsilon_0$  is the dielectric constant of vacuum. The photo-induced electron density is calculated from  $n_e = \beta F$  given the absorption coefficient  $\beta$  [cm<sup>-1</sup>]. For simplicity, the reflectivity of GaAs is not considered in this calculation. The transition dipole moment  $\mu$  is estimated using  $\mu = q \times r$ , where *q* is the charge and *r* is the distance between Ga and As atoms. For the calculation, we use an effective value of  $\mu = q \times r/2$ .

We consider the system of interest (the electron-phononplasmon composite system) to interact with the environment including the incoherent phonons, electrons, and impurities, and treat it as an open quantum system [26]. The time evolution of the quantum state is derived from the Lindblad-type master equation [27]:

$$i\hbar \frac{d}{dt}\rho(t) = [H,\rho(t)] + i\hbar D(\rho(t)), \qquad (4)$$

$$D(\rho(t)) = \sum_{i} \Gamma_i \bigg[ L_i \rho(t) L_i^{\dagger} - \frac{1}{2} \{ L_i^{\dagger} L_i, \rho(t) \} \bigg], \qquad (5)$$

where *H* denotes the total Hamiltonian  $(H = H_0 + H_I(t))$ , and  $\rho(t)$  the density operator  $\rho(t) = |\psi(t)\rangle \langle \psi(t)|$ . Moreover,  $D(\rho(t))$  and  $L_i$  denote the dissipator and the Lindblad operator, and  $\Gamma_i$  denotes the the decay rate. We define the phase relaxation of the electron, phonon, and plasmon states as the Lindblad operator  $L_i$  in Eq. (4); specifically,  $L_{el} = |e\rangle \langle e|$ ,  $L_{ph} = b^{\dagger}b$ , and  $L_{pl} = c^{\dagger}c$ .

The wave function  $|\psi(t)\rangle$  of the composite system is expanded using the basis sets of the electron, phonon, and plasmon subsystems,

$$|\psi(t)\rangle = \sum_{n,m} \left( c_{n,m}(t) \left| g, n, m \right\rangle + d_{n,m}(t) \left| e, n, m \right\rangle \right), \quad (6)$$

where  $|n\rangle$ , and  $|m\rangle$  denote the phonon and plasmon Fock states, respectively;  $c_{n,m}(t)$  and  $d_{n,m}(t)$  the time-dependent probability amplitudes of the *n* phonon and *m* plasmons correlated with the electronic ground and excited states, respectively. The initial state is set to an electronic ground state coupled with the states of zero phonons and zero plasmons,  $|\psi(0)\rangle = |g, 0, 0\rangle$ , which is a separable state.

We calculate the expectation value of the phonon nuclear coordinates using the time evolution of the density operator obtained from the master equation  $(\langle Q(t) \rangle = \text{Tr}[Q\rho(t)]]$ , where  $Q = \sqrt{\hbar/2M\omega_{\text{LO}}}(b^{\dagger} + b)$  with *M* the reduced mass of atoms per unit cell of GaAs). The coherent phonon behavior is governed by the coherence term of the density operator. In our model, we can evaluate the expectation value of the phonon coordinates by selecting the associated electronic state ( $|g\rangle$  or  $|e\rangle$ ). Therefore, we evaluate the expectation value under three conditions;  $\langle Q(t) \rangle = \langle Q_g(t) \rangle + \langle Q_e(t) \rangle$ ,  $\langle Q_g(t) \rangle = \sum_{i,j,m} \langle g, i, m | Q | g, j, m \rangle$ , and  $\langle Q_e(t) \rangle = \sum_{i,j,m} \langle e, i, m | Q | e, j, m \rangle$ .  $Q_e(t)$  and  $Q_g(t)$  are displacement in

the electronic excited  $|e\rangle$  and ground  $|g\rangle$  states, respectively. Note that  $|e\rangle$  and  $|g\rangle$  do not directly correspond to conduction and valence bands.

In the actual calculation, we used the Lindblad-type master equation implemented in the QuTiP solver of the Python package [28]. We used two electronic states and three Fock states for both the phonon and plasmon. The electron-phonon coupling constant  $\alpha$  is set to 0.01. The band-gap energy is set to  $\varepsilon_e - \varepsilon_g = 1.52$  eV which corresponds to the center of the optical frequency  $\Omega_0$ . The vibrational energy of the LO phonon is set to  $\hbar \omega_{\rm ph} = 36$  meV (approximately 8.7 THz). The high- and static-frequency dielectric constants are set to  $\epsilon_{\infty} = 11.1$  and  $\epsilon_s = 13.1$ , respectively [4]. The optical pulse parameter is set to  $\sigma = 30$  fs. The transition strength ( $\mu E_0$ ) is determined from settings of the laser pulse used in the previous work [29]. Furthermore, we set the decay rate of each phase-relaxation as  $\Gamma_{el} = 2/23$  (1/fs),  $\Gamma_{ph} = 2/3000$  (1/fs), and  $\Gamma_{\rm pl} = 2/150$  (1/fs). In this case, the coherent terms, calculated with Eqs. (3) and (4), decrease exponentially with  $\Gamma_i/2$ . The decay constant  $\Gamma_{\rm ph}$  is set based on the transient reflectance measurement of n-GaAs [15,30]. Additionally, the decay constant  $\Gamma_{el}$  is set based on the measured electronic decoherence time [21,31].

#### **III. RESULTS AND DISCUSSION**

Initially, we evaluated the time evolution of coherent phonon oscillation under single-pulse excitation  $(25.0 \,\mu\text{J/cm}^2)$ . Figure 1 illustrates the three conditions of the expectation value for the phonon nuclear coordinates  $(\langle Q_g(t) \rangle, \langle Q_e(t) \rangle, \text{ and } \langle Q(t) \rangle)$ , in which the non-vibrating component has not been removed.

In Fig. 1(a),  $\langle Q_g(t) \rangle$  shows a single damped oscillation at a frequency of 8.7 THz, which is attributed to LO phonons. In Fig. 1(b), the coherent oscillations at the electronic excited state ( $\langle Q_e(t) \rangle$ ) also show a single damped oscillation with a frequency of 7.5 THz, which is attributed to the lower branch of the LO-phonon-plasmon coupling ( $L_{-}$ ). The damping behavior is faster than that of the electronic ground state. The relative phase of these oscillations for the LO and LOPC modes is  $\pi/2$ ; it is clearly characteristic of Raman and absorption processes [19]. The different coherent oscillations indicate, depending on the electronic state, that the electronic and phonon states are entangled. From Fig. 1(c), the total coherent oscillation  $\langle Q(t) \rangle$  exhibits beating, consisting of two oscillations with frequencies 7.5 THz (LOPC) and 8.7 THz (LO phonons).

We performed Fourier transformation of Figs. 1(a)–1(c), with pump-probe delay ranging from 0.1 to 4.0 ps, respectively [Fig. 1(d)]. The Fourier spectra indicate that a LO-phonon oscillation is found only when we select the electric ground state. The  $L_{-}$  mode oscillation is found when the LO phonons are coupled to the electronic excited state. LO phonons play a dual role as bare phonons in the ground state and as plasmon-coupled mode in the excited state. The peak of the higher frequency  $L_{+}$  mode (approximately 11 THz) is too small to be seen in Fig. 1(d). We found that the dip in the total phonon spectrum at 8.5 THz is due to the phase difference between LO phonon and LOPC ( $L_{-}$ ) mode [19]. This dip has also been reported by experiment [15].



FIG. 1. Theoretical calculation of displacement  $\langle Q(t) \rangle$  of the LO phonons: (a) for the electronic ground state, (b) for the electronic excited state, and (c) for the sum of electronic ground and excited states as a function of the pump-probe delay. (d) Fourier spectra of each phonon expectation value obtained from the calculation; the black, red, and blue lines correspond, respectively, to the sum of the electronic ground and excited states, the electronic ground state, and the electronic excited state. The range of the Fourier transform is set with a delay of 0.1–4.0 ps. The nonvibrating component has not been removed. The peaks at 7.5 and 8.7 THz are due to the LO phonon-plasmon coupling oscillation and LO phonons, respectively.



FIG. 2. Experimental coherent phonon measurements (transient reflectance,  $\Delta R/R$ ) for n-GaAs (with a Si-doping density of  $1.0 \times 10^{18}$  cm<sup>-3</sup>). (a) Time evolution of transient reflectance ( $\Delta R/R$ ) taken at a temperature of 90 K. Coherent artifacts and nonvibrating background signals were removed. (b) Fourier spectra of transient reflectance ( $\Delta R/R$ ) in pump-probe delay times ranging from 0.15 to 2.50 ps.

Figure 2(a) presents the experimental transient reflectance measurements of n-GaAs (with a Si-doping density of  $1.0 \times 10^{18}$  cm<sup>-3</sup>) taken at 90 K using a Ti: sapphire laser with a center frequency of 800 nm. The coherent artifacts and nonvibrating background signals were removed. Additionally, the time origin of the pump-probe delay (horizontal axis) was set to 0 fs at the beginning of the oscillation after the coherent artifact. The oscillation behaviors in Figs. 2(a) and 1(c) are very similar. The Fourier spectrum obtained from the experiment [Fig. 2(b)] shows  $L_{-}$  and LO-phonon signals at approximately 7.6 and 8.7 THz, respectively, but no  $L_{+}$  signal was observed between 11 and 12 THz. The  $L_{+}$  signal is not observed in the weak excitation intensity region of the pump pulse.

We also evaluated from theory the excitation intensity dependence on the generation efficiency for the LO phonon and LOPC modes [Fig. 3(a)] up to a fluence of  $80 \,\mu J/cm^2$ . The oscillation intensity of the LO phonon shows nonlinear behavior as laser fluence increases [Fig. 3(b)] In contrast, the LOPC-mode oscillation intensity changes almost linearly up to about  $80 \,\mu J/cm^2$ . These features have also been observed in experiments [15]. The differences in phonon generation behavior can be explained with results from previous work [29] as being due to differences in quantum paths, namely, coherent phonon generation via impulsive stimulated Raman and impulsive absorption processes. The results show that the



FIG. 3. Comparison of the laser fluence of the pump pulse with the theoretical Fourier spectrum. We calculated the fluence between 10 and  $80 \,\mu\text{J/cm}^2$  in steps of  $5 \,\mu\text{J/cm}^2$ . (a) Fourier spectrum as a function of frequency (10 and  $80 \,\mu\text{J/cm}^2$  in steps of  $10 \,\mu\text{J/cm}^2$ ); (b) intensities of LO phonons (black circles) and L<sub>-</sub> (white circles) as a function of pump fluence.

coherent LO phonons behave as a bare LO phonon oscillation in the ground state and the LOPC coupled oscillation in the excited state.

Here we discuss briefly the relation between the present model and conventional Raman spectroscopy, although this model is focused on the transient reflectivity measurement with the pump-probe technique and does not include the emission process. In the Raman scattering of GaAs in an opaque region under visible light excitation, the ioninc Raman scattering [32,33] contribution is negligible and occurs through electron-phonon interactions via real electronic excited states. Since the electron-phonon coupling constant  $\alpha$  is very weak in GaAs crystals, there are two possible transition paths in the Raman scattering to the lowest order of the coupling constant. In the first path, the excited electron emits one LOPC mode in the excited state before transition to the ground state subspace. This gives rise to the scattered photon energy  $\hbar(\Omega - \omega_{\text{LOPC}})$ . In the second path, the electron first undergoes the transition to the ground state subspace, and subsequently emits one phonon in the ground state subspace. This corresponds to the energy of the scattered photon  $\hbar(\Omega - \omega_{\rm LO})$ . Here,  $\hbar\Omega$ ,  $\hbar\omega_{\rm LOPC}$ , and  $\hbar\omega_{\rm LO}$  are the energies of the excitation photon, the LOPC mode, and the LO phonon, respectively. Therefore, the present

model using electron-phonon interactions via electronic excited states would be consistent with Raman scattering.

To summarize, we proposed that the coexistence of LO and LOPC modes observed in the optical pumping experiments of GaAs crystal is attributed to the difference in the quantum paths—the LO mode by the impulsive stimulated Raman path and the LOPC mode by the impulsive absorption path. We calculated the probabilities associated with the LO phonons and LOPC modes in n-type GaAs by solving the Lindblad-type master equation for the simple model Hamiltonian describing the electron–phonon–plasmon system. We applied the low-temperature approximation in which the density of the thermally excited electrons from dopants is negligibly small and the phonon-plasmon coupling in the electronic ground state is negligible. The interpretation is consistent with the experimental data obtained. Our model provides a natural

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