Quantum interference between electron-phonon coupled states in bulk gallium arsenide

K. G. Nakamura[1,2], S. Hayashi[1,2], K. Kato[1,2], K. Norimatsu[1,2], M. Hada[1,3], and Y. Kayanuma[1,2]
[1]*Materials and Structures Laboratory, Tokyo Institute of Technology, Yokohama 226-8503, Japan*
[2]*CREST, Japan Science and Technology Agency, Kawaguchi 332-0012, Japan*
[3]*PRESTO, Japan Science and Technology Agency, Kawaguchi 332-0012, Japan*

Author e-mail address: nakamura@msl.titech.ac.jp

Abstract: We observed quantum interference between electron-phonon coupled states in a bulk GaAs by using two phase-locked femtosecond pulses and found that the electronic coherence remains within ~ 45 fs even at room temperature.
OCIS codes: (320.7120) Ultrafast phenomena; (160.6000) Semiconductor materials.

1. Introduction

Coherence is fundamental nature in quantum physics and a key issue in future device and information technologies. While both electronic and phonon coherence can be induced in solid material by irradiation of an optical pulse, they have been usually measured using different detection techniques such as four-wave mixing and transient reflectivity. The four-wave mixing experiments on GaAs show that the relaxation time for the electronic coherence in inter-band transition is 140 fs at 77 K [1]. On the other hand, coherent phonons have been observed by the transient reflectivity with the relaxation time of ~ 2 ps [2]. In this paper we demonstrate to measure both the electronic and phonon coherence simultaneously in the electron-phonon system of a semiconductor via interference of quantum states induced by two phase-locked femtosecond pulses.

2. Experimental configuration

Intensity interference of two electron-phonon coupled states induced by two phase-locked femtosecond pulses was measured via transient reflectivity using a pump and probe technique. A femtosecond pulse (pulse width of ~ 50 fs and center wavelength of 800 nm) from a Ti: sapphire oscillator was divided into two pulses using a beam splitter (9:1). The pulses with 90 % and 10 % energy are used as a pump and probe pulse, respectively. The pump pulse was introduced to a homemade Michelson-type interferometer to produce two phase-locked pulses (pulse 1 and pulse 2) [3]. Delay (t_{12}) between the two pulses was controlled by changing an optical-path length in one arm of the interferometer. A part of the phase-locked pulses wad detected by a spectrometer to detect a spectral interference and an optical intensity. Pump and probe pulses were focused on the sample using a lens, and the reflected pulse was collected using the same lens. Polarization of the pump and probe pulse was set orthogonal to each other to eliminate the coherent artifact. The probe pulse (pulse 3) was irradiated at delay (t_{23}), which was controlled with a scan delay unit, after irradiation of the pulse 2. The reflected pulse was detected using the electro-optic sampling technique [2]. All experiments have been done at room temperatures, and the sample used was a single crystal of n-type GaAs. Details of the experimental setup are described elsewhere [3,4].

3. Results and discussion

The intensity modulation in the reflected pulse, which is measured by varying t_{23} at fixed t_{12}, shows coherent oscillation due to longitudinal-optical (LO) phonons and LO phonon-plasmon coupling mode as found in Ref. [3]. Relaxation time of the LO phonons and the coupling mode are obtained to be 0.8 and 1.7 ps, respectively, by using analysis with two damped oscillations. Intensities of the LO phonons and the coupling mode was obtained from the corresponding peak intensity at 8.7 and 7.7 THz in the Fourier-transformed spectrum of the reflectivity, respectively.

Figure 1 (a) shows a change of the LO phonon intensity (phonon interferogram) obtained by varying t_{12} with a time step of ~ 500 attoseconds, which represents interference between the electron-phonon coupled states induced by two optical pulses. There are two distinct fringe patterns; a narrow fringe with a separation time of ~ 2.7 fs corresponding to the optical cycle and a wide fringe with a separation time of ~ 120 fs corresponding to the LO

phonon period. The narrow fringe disappears within ~ 80 fs and only the wide fringe remains at longer delay. The fringe patterns mean that the electronic coherence induced by the optical excitation is lost within ~ 80 fs and the phonon coherence is conserved after dissipation of the electronic coherence. Although a modulation in the optical intensity due to overlay of two pulses (shown in Fig. 1(a)) also affects the LO phonon intensity at early delays (< 50 fs), the electronic coherence can be recognized from the interference visibility. The interference visibility of the LO phonons keeps more than 0.8 until 60 fs and decreases to 0.5 at 70 fs but that of the optical intensity decreases to 0.5 within 25 fs (Fig. 1 (b)). This indicates that the bulk material (GaAs) keeps the electronic coherence within ~ 45 fs.

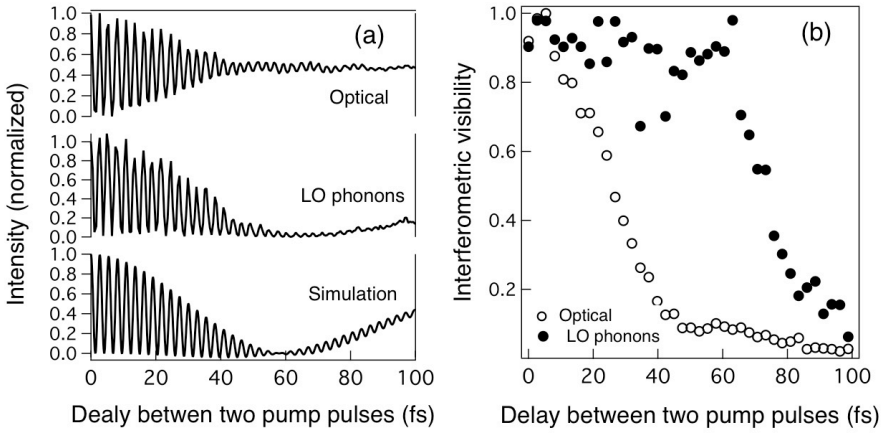

Figure 1 : (a) Intensity change of the LO phonons in GaAs (central panel) and optical intensity (upper panel) obtained by varying delay (t_{12}) of the two pump pulses. The bottom panel is simulation by eq. (1) in the manuscript with $\Omega = 2\pi\times370$ THz, $\omega = 2\pi\times8.7$ THz, and Gaussian damping with decay time of 80 fs . (b) Interferometric visibilities of the optical pulse interferogram and the LO phonon interferogram.

The characteristics in the interference pattern of the LO phonon intensity can be understood using a three-level model with the zero-phonon state in the electronic ground state $|g,0\rangle$ and the zero- and one-phonon states in the electronic excited state $|e,0\rangle$ and $|e,1\rangle$. A state induced by two phase-locked pulses can be obtained by solving time-dependent Schrödinger equation with rotating wave approximation. We calculate an expectation value of atomic displacement of phonons because the intensity of an oscillating part of the reflectivity [ΔR] is proportional to the atomic displacement. The electronic phase relaxation is taken into account phenomenologically in the density matrix formalism. [ΔR] is approximately obtained in the analytical form using a memory function $\Phi(t_{12})$ at resonance condition of the pump pulse to the energy gap between the electronic states as

$$[\Delta R] \propto 1+(\cos(\Omega t_{12})+\cos((\Omega+\omega)t_{12}))\Phi(t_{12})+\cos(\omega t_{12}), \quad (1)$$

where Ω is the central frequency of the pump pulse and ω is the phonon frequency. The second and third term in the right side of eq. (1) correspond to interference fringes due to electronic and phonon coherence, respectively. As shown in the bottom panel in Fig.1(a), the interference fringe pattern of the LO phonons is qualitatively reproduced using eq. (1) with a Gaussian decay form for the memory function $\Phi(t_{12})$.

[1] M. U. Wehner, M. H. Ulm, D. S. Chemla and M. C. Wegner "Coherent control of electron-LO phonons scattering in bulk GaAs," Phys. Rev. Lett. **80,** 1992-1995 (1998).

[2] G. C. Cho, W. Kutt and H. Kurz "Subpicosecond time-resolved coherent-phonon oscillation in GaAs," Phys. Rev. Lett. **65,** 764-766 (1990).

[3] J. Hu, O. V. Misochko, A. Goto and K. G. Nakamura "Delayed formation of coherent LO phonon-plasmon coupled modes in n- and p-type GaAs measured using a femtosecond coherent control technique," Phys. Rev. B **86,** 235145 (2012).

[4] H. Katsuki, J. C. Delagnes, K. Hosaka, K. Ishioka, H. Chiba, E. Z. Zijlstra, M. E. Garcia, H. Takahashi, K. Watanabe, M. Kitajima, Y. Matsumoto, K. G. Nakamura and K. Ohmori "All-optical control and visualization of ultrafast two-dimensional atomic motions in a single crystal of bismuth," Nature Commun. **4,** 2801 (2013).