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Depth-dependent Detection Mechanisms of Coherent Phonons in n-type GaAs

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Abstract. Transient reflectivity measurements at different probing wavelengths reveal detection mechanisms of coherent phonon and phonon-plasmon coupled modes of n-doped GaAs to be strongly depth-dependent due to the carrier depletion at the surface.

1 Introduction

Transient reflectivity measurements offer a general and sensitive approach to monitor coherent optical phonons. However, the mechanism of the reflectivity modulation by the coherent lattice oscillation remains hardly explored. Cho and coworkers measured the transient reflectivity response of GaAs for a few probe polarization angles with near infrared light [1,2], and attributed the detection of the LO phonon and LO phonon-plasmon coupled (LOPC) mode signals to the linear (Pockels) and non-linear (Franz-Keldysh) electro-optic effects. Their conclusions were inconsistent, however, with the weak electro-optic effect in GaAs [3,4] and with the depth-dependent phonon-plasmon coupling in doped GaAs [4,5]. For better understanding of the detection mechanism, a more systematic investigation of the probe polarization dependence at different critical points is required.

In the present study, we investigate the detection mechanisms of the coherent phonons of GaAs using 800 and 400 nm probe wavelengths. Strong light absorption at 400 nm near the E_1 gap enable highly surface sensitive reflectivity measurements of ultrafast LO phonon-plasmon dynamics. By analysing the probe-polarization-angle dependence of the coherent amplitudes, we demonstrate that different microscopic processes are operative in the detection of the LO and LOPC modes at different depths.

2 Experimental

The sample is (001) orientated n-type GaAs doped with $\sim 1\times10^{18}$ cm^{-3} Si. The maximum thickness of the depletion layer is 40 nm. For pump-probe reflectivity measurements we use 800 nm (1.55 eV) light to excite electron-hole pairs near the E_0 gap, and either 800 or 400 nm (3.1 eV) light to monitor the coherent phonon-plasmon dynamics in the bulk and surface regions. The probing depth is 110 (7.5) nm for 800 (400) nm light.

3 Results and Discussion

Figure 1a compares the oscillatory part of the reflectivity change ΔR/R of n-GaAs probed at 800 and 400 nm. When probed at 800 nm, the coherent oscillation is dominated by that of the lower (L-) branch of the LOPC mode at 7.6 THz, in which the LO phonon is coupled with the electron plasma in the n-doped bulk. The frequency of the L- mode, which is plotted as a function of the sum of doped and photoexcited carrier density in Fig. 1b, can be reproduced by the undamped LOPC frequency ω- [6]. By contrast, when the probe wavelength is 400 m, the coherent response is dominated by the bare LO phonon at 8.7 THz from the surface depletion region. In addition, we observe a shoulder peak, whose frequency shifts from TO to LO frequency limit with increasing photoexcited carrier density, as shown in Fig. 1b. The frequency cannot be reproduced by the undamped LOPC model, but is assigned as the LO phonons coupled with heavily damping hole plasma [7]. The LO-hole coupling dominates at the top surface of n-GaAs, because photoexcited holes are swept towards the surface by the built-in depletion field. Photoexcited electrons, by contrast, drift towards the bulk and give rise to the LO-electron coupled mode [4]. The amplitude of the LO-electron mode is much weaker than that of the LO-hole mode [Fig. 2b], confirming the high surface sensitivity of our reflectivity measurements with 400 nm probe light.

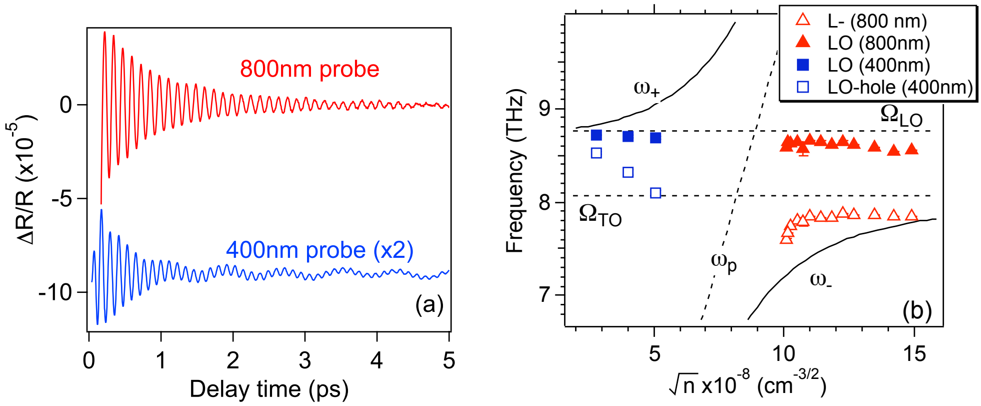

Fig. 1. (a) Oscillatory parts of ΔR/R response of n-GaAs probed with 800 and 400 nm light. (b) Freqiencies of the LO and LOPC modes as a function of carrier density for different probe wavelengths. We assume $n= n_{\mathrm{dope}}+n_{\mathrm{exc}}$ for 800 nm probe and $n=n_{\mathrm{exc}}$ for 400 nm probe, where n_{dope} and n_{exc} are chemically doped and photoexcited carrier densities.

When we rotate the polarization angle θ of the probe light, the coherent amplitudes of the bare LO, LO-electron (L- and L+) and LO-hole coupled modes exhibit interferences between the isotropic (θ-independent) component and the anisotropic component proportional to cos2θ, as shown in Fig. 2. We attribute the anisotropic and isotropic amplitudes to the reflectivity modulation via the dipole-allowed and dipole-forbidden Raman scattering processes. The cross sections of the latter processes are resonantly enhanced at the E_0 and E_1 critical points [3]. The relative contribution of the anisotropic and isotropic components depends on the mode as well as on the distance from the surface, because of the different Raman scattering mechanisms involved. We conclude that, while the Franz-Keldysh effect is efficient near the top surface, for the bulk LOPC mode it is overwhelmed by another dipole-forbidden process involving electron-phonon collective excitation [3,4].

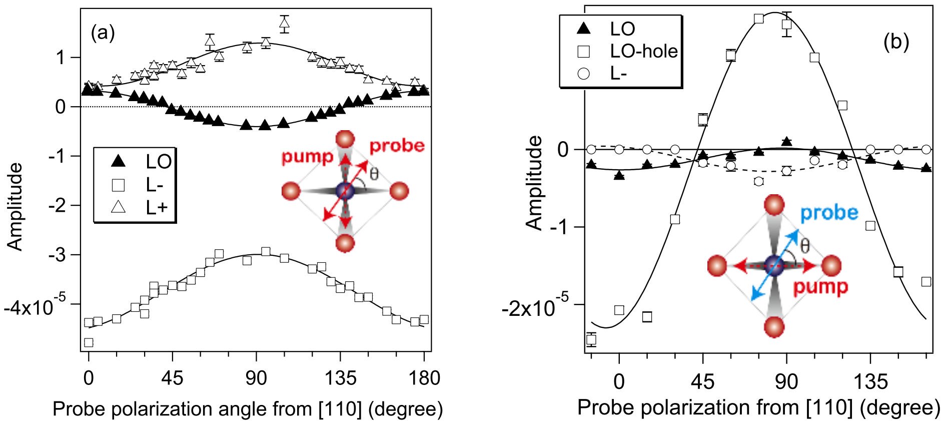

Fig. 2. Probe-polarization dependence of amplitude of coherent LO, LO-electron (L- and L+) and LO-hole modes of n- GaAs probed at 800 nm (a) and 400 nm (b). Insets show the polarization of pump and probe beams with respect to the crystal. Solid curves represent the fitting to A+B cos2θ.

4 Conclusion

We have investigated the depth-dependent detection mechanisms of the coherent LO and LOPC modes of n-GaAs in transient reflectivity measurements. We can describe the probe-polarization-angle dependence of the coherent amplitudes in terms of a coherent superposition of allowed and forbidden Raman scattering processes involved in the modulation of reflectivity by the lattice and electronic polarizations. With near ultraviolet light we can monitor exclusively the surface depletion layer. The observed coherent response demonstrates strong interaction between the lattice and spatiotemporal charge carrier dynamics under inhomogeneous carrier generation and sub-picosecond time scale non-diffusive transport.

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