The Dephasing of Coherent Phonons in GaAs (100)

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Abstract

The dephasing of coherent LO phonons due to excess photoexcited carriers in the near surface region of GaAs (100) crystal is investigated with time-resolved second-harmonic generation technique.

Summary

The development of ultrashort laser system makes it possible to generate and detect coherent phonon modes through impulsive Raman scattering, dispersive excitation or rapid field screening mechanisms [1-3]. Those experiments used pump-probe technique to create coherent phonon and monitor its free-induction decay by measuring the change in either the transmission or reflection. For example, coherent LO phonon in GaAs (100) has been intensively studied with reflective electro-optic sampling (REOS) technique [3]. It was found that bare LO phonon mode (8.8 THz) with 4 ps dephasing time could be launched for low photoexcited carrier density ($<1 \times 10^{18}$ /cm³). The main dephasing process is to be via one longitudinal optic phonon and one longitudinal acoustic phonon. However, the plasmon-LO phonon coupling mode (8.0 THz) with much shorter dephasing time 0.4 ps appears for higher photoexcited carrier density ($\sim 5 \times 10^{18}$ /cm³). This observation implies that the injected carrier density plays a significant role in the dephasing process of coherent phonons. Since carrier-phonon interaction is one of the most fundamental scattering processes in semiconductors, in this report, we proposed to pre-inject free carriers into the sample before the launch of coherent phonons, then monitor the pump-induced coherent phonon dephasing with time-resolved second-harmonic generation (TRSHG) technique.

TRSHG has been demonstrated to be surface sensitive and suitable to probe phonon and electronic spectroscopy of surfaces and interfaces [4]. The TRSHG experiments were carried out in air. The GaAs(100) sample was highly n-doped (3×10^{18} Si/cm³). The depletion field was calculated to be 740 KV/cm over the topmost 140 Å. In the experiments, three laser pulses, the pre-pump, pump and probe pulses, were derived from 25 fs, 80 MHz, 800 nm output of a Ti:Sapphire oscillator. The pre-pump was set collinear to the pump and 165 fs ahead the pump to photoexcite excess free carriers in the sample before the pump-probe measurement. The probe was passed through a variable delay line. The SH of the probe was then detected as a function of the time delay between the pump and probe. In the measurements, the pre-pump and pump were kept

to *s*-polarized and the probed SH was detected in *p*-in and *p*-out geometry. The pump-induced change in the probe SH was obtained by processing the PMT output with a lock-in amplifier synchronized to the mechanical chopping rate of the pump. The SH measures the nonlinear optical response in the topmost 200 Å of the GaAs sample due to both a bulk dipole-allowed SH susceptibility and DC-field induced SH in the depletion region.

First of all, we reduced the pump injected carrier density to 2×10^{17} /cm³ to make sure the oscillatory SH signal was mainly due to coherent LO phonons. Then we varied the pre-pump power and monitored the change in the TRSHG signal. The results are shown in Figure 1A. The pre-injected carrier densities for curves b, c and d were calculated to be 7×10^{16} , 2×10^{17} , and 4×10^{17} /cm³ respectively. The background SH signal changed dramatically when the pre-injected carriers were introduced. This phenomenon can be assigned to the

Figure 1 (A) Autocorrelation curve (marked as AC) indicates the delay time of the pre-pump (at -165 fs) and the pump (at 0 fs). TRSHG spectra are shown for different pre-injected carrier density: a: none, b: 7×10^{16} , c: 2×10^{17} , d: 4×10^{17} /cm³. The inset shows the Fourier power spectra of the oscillatory parts, where only LO phonon peak was clearly observed. (B) The oscillatory parts (dots) and the fitting curves (solid lines) are shown together.

Figure 2 The oscillation amplitude of the coherent LO phonon decreases as pre-injected carrier density increases. The dephasing time shows the same trend. The linear fitting curves to the data are shown as the dash lines.

electronic response of the nonlinear susceptibility $\chi^{(2)}(2\omega)$ to the pre-injected carriers in the topmost 200 Å region. The oscillatory parts are then fitted with

$$S(t) = A e^{-t/\tau_{LO}} \cos(2\pi f_{LO} t + \varphi),$$

where A is the oscillation amplitude, τ_{LO} is the dephasing time constant, f_{LO} is the phonon frequency (8.8 THz), and φ is the initial phase of the coherent LO phonon mode. The fitting was shown in Figure 1B. Obviously, our results demonstrated the pre-injected carriers could easily destroy the constant phase relationship of the coherent LO phonons.

Figure 2 plotted the fitting parameters: A and τ_{LO} vs. the pre-injected carrier density. The coherent phonon amplitude reduced a factor of 2 when extra $4 \times 10^{17} / \text{cm}^3$ carriers were presented in the near surface region. According to the depletion field screening mechanism [3], a rapid screening of the depletion field by photoexcited carriers is responsible for launching the coherent LO phonons in the surface space-charge region of GaAs. The pre-injection of carriers is expected to partially screen the depletion field, and therefore weaken the launch of coherent phonons through the pump.

At low carrier densities ($\sim 10^{17} / \text{cm}^3$), the oscillations decay with a single exponential from which the dephasing time was deduced to be ~ 4 ps [3]. However, our measurements showed the dephasing time was only 1.33 ± 0.07 ps when the pump carrier density was $2 \times 10^{17} / \text{cm}^3$. The dephasing time reduced to 1.08 ± 0.07 ps as the pre-pump carrier density turned to $4 \times 10^{17} / \text{cm}^3$. The disagreement might be due to the different probing depth of linear reflectivity (absorption length $\sim 1 \mu\text{m}$) and reflected SH (coherent length ~ 200 Å), where the later is more sensitive to the inhomogeneous carrier distributions in the near surface-

space region. The shorter dephasing time indicates a much stronger carrier-phonon scattering happened in the near surface-space region.

The dephasing dynamics of coherent phonon in bulk GaAs has been investigated by Coherent Anti-Stokes Raman Scattering (CARS) technique [5]. The dephasing of coherent LO phonons is controlled not only by the decay of the optical phonons into acoustic phonons, but also by scattering with in the optical branch and any other phase-destroying collision processes. The measured dephasing rate $1/\tau$ includes contributions from all dephasing phenomena, such as elastic scattering within the phonon branch (pure dephasing rate $2/T_2^*$), as well as from the anharmonic decay of the optical into two or more acoustic phonons (decay rate $1/T_1$). However, the thermalization of photoexcited hot carriers might play the most important role in the observed dephasing rate since the most efficient energy relaxation process for photoexcited carriers is inelastic carrier-phonon scattering in GaAs. The relevant interaction is the long-range Fröhlich interaction. The relaxation time for photoexcited electrons to emit a LO-phonon due to the polar Fröhlich coupling turns out to be 130 fs in bulk GaAs [6]. In fact, these emitted LO phonons are *incoherent* comparing with those coherent LO phonons. It is no surprise that these thermal phonons can easily break the phase relation of coherent LO phonons via phonon-phonon scattering. We believe that this scattering process has some contribution to the observed short dephasing time.

In conclusion, the dephasing process of coherent LO phonons in GaAs was accelerated by the pre-injected carriers through carrier-phonon scattering and incoherent phonon-phonon scattering. This experiment allows one to directly investigate the influence of carriers on the dephasing mechanisms of phonons in semiconductors and their nanostructures.

Reference

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