Dynamic longitudinal-optical phonon decay via transient electron-phonon interactions in low-temperature-grown GaAs

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We present results on dynamic decay of coherent longitudinal-optical phonons in a low-temperature-grown GaAs. We show that the observed behavior originates from transient electron-phonon scatterings which reflect the depletion of carriers from the surface via carrier trapping and diffusion processes. The electron longitudinal-optical (LO)-phonon scattering rate of around $8.6 \times 10^{-18}$ ps$^{-2}$ cm$^3$ is obtained from the excitation density dependence of the scattering rate. © 2006 American Institute of Physics. [DOI: 10.1063/1.2188590]

Developments in femtosecond laser techniques have enabled the coherent excitation and time-resolved mapping of lattice vibrations. Recent application of coherent phonons has extended to such vigorous fields as direct observation of coherent atomic motions with time-resolved x-ray diffraction, and the ultrafast structural changes by the excitation of large amplitude coherent phonons. The redshift of coherent phonon frequency observed in Bi for intense excitations with lattice displacements up to several percentage of coherent phonon frequency observed in Bi for intense excitations with lattice displacements up to several percentage of the interatomic distance was explained by the anharmonicity of the lattice potential, while a similar behavior in tellurium was interpreted by the electronic softening of lattices. It was also suggested that coherent phonons can be used to forcibly drive precise transformations in carbon nanotubes.

Coherent lattice vibrations are generated by ultrashort pulses under mechanisms such as, surface field screening in bulk GaAs, dispersive excitation of coherent phonons in semimetals, or impulsive stimulated Raman scattering for transparent materials. Regarding decay of the excited coherent lattice modes, anharmonic phonon decay processes and electronic, hole-type carrier-phonon interactions were studied.

In this letter, we report on dynamic decay rate of coherent longitudinal optical (LO) phonon oscillations in low-temperature (LT)-grown GaAs, in which the phonon scattering rate induced by carrier-phonon interactions depends strongly on transient carrier dynamics. We note that LT-GaAs material has been studied extensively for its ultrafast optoelectronic applications due to its reduced carrier lifetime.

Using a Kerr-lens mode-locked Ti:Sapphire laser with the pulse width 20 fs and with the center wavelength around 780 nm, we have performed time-resolved reflective electro-optic sampling measurements on a LT-GaAs layer. In the experiments, femtosecond pulses are split into a pump beam and a weaker probe beam. The pump pulse excites coherent LO phonons in GaAs via the generation mechanism of the ultrafast surface-field-screening of photoexcited carriers. Then, the polarization rotation of the linearly polarized probe beam, which follows the collective lattice motion, is measured as a function of time delay between the pump and probe pulses by the polarization sensitive technique of the electro-optic sampling.

A GaAs layer with the thickness of 1 μm was LT grown at 225 °C after the ZnSe buffer layer growth on a semi-insulating (001) GaAs substrate in a Riber R&D molecular-beam epitaxy machine equipped with both III-V and II-VI growth chambers. The transient absorption measurement to get information on carrier lifetime was also performed after etching the GaAs substrate. The sample temperature was kept at 80 K, and the photoexcitation density was varied.

Figure 1(a) shows the coherent phonon oscillations observed for the LT-GaAs at several pump intensities of $I_0$, 6 $\times$ $I_0$, and 20 $\times$ $I_0$, where $I_0$ corresponds to the photoexcited carrier density of about $3 \times 10^{16}$ cm$^{-3}$. Figure 1(b) is the fast Fourier transform spectra of the respective phonon oscillations in Fig. 1(a). For the case of the highest excitation of $I=20 \times I_0$ with $n \approx 6 \times 10^{17}$ cm$^{-3}$, beatings from the existence of two frequency components are observed, which disappearing at delay times longer than 3 ps. At the excitation density for which the plasmonic frequency is near the LO phonon frequency or at the higher density, the electronic charge oscillations efficiently couple with the LO phonon motion. In this case, it is known that the coupled mode energy of the electronic plasmon and LO phonon moves toward the transverse optical (TO) frequency.

As is expected, the coupled plasmon-LO phonon mode is observed at around 7.86 THz for the highest excitation in the Fourier transformed spectra, with the linewidth being an order of magnitude broader than that of the unscreened LO mode at 8.82 THz. Thus, the initial beatings in time domain disappear with the decay of the coupled plasmon-phonon mode. Two origins for fast coupled mode decay as compared to the un-
Coexistence of the unscreened mode as well as the coupled plasmon-phonon mode is due to the spatial inhomogeneity of the optically excited carrier density within the laser spot.

As we go to lower excitation density regime of \( I \lesssim 6 \times I_0 \), the Fourier transform spectra do not show any signature of the coupled mode near TO phonon frequency and also do not reveal any significant differences between the spectrum at \( I = I_0 \) and that at \( I = 6 \times I_0 \). However, as we give attention to the decaying profiles of the phonon oscillations in time domain, the initially fast decay at \( I = 6 \times I_0 \) becomes obvious as compared to the case of \( I = I_0 \).

It is interesting that the LO phonon decay rate at \( I = 6 \times I_0 \) is not monotonic, but can be changing with time delay. When we plot the phonon amplitude in log scale as a function of time delay in Fig. 2, where the slope of the curve corresponds to the phonon decay rate, the manifestation of dynamic decay rate becomes more clear. While the decay rate converges to a constant value (0.087 ps\(^{-1}\)) with increased time delay, the initial decay rates are dynamic and much faster.

The observed dynamic behavior of the LO phonon decay can be explained when we consider the phonon decay channel of the electron-LO phonon scatterings. In polar materials, such as GaAs, the LO phonon mode efficiently couples to electrons via the deformational potential interactions. The electron and hole carriers which are launched by pump pulse are expected to contribute to the LO phonon decay rate via carrier-phonon interactions which will scale with the carrier density. In case of LT-GaAs, where the carrier decay time can be comparable to or even shorter than the intrinsic phonon decay time, the time-dependent LO phonon decay rate can be comparable to or even shorter than the intrinsic phonon decay time, the time-dependent LO phonon decay rate converges to a constant value with increasing time delay, the initial decay rates are dynamic and much faster.

The fitting parameter of \( \Gamma_0 \) did not change much with excitation densities, is longer than the pump beam intensity. Solving the differential equation of \( \frac{dA}{dt} = -\Gamma(t) \times A \) for phonon amplitude \( A \), we get the solution such that

\[
\ln A = \text{const.} - \gamma_0 t + \Gamma_0 \times n_0 \times \exp(-t/T_d),
\]

where \( \Gamma_0 \) is the phonon decay rate without the contribution of electron-phonon interactions, and \( \Gamma_0 \), \( T_d \) being the electron-LO phonon scattering constant, the electronic density disappearance time, respectively. And, \( n_0 \) is the initial photoexcited electron density which will be proportional to the pump beam intensity. Solving the differential equation of \( \frac{dA}{dt} = -\Gamma(t) \times A \) for phonon amplitude \( A \), we get the solution such that

\[
\ln A = \text{const.} - \gamma_0 t + \Gamma_0 \times n_0 \times \exp(-t/T_d),
\]

Assuming that the effect of spatial carrier density inhomogeneity is not dominant, we have fitted the log-scale amplitude \( \ln A \) obtained at \( I = 6 \times I_0 \) with Eq. (2) and obtained fitting parameters; \( \Gamma_0 = 0.087 \text{ ps}^{-1} \), \( \Gamma_0 = 8.3 \times 10^{-18} \text{ ps}^{-1} \text{ cm}^3 \), and \( T_d = 0.85 \text{ ps} \). Regarding the obtained electron density disappearance time of \( T_d = 0.85 \text{ ps} \), we suggest two kinds of channels; one is the decay of carriers by the recombinasion and/or trapping to defect states, and the other is the diffusion of carriers off the surface region where coherent phonons are generated. We note that coherent LO phonons are dominantly generated near the surface region for the generation mechanism of the ultrafast surface field screening which is the case for GaAs.

To get access to the carrier decay time of the LT-GaAs, we have performed time-resolved transmission measurements after etching the GaAs substrate. As is shown in the inset of Fig. 2, the estimated carrier lifetime of 2.4 ps, which did not change much with excitation densities, is longer than the fitting parameter of \( T_d = 0.85 \text{ ps} \) for the electron density disappearance time at \( I = 6 \times I_0 \). This discrepancy is understandable when we take into account the diffusion of carriers off the surface region. The transmission measurements should not be sensitive to carrier diffusions along the growth direction, while the interaction rate of carriers with coherent phonons must be sensitive to carrier densities near the surface, as the density of LO phonon excitation is dominant near the surface.

\[
\Gamma(t) = \Gamma_0 + \Gamma_0 \times n_0 \times \exp(-t/T_d),
\]

where \( \Gamma_0 \) is the phonon decay rate without the contribution of electron-phonon interactions, and \( \Gamma_0 \), \( T_d \) being the electron-LO phonon scattering constant, the electronic density disappearance time, respectively. And, \( n_0 \) is the initial photoexcited electron density which will be proportional to the pump beam intensity. Solving the differential equation of \( \frac{dA}{dt} = -\Gamma(t) \times A \) for phonon amplitude \( A \), we get the solution such that

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\ln A = \text{const.} - \gamma_0 t + \Gamma_0 \times n_0 \times \exp(-t/T_d),
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be fitted well by the dynamic phonon decay model of Eq. (2), proportional to the existing carrier density. Each curve could be fitted well by the phonon scattering probability with carriers which is initial slope becoming steeper with the higher excitations.

The dependence on excitation density of the fitting parameter $T_{\text{el}}$ of Fig. 3(a) is plotted in the inset of Fig. 3(a). We find that $T_{\text{el}}$ increases with decreasing excitation density, reaching $T_{\text{el}} = 2.2$ ps at the lowest density of $I_0$. This behavior on carrier density can be understood as the effect of the increased diffusion rate of carriers off the surface region, with the higher photoexcitations.21

Though the surface carrier density undergoes different disappearance rates with excitation intensities, as inferred from the inset of Fig. 3(a), the initial carrier density shortly after the pump pulse is proportional to the excitation intensity. Figure 3(b) shows an initial decay rate of coherent phonons deduced from the fittings, as a function of the carrier excitation density by the pump pulse. We confirm that the initial decay rate is linearly proportional to the initial carrier density. Furthermore, the consistency of the proportionality value of $8.6 \times 10^{-18}$ ps$^{-1}$ cm$^3$ from Fig. 3(b) with the value of $\Gamma_{\text{el}} = 8.3 \times 10^{-18}$ ps$^{-1}$ cm$^3$ obtained from the single curve at $t = 6 \times I_0$ of Fig. 2, supports the proposed dynamic decay model, especially by the mechanism of transient electron-LO phonon scatterings in LT-GaAs.

In summary, we have performed coherent LO phonon measurements in a LT-GaAs, where the carrier lifetime is shorter than the intrinsic phonon decay time. As the carrier density undergoes the ultrafast depletion from the surface via carrier trapping and diffusion processes, the coherent LO phonon decay rate is reduced due to the disappearance of the transient electron-phonon scattering channel. The electron LO-phonon scattering rate of around $8.6 \times 10^{-18}$ ps$^{-1}$ cm$^3$ is obtained from the excitation dependence of the scattering rate.

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References:

FIG. 3. (a) Log-scale amplitudes ($\ln A$’s) as a function of time delay at excitation intensities of $I_0$, $2 \times I_0$, and $6 \times I_0$. Inset shows $T_{\text{el}}$’s as a function of excitation density obtained from fittings with Eq. (2). (b) Initial decay rate as a function of the excitation density.