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Coherent time-resolved investigation of LO-phonon dynamics in GaAs

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The dynamics of coherently excited LO phonons is investigated in GaAs using an infrared time-resolved coherent anti-Stokes Raman scattering technique. The LO-phonon dephasing time is precisely measured as a function of crystal temperature. The results reveal that the relaxation is dominated by the intraband decay into a transverse-acoustic phonon and a LO phonon at the L critical point of the Brillouin zone. The frequently proposed decay route into two longitudinal-acoustic phonons is found to be incompatible with results from our temperature-dependent measurements.

Phonons strongly influence most of the fundamental physical properties of semiconductors. Among these is the central problem of hot-carrier dynamics which is mainly governed by carrier-carrier and carrier-phonon interactions. Except at very low temperatures, the latter process is dominated by the strong Fröhlich coupling of carriers with the longitudinal-optic (LO) phonons. Recently, the role of the LO phonons in the energy relaxation of electrons has been extensively investigated in bulk GaAs and in GaAs quantum wells,[1-9] yielding evidence for a slowing down of the electron relaxation at high electronic densities due to the hot-phonon effect (i.e., the effect of the nonequilibrium LO phonons generated by the hot-electron relaxation). The dynamical properties of the LO phonons directly govern this accumulation effect and are thus of central interest for the understanding of relaxation and thermalization of electrons in GaAs.[1,5,6]

The LO phonon dynamics in GaAs was investigated using both time-resolved and frequency-resolved techniques. In the time domain, time-resolved incoherent Raman spectroscopy has been recently used to measure the population relaxation time (T_1) of the LO phonons for a few specific temperatures.[7-9] In these measurements one probes the nonequilibrium population of incoherent phonons indirectly created by cascade relaxation of photoexcited electrons and, hence, the measured decay of the phonon population can be influenced by the hot-electron dynamics under certain experimental conditions.[4] Furthermore, the smallness of the incoherent signal limits the dynamics and, thus, the precision of the measurement. In the frequency domain, linewidth measurements were performed by spontaneous Raman spectroscopy,[10-12] but with a limited precision, particularly at low temperatures, leading to some discrepancy in the experimental results. These limitations, together with the frequently limited temperature range of the investigations have precluded an unambiguous identification of the LO phonon relaxation channel, and further experiments are clearly required to determine their dynamics.

In spite of their large dynamics of measurement and their consequent high accuracy, time-resolved coherent nonlinear techniques were not yet applied to the investigation of the LO phonon dynamics in GaAs. In order to obtain more precise and extensive information on the dynamics of this excitation, we have thus performed a direct time-domain determination of its dephasing time (T_2) using a time-resolved coherent anti-Stokes Raman scattering (CARS) technique. Here, a coherent phonon population is directly created by coherent Raman scattering of two time-coincident picosecond pulses of frequencies ω_L and ω_S, with $\omega_L - \omega_S = \omega_{\text{phonon}}$. The loss of coherence of this excitation is then followed by coherent anti-Stokes Raman scattering at $\omega_{\text{AS}} = \omega_P + \omega_{\text{phonon}}$ of a time-delayed probe pulse of frequency ω_P.

In this technique, the excitation is generated in the bulk of the crystal, which requires at least two picosecond pulses (if $\omega_L = \omega_P$) operating in the transparency region of the sample, i.e., in the near infrared for GaAs ($E_{\text{gap}} \sim 1.52$ eV). These infrared pulses were produced by a modified version of the passively mode-locked Nd:glass laser system already described and utilized for visible CARS experiments.[13] The initial beam, delivering 5-ps pulses of 5 mJ at 1.054 μm is split into three parts. The first one ($\sim 10\ \mu$J), after passing through a variable delay, is used as the probe beam (ω_P). The second one ($\sim 30\ \mu$J) is utilized as one of the excitation beams (ω_L), while the third one is frequency shifted by stimulated Raman scattering in $CHBr_2Cl$ to generate the ω_S pulse ($\sim 10 \mu$J) at $\sim 1.084\ \mu$m ($\omega_L - \omega_S = 295\ \text{cm}^{-1}$, the LO phonon frequency in GaAs). After polarization adjustment, the three beams are slightly focused into the sample over a diameter of $\sim 500\ \mu$m. The sample is a high-quality crystal of semi-insulating GaAs (carrier concentration $< 10^{15}$ electrons/cm^3) with (100) surfaces and 625 μm thickness. It was placed in a variable-temperature cryostat and was oriented near the Brewster angle for the incident beams. The crystallographic axes parallel to the crystal surface were oriented at 45° with respect to the scattering plane. This geometry with the three incident pulses and, thus, the coherent signal polarized in the incident plane, minimizes multiple reflections from the crystal surfaces. The generated anti-Stokes signal is detected by an S1 photomultiplier after spatial and spectral filtering. The temporal resolution of the system is limited to 2 ps because of residual reflections on the sample surfaces, but could be easily improved using a thinner crystal.

In this experiment, we have limited the focusing of the beams in order to avoid any complication arising from strong photoexcitation of electrons by two-photon absorption[14] and by residual one-photon absorption from impurities.[15] The creation of an electron plasma, even for relatively low densities, $\leq 10^{17}$ electron/cm^3, strongly modifies the statics and the dynamics of the LO phonons because of the phonon-plasmon interaction[16,17] and care must be taken to avoid any spurious effects on the measured decay time. We have verified that the density of optically created carriers is too small to influence our measurements by checking the independence of the measured decay time on the laser intensity.

The observed CARS signal is plotted on a logarithmic scale as a function of probe delay in Fig. 1, for a crystal temperature of 77 K. After an initial fast rise and fall essentially limited by the system response function and due to the electronic nonlinear response of the crystal, we observe an exponential decrease of the coherent signal over 4 orders of magnitude indicating a Lorentzian (homogeneous) broadening of the line. The slope of the decay directly measures the dephasing time $T_2/2=6.4\pm0.4$ ps [i.e., a full width at half maximum (FWHM) of 0.83 ± 0.05 cm^{-1}]. This $T_2/2$ value is in good agreement with the energy relaxation time T_1 ($T_1=7\pm1$ ps) measured in time-resolved Raman experiments[7,9] and with the Raman data reported in Ref. 7.

The observed decay was exponential over several decades for all temperatures investigated, ranging from 6 to 215 K. A maximum value of $T_2/2=9.2\pm0.6$ ps is measured at 6 K. The measured temperature dependence of the LO phonon dephasing rate $\Gamma=2/T_2$ is shown in Fig. 2. This dependence provides the signature allowing us to discriminate between the possible decay channels.

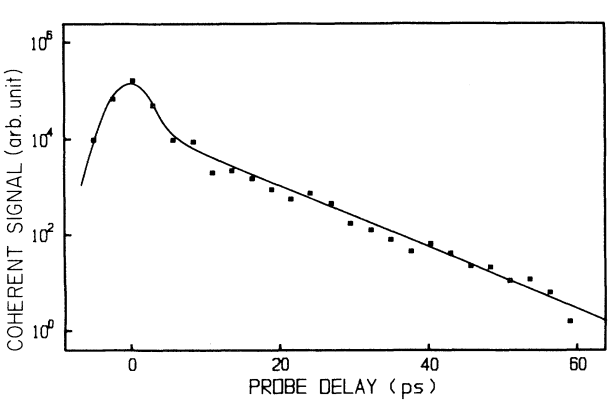

FIG. 1. Coherent anti-Stokes signal from the LO phonon in GaAs at 77 K, plotted on a logarithmic scale as a function of probe delay in ps. A dephasing time of $T_2/2=6.4$ ps is measured from the exponential decay of the signal.

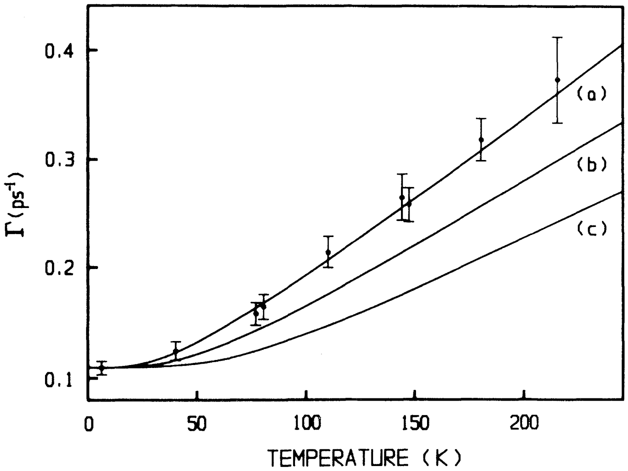

FIG. 2. Measured dephasing rate Γ for the LO phonon in GaAs plotted vs crystal temperature. The solid lines are calculated for a LO phonon decay into (a) a TA(L) phonon and a LO(L) phonon, (b) a TA(X,K) phonon and a LA(X,K) phonon, and (c) two LA phonons of half the LO phonon energy (see text).

In good quality crystals, phonon relaxation is a consequence of anharmonic interactions between phonons. Three-phonon interactions, which are governed by the third-order anharmonicity, are the most probable processes contributing to the phonon lifetime. Since pure dephasing involves higher-order anharmonicity, the relation $T_2/2=T_1$ holds for the three-phonon mechanisms, in agreement with our experimental results. For LO phonons in GaAs energy conservation only allows third-order down-conversion processes where an initial phonon splits into two lower-energy phonons. As the efficiency of these mechanisms strongly depends on the density of final two-phonon states, the dominant contributions are expected to come from relaxation channels involving zone-edge phonons. On this basis and taking into account the measured phonon dispersion curves[18,19] two relaxation channels satisfying energy, wave vector, and symmetry conservation might be expected to give an important contribution to the energy relaxation of the LO phonons. The first channel is intraband decay into a transverse-acoustic (TA) phonon ($\nu_{\mathrm{TA}}\sim 60$ cm^{-1}) and a LO phonon ($\nu_{\mathrm{LO}}\sim 235$ cm^{-1}) of opposite wave vector at the L critical point of the Brillouin zone. The relevance of this mechanism can be tested by comparing the predicted and measured temperature dependence of the damping rate. For such a down-conversion process, the theoretical variation of the damping rate Γ with temperature is given by[20]

$$\Gamma=\gamma_0[1+n(\nu_{\mathrm{TA}})+n(\nu_{\mathrm{LO}})]\ , \qquad (1)$$

where $n(\nu)$ is the occupation number for the ν phonon. γ_0 is an effective anharmonic constant which is fixed by the low-temperature measurement at 0.11 ps^{-1} (the occupation numbers are negligible at 6 K). The precision of this low-temperature measurement is thus essential for the discrimination of the relaxation channels as it imposes the only parameter in the comparison between theory and experiment. Using this expression, we obtain curve (a) in Fig. 2 which perfectly reproduces the experimental data suggesting a dominant contribution of this intraband decay route. Furthermore, extrapolation of this theoretical curve predicts a LO phonon linewidth of 2.6 cm^{-1} at 300 K, in good agreement with the most recent Raman measurements.[8,12]

The second mechanism is relaxation of the LO phonon into a TA phonon ($\nu_{\mathrm{TA}} \sim 80\ \mathrm{cm}^{-1}$) and a longitudinal-acoustic (LA) phonon ($\nu_{\mathrm{LA}} \sim 215\ \mathrm{cm}^{-1}$) near the K and X points of the Brillouin zone.[18,19] The temperature dependence of the associated damping rate is given by an expression similar to Eq. (1) (changing ν_{LO} into ν_{LA}). Taking into account only this mechanism and noting that γ_0 is again fixed by the low-temperature measurement, we obtain curve (b) in Fig. 2. This curve clearly deviates from the experimental data as the temperature increases, and only gives a poor description of our measurements. In fact, if we take simultaneously into account the two previous relaxation channels, the best fit is obtained for a negligible contribution of the last process suggesting that the first relaxation channel is dominant. As the density of two-phonon states is expected to be comparable for these two mechanisms the fact that the measured temperature dependence is only consistent with the first one suggests that the cubic anharmonic constant is greater for an intraband process involving two LO phonons.

The relevance of the proposed relaxation channel is also supported by nonequilibrium two-phonon Raman spectra.[15] They reveal a strong enhancement of the LO(L)-TA(L) and TO(L)-TA(L) difference bands at the L point of the Brillouin zone after photoexcitation of electrons, suggesting that the near-zone-center LO phonons generated by hot-electron decay relax with creation of TA(L) phonons.

Finally, we emphasize that the frequently invoked LO phonon decay route into two LA phonons with half the LO phonon energy and opposite wave vector is not supported by our experimental data. For this mechanism the theoretical temperature dependence of the decay rate can be written as

$$\Gamma = \gamma_0[1 + 2n(\omega_{\mathrm{LO}}/2)] \ . \qquad (2)$$

Using this expression we calculate a temperature variation of Γ by far slower than the measured one [curve (c) in Fig. 2]. In fact, the phonon dispersion curves clearly show that this down-conversion process is associated with a very low density of final two-phonon states and is thus much less likely than the previous ones. Moreover, the Γ_{15} component of the two-phonon spectrum, satisfying symmetry conservation for relaxation of the LO mode is mainly due to combination bands rather than to overtone modes.[21,22] This decay channel is thus theoretically very unlikely in GaAs and, as confirmed experimentally, can be neglected.

In conclusion, the LO phonon dephasing time in GaAs is investigated by use of an infrared time-resolved CARS. The results improve the precision of the previously available data, particularly at low temperature, allowing the discrimination between the possible dephasing processes. As expected from the theory, pure dephasing effects are negligible and the experimental results are interpreted in terms of population relaxation. Comparison between the theoretical and experimental temperature dependence of the dephasing rate shows that the LO phonon relaxation in bulk GaAs is dominated by the intraband decay into a TA phonon and LO phonon at the L critical point of the Brillouin zone. The contribution of the frequently cited decay channel into two LA phonons is found to be negligible as expected from simple theoretical considerations. Similar coherent experiments could yield some additional insight into the LO phonon dynamics under different conditions in bulk GaAs and, in particular, in the presence of a high-density electron or hole plasma, or in more complex media such as GaAs quantum wells or $\mathrm{Ga}_{1-x}\mathrm{Al}_x\mathrm{As}$ alloys.

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