Temperature-dependent electron-lattice thermalization in GaAs

N. Del Fatti, P. Langot,* R. Tommasi,† and F. Vallée
Laboratoire d'Optique Quantique du CNRS, Ecole Polytechnique, 91128 Palaiseau cedex, France
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Thermalization of hot electron with the lattice is precisely investigated in bulk GaAs in the temperature range 50–300 K using a high-sensitivity two-color femtosecond absorption saturation technique. The results show that for lattice temperatures larger than 100 K, the photoexcited electron gas eventually thermalizes with the lattice with a characteristic time close to the LO phonon lifetime as a consequence of the nonequilibrium hot-phonon effect. For lower temperatures, the electron-lattice thermalization time is found to be longer than the LO phonon lifetime in agreement with numerical simulations of the coupled carrier-LO phonon dynamics. [S0163-1829(99)09807-0]

I. INTRODUCTION

Redistribution mechanisms of the energy of carriers between themselves and with the lattice are important issues in the electronic properties of semiconductors.[1,2] In polar compounds, LO phonons play a central role in the latter processes, mediating carrier-lattice energy exchanges for not-too-low carrier temperatures. The efficiency of this energy-transfer channel is, however, limited by carrier-LO phonon energy exchanges and by the energy redistribution processes between the lattice modes via the lattice potential anharmonicity.[1–4] As the intrinsic lifetime of LO phonons is of the order of a few picoseconds,[5] they can be driven out of equilibrium during fast excess energy relaxation of nonequilibrium electrons (nonequilibrium phonon effect).[2,6–12] This bottleneck effect leads to a drastic slowing down of the electron-lattice thermalization whose dynamics is then related to that of the saturation process, i.e., energy redistribution between the lattice modes and/or electron-gas–LO-phonon energy exchanges.

The nonequilibrium phonon effect has been extensively investigated in various bulk and quantum-confined semiconductor systems. Direct correlation between the final characteristic electron-lattice thermalization time τ_{th} and the LO phonon lifetime τ_{LO} has, however, been only recently demonstrated by precisely investigating hot- and cold-electron thermalization dynamics at room temperature for low-photoexcited carrier densities ($\leqslant 10^{17}$ cm^{-3}) in bulk GaAs.[10,12,13] In these conditions, the energy exchanges between the electron gas and the LO phonons occur on a much faster time scale than τ_{LO} and, as a quasiequilibrium establishes between the two systems, τ_{LO} eventually governs the final electron-lattice thermalization (i.e., $\tau_{th} \sim \tau_{LO}$).[12] In contrast, at very low temperatures hot electrons have been shown to cool down to the lattice temperature on a time scale much longer than τ_{LO}.[2,6–9] In this regime, slowing down of the electron-gas–LO-phonon energy exchanges and additional bottleneck effect due to second-generation phonons (i.e., phonons created by nonequilibrium LO phonon decay)[14,15] play crucial roles.

We have investigated this change of behavior of the electron-lattice thermalization dynamics by extending our previous room-temperature measurements in bulk GaAs (Ref. 12) to lower temperatures, in the range 50–300 K. The results show that the electron-lattice thermalization time is comparable to the LO phonon lifetime for lattice temperatures down to 100 K and becomes much longer for lower temperatures, in agreement with numerical simulations of the carrier-relaxation dynamics.

II. EXPERIMENTAL TECHNIQUE

Measurements were performed using a high sensitivity two-wavelength femtosecond absorption saturation technique. The pump wavelength is chosen to photoexcite ''hot'' electrons with an average initial energy E_{ex} about one LO phonon energy larger than their average thermal energy E_{th} at the lattice temperature $E_{ex}-E_{th}(T_L) \sim E_{LO}$. This moderate electron excess energy precludes any spurious effects due to electron intervalley scattering.[16] After photoexcitation, electrons and holes internally thermalize within each band in few hundred femtoseconds. Electron-lattice thermalization is followed by measuring the transient transmission change of a probe pulse whose wavelength is chosen so that the sample absorption change is dominated by the electron response.[12,17]

The femtosecond pump and probe pulses are created by frequency conversion of a high power femtosecond Ti:Al_2O_3 oscillator. The details of the experimental system were described elsewhere.[18] We simply recall here that the initial pulses are first frequency broadened in an optical fiber and two synchronized, independently tunable 100-fs pulses are selected using two grating pair compressors. This system permits very high sensitivity two-wavelength measurements using a standard pump-probe setup and thus to precisely monitor the electron cooling dynamics.[12]

Measurements were performed in a 0.2-μm-thick molecular-beam epitaxy (MBE) grown intrinsic GaAs sample with $Al_xGa_{1-x}As$ cladding layers and antireflection coating. It is glued on the cold finger of a helium-cooled cryostat permitting sample temperatures in the range 30–300 K.

III. RESULTS AND DISCUSSION

The measured transient differential transmission $\Delta T/T$ is shown in Fig. 1 for a carrier density of 1×10^{17} cm^{-3} and

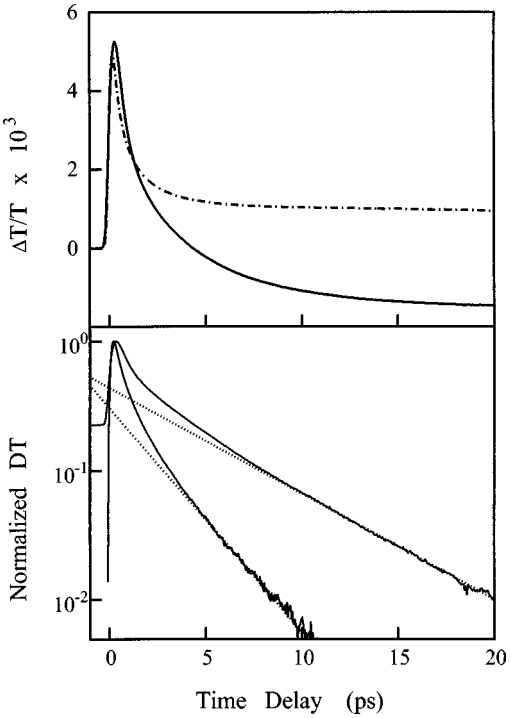

FIG. 1. (a) Transient change of transmission $\Delta T/T$ in GaAs for a carrier density of 1×10^{17} cm^{-3} and lattice temperatures T_L of 100 K (full line) and 190 K (dash-dotted line). (b) Time dependence of the normalized transmission difference $DT=\Delta T/T-(\Delta T/T)_{QE}$ on a logarithmic scale. $(\Delta T/T)_{QE}$ is the quasi equilibrium $\Delta T/T$ measured at $t=40$ ps. The dotted lines correspond to exponential decays with time constants τ_{th} of 2.5 ps ($T_L=190$ K) and 5.3 ps ($T_L=100$ K).

sample temperatures of 100 and 190 K. The pump wavelength λ_{pump} has been modified to account for the band-gap shift with temperature and is, respectively, 775 and 780 nm. High-energy electron states in the tail of the electron-gas distribution being probed ($\lambda_{probe}\sim805$ nm), electron-gas cooling corresponds to a decrease of the probed state occupation number and thus of $\Delta T/T$. After a fast transient, the measured transmission change slowly decays to its long-term value $(\Delta T/T)_{QE}$ that corresponds to a quasiequilibrium situation between the carriers and the lattice.[19] The amplitude of $(\Delta T/T)_{QE}$ actually results from both absorption reduction due to band filling and absorption increase due to many-body effects.[17] For a fixed carrier density, the former contribution decreases with temperature due to reduction of the occupation numbers of the probed states concomitant with reduction of the carrier average energy. Lowering the sample temperature thus leads to a decrease of $(\Delta T/T)_{QE}$ that eventually becomes negative as the many-body effect contribution becomes dominant (Fig. 1). This contribution, however, essentially introduces an offset on the measured signal[17,20] whose temporal behavior is thus dominated by the electron thermalization dynamics.

The picosecond relaxation of $\Delta T/T$ to $(\Delta T/T)_{QE}$ shows a slow cooling of the electron gas to the lattice temperature. For the investigated low-carrier density, screening of the Fröhlich interaction and plasmon-phonon hybridization play a minor role and this behavior can be attributed to overpopulation of the LO phonon modes coupled with the hot electrons, i.e., a hot phonon effect.[12] The characteristic long-term electron-lattice thermalization time can be estimated by plotting the transmission difference $DT=\Delta T/T-(\Delta T/T)_{QE}$ on a logarithmic scale as a function of the probe-time delay (Fig. 1). After a fast decay, DT decreases almost monoexponentially with a time constant τ_{th} rising from ~2.5 ps at 190 K to ~5.3 ps at 100 K. A similar behavior has been observed over the entire investigated temperature range for photoexcited carrier densities smaller than $\sim10^{17}$ cm^{-3}. The τ_{th} values determined for sample temperature ranging from 50 to 300 K are plotted in Fig. 2 and show a large increase of the electron-lattice thermalization time for temperatures lower than 100 K.

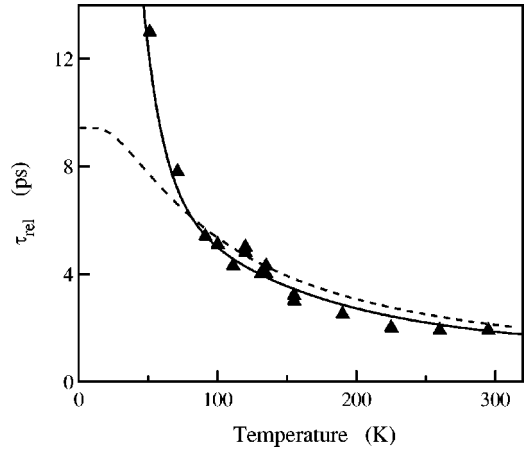

FIG. 2. Lattice temperature dependence of the measured long delay electron-gas cooling time τ_{th}. The full line is computed using simulations of the coupled LO phonon-carrier dynamics and the dashed line shows the LO phonon dephasing time $T_2/2$.

The observed behavior can be understood using a simplified rate equation model describing energy exchanges between the electron-gas and LO phonon subsystems, and their coupling with the thermal bath (other lattice modes). During cooling of the electron gas, zone center LO phonons are created in a wave-vector range limited by momentum and energy conservations. These decay into lower frequency phonons with their intrinsic lifetime τ_{LO}, due to lattice anharmonicity. As a crude approximation, assuming that a temperature T_{LO} can be defined for the zone center LO phonons interacting with the electron gas at temperature T_e, the electron and LO phonon cooling dynamics is described for small excess temperatures (i.e., T_e-T_L, $T_{LO}-T_L\ll T_L$) by

$$\frac{\partial T_e}{\partial t}=\frac{T_{LO}-T_e}{\tau_{e\text{-}LO}}, \tag{3.1a}$$

$$\frac{\partial T_{LO}}{\partial t}=\frac{C_e}{C_{LO}}\frac{T_e-T_{LO}}{\tau_{e\text{-}LO}}-\frac{T_{LO}-T_L}{\tau_{LO}}, \tag{3.1b}$$

where T_L is the lattice temperature (second-generation phonons are neglected here), $\tau_{e\text{-}LO}$ the electron–LO-phonon energy exchange time, C_e the electron-gas heat capacity, and C_{LO} the heat capacity of the LO phonons coupled with the electron gas.[3,4,12]

For not-too-low temperatures, electron–LO-phonon energy exchanges are much faster than LO phonon energy decay to the lattice, $\tau_{e\text{-}LO}\ll\tau_{LO}$ (Fig. 3). Here $\tau_{e\text{-}LO}$ has been numerically computed for an electron density of

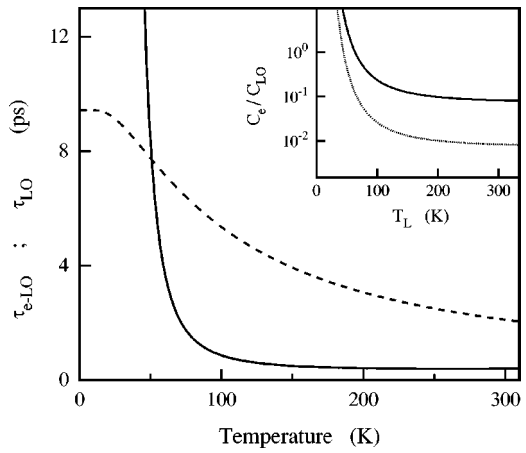

FIG. 3. Temperature dependence of the electron–LO-phonon energy exchange time $\tau_{e\text{-}LO}$ calculated for an electron density of 1×10^{17} cm^{-3} and of the LO phonon dephasing time $T_2/2$. The inset shows the temperature dependence of the ratio of the electron and LO phonon heat capacities computed using Eq. (3.3) and for carrier densities of 1×10^{17} cm^{-3} (full line) and 1×10^{16} cm^{-3} (dotted line).

1×10^{17} cm^{-3} taking into account screening and Pauli exclusion principle.[12] The calculated $\tau_{e\text{-}LO}$ values are almost identical to the ones that can be obtained in the small excess temperature limit from the usual formula for the average rate of electron-energy loss.[1,2] In this regime, analogous to the room-temperature one, the two strongly interacting subsystems thus quickly reach a quasiequilibrium $T_e \sim T_{LO} > T_L$ and their subsequent cooling is governed by LO phonon decay. Using Eqs. (3.1) one can show that the coupled subsystems thus cool to the lattice temperature with a characteristic time τ_{th}:[3,4,12]

$$\tau_{th} \approx (1 + C_e/C_{LO})\ \tau_{LO}, \tag{3.2}$$

depending on the ratio of the heat capacity of the electron-gas and LO phonon subsystem, with

$$C_{LO} \approx \frac{(E_{LO})^2 k_M^3}{6\pi^2 k_B T_{LO}^2} n_{LO}(1+n_{LO}). \tag{3.3}$$

For a finite electron temperature, there is no clear cut-off for the maximum wave vector k_M of the LO phonons interacting with the electron gas and C_{LO} is thus not precisely defined here. Using numerical simulation of the nonequilibrium LO phonon population dynamics,[12] k_M can however be estimated to be of the order of 5×10^6 cm^{-1}. The ratio C_e/C_{LO} computed with this approximation is plotted in Fig. 3 for identical electron and LO phonon temperatures and electron densities of 1×10^{17} and 1×10^{16} cm^{-3}. For low-electron densities and not-too-low temperatures, C_{LO} largely exceed C_e. In this regime, most of the excess energy of the coupled system is stored in the LO phonons and it thus cools down with a time constant τ_{th} almost identical to τ_{LO}.

In intrinsic GaAs in absence of injected carriers, scattering of zone center LO phonons is dominated by population decay processes and their lifetime and dephasing time can be identified ($\tau_{LO} = T_2/2$).[5,13] The temperature dependence of the latter has been investigated in semi-insulating GaAs using a time-resolved coherent anti-Stokes Raman spectroscopy technique[5] and is reproduced in Fig. 2 by the dashed line. For lattice temperatures higher than 100 K, the measured τ_{th} and τ_{LO} are comparable in the above model, showing that the final electron-lattice thermalization is governed by LO phonon decay. The decay time τ_{th} is, however, found to be slightly smaller than τ_{LO} (Fig. 2), in contrast to the prediction of Eq. (3.2). A similar behavior has been observed in previous room-temperature studies and can be ascribed to additional energy transfer to the photoexcited holes, via electron-hole and nonequilibrium phonon-hole interactions that have been neglected here.[12]

For lower lattice temperatures ($T_L < 100$ K), the LO phonon electron-gas energy exchange time, $\tau_{e\text{-}LO}$, strongly increases due to reduction of the number of electrons above the threshold for LO phonon emission as the electron temperature decreases (Fig. 3). This, together with the reduction of the LO phonon heat capacity as compared to the electronic one, is responsible for the observed strong slowing down of the electron-gas cooling at low temperatures. In this regime, no quasiequilibrium is established between the electrons and LO phonons and a large part of the excess energy stays in the electron gas. τ_{th} is then no more limited by the LO phonon lifetime and becomes much larger than τ_{LO} (Fig. 2).

To better describe our results we have performed simulations of the carrier-relaxation dynamics by numerically solving the coupled LO phonon-carrier Boltzmann equations for electrons and heavy and light holes.[12,17] All carrier-carrier scattering processes have been considered including those inducing intervalence-band transfers. Hole-optical phonon polar and nonpolar interactions have been considered for both intra and intervalence-band mechanisms as well as intervalence-band transfer assisted by acoustic phonons in the energy equipartition approximation. Carrier-acoustic phonon energy exchanges were neglected as the minimum lattice temperature investigated here is 50 K.[2] Dynamic screening in the plasmon pole approximation has been used for the LO phonon polar interactions with electron and light holes. The nonequilibrium LO phonon effect has been considered both for electrons and light holes but has been neglected for heavy holes in agreement with recent experimental investigation of cold-hole heating.[17] The long delay electron-gas cooling times extracted from the computed absorption change are in very good agreement with the measured ones (Fig. 2, full line) with in particular the same increase of τ_{th} for temperatures lower than 100 K ($\tau_{LO} = T_2/2$ has been used in the simulations).

IV. CONCLUSION

Using a two-color femtosecond technique we have investigated the cooling dynamics of a hot photoexcited electron gas to the lattice in GaAs for low-carrier densities ($\leqslant 10^{17}$ cm^{-3}) in the temperature range 50–300 K. The results show that for lattice temperatures higher than $\approx$100 K the final electron-gas cooling dynamics is limited by the LO phonon lifetime, in agreement with a simple rate equation model. In this strong coupling regime of the electron gas with the LO phonons, the two systems quickly reach a thermal quasiequilibrium where most of their excess energy is stored in the LO phonon subsystem. The electron cooling

time thus reflects decrease of the common temperature due to energy losses of the LO phonon subsystem to the other phonon modes. This correlation between the electron cooling time and the LO phonon lifetime also provides a new way of determining this lifetime in polar semiconductors.[13]

At lower temperatures, electron-gas cooling times much larger than the LO phonon lifetime have been measured. This additional slowing down reflects reduction of both the electron-gas–LO-phonon energy exchanges due to blocking of the LO phonon emission by electrons and of the heat capacity of the LO phonons coupled with the electron gas. These results are in very good agreement with numerical simulations of the coupled carrier-LO phonon dynamics indicating, in particular, that second-generation phonon effects, important at very low temperatures and high carrier densities,[15] are negligible here.

*Permanent address: CPMOH, Univ. Bordeaux I, 351 Cours de la Libération, 33405 Talence cedex, France.

†Also at: Istituto di Fisica Medica and Istituto Nazionale di Fisica della Materia, c/o Università degli Studi di Bari, Via Orabona 4, 70126 Bari, Italy.

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