Hot phonons in InAs observed via picosecond free-carrier absorption

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The transient infrared absorption of hot electrons is studied in *n*-type InAs at a wavelength of 6.5 μm. The electron gas is heated by a picosecond infrared pulse to a maximum electron temperature of 560 K. The absorption of hot electrons exceeds the original value substantially and relaxes within 100 ps. Excess LO phonons created by the cooling process are important for the observed enhancement of free-carrier absorption. The free-carrier studies presented here give valuable information on the distribution function of hot phonons.

The stationary free-carrier absorption of polar semiconductors has been investigated in a variety of *p*- and *n*-type doped samples to elucidate the magnitude and the wavelength dependence of the absorption coefficient.[1,2] The experimental results have been analyzed by the simple Drude formula or by more extended quantum-mechanical calculations, which demonstrate the important role played by phonons or lattice imperfections.[3,4] Absorption of free electrons is a three-particle interaction as depicted schematically in the inset of Fig. 1. Absorption of the photon promotes the electron to a virtual level in **k** space, from where it is scattered to a final state in the conduction band by the third particle. Various scattering mechanisms have been considered in the literature, e.g., coupling to longitudinal-optical (LO) and acoustic phonons or scattering by charged impurities. In addition, the influence of many-body effects, e.g., screening of the polar electron-phonon interaction, and collective phenomena, such as coupling of the light wave to plasmons, have been treated theoretically.[5,6] In most cases, the carrier distribution and the lattice are assumed to be in thermal equilibrium at temperatures close to $T=0$.

There is very little experimental knowledge of the absorption of hot free carriers in polar III-V compound semiconductors. The absorption cross section of electrons for the case of different temperatures of the carriers and the lattice is not known. In addition, the effect of a nonequilibrium phonon population—created by carrier cooling—on the strength of the absorption has never been considered. In this paper, we report on the first picosecond study of the transient free-carrier absorption of hot electrons. Substantial changes of the absorption of hot electrons in InAs are detected. The excess absorption returns on a time scale of approximately 100 ps to the initial steady-state value. The theoretical analysis gives convincing evidence for a nonthermal phonon distribution.

The InAs crystal investigated in our experiments is *n* type with an electron density of 1.5×10^{18} cm^{-3}. The sample of a thickness of 300 μm is held at temperatures of 10 and 70 K in the different experiments.

Picosecond infrared pulses tunable between 3.5 and 9.5 μm are generated by parametric down conversion in a nonlinear $AgGaS_2$ crystal. Two pulses from a near-infrared dye laser and a mode-locked neodymium-doped yttrium aluminum garnet (Nd:YAG) laser produce the difference frequency.[7] The pulse duration amounts to 8 ps. The infrared pulses are split in an intense excitation pulse of an energy of 300 nJ and a probe pulse, which is weaker by 2 orders of magnitude. The polarization of the pump pulse is chosen perpendicular to that of the probe pulse to suppress the coherent artifact known to occur around zero delay time for parallel electric field vectors of the two pulses.[8]

The steady-state absorption spectrum of the InAs sample—measured with a standard infrared spectrometer—is depicted in Fig. 1 in the wavelength range from 2 to 10 μm for a sample temperature of 70 K. At wavelengths below 2.5 μm, the band to band absorption rises steeply. At longer wavelengths λ, the absorption of

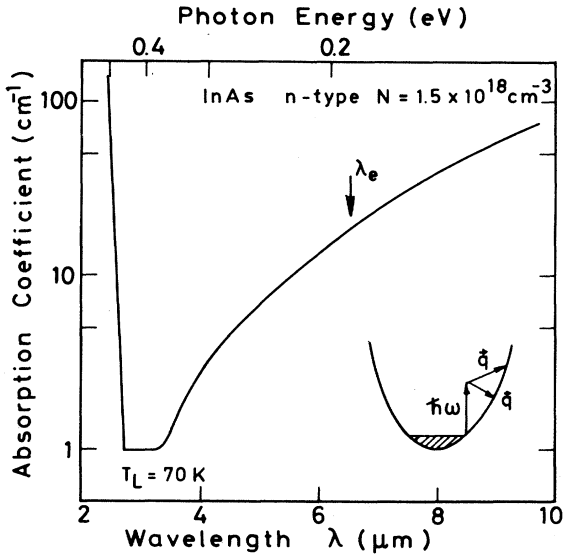

FIG. 1. Infrared absorption spectrum of the *n*-type doped InAs sample at a lattice temperature of $T_L=70$ K. The wavelength λ_e of the picosecond pulses is marked in the figure. The inset shows schematically the free-carrier absorption process.

the free electrons increases proportional to λ^3 (Ref. 9). The spectral position of the picosecond pulses, λ_e, is marked in Fig. 1.

In our picosecond experiments, the sample is excited by an intense pulse at 0.19 eV ($\lambda_e = 6.5\ \mu$m). The photon energy is less than half of the band gap ($E_G \simeq 0.5$ eV) of the doped InAs sample at a lattice temperature of $T_L = 70$ K. As a consequence, no excess electron-hole pairs are created via two-photon absorption. The absorbed infrared photons heat the electron gas and cause a redistribution of the carriers within the conduction band. The resulting change of free-carrier absorption is monitored by a weak delayed probe pulse at the same wavelength. This type of investigation offers substantial advantages in comparison to experiments at shorter wavelengths, where an electron-hole plasma is generated. (i) The carrier distribution consists of electrons in the conduction band only. This fact simplifies the analysis of the data. Just one type of carrier with its specific absorption and scattering mechanisms has to be considered. (ii) The carrier density is constant in the experiment and consequently recombination processes as well as density-dependent many-body effects, like band-gap renormalization, may be neglected.

Experimental results are presented in Figs. 2(a) and 2(b) for lattice temperatures of $T_L = 70$ and 10 K, respectively. The normalized absorption change $\Delta A = -\ln(T/T_0)$ is plotted versus the delay time between pump and probe pulses (points); T_0 and T are the transmissions of the sample before and after excitation. The density of the absorbed photons is approximately 5×10^{17} cm^{-3}. In Fig. 2, the absorption rises rapidly to $\Delta A \simeq 0.15$ at the peak of the curves. At $T_L = 70$ K, the absorption change decays within 70 ps simultaneously with the cooling of the carriers. The complete relaxation of ΔA confirms that no excess carriers are created; the latter would give rise to a long-lived additional absorption. The results for a lattice temperature of $T_L = 10$ K are shown in Fig. 2(b). The decay of the enhanced absorption is somewhat slower extending over a time interval of 100 ps.

We now analyze the data within the framework of the quantum-mechanical theory of free-carrier absorption. The three-particle interaction process, which underlies the absorption mechanism, is described in second-order perturbation theory. The total transition rate $R_{\rm tot}$ of electrons[10] from their initial states $\varepsilon(k)$ to their final states $\varepsilon'(k')$ is given by $R_{\rm tot} = R_+ + R_-$ with

$$R_\pm = (V/2\pi^2\hbar^3)\int d^3k \int d^3k' (|H_{k'k}|^2_\pm |H_{k''k'}|^2/\omega^2)\delta(\varepsilon' - \varepsilon - \hbar\omega \pm \hbar\omega_{\rm LO}) f(k)[1-f(k')] \ . \tag{1}$$

Here $H_{k''k'}$ is the matrix element of the electron-photon interaction, $H_{k'k}$ describes the scattering of the electrons by phonons (or impurities), where + and − signs stand for phonon emission or absorption, respectively; $f(k)$ and $f(k')$ are the Fermi distribution functions of the initial and final states.[11] The delta function $\delta(\varepsilon' - \varepsilon - \hbar\omega \pm \hbar\omega_{\rm LO})$ assures energy conservation ($\hbar = h/2\pi$ with h Planck's constant, V volume of the crystal). The total transition rate is obtained by integrating over all initial and final states. The standard expression for the dipole interaction of photons and electrons is used for the matrix element $H_{k''k'}$, which is proportional to the momentum matrix element of the electrons.[10]

In polar semiconductors, electron–LO-phonon scattering is the most effective coupling mechanism providing momentum conservation in the absorption process for $\hbar\omega > \hbar\omega_{\rm LO}$. The corresponding matrix element[12] is given by Eq. (2):

$$|H_{k'k}|^2_\pm = (2\pi e^2 \hbar\omega_{\rm LO}/V)(1/\epsilon_\infty - 1/\epsilon_0)(1/q^2) \times [q^2/(q^2+q_s^2)]^2 (N_q + \tfrac{1}{2} \pm \tfrac{1}{2}) \ . \tag{2}$$

Here $\mathbf{q}$ is the wave vector of the LO phonon of energy $\hbar\omega_{\rm LO}$, N_q is the occupation factor of the modes with $|\mathbf{q}| = q$, and ϵ_0 and ϵ_∞ represent the dielectric constants of the semiconductor at zero and optical frequencies, respectively. Screening of the polar interaction has to be taken into account for electron densities in the range of 10^{18} cm^{-3}. In our model, static Thomas-Fermi screening characterized by the screening wave vector q_s is considered. As discussed below, it alters the carrier-phonon coupling. In the parabolic band approximation, one obtains for the total free-carrier absorption coefficient $\alpha_{\rm FC}(\omega) = \alpha_+ + \alpha_-$, where

$$\alpha_\pm = (C/\hbar\omega^3)\int_0^\infty dq\, q^3 |H_{k'k}|^2_\pm \times \int_{|k_{\rm min}|_\pm}^\infty dk\, k f(k)[1-f(k+q)] \ . \tag{3}$$

The second integral of Eq. (3) depends parametrically on q via $|k_{\rm min}|_\pm = |m(\omega \mp \omega_{\rm LO})/\hbar q - q/2|$ and via $f(k+q)$. According to Eqs. (2) and (3), $\alpha_{\rm FC}$ depends upon the strength of the electron-phonon interaction and on the phonon occupation numbers N_q (Ref. 13). C is a constant containing material parameters.

Two mechanisms lead to an increase of absorption with electron temperature. First, the distribution functions $f(k)$ and $f(k+q)$ depend on the electron temperature T_e, i.e., the value of the second integral in Eq. (3) changes with time because of the redistribution of carriers within the conduction band. Second, the cooling of a hot carrier distribution is connected with the emission of LO phonons and may lead to a transient nonequilibrium population N_q of the LO-phonon branch.

The transient electron temperature after picosecond infrared excitation and the momentary phonon populations

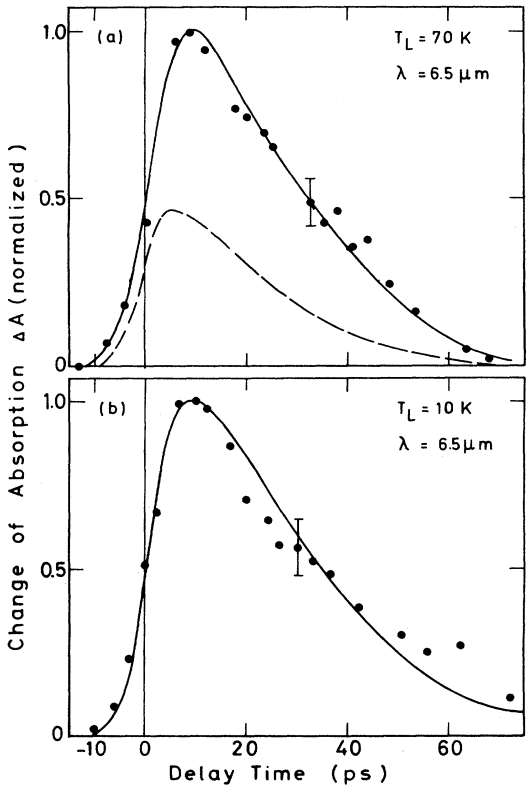

FIG. 2. (a) Transient rise $\Delta A = -\ln(T/T_0)$ of the free-carrier absorption at 6.5 μm vs the delay time between excitation and probe pulses (T_0 and T, transmission of the sample, respectively, before and after excitation). Lattice temperature is 70 K. Solid line: Theoretical transient absorption calculated for phonon excess populations and redistribution of hot electrons in the conduction band. Dashed line: Calculation without hot phonon effects. (b) Transient absorption change for a lattice temperature of $T_L = 10$ K. Experimental points and calculated curve.

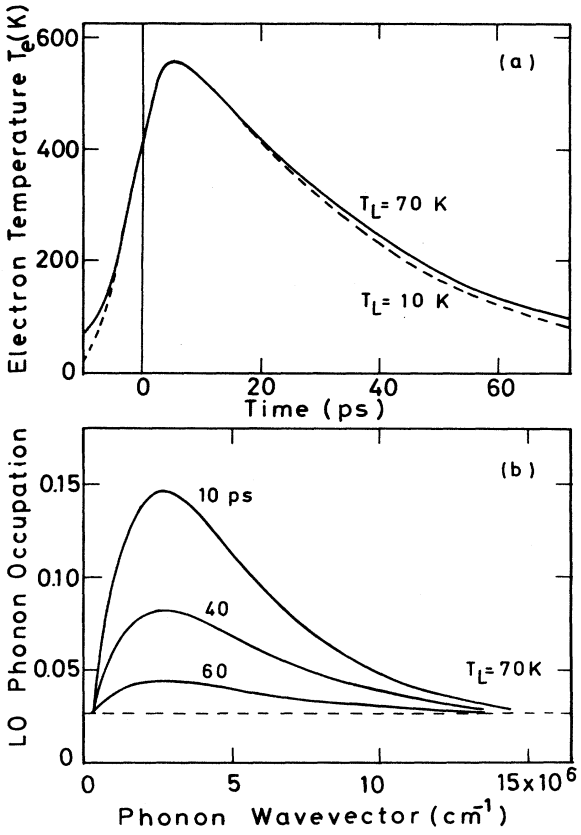

FIG. 3. (a) Calculated time dependence of the electron temperature T_e for two lattice temperatures of $T_L = 10$ and 70 K. (b) Transient phonon spectra generated by carrier cooling for delay times of 10, 40, and 60 ps after excitation at 6.5 μm. The dashed line gives the equilibrium population of the LO-phonon branch for a lattice temperature of $T_L = 70$ K.

N_q are calculated numerically for carrier cooling involving hot phonon effects.[14] This approach consists of two coupled differential equations for the change of energy of the carrier distribution and the Boltzmann equation for the phonon populations N_q. The solution gives the electron temperature T_e and the phonon populations for various q values as a function of time. In our calculations, the Boltzmann equation is solved for 450 q values distributed equidistantly over the Brillouin zone. The phonon energy is $\hbar\omega_{LO} = 0.022$ eV and the q dispersion of the LO-phonon energy is neglected. The phonon lifetime is taken to be 7 ps, as known for GaAs.[15]

The results of the calculation are presented in Fig. 3. The time dependence of the carrier temperature T_e is shown in Fig. 3(a) for lattice temperatures of 10 and 70 K. For an excitation density of 5×10^{17} infrared photons per cm^3, the peak electron temperature rises to 560 K. The cooling of the electron gas does not depend strongly on the lattice temperature for $T_L \leq 70$ K. In Fig. 3(b), the transient phonon population N_q is plotted versus the phonon wave vector q for various delay times after excitation. The q interval depicted corresponds to approximately 15% of the Brillouin zone. The dashed line in Fig. 3(b) represents the small equilibrium phonon population for a lattice temperature of $T_L = 70$ K. Strong nonequilibrium populations of phonons with q around 3×10^6 cm^{-1} occur in the picosecond experiment, especially at early times, where the carrier temperature and, consequently, the phonon emission rate is high. At later times, the phonon distribution broadens because of preferential emission of phonons with larger q values by a carrier distribution of lower electron temperature T_e.

The transient values of T_e and N_q are taken to calculate the free-carrier absorption $\alpha_{FC}(\omega)$ [see Eq. (3)]. Finally, the time dependence of α_{FC} is convoluted with an infrared probe pulse of a duration of 8 ps. The result for a lattice temperature of $T_L = 70$ K is presented in Fig. 2(a) (solid curve). The calculated absorption change has a maximum value of $\Delta A \simeq 0.2$. This result is in good agreement with our experimental findings considering the experimental accuracy and theoretical limitations. It should be noted that the calculation accounts very well

for the time dependence of ΔA found in our measurement.

In order to determine the contribution of the hot phonons to the transient absorption, we have performed a calculation without hot phonons, i.e., the phonon system is assumed to remain in equilibrium [$T_L = 70$ K, dashed line in Fig. 3(b)]; only the effect of carrier redistribution within the conduction band is considered. The result of this calculation is shown in Fig. 2(a) (dashed line). Now, we find a substantially smaller increase of absorption and a faster decay of the transient α_{FC}. This fact clearly demonstrates the important role of the excess phonons for the explanation of the observed transient free-carrier absorption.

The result of the calculation (including excess phonons) for a lattice temperature of $T_L = 10$ K is presented in Fig. 2(b) (solid line). Here, the initial phonon density is negligible and the excess phonons created by cooling lead to the increase of hot-electron absorption. The observed longer decay time stems from the larger deviation from equilibrium at later times.

Of interest are the findings at room temperature. At $T_L = 300$ K, the equilibrium population of the LO-phonon branch ($N_q \simeq 0.64$) is high with a relatively small addition during carrier cooling. Thus, one expects a smaller effect on the free-carrier absorption. This prediction is confirmed by our measurements. The transient hot-electron absorption increases by 0.03 only.

A comment should be made on the importance of the screening of polar interaction. Without screening, one finds a very fast cooling of the carriers within 15 ps and a strongly reduced maximum of the electron temperature of $T_e \simeq 200$ K. The maximum of the phonon spectrum [cf. Fig. 3(b)] occurs at small q values around 3×10^5 cm^{-1} and the width of the spectrum is reduced to approximately 2.5% of the Brillouin zone. For a very short time, N_q reaches values as high as 1.0. These phonons of small q vectors do not couple efficiently in the absorption process. As a result, the change of α_{FC} is very small, i.e., less than 2%. The calculations demonstrate that screening is essential to account for the observed slow cooling and for the measured magnitude of the excess absorption.

In summary, we have demonstrated for the first time that the absorption of free electrons in polar semiconductors increases with the temperature of the hot electrons. The excess LO phonons generated by carrier cooling enhance the absorption process. Our investigations suggest that transient free-carrier absorption—measured on a time scale of picoseconds—offers a promising new tool to study hot phonons in semiconductors.

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