Interaction of an Electron-Hole Plasma with Optical Phonons in GaP

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The interaction of an optically induced nonstationary electron-hole plasma with coherently excited optical phonons in GaP has been investigated through time-resolved coherent anti-Stokes Raman spectroscopy. Nonexponential optical-phonon dephasing is observed in the presence of the electron-hole plasma. A model describing the interaction between the optical phonons and the electron-hole plasma is presented. A comparison of results from the theoretical model and from the experimental data indicates good agreement.

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Investigations in the frequency domain of plasma-phonon interactions in GaP have previously been performed for both a one-component electron plasma [1] and a two-component electron-hole plasma [2]. In the case of a one-component plasma, a stationary electron plasma can be easily maintained using an *n*-doped semiconductor held at a fixed ambient temperature. In the present case, an electron-hole plasma is formed in a pure crystal through two-photon absorption (TPA) of incident pulsed laser radiation. This method of photoexcitation may produce a plasma which is nonstationary during the experimental observation period. It is, therefore, appropriate to conduct time-resolved observations to provide information on the dynamics of the interaction between the two-component plasma and the phonons. Recently, time-resolved coherent anti-Stokes Raman scattering (TR-CARS) has been widely utilized to measure the dephasing rate of near-zone-center longitudinal optical (LO) phonons in GaP [3], and to determine its dependence on the ambient temperature. It has also been shown that the dynamics of the LO phonon dephasing changes in the presence of high concentrations of acoustic phonons [4]. However, the dynamics of the LO phonon dephasing in the presence of a two-component plasma has received only limited attention. An understanding of the plasma–LO-phonon interaction is particularly important when the intensity of the incident laser pulse, used to inject the two-component plasma, approaches the (bulk) damage threshold of the material.

We report here the results of an application of TR-CARS to study the temporal evolution of the interaction of a two-component plasma and LO phonons in GaP. The investigation provides, for the first time, direct observation of the transient dynamics of the plasma-phonon interaction. We observe an increase in the instantaneous dephasing rate of the LO phonons during the first 150 ps of the interaction. The dephasing rate increases as the incident laser irradiance increases (i.e., with increasing plasma density). Furthermore, we observe that the increase in the dephasing rate becomes negligible after 600 ps. These effects are in good agreement with the prediction of a theoretical analysis.

The phonons and two-component plasma are excited using a dual synchronously amplified picosecond laser system operating at 1 kHz [5] with outputs at the frequencies ω_l and ω_s ($\omega_l = 2.137$ eV and $\omega_s = 2.087$ eV). The general TR-CARS apparatus is shown in Fig. 3 of Ref. [3] with the exception of an additional beam used to inject the two-component plasma in the mutual interaction volume. The difference in output frequencies of the two laser beams ($\omega_l - \omega_s$) is resonantly tuned to the LO phonon frequency (50 meV) and a variably time delayed part of the ω_l beam is used to probe the dephasing of the LO phonons; note that the probe frequency $\omega_p = \omega_l$. An additional synchronized laser pulse with frequency ω_s is used to "inject" a nonstationary electron-hole plasma (NEHP) via two-photon absorption ($2\omega_s$) with an initial excess energy of 1.05 eV above the direct gap of 2.7 eV. The injection laser pulse arrives at the interaction volume at a fixed time after the excitation of the LO phonons. The NEHP density is varied by changing the incident irradiance of the injection laser pulse. The intensities of the ω_l, ω_s, and ω_p beams are kept sufficiently weak to avoid additional TPA and intense LO phonon generation that could cause nonexponential dephasing of the LO phonons [4]; thus pulse energies for phonon excitation and probing are kept below 1 nJ, whereas the injection pulse energies are varied from 5 to 500 nJ. The four beams are focused into a high purity ($< 10^{15}$ N impurities per cm^3) GaP crystal maintained at 5 K. The experiment is carried out by alternately measuring the TR-CARS signal with and without the presence of the NEHP at a set of fixed probe delay times in the interval between -50 and 200 ps, to minimize long-term laser fluctuations.

Figure 1 displays typical experimental results on the intensity of the TR-CARS signal as a function of the probe delay time for three values of the irradiance of the injection laser pulse. The upper curve, which represents the LO phonon dephasing without NEHP injection, clearly shows exponential dephasing. The lower two curves indicate LO phonon dephasing in the presence of the NEHP.

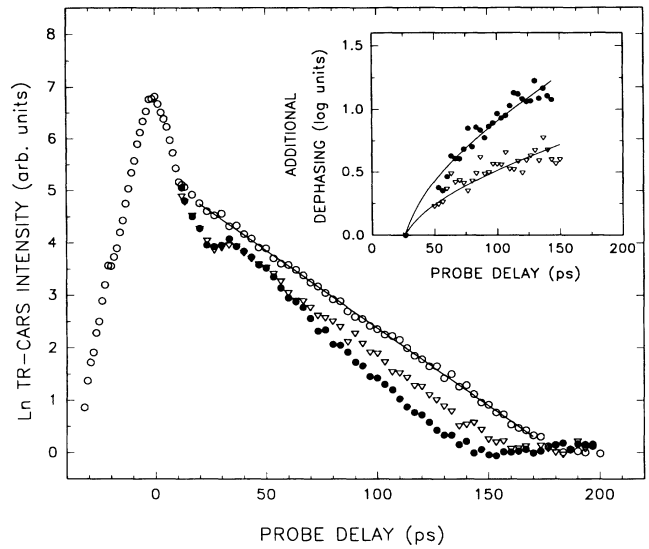

FIG. 1. Observed TR-CARS intensity as a function of the probe delay time for three different NEHP injection pulse irradiances: no injection (open circles), 4.1×10^3 MW/cm^2 (closed circles), and 2.4×10^3 MW/cm^2 (open triangles). Inset: The additional dephasing of the LO phonons as a function of probe delay time for two injection pulse irradiances. The solid curves are the result of fitting the theoretical model to the data to determine the plasma density n. The resulting values for the density are $n=2.1\times10^{17}$ cm^{-3} (closed circles) and $n=1.2\times10^{17}$ cm^{-3} (open triangles).

The dip at ~26 ps delay is consistent with the timing of the injection pulse, but its origin is not yet understood. An increase in the dephasing rate, due to the presence of the NEHP, follows the injection pulse. The inset of Fig. 1 illustrates the difference between the natural logarithm of the TR-CARS signal observed with the NEHP and that observed without the NEHP as a function of delay time, and indicates the additional dephasing resulting from the plasma-phonon interaction. The additional dephasing, depicted in the inset of Fig. 1, represents a time-dependent interaction between the LO phonons and the NEHP.

We now present a synopsis of a theoretical model which we apply to the experimental observations. A more detailed description of the theoretical model will be presented elsewhere. The interaction of the LO phonons and the NEHP can be described in terms of a coupled vibrational system consisting of the LO phonons and the electron-hole plasma interacting by way of a local polarization field. The equations of motion of such a system are given by

$$\ddot{W}+\omega_t^2W+\Gamma\dot{W}=(e^*/M)E\,, \quad (1)$$

$$m_e\ddot{X}_e=-(1/\tau_e)m_e\dot{X}_e-\eta(\dot{X}_e-\dot{X}_h)+eE\,, \quad (2)$$

$$m_h\ddot{X}_h=-(1/\tau_h)m_h\dot{X}_h-\eta(\dot{X}_h-\dot{X}_e)-eE\,, \quad (3)$$

$$E+4\pi\left[\frac{e^*}{M}E+\frac{e^*}{V_0}W+ne(X_e-X_h)\right]=0\,, \quad (4)$$

where W is the amplitude of the relative interatomic lattice displacement; ω_t is the TO phonon frequency; Γ is the spontaneous anharmonic phonon decay rate; E is the local electric field; M and e^* are the reduced mass and effective charge of Ga and P, respectively; $\dot{X}_e,\dot{X}_h$ and m_e,m_h are the drift velocities and effective masses of the electrons and holes, respectively; τ_e and τ_h are the momentum relaxation times of the electrons and holes due to scattering by acoustical phonons; and η is due to electron-hole Coulomb scattering.

In order to solve Eqs. (1)-(4) we adopt the following assumptions: (a) The plasma frequency $\omega_p\ll\omega_{\rm LO}$; this corresponds to a low plasma density under which the optical lattice vibrations are only slightly perturbed. The plasma density estimated in the present experiment is $n\sim10^{17}$ cm^{-3}, and thus $\omega_p\sim10^{13}$ s^{-1} and $\omega_{\rm LO}\sim10^{14}$ s^{-1}. (b) $1/\omega_{\rm LO}\ll\tau_{e\text{-}e},\tau_{e\text{-}h},\tau_{h\text{-}h}$; this indicates slow carrier-carrier scattering compared to the period of an LO phonon oscillation. For electrons and holes in GaP, $\tau_{e\text{-}e},\tau_{e\text{-}h},\tau_{h\text{-}h}\sim10^{-13}$ s^{-1} [6]. Under these conditions, the coupled system yields a solution for the temporal evolution of the coherent phonon intensity $|A(t)|^2$ in the presence of the plasma. The result can be expressed in the following form:

$$|A(t)|^2=|A(0)|^2\exp\left[-\Gamma t-\beta\int_0^t\gamma(t')dt'\right]\,, \quad (5)$$

where $A(0)$ is the coherent phonon amplitude at $t=0$, $\beta=(1-\varepsilon_\infty/\varepsilon_0)\omega_p/\omega_l$ is the coupling constant between the LO phonons and the NEHP, and $\gamma(t)$ is the time-dependent damping of the plasma oscillations. In the absence of a NEHP, the solution reduces to the standard exponential LO phonon dephasing solution with decay rate Γ [3]. See the upper curve of Fig. 1. In the presence of the NEHP, an additional dephasing term appears representing an increase in the total dephasing rate. The additional dephasing due to the plasma is represented by the results shown in the inset to Fig. 1.

In order to arrive at an expression for $\gamma(t)$, it is necessary to determine the temporal dependence of the carrier temperature. After an initial carrier cascade due to the Fröhlich interaction (< 2 ps) [6,7], the free carriers form a plasma at energies below that of the $k\approx0$ LO phonon and the phonons generated as a result of inter-X-valley electron scattering ("XX scattering"). Starting at this point of the cooling of the plasma, we have calculated further cooling as the carriers undergo additional LO phonon and XX phonon scattering assisted by carrier-carrier scattering, deformation potential (DA) scattering, and piezoelectric (PA) scattering from acoustical phonons [6]. The result of this calculation appears as the upper left inset to Fig. 2. It can be seen from Fig. 2 that the plasma cools from a starting temperature of approximately 150 K to a temperature of 30 K within the first 25 ps after injection. The initial rapid cooling is due to LO phonon emission and XX scattering. The cooling rate then decreases as DA and PA scattering become the

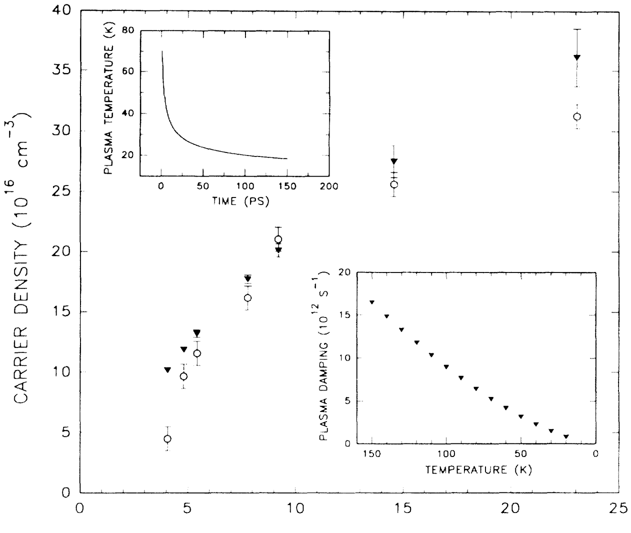

FIG. 2. The carrier density as a function of incident injection pulse irradiance as determined from the theoretical model (open circles) and that obtained by TPA (closed triangles). Upper left inset: The calculated plasma temperature as a function of time for a starting temperature of 150 K. Lower right inset: The damping of the plasma as a function of the plasma temperature between 150 and 20 K as calculated from Eq. (6).

dominant mechanisms of energy relaxation. In order to determine the plasma damping, it is necessary to consider only those carrier interactions which do not conserve current. Assuming a nearly parabolic energy dispersion for the electrons and neglecting contributions due to the anharmonicity of the hole band, the expression for the plasma damping reduces to

$$\gamma(t)=\frac{1}{\tau_{e\text{-}h}}=n\sigma v=n(4\pi r_D^2)\left(\frac{3k_BT_c}{m^*}\right)^{1/2}, \qquad (6)$$

where n is the plasma density, σ is the scattering cross section, v is the thermally controlled drift velocity of the carriers, $T_c(t)$ is the plasma temperature as given in the upper inset to Fig. 2, $r_D(\text{Å})=23(T_c/10)$ is the Debye screening radius at temperature T_c [6], k_B is the Boltzmann constant, and m^* is the reduced mass for electrons and holes. Substituting Eq. (6) into (5) and using the derived carrier temperature T_c from Fig. 2 yields an expression for the dependence of the additional phonon dephasing on the carrier density n. A best fit is then made to the additional dephasing using the theoretical expression to determine the carrier density n. The resulting additional dephasing is shown in Fig. 1. The one-parameter fit to the data indicates good agreement with the data and yields $n=2.1\times10^{17}$ and 1.2×10^{17} cm^{-3} for the illustrative case.

Whereas the temporal evolution of the additional dephasing indicates good agreement between the theoretical model and the data, it is necessary to determine the reliability of the fitted parameter, i.e., the carrier density n. An alternate evaluation of the carrier density n can be obtained through a calculation of the number of carriers generated through TPA in the mutual interaction volume of the three laser beams. Only carriers excited in the interaction volume are available for interaction, since transport of carriers out of the volume is negligible during the 150-ps interaction time. The sample thickness used in the experiment was 3 mm with all laser beams having a focal spot size of 30 μm. Under these conditions, the spatial variation in the profile of the beam between the crystal face and the interaction volume due to focusing is small and can be neglected. Thus, the beam can be considered to be collimated inside the crystal. The irradiance of a collimated laser pulse as a function of propagation distance through an absorbing medium has been given by Van Stryland *et al.* [8]. We apply this result to determine the energy absorbed in the interaction volume due to TPA which in turn leads to the number of generated carriers. The main part of Fig. 2 is a comparison between the carrier density obtained from fitting Eq. (5) and that calculated to arise from the TPA.

It is clear that we have observed additional phonon dephasing due to the interaction of lattice vibrations and plasma oscillations. The model is in good agreement with the observed dephasing of the LO phonons, which depends on the plasma density and plasma damping. Moreover, the plasma density as obtained from the model and that calculated independently from TPA are also in good agreement. Since ballistic transport at the thermal velocities does not result in significant carrier transport out of the interaction volume within the first 200 ps of the observation, we eliminate changes in the plasma density as a source of the change in the magnitude of the additional phonon dephasing. The plasma damping results from electron-hole scattering as described in Eqs. (5) and (6). The plasma oscillations are damped by any carrier-scattering event which does not conserve current. The assumption that electron-hole scattering dominates the damping appears valid for the conditions of the experiment, although the deviation seen in Fig. 2 may be due to effects of plasma damping due to hole-hole scattering in a nonparabolic and nonspherical hole band at higher densities. The temporal dependence of the magnitude of the scattering is due to the cooling of the plasma which decreases the plasma velocity and thus the scattering cross section [see Eq. (5)]. For the convenience of the reader we include, in the lower inset to Fig. 2, the temperature dependence of the plasma damping as obtained from Eq. (6) and the fitted values of the carrier density.

In order to investigate the effect of carrier transport, the NEHP is preinjected at times ranging from 0 to 600 ps before the LO phonon generation. Carriers which are present in the interaction volume at the time of LO phonon generation are reexcited by the pump pulses through single-photon free-carrier absorption and thus effectively act as carriers injected at $t=0$ ps. Carriers which have transported out of the volume no longer participate in the interaction causing the initial carrier density to decrease as the transport out of the region increases. The observed additional phonon dephasing is negligible once preinjection is greater than 600 ps, at which time, apparently, all carriers have left the interaction volume. Further analysis of this effect will be presented in a later publication. Kardontchik and Cohen have reported negligible damping of a two-component plasma in GaP as observed through incoherent Raman scattering with laser pulses of 2 ns duration and probe delays also of the order of several nanoseconds [2]. Thus their findings support our observation that the plasma-phonon interaction ceases for delay times greater than of the order of nanoseconds. Prior to this explanation of Kordontchik and Cohen's statement that the plasma was "undamped" [2], we had speculated that the damping of a one-component plasma [1] and that of a two-component plasma [2] is fundamentally different. However, a comparison of the present plasma damping rate and those reported earlier [1] on a one-component plasma indicate no appreciable difference. Comparing high purity samples with carrier concentrations of the order of 10^{17} cm^{-3}, we find from Figs. 1 and 6 of Ref. [1] that $\gamma \sim 100$ cm^{-1} ($\sim 9\times 10^{12}$ s^{-1}), and typical values from the inset of Fig. 2 of the present experiment are $\gamma \sim 10\times 10^{12}$ s^{-1} at $T\sim 100$ K.

In conclusion, we have observed, for the first time, an interaction of an optically induced two-component electron-hole plasma with LO phonons in GaP using the TR-CARS technique. A time-dependent increase in the dephasing rate of the LO phonons is observed to arise from the coupling of the vibrational modes of the plasma and those of the phonons. It is demonstrated, on experimental and theoretical grounds, that the increase of the dephasing rate depends on the density and on the damping of the plasma. The temporal dependence of the increased dephasing arises from a nonstationary plasma temperature, and the diffusion of carriers out of the excitation volume. Plasma damping is readily observable for temporal resolution of the order of picoseconds. The present experimental technique makes it possible to observe, directly in the time domain, the interaction of a plasma with optical phonons. Several of the details of the interaction are analytically recovered from the experiment.

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