# Direct observation of the strength of plasmon-longitudinal optical phonon interaction in *n*-type GaAs

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(Received 6 April 2006; accepted 22 June 2006; published online 3 August 2006)

The screening of longitudinal optical phonons by plasmons is investigated by time-resolved visible pump-mid-infrared probe transmission measurements in a series of light to highly doped *n*-type GaAs wafers. The reduced relaxation of photogenerated carriers is strongly correlated to the coupling between longitudinal optical phonons and background plasmons as suggested by the variation of the phonon strength over the doping range. The critical plasmon density at which the optical phonons were effectively screened, thereby reducing the carrier relaxation, was determined to be $N_c \sim 1\times10^{18}$ cm$^{-3}$. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2236300]

Ultrafast laser techniques provide a unique means of understanding carrier dynamics in semiconductors. For polar semiconductors, while near-absorption edge relaxation of the carriers is well understood, above gap dynamics can be far more complex due to intra- and intervalley scattering phenomena. The subsequent relaxation of hot carriers generated by photons having an energy in excess of the band gap energy ($h\nu > E_g$) to the band edge occurs as a result of carrier-carrier and carrier-lattice interactions,[1] primarily through polar optical mode scattering.[2] In GaAs, the interactions between carriers and lattice vibrations are strongest with the longitudinal optical modes[2,3] and lead to hybrid states between longitudinal optical (LO) phonons and plasmons. The coupled hybridization of the LO plasmon has two branches, either plasmon like ($\omega_{L-}$) or phonon like ($\omega_{L+}$) modes at low carrier densities,[4] and vice versa at high carrier densities after a doping density on the order of $N\sim1\times10^{18}$ cm$^{-3}$ and greater.[5] At this doping level, LO phonons are not a well defined elementary excitation in GaAs. The coupled mode[5] and GaAs band structure are shown in Fig. 1. By studying *n*-type GaAs samples of various doping at low photoexcitation densities, and observing the time evolution of above band gap generated hot carriers, the effect of coupling between background plasmons due to doping and longitudinal optical phonons can be resolved thereby allowing for the determination of the critical density at which screening effects on LO phonons become dominant.

In this letter, the slowing of carrier relaxation in doped *n*-type GaAs is investigated and attributed to the decrease in strength of the polar optical mode scattering with increasing doping density. The disappearance of the pure LO-phonon in highly doped *n*-type GaAs due to background plasmon screening gives rise to the slow relaxation of photogenerated carriers through coupling effects which can adversely affect the spectrum of diode lasers and the performance of semiconductor electronic devices.[6]

In order to probe the free carrier-LO lattice interaction, femtosecond pulses from the primary and frequency doubled components of a mode locked Er:fiber laser were used. The laser consists of a mode locked, Er:fiber oscillator with a pulse repetition frequency of 48 MHz. The output pulses from the oscillator are amplified in an erbium doped fiber amplifier, where the fundamental output centered at 1560 nm is frequency doubled. The frequency doubled component, at 780 nm (1.59 eV) with a pulse duration $\tau\sim120$ fs with an average power $P_{\text{avg}}\sim5$ mW was used to excite carriers to cause a small perturbation to the equilibrium density in the $\Gamma$ valley to a minimum energy of 130 meV above the band edge. The fundamental component, $\lambda\sim1560$ nm, $P_{\text{avg}}\sim5$ mW, was used to probe the change in free carrier absorption due to the photogenerated electrons relaxing to the bottom of the $\Gamma$ valley. The polarizations of the pump and probe beams were set orthogonal to each other to avoid coherent artifacts at zero delay time. The probing beam was passed through a variable optical delay line, focused to a 150 $\mu$m spot size at normal incidence to the sample and was subsequently detected upon transmission using a commercial germanium photodetector. The pump beam was mechanically chopped at 400 Hz and focused to a spot size of $\sim300$ $\mu$m at the sample surface. Filters were used to separate the wavelength region of interest in both arms.

Our samples were comprised of five *n*-type GaAs(100) commercial wafers with various doping concentrations ($N_d$) in order to span the extent of the LO phonon-plasmon coupling modes: $N_d=1\times10^{17}$, $6.6\times10^{17}$, $2.1\times10^{18}$, $3.0\times10^{18}$, and $4.1\times10^{18}$ Si/cm$^3$. Each sample was ground down on its backside and polished to achieve a nominal thickness of 350 $\mu$m. The transmitted probe beam intensity was measured with a power meter and determined to be the same for all samples.

The differential transmission ($-\Delta T$) vs delay time ($\tau$) profiles were measured for each wafer at room temperature and are displayed in Fig. 2. The salient features of these profiles are a fast rise time and a slow decay time. The measurements show that the change in transmission is slowed with increasing doping density. Above $6.6\times10^{17}$ cm$^{-3}$ doping the rise times dramatically increase. This is indicative of a slow relaxation of the photoexcited carriers before they recombine with holes in the valence band as indicated by the decreasing absorption level of the infrared probing beam. The change in the peak transmission level before and after the arrival of the visible pulse is similar for all samples of various doping levels, indicating that the intrinsic level of

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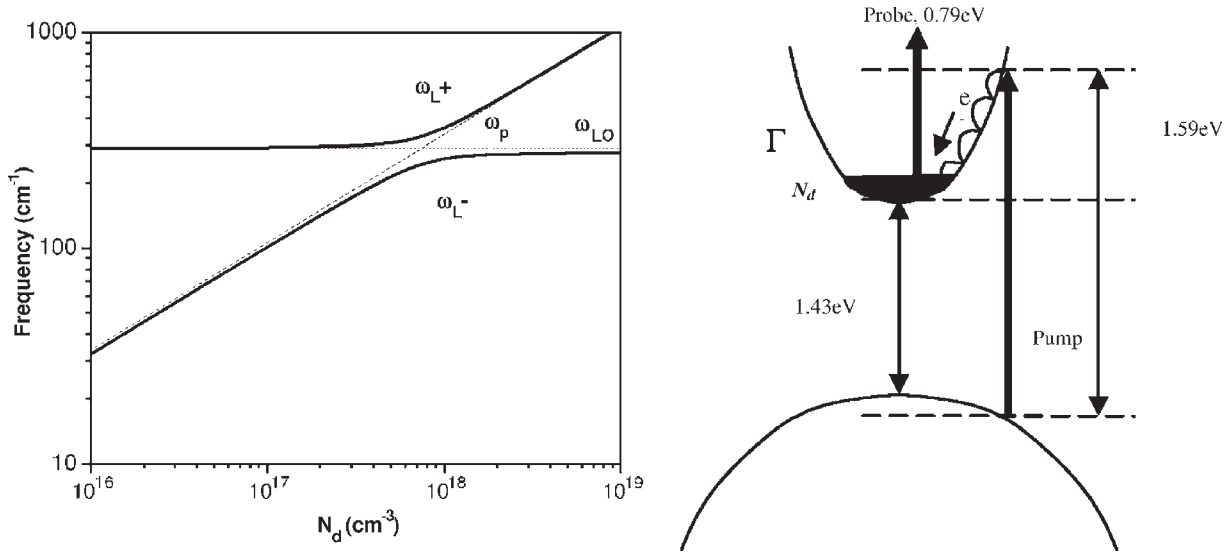


FIG. 1. Frequencies of the mixed longitudinal optical phonon-plasmon modes ($\omega_{L+}$ and $\omega_L$) for *n*-type GaAs as a function of the doping density ($N_d$) as given by Eq. (2), and the band structure for GaAs depicting the photoexcited carrier dynamics after the arrival of the pump and probe pulses.

doped carriers does not contribute to the absorption.

The rise times for each curve were fit to extract the rate at which the carrier relaxation slowed due to screening of LO phonons by background plasmons. The relaxation to the band edge due to LO-phonon interactions typically occurs on time scales less than one picosecond[2,7,8] as had been observed for undoped GaAs samples probed with ultrahigh intensity laser pulses, where coupling between the modes becomes important after a certain threshold in the photogenerated carrier density.[9] In *n*-type GaAs, the LO phonon-plasmon coupling modes have been previously examined using time-resolved Raman scattering[10,11] and time-resolved reflectivity measurements.[12–14] The dephasing time for coherent LO phonons is ~4 ps or less at room temperature for low excitation densities ($<1\times10^{18}$ cm$^{-3}$).[14,15] Because this time is longer than the LO phonon-plasmon thermalization time (~1 ps)[7,14] large nonequilibrium optical phonon populations can exist during carrier relaxation for excitation concentrations from $5\times10^{16}$ to $1\times10^{18}$ cm$^{-3}$, which can inhibit the energy relaxation of photogenerated carriers[16,17] Also, at high plasmon densities ($>6\times10^{17}$ cm$^{-3}$), screening effects can reduce the rate of relaxation of carriers in the Γ valley due to a reduction in the phonon emission rate.[18] Our time-resolved pump-probe measurements show that the rise times increase from ~300 fs to ~2.5 ps over the doping range of the samples. Specifically, we measure $\tau_{\text{rise}}=320$ fs±13 fs for $N_d=1\times10^{17}$ cm$^{-3}$, $\tau_{\text{rise}}=550$ fs±132 fs for $N_d=6.6\times10^{17}$ cm$^{-3}$, $\tau_{\text{rise}}=940$ fs±255 fs for $N_d=2.1\times10^{18}$ cm$^{-3}$, $\tau_{\text{rise}}=2.1$ ps±0.67 ps for $N_d=3.0\times10^{18}$ cm$^{-3}$, and $\tau_{\text{rise}}=2.7$ ps±1.14 ps for $N_d=4.1\times10^{18}$ cm$^{-3}$. Any carrier-carrier interactions present are dominated by the background plasmons (doping level), due to the low excitation densities that we photoexcite the samples with ($\sim1\times10^{14}$ cm$^{-3}$).

The rise times that were extracted from the various *n*-type GaAs samples are normalized to the unscreened lifetime of the $1\times10^{17}$ cm$^{-3}$ doped sample and are plotted versus the doping density in Fig. 3. Previously, efforts have been made to characterize electron scattering rates by strongly coupled LO phonon-plasmon modes in polar semiconductors within the random-phase approximation for the dielectric response.[19,20] It has been shown that at room temperature the energy loss rate due to the individual carrier motions becomes nearly equal to that due to the coupled mode at carrier densities[21] $\sim1\times10^{17}$ cm$^{-3}$. As the doping concentration increases, the plasmon mode couples to the LO phonon mode as the plasma frequency passes through the LO phonon frequency. The eigenfrequencies corresponding to the two branches of the coupled LO phonon-plasmon modes are given by the zeros of the total dielectric function. With increasing carrier density,[5] the $\omega_{L-}$ mode goes from being plasmon like to phonon like, and can dominate the relaxation time of the oscillations due to the longer dephasing time of

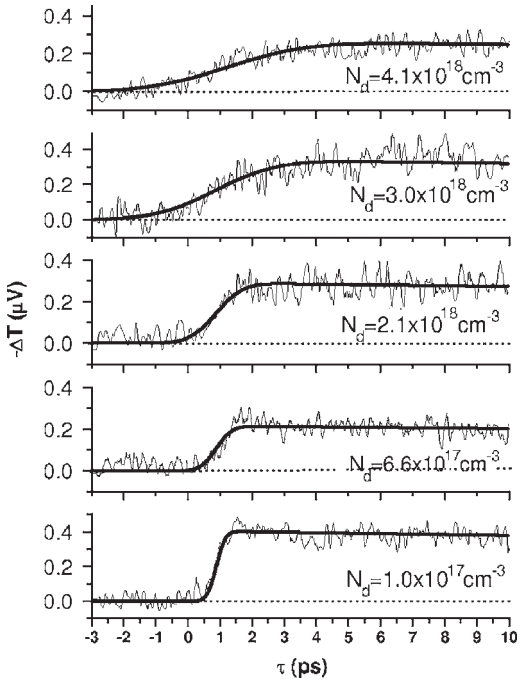


FIG. 2. Visible pump-mid-infrared probe differential transmission curves for various dopant density *n*-type GaAs wafers. The data were modeled with convolution of a Gaussian and exponential function of the form: $e^{-t/\tau_d}\cdot[1+\text{erf}(t/\tau_r-\tau_r/2\tau_d)]$ where $\tau_d=150$ ps (bold lines). The rise times increase with increasing dopant level. The pump wavelength is 780 nm (1.59 eV), the probe wavelength is 1560 nm (0.79 eV).

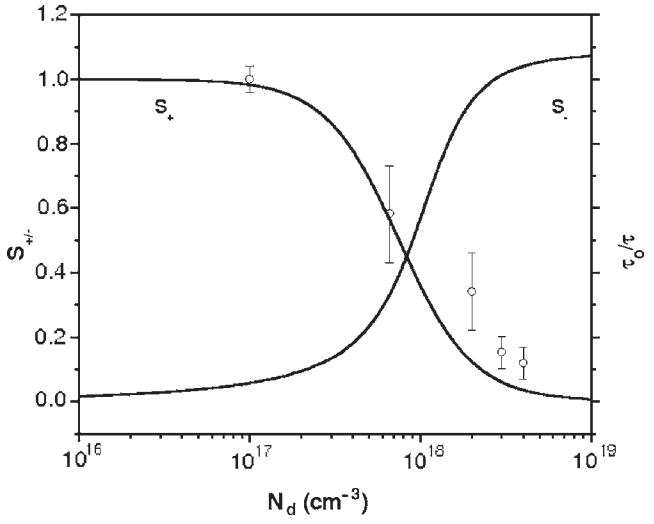


FIG. 3. Phonon strengths for both the $\omega_{L+}$ and $\omega_{L-}$ mode as given by Eq. (1) (solid line), and rise times extracted from Fig. 2 (data points) plotted with respect to dopant density for *n*-type GaAs. The rise times were normalized to the unscreened lifetime as extracted from the $1\times10^{17}$ cm$^{-3}$ doped *n*-type GaAs wafer, namely $\tau_0/\tau$ where $\tau_0=320$ fs for the $1\times10^{17}$ cm$^{-3}$ doped sample.

LO phonons (~4 ps).[12] At the same time the $\omega_{L+}$ mode goes from phonon like to plasmon like, where, due to carrier-carrier interactions, screening effects can also reduce relaxation rates (Fig. 1).[12,18] Both effects can be depicted by examining the relative phonon strength[3,22] in the two coupled modes:

$$S_{\pm}=\frac{\frac{\omega_l}{\omega_t}\left[\left(\frac{\omega_l}{\omega_t}\right)^2-1\right]\frac{\omega_{\pm}}{\omega_t}}{\left[\left(\frac{\omega_l}{\omega_t}\right)^2-1\right]\left(\frac{\omega_{\pm}}{\omega_t}\right)^4+\left(\frac{\omega_p}{\omega_t}\right)^2\left[1-\left(\frac{\omega_{\pm}}{\omega_t}\right)^2\right]^2}, \quad (1)$$

where, $\omega_p$ is the plasma frequency with $\varepsilon_\infty=11.3$ and $m^*=0.07m_0$, the LO phonon frequency $\omega_1=291$ cm$^{-1}$ and TO phonon frequency $\omega_t=268$ cm$^{-1}$ for GaAs. The coupled mode frequencies are given by

$$\left(\frac{\omega_{\pm}}{\omega_t}\right)^2=\frac{1}{2}\left\{\left(\frac{\omega_l}{\omega_t}\right)^2+\left(\frac{\omega_p}{\omega_t}\right)^2\pm\left[\left(\left(\frac{\omega_l}{\omega_t}\right)^2+\left(\frac{\omega_p}{\omega_t}\right)^2\right)^2-4\left(\frac{\omega_p}{\omega_t}\right)^2\right]^{1/2}\right\}. \quad (2)$$

In Fig. 3, the phonon strength [Eq. (1)] for both the $\omega_{L+}$ and $\omega_{L-}$ mode [Eq. (2)] along with rise times extracted from Fig. 2, are plotted with respect to the doping density ($N_d$). After a doping density of about $1\times10^{18}$ cm$^{-3}$ the phonon strength and plasmon strength for each mode interchanges. The long rise times (>2 ps) observed in the heavily doped samples is directly related to the dephasing time of the hybrid LO phonon-plasmon mode which has been shown to be dominated by the longer dephasing time of the LO phonon[12,14] for moderate excitation densities. At the same time, the increase in plasmon character of the upper branch of the LO phonon-plasmon mode with increasing carrier density can prolong the LO phonon dephasing rate,[12] which can result in a longer relaxation rate for the photoexcited carriers. The carrier level density for the interchange in strengths in the coupled modes agrees well with our observations for *n*-type GaAs where the rise times changed dramatically above the $6.6\times10^{17}$ cm$^{-3}$ doping level.

In conclusion, we investigated the screening of the LO phonons by plasmons in variously doped *n*-type GaAs wafers using time-resolved visible pump and mid-infrared probe transmission spectroscopy. Our results show that at low photogeneration ($<10^{14}$ cm$^{-3}$) the critical doping density at which the strength of the coupling between LO phonons and plasmons slow the carrier relaxation rates is on the order of $N_c\sim1\times10^{18}$ cm$^{-3}$. The slowing rate of the hot carriers with respect to increasing doping level, as extracted by our measurements, agrees well with calculated values for the phonon strength of the coupled modes due to polar optical phonon-plasmon coupling, where LO phonons do not significantly contribute to the energy relaxation of hot carriers above $N_c\sim1\times10^{18}$ cm$^{-3}$. The lack of LO phonons that participate in relaxation of carriers due to the hybridization of the longitudinal modes at high carrier concentrations, may result in adverse effects in semiconductor laser devices.

This work was supported under DOD No. W911NF-04-1-0023. The authors would like to acknowledge Dr. Wei Cai's helpful suggestions and Dr. Nathan Ockman's valuable review of the first draft of our paper.

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