Subpicosecond Time-Resolved Coherent-Phonon Oscillations in GaAs

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Coherent LO phonons are excited at the surface of bulk GaAs with femtosecond laser pulses. They are observed for the first time through electro-optic modulations of the transient reflectivity. The corresponding signal oscillations are superimposed upon an additional longitudinal polarization feature which decays exponentially and whose rise time is sufficiently short to act as a driving force for the coherent phonons. The components of the coherent phonon states dephase relative to each other in the order of picoseconds depending on the density of optically excited electron-hole pairs.

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A number of time-resolved transient reflectivity experiments with picosecond and femtosecond pulses have been applied to the study of carrier relaxation and transport in GaAs at excitation densities of 10^{18} cm^{-3} and above.[1-3] In this Letter we report on studies at densities below 10^{18} cm^{-3}, using 50-fs pulses from a ring dye laser and a novel detection scheme capable of resolving reflectivity changes of only $\Delta R/R_0 \sim 10^{-7}$. In addition to carrier-induced refractive-index changes, we observe electro-optic contributions due to depolarization effects introduced by the optically excited free carriers. This charge separation in surface space-charge fields was first observed by Dwayne Miller *et al.*[4] and can be exploited to generate freely propagating electromagnetic waves, as demonstrated by Zhang *et al.*[5] In this Letter we report on the observation of signal oscillations in the transient re- flectivity of GaAs as a result of coherent phonons generated by the ultrafast screening of the surface space-charge field via optical carrier injection.

Transient pump-probe reflectivity experiments, where reflectivity changes induced by an excitation pulse are interrogated by a strongly attenuated probe beam, are performed with 50-fs pulses at 2 eV. Both pulses are derived by splitting the output of a dispersion balanced ring dye laser[6] working at a 100-MHz repetition rate with an optical output of approximately 20 mW per beam. Pump and probe beams are focused noncollinearly onto the sample to a spot diameter of approximately 20 μm. The reflected probe signal and a part of the input probe signal are detected with large-area silicon pin photodiodes. The difference signal was fed directly into a specially developed computer comprising a high-speed data-acquisition unit and a 12-bit 4-MHz analog-to-digital converter.[7] The optical delay was accomplished using a retroreflector fitted onto a shaker working at a rate of up to 200 Hz. Signal averaging has been performed over 10^5 successive scans in a time interval of less than 10 min. This allowed us to detect signal changes $\Delta R/R_0$ of a few 10^{-7}, which is crucial for quantitative investigation of phonon-induced reflectivity changes ($\Delta R/R_0 \leq 10^{-5}$).

In Fig. 1(a) time-resolved reflectivity changes of bulk

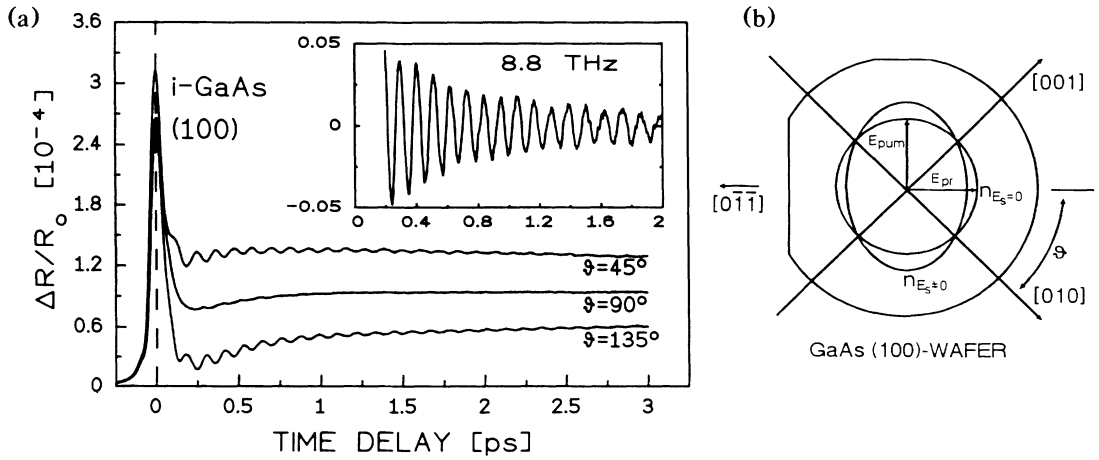

FIG. 1. (a) Time-resolved reflectivity changes of (100)-oriented intrinsic GaAs at an excitation density of $N = 10^{18}$ cm^{-3} for orthogonal polarization of pump and probe beam. (b) The parameter ϑ is the angle between probe polarization and [010] crystal axis. Also shown is the projection of the index ellipsoid on the surface of a (100)-oriented wafer in the case of zero and nonzero applied field in the [100] direction, e.g., perpendicular to the surface.

(100)-oriented semi-insulating GaAs are shown for three different angles ϑ between the probe beam polarization and the [010] crystal axis [Fig. 1(b)]. The orthogonally polarized beams are kept close to normal incidence. In the case of $\vartheta=90°$ the reflectivity rises during and peaks towards the end of the excitation pulse. After passing through a minimum at a time delay of 200 fs the reflectivity rises again to a quasistationary value on a picosecond time scale, without any periodic oscillations. The temporal evolution of the reflectivity transient is entirely due to susceptibility changes induced by the optically excited electronic carriers, their thermalization, and relaxation down to the band edges as well as intervalley transfer.[1,3] Rotating the sample to $\vartheta=45°$ [Fig. 1(b)] results in a shift of the entire reflectivity signature to higher values. The signal response for positive time delays shows a periodic modulation with an oscillation frequency of 8.8 ± 0.15 THz, exactly matching the frequency of the Γ_{15} LO phonon in GaAs.[8] In the case of $\vartheta=135°$ a complementary shift to lower reflectivity changes together with an additional phase shift of π in the phonon-induced oscillations is observed.

The distinct dependence of reflectivity modulation on probe beam polarization demonstrates the electro-optic nature of this effect, which is well known for cubic zinc-blende group materials with $\bar{4}3m$ crystal symmetry and has been widely used for "electro-optic sampling" in integrated circuits.[9] The longitudinal electric field, which accompanies the LO-phonon vibration, modulates the linear optical susceptibility through the first-order electro-optic effect. The corresponding distortion of the spherical index ellipsoid due to a longitudinal electric field E is shown in Fig. 1(b). The electro-optic refractive-index change Δn_{eo} is given by $\Delta n_{eo}=0.5n^3 r_{41}\Delta E$, with n the index of refraction, r_{41} the electro-optic coefficient, which for GaAs has a value[9] of 1.6×10^{-10} cm/V, and ΔE the longitudinal surface field changes. Neglecting changes in the absorption coefficient the reflectivity of a bulk sample at normal incidence is given by $R=(n-1)^2/(n+1)^2$. With $\Delta R=(\partial R/\partial n)\Delta n_{eo}$ the measured reflectivity changes thus provide a direct measure of the surface electric-field changes.

To gain more insight into the generation process of a coherent phonon mode we discuss the driving terms in the classical equation of motion for the coherent phonon amplitude $\langle Q\rangle$, given by[10]

$$\mu(\ddot{Q}+\Gamma\dot{Q}+\omega_{LO}^2 Q)=\hat{\mathbf{e}}_{LO}\left(\bar{R}\mathbf{E}_l\mathbf{E}_s-\frac{4\pi e^*}{\varepsilon_\infty}\mathbf{P}_{nl}\right), \quad (1)$$

where μ is the reduced lattice mass, Γ a phenomenological damping rate, ω_{LO} the phonon frequency, $\hat{\mathbf{e}}_{LO}$ the unit polarization vector of the LO phonon, ε_∞ the high-frequency dielectric constant, $\bar{R}$ the Raman tensor, e^* the effective lattice charge, and $\mathbf{P}_{nl}$ a nonlinear polarization. The first term of the driving force on the right-hand side is given by the vibronic Raman interaction, with $\omega_l-\omega_s=\omega_{LO}$. Since in our case ω_l and ω_s are contained within the same pulse of duration less than the phonon cycle, this term leads to an impulsive stimulated-Raman-scattering (ISRS) process.[11] As long as no free carriers are created, the nonlinear electronic polarization term $\mathbf{P}_{nl}$ is determined by the instantaneous nonlinear optical response functions $\chi^{(2)}$ and $\chi^{(3)}$. In the presence of electrons and holes, however, new nonlinear impulsive driving terms become operative such as diffusion in large carrier density gradients and charge separation in a surface space-charge field.[4] The depolarization field can also result from surface boundary conditions.[12] A direct time-resolved measurement of the longitudinal surface field resulting from the various polarization terms is accomplished via electro-optic sampling. To separate field contributions from carrier-induced refractive-index changes, the probe beam polarization, set to $\vartheta=90°$, is split into two orthogonally polarized components, sampling the reflectivity changes of the $[0\bar{1}1]$ and [011] directions, respectively. The difference $\Delta R_{eo}/\Delta R_0$ of these two signals is directly related to the time evolution of the electric-field change $E(t)$ via the electro-optic tensor element r_{41}.

In Fig. 2(a) the temporal evolution of the electro-optically induced reflectivity changes and of the related longitudinal electric field is displayed on a subpicosecond time scale. An impulsive initial field change of up to 40 kV/cm with an extrapolated rise time of 80 fs, much shorter than a single-phonon vibration period, is observed. This rise is already modulated by the phonon-induced oscillations towards the end of the pump pulse. The signal reaches a maximum value at 140 fs after the excitation and subsequently decays on a picosecond time scale. To study the exact nature of the nonlinear electric shock-wave launching the coherent phonons we have performed additional three-pulse experiments. 10 ps before the actual pump-probe experiment any surface space-charge field should be reduced by injection of free electron-hole pairs[13] with a femtosecond pulse derived

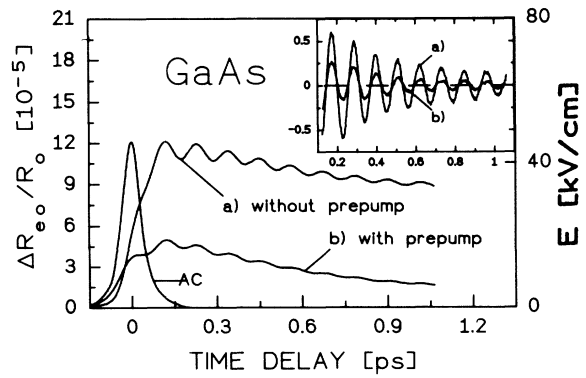

FIG. 2. Electro-optic signal changes at the GaAs surface at (a) an excitation density of approximately 10^{18} cm^{-3} and (b) with prepump excitation ($>10^{18}$ cm^{-3}) at a time delay of $\Delta t=-10$ ps. Inset: Oscillatory signal part.

from the second output of our colliding mode-locked (CPM) laser. As expected and demonstrated in Fig. 2(b) the magnitude of the electric-field transient is strongly reduced. However, the strict proportionality between initial maximum field change and phonon oscillation amplitude remains, as can be seen from the phonon-induced signal modulations shown in the inset of Fig. 2. This prepulse experiment confirms the surface space charge as the decisive source term. The initial field distribution is radically altered by the opposite drift of the optically injected electrons and holes. The accompanying depolarization field then acts as the nonlinear driving force for the coherent phonons. The dielectric relaxation in the picosecond time domain is enhanced in the presence of additional free carriers, indicating the restoration of the initial near-zero-field conditions on a much faster time scale.

Contributions of nonlinear $\chi^{(2)}$ and $\chi^{(3)}$ effects to the driving force of LO phonons can be ruled out by symmetry considerations. Distinct polarization configurations are necessary to couple the pump laser field to the nonvanishing components of the second- and third-order nonlinear susceptibility.[14] At fixed sample orientation and probe beam polarization, however, we observe the transient field and phonon oscillation signal to be totally independent of pump polarization within experimental reproducibility ($< 5\%$). Coherent phonons launched by ultrafast surface field transients have also been observed in femtosecond reflectivity experiments on GaAlAs and InP at our low CPM power levels. The isotropic excitation dependence of III-V compounds is in contrast to the distinct polarization sensitivity of impulsive stimulated Raman scattering observed in organic molecules.[11]

We have used the striking visualization of coherent phonons in time domain for a preliminary study of the dephasing time $T_2 = \Gamma^{-1}$ as a function of optical excitation density. At densities close to 10^{17} cm^{-3} we observe an initial dephasing time of 2.0 ps at room temperature. Increasing the density to 10^{18} cm^{-3}, the dephasing time decreases to 0.7 ps. It is interesting to note that our results on dephasing show a similar density dependence as in the work of Kash and Tsang.[15] There, the decay of nonequilibrium incoherent phonons is monitored in time-resolved Raman experiments at low temperature. Our present subpicosecond results of *dephasing times* at 300 K fit very well with their measured phonon *lifetimes* of 3.5 ps at 300 K and a carrier density of 2×10^{16} cm^{-3}. This close correlation of dephasing times and lifetimes of incoherent phonons demonstrate that, even in the presence of simultaneously photoexcited carriers, dephasing is primarily governed by phonon decay. However, we do observe deviations from the strictly monoexponential dephasing at higher densities, which cannot be correlated to a simple decay mechanism anymore. More-detailed investigations of the dephasing of coherent phonons as a function of excitation and *n*- and *p*-type doping density will be published in a forthcoming paper.

In conclusion, we have observed subpicosecond coherent LO-phonon oscillations in GaAs directly through modulations in the reflectivity signal via an electro-optic effect. They are driven by an initial electric "shock wave" associated with the charge separation of the photogenerated carriers in the presence of a strong surface space-charge field. Our results open the way to study directly in the time domain the interaction of coherent phonons with optically excited carriers and incoherent phonons created by the relaxation of hot carriers. In addition, a study of the initial electric-field transient as a function of excitation density and excess carrier energy will reveal details of ultrafast charge separation approaching the ballistic transport regime.

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