Coherent phonon-plasmon modes in GaAs:$Al_xGa_{1-x}As$ heterostructures

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Time-resolved transient Kerr reflectivity was used to study coherent phonon-plasmon excitations in GaAs:$Al_xGa_{1-x}As$ heterostructures containing a two-dimensional electron gas. Ultrafast laser pulses of 65 fs duration were used in a balanced optical bridge to observe the oscillatory component of the reflectivity changes directly. This was found to be strongly dependent on the excitation photon energy, showing a crossover in the spectral amplitude of the longitudinal and transverse-optic-phonon frequencies as the excitation energy was varied. The energy dependence reflects variations in the phonon-plasmon coupling with depth, and hence local electron density, which is in very good agreement with recent predictions. [S0163-1829(96)50324-3]

Coherent optic phonons can be created when an ultrafast laser pulse is incident on the surface of a semiconductor.[1] There are various methods of detection, including Raman scattering,[2] surface deformation measurement,[3] infrared absorption,[4] and bolometry,[5] but one of the most direct ways of studying the generation and dephasing of such a phonon population is the use of time-resolved reflectivity,[6–8] a comprehensive review of which is given by Kütt *et al.*[9] By this method, a high-intensity pump pulse of sub-100-fs duration excites the lattice via photogenerated carriers, and then a low-intensity probe pulse of similar duration is used with a variable time delay to measure the reflectivity response. Using these techniques, many observations have been made of coherent longitudinal-optic phonons in bulk semiconductors, and some studies of bulk semiconductors have seen beating between the longitudinal (LO) and transverse (TO)-optic phonon frequencies at high excitation densities,[9] which was ascribed to a phonon-plasmon mode. Recently, more evidence of phonon-plasmon coupling in multiple quantum wells has been reported,[10] where plasmon activation requires an injected photocarrier density.

In this paper we report time-resolved reflectivity observations on a modulation-doped semiconductor heterostructure containing a two-dimensional electron gas at room temperature, and show that by varying the excitation energy we can probe the phonon response of different regions of the layered structure. We find that in the flat-band regions no coherent optic phonons are created, while in regions of higher local field close to the two-dimensional electron gas, phonon-plasmon-coupled modes are activated, which are dependent on excitation energy. The energy variation probes regions of different local electron density, and the response agrees well with a recent model.[11] In this case, however, the plasmon oscillations are determined by the existing electron density, rather than the photocarrier density, and so the response depends upon the excitation energy rather than the intensity as in Refs. 9 and 10.

An incident laser pulse couples to the lattice principally by two mechanisms: direct Raman excitation of the lattice by the incoming field, and indirectly via the creation of photocarriers, which subsequently interact with the lattice polarization. In both cases, the use of a laser pulse which is significantly shorter than the phonon period ensures that the generated phonon distribution is initially coherent, and is usually composed of near-zone-center longitudinal-optic phonons.

When there is a significant carrier density created, the lattice Raman contribution is negligible, and the lattice response is determined solely by the action of the carrier population on the lattice polarization. Although subsequent cooling and thermalization of the hot-electron-hole photoplasma leads to further emission of *incoherent* optic and acoustic phonons these are not the concern of the present work. The ultrafast *coherent* phonon generation and dephasing processes response are dramatically affected by the local static field distribution, as seen in previous studies which employed a surface gate to increase the induced phonon amplitude by increasing the surface field.[9] The local field separates the *e*-*h* pairs, creating a local dipole polarization which interacts with the lattice polarization, generating the phonons.

At the same time as the optic-phonon generation, the polarization will generate plasmon excitations, which can themselves couple to the phonon modes. These coupled excitations have been observed in Raman scattering experiments which are not time resolved, and hence do not give any information about the coherence of the system. The phonon-plasmon excitation coherence has been predicted to be very sensitive to density fluctuations such as those due to dopant distributions in lightly doped semiconductors, and excitation field inhomogeneities due to the lateral intensity profile of a focussed laser spot. This has been suggested as one reason for the previous lack of direct observation of these modes in time-resolved reflectivity experiments.[11,12]

A recent model has demonstrated the relationship of phonon-plasmon coupling to photocarrier density in GaAs, and shown that if the effect of a Gaussian beam intensity profile is taken into account, then the Kerr reflectivity response should shift from predominantly at the LO frequency (unscreened at low density) to mostly the TO (screened LO) frequency, with strong beating between the two. These reflect the density-independent extrema of the coupled-mode excitation spectrum, and represent the modes observable by Kerr reflectivity. Other intermediate-density modes are excited, but their Kerr responses average out. Experimental studies of bulk material have seen TO-LO beating at very high excitation densities,[9] and a recent work on multiple

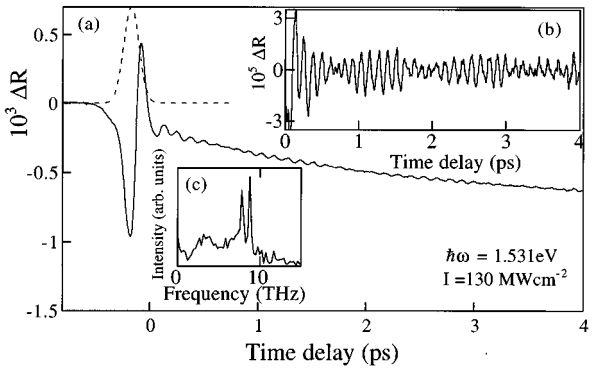

FIG. 1. (a) Transient reflectivity measured through the Kerr rotation, ΔR, for a heterostructure containing a 2DEG at 300 K using a photon energy of 1.531 eV. The pump-and-probe pulses are cross polarized to null out the large artifact at zero delay. (b) Oscillatory phonon component extracted from (a) exhibits strong beating. (c) Fourier transform of the oscillatory component showing peaks at the LO- and TO-phonon frequencies of bulk GaAs.

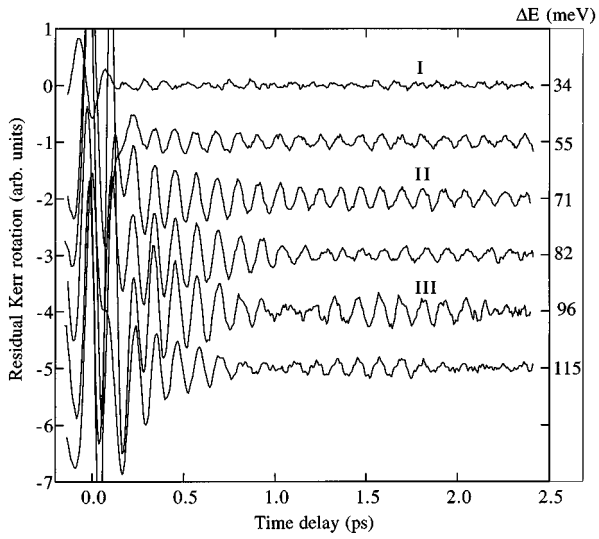

FIG. 2. The dependence of the oscillatory component of the Kerr rotation on the pump pulse excitation energy. The excess energy, ΔE, is measured relative to the bulk GaAs band gap at room temperature. The laser bandwidth FWHM is 29 meV. The labels *I*, *II*, and *III* refer to the excitation regions labeled in Fig. 3.

quantum wells of various widths has shown the predicted density-dependent TO-LO beating[10] and some evidence of the LO to TO frequency shift, in good agreement with the model.

The phonon-plasmon modes satisfy the equations[11]

$$\ddot{P}+\gamma_{\rm el}\dot{P}+\omega_{\rm pl}^2 P=\frac{e^2N(r)}{\epsilon_\infty\mu}[E^{\rm ext}-4\pi\gamma_{12}Q],$$

$$\ddot{Q}+\gamma_{\rm ph}\dot{Q}+\omega_{\rm LO}^2 Q=\frac{\gamma_{12}}{\epsilon_\infty}[E^{\rm ext}-4\pi P],$$

where P is the electronic polarization, Q is the normalized lattice displacement, $\omega_{\rm pl,LO}$ are the plasmon and phonon energies, $E^{\rm ext}$ is the driving field, and $\gamma_{12}=\omega_{\rm TO}\sqrt{(\epsilon_0-\epsilon_\infty)/4\pi}$ is the electric-field–phonon coupling.

In the experiments reported here, the semiconductor material used was a standard modulation-doped GaAs:$Al_{0.33}Ga_{0.67}As$ heterostructure, with a two-dimensional electron gas of sheet density 5×10^{11} cm^{-2} approximately 80 nm below the surface, and the measurements were made at room temperature. Similar results were seen in other samples of comparable electron density and mobility. Laser pulses of approximately 65 fs duration were generated in a Ti:sapphire system, and used in a high-intensity pump, low-intensity probe arrangement. Optic phonons have associated piezoelectric fields which alter the refractive indices in the $\langle 110\rangle$ directions, and hence the reflection coefficients in these directions. Thus the phonons will tend to rotate the polarization of the incoming [100] oriented probe pulse (a dynamic Kerr rotation). The difference signal between the reflectivities in the [110] and [$\bar{1}$10] directions is measured in a balanced optical bridge which gives a sensitivity of the order 10^{-5}.

Figure 1 shows a typical difference reflectivity response. There is an initial coherence spike, caused by the overlap of pump and probe pulses, followed by a gradual change in reflectivity over a few picoseconds. The duration of the pump pulse is shown dashed at time zero. Superimposed on the gradual change is the oscillatory component due to the phonons, the Fourier transform of which is shown in the inset. The gradual background change in difference reflectivity is due to the long-term screening of the surface depletion field by diffusion of the photocarriers and associated re-equilibration of the two-dimensional electron gas (2DEG). The oscillation frequencies in all cases studied here were found to be power independent, demonstrating that the injected carrier density does not strongly influence the phonon generation, which is largely determined by the doped carrier distribution.

Figure 2 shows the dependence of the oscillatory component on the excitation energy. At low energies (but above the GaAs band gap), there are no oscillations. At slightly higher energy, oscillations are seen with a single frequency corresponding to the LO bare energy. At higher energies still, the oscillations show strong beating between the LO and TO frequencies, as also shown in Fig. 1. This energy dependence can be explained with reference to the band structure of this material, shown schematically in Fig. 3.

At low energies, excitation is predominantly in the GaAs region towards the substrate, which is approximately flat band, shown as *I* in the figure. Although photocarriers are generated, there is no local field to create a polarization by separating the *e*-*h* pairs, and hence no creation of coherent phonons. This convincingly illustrates the advantage of time-resolved techniques over spectral methods, such as Raman spectroscopy, that unavoidably probe a much larger sample volume including the GaAs substrate. As the energy is increased, excitation occurs near the two-dimensional electron gas towards the GaAs:$Al_xGa_{1-x}As$ interface (region *II*), where there is a local field which mediates excitation of LO phonons. At higher energies, excitation takes place with average position closer to the interface, but in a region where there is sufficient local field to create a electronic polarization (*III*). The photoexcited electrons move towards the

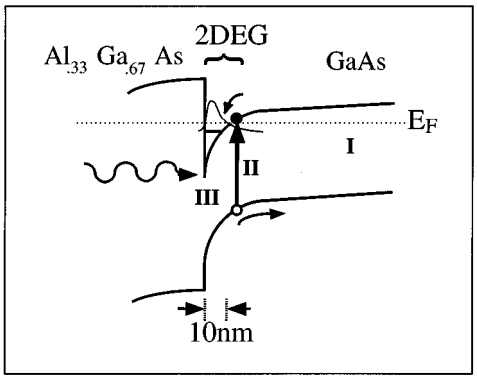

FIG. 3. A schematic band diagram of the heterostructure. *I* is the region with lowest above-gap excitation energy, in a nearly flat-band condition. *II* is the next highest energy excitation region—injecting electrons just above the Fermi energy in the tail of the two-dimensional electron gas. *III* Higher energy excitation into states which are close to the interface and near the peak electron density of the confined electron gas.

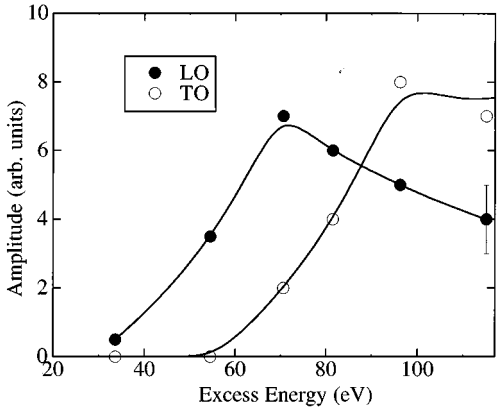

FIG. 4. The evolution of the LO and TO amplitudes with excess excitation energy measured relative to the bulk GaAs bandgap.

2DEG, while the holes move away, and the result is strong optic-phonon and plasmon generation in regions *II* and *III*.

The relatively high local density of the electron gas screens lateral inhomogeneities, but is sufficiently localized in the direction perpendicular to its plane that the local fields remain effective at carrier separation and thus vertical phonon and plasmon excitation. A plot of the evolution of the spectral amplitudes of the LO and TO components with energy is shown in Fig. 4. Both components initially rise with increasing excitation energy, reach a maximum and fall, with the LO frequency peaking at lower excitation energy than the TO. The rise and fall of the LO component, and rise of the TO, are superficially very similar to the recent predictions by Kuznetsov and Stanton[11] of the spectral dependence as a function of density, showing the predominance of LO-phonon excitation at low local electron density changing to a plasmon-coupled mixed mode, with the TO (screened LO) energy dominating at higher density.

The similarity of spectral development in these observations as a function of energy, and the predictions of Kuznetsov and Stanton of the development as a function of density, is due to the link between excitation depth, energy and local electron density in these samples. As can be seen in Fig. 3, as the energy is increased, the excitation position moves towards the interface because the photon energy now becomes sufficient to access unoccupied electron states at these shallower depths. These photoinjected electrons experience regions of higher local electron density and larger static fields. Over the same energy range, the optical penetration depth beyond the interface, the total injected carrier density, and the probe detection sensitivity change relatively slowly. In these experiments, the subbands concerned are already occupied, and so the injected density plays little part in the observed oscillations, other than determining the size of the initial impulsive dipole stimulus, in conjunction with the local static field. As noted by Kuznetsov and Stanton,[11] the reflectivity change measures the lattice displacement, and the observation of oscillations requires that these be coherent. We similarly infer that the phonon-plasmon modes excited in these heterostructures are initially coherent.

Although in this case the beam profile at the focus leads to no concomitant spread in electron density (the photon energy is spatially homogeneous), the use of a sub-100-fs pulse results in a transform-limited spread of photon energies and penetration depths, and hence a similar averaging over density as predicted by Kuznetsov and Stanton,[11] and observed by Dekorsy *et al.*[10] The spread in this case is also approximately Gaussian, but this must be convoluted with the density distribution in the two-dimensional electron gas to estimate the detailed response in the manner of Kuznetsov and Stanton.[11] The falloff of the TO amplitude at higher excitation energy was not predicted, but is probably due to the decrease in electron density near the GaAs:$Al_xGa_{1-x}As$ interface, which is the region being accessed at the higher energies, as can be seen in Fig. 3.

The dephasing processes of coherent optic-phonon populations have been the subject of much recent discussion. The principal decay route is believed to be via one longitudinal-optic and one longitudinal-acoustic phonon.[2,13] The measured dephasing times of the phonon-plasmon modes reported here suggest that the dephasing process follows a similar mechanism, although a measurement of the temperature dependence is needed to confirm this. The measured time (>4 ps) is slightly longer than the bulk response, which is in agreement with Ref. 10, suggesting that the heterostructure or the plasmon coupling may affect the decay process. The more likely case is believed to be that the plasmon coupling is stabilizing the mode, increasing the decay time.

In conclusion, we have shown that coherent coupled phonon-plasmon modes are observable at room temperature in a modulation-doped semiconductor heterostructure, where different phonon generation depths can be accessed by using different laser wavelengths. The observations that the phonon generation processes can be tailored by the local-field distribution on a nanometer scale opens the possibility of engineering the phonon response of an ultrafast optoelectronic system by a combination of adjustment of the excitation energy and design of the nanostructure, continuing the possibilities of phonon engineering.[6,14,15]

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