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Role of interface optical phonons in cooling hot carriers in GaAs-AlAs quantum wells

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We present an experimental study of the nonequilibrium phonon population generated by the relaxation of hot carriers photoexcited in GaAs-AlAs quantum wells. The optical phonon population is measured by anti-Stokes Raman scattering performed in the plane of the layers. The measured occupation numbers suggest that interface phonon modes contribute significantly to the relaxation dynamics over the lifetime of an electron-hole pair. These results are explained on the basis of the interplay between emission and reabsorption of hot phonons, which leads to a large population of interface phonons under steady-state conditions.

The interaction of electrons and optical phonons plays a central role in our understanding of the transport and optical properties of semiconductors. In a system with reduced dimensionality, the electron-phonon interaction has to be modified substantially to account for the quantization of electron motion and the anisotropic nature of the phonon propagation.[1] The presence of interfaces in semiconductor superlattices or single quantum wells gives rise to a similar confinement of optical phonons when the optical frequencies in both materials are different, which is the case for GaAs-AlAs heterostructures.[2] Additional modes have also been found at frequencies between the transverse- and longitudinal-phonon frequencies of the host materials and described as interface modes.[3] The crucial contribution of the interface phonons to electron-phonon scattering rates was initially underestimated owing to the misleading perception that they were strongly localized at the heterointerface. For the values of in-plane momentum relevant to coupling with carriers, the electrostatic potential of interface modes is largely delocalized over the whole heterostructure and thus can make a sizable contribution.[4,5]

Many recent optical studies have concentrated on the early phase of the dynamics of a photoexcited electron-hole plasma taking advantage of the newly developed femtosecond laser sources.[6,7] A direct study of nonequilibrium phonons generated during the latter stage of carrier relaxation has mostly escaped any experimental investigation in two-dimensional systems. Extensive studies on the generation of nonequilibrium optical phonon population have also been performed by subpicosecond Raman scattering in bulk GaAs.[8–10]

In this study, we investigate the nonequilibrium optical phonon populations in multiple-quantum-well structures (MQW's) by performing anti-Stokes Raman scattering under continuous photoexcitation. We find a strong contribution of both AlAs- and GaAs-like interface modes to the nonequilibrium phonon population. Surprisingly, we also observe a large population of AlAs-confined optical phonon modes. Our measurements demonstrate conclusively that a quasiequilibrium is not reached during the lifetime of the photoexcited electron-hole plasma and that a Planck distribution function cannot describe the occupation of the optical phonon modes.

We have studied two undoped GaAs-AlAs MQW's grown by molecular-beam epitaxy on a [001]-oriented semi-insulating GaAs substrate. Each structure consists of 60 periods of a double quantum-well unit with nominal thicknesses 100 and 50 Å. The width of the AlAs layer separating the two wells is either 15 or 50 Å. Each unit is isolated from the previous one by a 100-Å AlAs layer. The sample is mounted on the cold finger of a helium cryostat and kept at 10 K. Raman-scattering experiments are performed by focusing the laser light through a microscope objective on the cleaved edge of the sample. In this scattering geometry, we can record spectra for both polarized $x'(y',y')\bar{x}'$ and depolarized $x'(z,y')\bar{x}'$ inelastic light scattering, where z is defined by the growth direction and x' is the crystalline axis [110]. Further experimental details have been described elsewhere.[11]

In Fig. 1 we display a set of typical resonant Raman spectra for Stokes phonons. The laser energy is tuned in resonance with optical transitions between a split-off valence subband and a conduction subband at which a strong enhancement of the intraband Fröhlich-mediated

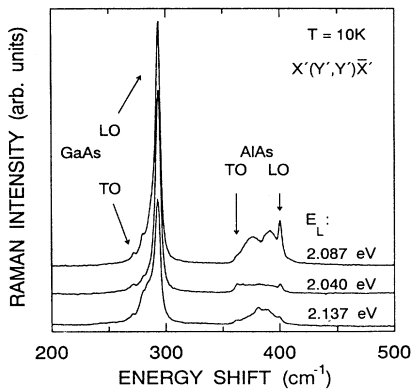

FIG. 1. Polarized Raman-scattering spectra of MQW's with 15-Å barrier. Broader features on the low-energy side of confined LO modes are identified with the interface modes of the structure. Incident photon energies at 2.087 and 2.137 eV correspond to an outgoing resonance for AlAs-confined LO phonons associated with the second and third electronic subbands of the structure, respectively.

scattering takes place. At frequencies 272 (362) and 294 cm^{-1} (400), one recognizes the optical phonon peaks of transverse and longitudinal modes of GaAs (AlAs), respectively. One should note that the quantization energies are so small that it is not possible to resolve the different confined optical modes. Within the limits set by those frequencies, the broad features are identified as the interface optical phonon modes[3] arising from the four different AlAs-GaAs interfaces. These spectra illustrate the sensitivity of the spectral profile of interface modes on the particular resonance frequency. A more complete description of the resonant behavior of interface modes and its relation to the symmetry of the resonant level is beyond the scope of this paper.

In Fig. 2 we present two Raman spectra of anti-Stokes phonons taken with different excitation energies. The main spectral features are related to the previous Stokes spectra: dominant in all the spectra is the LO phonon peak at 294 cm^{-1}; other less prominent peaks, but nevertheless important modes, are the AlAs- and GaAs-like interface modes and a phonon peak at the frequency of the AlAs-confined LO phonon. At a lattice temperature of 10 K, the thermal occupation of all optical modes is infinitesimal and, thus, the observed signal originates from a nonthermal occupation. The measured anti-Stokes Raman spectra represent an average over time of the optical phonons generated during the thermalization of the photoexcited electron-hole plasma. It is important to point out that only a subset of those modes is observable as conservation of energy and momentum in a Raman-scattering process restrict the accessible values in momentum space. The spectra, although they were taken under similar excitation conditions, differ qualitatively in the spectral region pertaining to the AlAs-phonon frequencies (see inset of Fig. 2). The spectra in Fig. 2 clearly demonstrate the presence of nonequilibrium phonons at a frequency corresponding to confined optical phonons in the AlAs barrier separating the two wells. Owing to its larger confinement energy, the first even-order-confined mode in the 15-Å barrier is found at 400 cm^{-1} whereas the same mode in the 100-Å AlAs layers occurs at 405 cm^{-1}. One also notes the spectral shift and broadening of the feature associated with AlAs-like interface modes. These differences are significant and reveal an essential aspect of a resonant Raman process: *Phonon modes are enhanced selectively depending on the specific nature of the resonant electronic transition.*

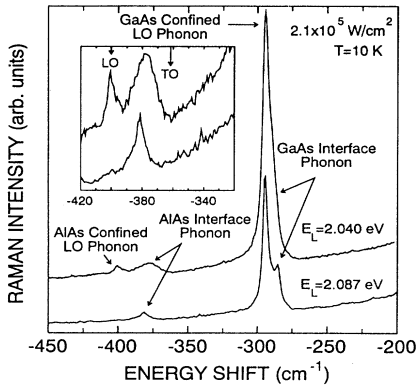

FIG. 2. Anti-Stokes Raman spectra obtained at a lattice temperature of 10 K for two excitation energies E_L. Inset shows an expanded view of both spectra for AlAs-like optical phonon modes. The spectra at these excitation energies correspond to incoming resonances of the Raman-scattering cross section.

Before we proceed to a quantitative analysis of the nonequilibrium phonon modes observed in the anti-Stokes spectra, the eventual influence of a local lattice heating due to the focused laser beam ought to be carefully considered. This effect is disregarded for the following reasons. Any significant lattice heating would lower the confined optical phonon frequencies. As the laser power density was decreased by more than an order of magnitude (from 4×10^5 to 1×10^4 W/cm^2), the Stokes Raman spectra remained unchanged implying an upper limit of 35 K to a local lattice heating. We also measured luminescence spectra under experimental conditions identical to those of the Raman measurements. The spectra are displayed in Fig. 3 for two values of the laser power density. At the lowest power density ($P_0/20$), the luminescence spectrum of the narrow well is essentially that of an excitonic gas. At a power density P_0, a fit to the high-energy tail of the luminescence yields a value of 120 ± 10 K. This value represents the average temperature of the photoexcited electron-hole plasma. Finally, the absence of a measurable population of TO phonons in the anti-Stokes spectra strongly argues against a local lattice heating, which is estimated to be less than 60 K with our signal over noise ratio (note that along [110] TO modes are Raman active in backscattering geometry).

The data in Fig. 2 provide direct experimental evidence for the presence of hot phonons at an energy corresponding to confined optical phonons in the 15-Å AlAs barrier. Relaxation of carriers across this potential barrier necessarily involves an inelasting scattering process mediated by LO-phonon emission or interface roughness. Because of the large penetration of the electronic wave functions inside the barrier, phonon-assisted tunneling of electrons across the barrier is also expected to occur via the emission of a confined optical phonon in the barrier. Consequently, we attribute the origin of hot phonons at 400

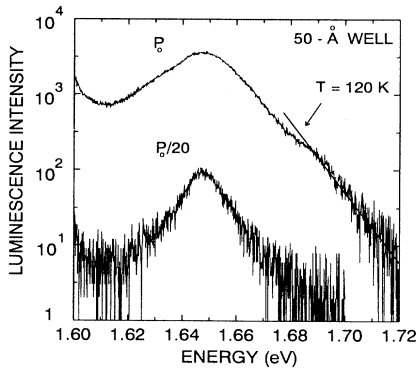

FIG. 3. Luminescence spectra taken under the same conditions as previous Raman spectra. Power density $P_0=2.1\times10^5$ W/cm^2 is identical to that of the Raman measurements.

cm^{-1} to phonon-assisted tunneling across the barrier. This interpretation is corroborated by the absence of any AlAs-like optical phonon modes in the anti-Stokes Raman spectra of the second MQW structure whose barrier width is 50 Å. This wide AlAs barrier effectively inhibits optical phonon-assisted tunneling processes. We also point out that optical phonon-assisted tunneling has already been observed by subpicosecond time-resolved luminescence spectroscopy in a similar structure.[12] The nature of the optical phonons associated with the tunneling process was not clearly identified but the data suggested that AlAs-like optical modes were also contributing in addition to GaAs-confined optical modes.[13]

In order to compare the coupling strength of interface and confined optical phonon modes to the electron gas, one has to take account of their relative Raman-scattering efficiencies. Away from any optical resonance, it is well established that the ratio of the Stokes and anti-Stokes scattering cross sections is proportional to the ratio $n(\omega)+1$ over $n(\omega)$ where $n(\omega)$ is the Bose-Einstein factor.[14] Close to a resonance, this simple relation is no more valid owing to the laser frequency dependence of the Raman-scattering cross section. The following expression is then substituted to it:

$$I_S(\omega_{\rm inc}=\omega_I,\omega_S=\omega_I-\Omega_P) = [(\omega_I-\Omega_P)^4/\omega_I^4]\{[n_S(\Omega_P)+1)]/n_{\rm AS}(\Omega_P)\} \times I_{\rm AS}(\omega_{\rm inc}=\omega_I-\Omega_P,\omega_S=\omega_I)\ , \quad (1)$$

where $I_S(\omega_{\rm inc},\omega_S)$ is the Raman-scattering intensity for an incident photon of frequency $\omega_{\rm inc}$ and scattered photon of frequency ω_S [the subscript S (AS) refers to a Stokes (anti-Stokes) scattering process] and $n_S(\Omega_P)$ is the occupation number for a phonon mode Ω_P. In order to derive this expression, one follows the same approach given in Ref. 14 and assumes that the anti-Stokes Raman-scattering cross section remains proportional to the occupation number of a phonon mode driven out of equilibrium with the lattice. The validity of this expression was tested by raising the lattice temperature (220 K) in order to create a thermal population of optical phonon modes. The laser frequency was shifted accordingly to resonate with the same optical transition. In Fig. 4 we have superimposed the anti-Stokes Raman spectrum and the corresponding Stokes spectrum corrected by the prefactor in Eq. (1). For this comparison, it is crucial to record the Stokes spectrum at a photon frequency upshifted by the phonon frequency. A good agreement over the entire spectral region corresponding to AlAs-like phonon modes is obtained assuming that the temperature of the phonon distribution is identical to the lattice temperature. The Raman spectra of Fig. 4 also reveal that in our backscattering geometry TO phonon modes can be observed and are populated according to a Planck distribution with the same temperature. The thermal phonon population of Fig. 4 is also much broader than the distribution of Fig. 2: this comparison emphasizes further the different nature of both distributions.

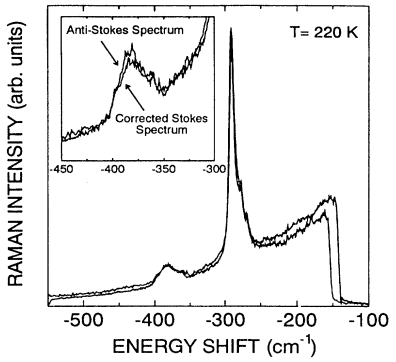

FIG. 4. Comparison between anti-Stokes ($h\omega_L=2.035$ eV) and corrected Stokes ($h\omega_L=2.082$ eV) spectra taken at a lattice temperature of 220 K. Inset shows an expanded view for energy shifts pertaining to AlAs-like interface modes.

A determination of the phonon occupation number is thus fully justified experimentally. Since anti-Stokes phonon modes exhibit a stronger incoming resonance, Eq. (1) was only applied in this case. In Table I, the derived phonon occupation numbers are given for each phonon mode observed in Fig. 2. The occupation number of AlAs-like optical phonons is, however, smaller than that of GaAs-like phonons and depends on the specific frequency of the interface phonon mode. For a mode with identical phonon frequency, we find that the occupation number is independent of the laser wavelength. These results clearly demonstrate that interface phonon modes reach an occupation number comparable to that of the GaAs-confined optical phonon modes during the cooling phase of the photoexcited electron-hole pairs.

We will now present a qualitative description of our results. Under our excitation conditions, the electron-hole pairs are directly generated into the quantum wells. The subsequent relaxation of those carriers will thus predominantly involve the coupling to optical phonon modes with the frequencies of GaAs vibrations. The initial relaxation of the photoexcited carriers is dominated by the

TABLE I. Occupation number of optical phonon modes for the resonant excitation energies of Fig. 2. Error bars are estimated on the basis of the deviation from the mean value of several data sets. Phonon energies are given in parentheses.

Excitation energy (eV)	GaAs-LO confined	AlAs-LO confined	GaAs-like interface	AlAs-like interface
2.040	0.037±0.002 (294 cm^{-1})	0.011±0.0015 (400 cm^{-1})	0.043±0.002 (285 cm^{-1})	0.011±0.001 (378 cm^{-1})
2.087	0.038±0.002 (294 cm^{-1})		0.043±0.002 (285 cm^{-1})	0.021±0.002 (381 cm^{-1})

polar-optical phonon interaction with emission of LO phonons in the case of electrons. For holes, the optical deformation potential is an additional scattering mechanism which will drive transverse- and longitudinal-phonon modes out of equilibrium. The absence of transverse optical phonons, even though Raman active, has an intrinsic origin. First, TO phonon modes have a much weaker scattering cross section than LO confined optical phonon modes at the energy gap corresponding to the split-off valence band. Second, the TO phonon modes emitted when scattering with a heavy hole have a much larger in-plane momentum (2×10^7 cm^{-1}) than allowed by momentum conservation and therefore cannot contribute to the anti-Stokes Raman spectra. *Hot phonon effects,* such as those described by Pötz and Kocevar[15] for picosecond pulse excitation, are also expected to play a role under our experimental conditions. Under continuous excitation, one expects that a stationary state establishes itself and therefore that all optical phonon modes reach an internal equilibrium with the electron-hole plasma since the modes more strongly coupled to the electron-hole gas are also more effectively reabsorbed. This interplay between emission and reabsorption leads to an enhanced effectiveness of more weakly coupled modes and eventually results in a heated Planck distribution with a temperature determined by that of the electron-hole gas. Simulations based on the solution of a time-dependent Boltzmann equation support this picture for pulsed and quasicontinuous photoexcitation.[15]

The existence of a quasiequilibrium for the phonons is, however, not supported by our experimental results. If one were to ascribe an effective temperature to each phonon mode according to a Planck distribution, its value would range from 120 to 141 K for AlAs- and GaAs-like optical phonon modes. A large occupation number of the AlAs-like phonon modes is rather surprising since it does not reflect their weaker coupling strength to electrons in a structure with wide quantum wells. This situation is analogous to that of holes for which the generation rates of LO phonons via the Fröhlich interaction is stronger than that of TO phonons via the optical deformation potential. Nevertheless, the weaker scattering process contributes significantly to the energy-loss rate of holes on a picosecond time scale as shown experimentally by Zhou, Leo, and Kurz[16] and theoretically by Pötz and Kocevar.[15] On a longer time scale determined by the lifetime of the electron-hole gas, optical phonon modes coupled to the electron-hole plasma tend to reach the same effective temperature and eventually realize an internal equilibrium. Whether this situation is realized experimentally depends on the carrier lifetime and on the optical phonon lifetimes as was discussed early by Kocevar.[15] This interpretation is also consistent with the weak dependence of the phonon occupation number as a function of the power density. As the incident power density was raised from 2.1×10^5 W/cm^2 to twice that value, the occupation number of confined optical phonons increased by about 30%. In the case of a picosecond pulsed excitation, a linear increase would be expected as the phonon generation rate is directly proportional to the initial carrier density in the low density limit (less than about 10^{17} cm^{-3}) where screening of the polar optical phonon coupling can be neglected. Under continuous excitation, the average phonon occupation number of nonequilibrium optical phonon modes will steadily follow a temperature increase of the electron-hole plasma.

Finally, we discuss the role of interface phonons in cooling hot carriers. Although we have measured a large occupation number for interface modes, their effectiveness in dissipating energy away from the carriers is also connected to their lifetime through their anharmonic decay into acoustic and optical phonon modes.[17] Quite generally, we expect optical phonon modes with a shorter lifetime to be more effective in dissipating energy from the carriers to the thermal bath. Insight into the magnitude of interface phonon lifetime may be gained from a recent work by Gupta and Ridley[18] who predict a slight increase of this lifetime of at most 50% for wave vectors of interest to electron-phonon coupling. To our knowledge the lifetime of interface modes has not yet been experimentally measured; besides, a twofold increase of the occupation number of one of the AlAs-like interface phonons (381 cm^{-1}) is consistent with a larger mode lifetime. Thus, our results suggest that some interface modes effectively dissipate energy to the lattice.

In conclusion, we have performed a study of a nonequilibrium phonon population in GaAs/AlAs MQW's. Our results demonstrate the dominant influence of phonon reabsorption on an optically excited electron-hole gas and imply that a quasiequilibrium among the optical phonon population is not realized under steady-state conditions.

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