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Time-resolved Raman scattering of nonequilibrium LO phonons in GaAs quantum wells

K. T. Tsen
Physics Department, Arizona State University, Tempe, Arizona 85287

R. P. Joshi and D. K. Ferry
Center for Solid State Electronic Research, Arizona State University, Tempe, Arizona 85287

H. Morkoc
Coordinated Science Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

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Time-resolved resonant Raman spectroscopy has been used to study the properties of nonequilibrium GaAs LO phonons generated as a result of the cascade of photoexcited carriers in GaAs quantum wells. The average population relaxation time of these LO phonons, which are responsible for hot-phonon effects in GaAs quantum wells, is directly measured to be 8 ± 1 ps at $T\simeq10$ K. These experimental results should help determine quantitatively and accurately the role hot phonons play in the hot-carrier dynamics of multiple-quantum-well structures.

Recently, there has been considerable interest in properties of hot electrons and holes created either by photoexcitation or by the application of an electric field in semiconductors.[1] This stems in part from the necessity to understand ultrasmall, high-field devices and also from the motivation of a better understanding of the fundamental interactions such as electron-electron and electron-phonon scattering. Techniques involving excitation and probing with laser pulse trains having pulse duration of the order of picosecond,[2,3] luminescence,[4,5] and steady-state methods[6,7] have been employed to deduce cooling curves of the hot-carrier system. All of these experiments show that the cooling of the plasma is much slower than is expected from a simple consideration of the bulk electron–longitudinal-optical-phonon interaction. Although many possibilities have been suggested for this observed slow cooling rate, it is generally believed that hot-phonon effects[8] (i.e., effects resulting from the presence of phonon distributions, particularly LO phonons, whose occupation probabilities exceed that at thermal equilibrium) play a central role in the process. For example, in GaAs quantum wells, Shah, Pinczuk, Gossard, and Wiegmann[6] have attributed anomalously low electron-energy loss rates to the presence of nonequilibrium LO phonons. Monte Carlo calculations of Lugli and Goodnick[9] have demonstrated that inclusion of hot phonons, generated as a result of the cascade of photoexcited carriers in GaAs quantum wells, reduces the electron relaxation rate in qualitative agreement with available experimental results. Recent theoretical calculations of Das Sarma, Jain, and Jalabert[10] have also emphasized the need for hot-phonon effects for a reasonable agreement between their results and existing experimental measurements. The necessity of measuring the lifetime of LO phonons in GaAs quantum wells directly and independently is justified by the observation that in order to fit the data satisfactorily, different values of LO phonon lifetime have been assumed.[6,7,9–11] In view of this, we describe in this paper a novel technique for the determination of average population relaxation time of nonequilibrium LO phonons created as a result of the cascade of photoexcited carriers in GaAs quantum wells. These LO phonons are responsible for hot-phonon effects. We observe the generation of LO phonons in GaAs quantum wells during the interaction of photoexcited hot electrons and holes with the lattice, and we obtain the average relaxation time of a nonequilibrium population of LO phonons.

The technique is based on a Raman-scattering version of the pump-and-probe scheme. However, previous studies have concentrated either on off-resonance scattering of LO phonons in GaAs (Refs. 12 and 13) and $Al_xGa_{1-x}As$,[14] or on resonant scattering (at the $E_0+\Delta_0$ energy gap) of electrons between different subbands in GaAs quantum wells.[15] In these measurements, conservation of the wave vector during the light-scattering process imposes a limitation on the excitations that can be probed by the technique. In contrast to all of the previous work, our present experimental method probes the excitations with phonon energy in resonance with the E_0 fundamental gap of semiconductors. Under such conditions, it has been demonstrated[16] that the validity of wave-vector conservation during the light scattering process depends critically on the number of impurities or the degree of interfacial roughness in GaAs quantum wells. This violation of conservation of wave vector allows us to investigate the properties of LO phonons active in hot-phonon effects in GaAs quantum wells which are otherwise inaccessible by the light scattering technique.

The undoped GaAs-$Al_xGa_{1-x}As$ multiple quantum-well structures (MQWS) studied in this work were grown by molecular-beam epitaxy on a (001)-oriented undoped GaAs substrate. It consists of approximately 50 periods of 10-nm-thick $Al_xGa_{1-x}As$ ($x\simeq0.3$) and 5-nm-thick GaAs layers. The sample was kept in contact with a constant flow of cold He gas (~10 K). The experiments are carried out with two independently tunable dye lasers (DCM and Pyridine 2) which are synchronously pumped by the second harmonic of a cw mode-locked yttrium aluminum garnet laser at a repetition rate of 76 MHz. The pulse width of the dye lasers is ~ 3 ps and the time jitter between the two dye lasers is estimated to be ~ 2 ps from a cross-correlation measurement. The pump pulse which excites the electron-hole plasma is chosen to operate at ~ 1.86 eV, so that only the first (or lowest) conducting subband is populated with the electrons. The photon energy of probe pulse is set at ~ 1.68 eV in resonance with the heavy-hole and first-conduction-subband exciton. These two beams after being suitably polarized are made to overlap with a spot size of about 100 μm on the surface of the sample. The ratio of the intensity of the pump to the intensity of the probe pulses is kept at about 1:1. The generated electron-hole pair density is estimated from the power density and the absorption coefficient of the pump pulse and is given by $\sim 3\times 10^{11}$ cm^{-2}. The backward-scattered Raman signal is detected by a standard photon-counting Raman system. We have measured both the polarized Raman spectrum which is usually denoted by $z(x,x)\bar{z}$ and the depolarized Raman spectrum which is usually denoted by $z(x,y)\bar{z}$, where $x=[110]$ appearing in the first position within the parentheses refers to the polarization of the incident light and x or $y=[1\bar{1}0]$ appearing in the second position within the parentheses refers to the polarization of the scattered light, and $z=[001]$ $(\bar{z})$ is the direction of propagation of the incident (scattered) light. Because there is no essential difference between these two spectra, except that the former is much stronger than the latter, we shall only present the polarized Raman spectra.

Figure 1 shows a polarized Raman spectrum of the sample taken without the time delay between the pump and the probe pulses. In addition to two sharp peaks at ~ 284 and 296 cm^{-1} corresponding to GaAs-like and GaAs LO phonons with wave vectors perpendicular to the layers (i.e., confined phonons), respectively, a relatively broad structure around 287 cm^{-1} is seen. Based on the following argument, we attribute this broad feature to Raman scattering from LO phonons which are responsible for hot-phonon effects in GaAs quantum wells. Under our current experimental conditions, the electrons are photoexcited to the first conduction subband with excess energy ~ 0.18 eV; in a polar semiconductor such as GaAs, these hot electrons will thermalize to the bottom of the subband by losing their energy to LO phonons.[12] Therefore, at very low temperature, these nonequilibrium LO phonons should show up in the anti-Stokes Raman spectrum. This expectation is indeed observed. In Fig. 2, an anti-Stokes Raman spectrum is shown with the time delay $\Delta t=0$ ps. We notice that in this spectrum a similar broad structure centered around 287 cm^{-1} is observed and this broad structure has been found to disappear when the probe pulse alone is present. We can rule out the possibility that this broad feature is the result of scattering of light from the photoexcited electron-hole plasma, because both the shape and the peak position do not change significantly as the injected density is varied by a factor of three. Furthermore, we can also disregard the explanation that it might come from Raman scattering of nonequilibrium GaAs-like or AlAs-like LO phonons, since the photon energy of both the pump and the probe pulses is less than the energy gap of $Al_xGa_{1-x}As$ layers.[17]

In a typical Raman backscattering geometry, scattering from the GaAs LO phonons with sizable wave-vector components[18] parallel to the layers of GaAs-$Al_xGa_{1-x}As$ MQWS is forbidden by the conservation of wave vector. However, if there is a breakdown in wave-vector conservation, this scattering becomes allowed. In cases where the Fröhlich interaction dominates over the deformation potential interaction, a small number of impurities can lead to a strong relaxation of the wave-vector selection rule.[19-22] We believe that this is what is happening in our experiments. Either the defect or interface roughness in GaAs quantum wells causes a breakdown in the wave-

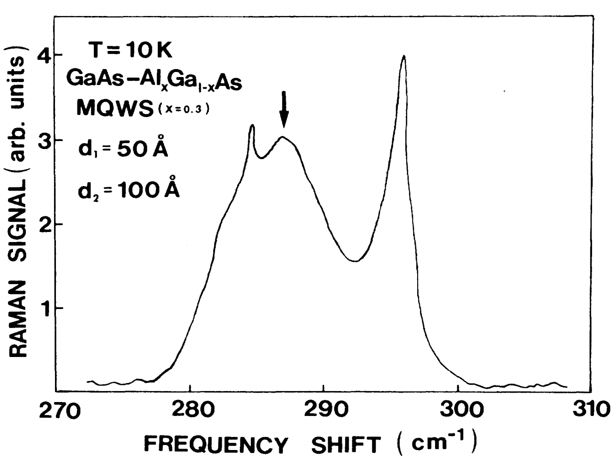

FIG. 1. Resonant Stokes Raman spectra of a GaAs-$Al_xGa_{1-x}As$ MQWS with aluminum concentration $x \simeq 0.3$, well-thickness $d_1 \simeq 50$ Å, barrier width $d_2 \simeq 100$ Å taken with $\Delta t=0$ ps and at $T \simeq 10$ K. The sharp peaks at $\simeq 284$ cm^{-1} and 296 cm^{-1} are Raman scattering signal from GaAs-like and GaAs LO phonons, respectively. The broad structure indicated by the arrow corresponds to scattering of light by the GaAs LO phonons which have wave vectors almost parallel to the layers of GaAs-$Al_xGa_{1-x}As$ MQWS. See text for discussions.

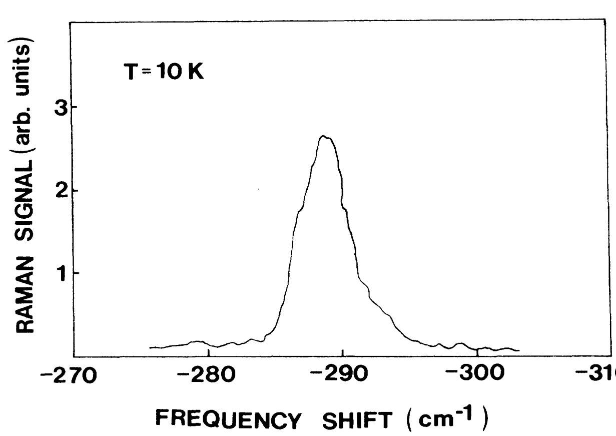

FIG. 2. Resonant anti-Stokes Raman spectrum of the sample described in Fig. 1 taken with $\Delta t=0$ ps and at $T \simeq 10$ K shows the presence of the nonequilibrium LO phonons with wave vectors almost parallel to the layers. This broad structure disappears when the pump beam is absent.

vector conservation during light scattering process.

This interpretation is further supported by the data shown in Fig. 3 in which the peak intensity of the broad structure centered around 287 cm^{-1} is plotted against the photon energy of probe pulse. The resonance curve shows a maximum for the outgoing channel. This asymmetry between incoming and outgoing channels is typical for impurity-induced scattering in bulk materials.[22,23]

We show in Fig. 4 the logarithm of the integrated anti-Stokes Raman signal of observed broad structure centered around ~ 287 cm^{-1} versus the time delay between the pump and the probe pulses. The rise of the signal indicates the generation of LO phonons active in hot-phonon effects in GaAs quantum wells by the thermalizing electrons through an intrasubband scattering process, whereas the decrease of the signal corresponds to the relaxation of these nonequilibrium excitations. From the decaying part of the signal, the average population relaxation time of these LO phonons is determined to be 8 ± 1 ps.[24]

The initial decay of the hot photoexcited carriers occurs from a phonon cascade process,[12,13] which leads to the buildup of the nonequilibrium phonon population. When this population is sufficiently large, a phonon "bottleneck" forms in which the carrier cooling is slowed by the effective balancing of emission and absorption processes. This bottleneck is formed at a time corresponding to the peak in Fig. 4. For longer times, the decay is characterized by the coupling between the electron-hole-phonon system, and the loss of LO phonons from the distribution is partially balanced by the new phonons emitted by the carriers. The net result is that the "observed" decay will be slightly slower by an amount that depends upon the excitation intensity of the laser. This effect can lead to different measured decay times for the LO phonons, but the true time is masked by the carrier-phonon coupling. Overlaid in Fig. 4 is the calculated decay of the nonequilibrium LO phonon modes for the value of lifetime which best fits the experimental data, using an analytic formulation for the quantum-well system,[25] which incorporates all of these effects. This calculation yields an experimentally observable decay time of 8 ps, when the value of actual phonon lifetime (7.8 ps) is used.

This value compares favorably with the LO phonon lifetime deduced from a parameter fit by Shah *et al.*,[6] and justifies the assumption made by Lugli and Goodnick[9] and Cai, Marchetti, and Lax[11] in their theoretical simulation; however, it is about a factor of two larger than the LO phonon lifetime needed to reasonably explain available experimental results with the theoretical calculations by Das Sarma *et al.*[10]

It is not clear at this moment why wave-vector conservation breaks down during a light scattering process in which the photon energy of the probe pulse is set at E_0 and not at the $E_0+\Delta_0$ energy gap of GaAs. One possible explanation is that because of much larger resonance enhancement at E_0 than at $E_0+\Delta_0$, the light scattering process becomes more "sensitive" to imperfection in the samples.

We notice that under our experimental conditions the wave-vector selection rule has been violated during the light scattering process; nevertheless, we believe that the cascade of photoexcited carriers should still obey the conservation of momentum. This, together with the argument made in Ref. 18, explains why nonequilibrium LO phonons with wave vector perpendicular to the layers (i.e., confined phonons) are not detected in our measurements.

In summary, we have developed a new time-resolved resonant Raman scattering technique to study the properties of nonequilibrium LO phonons in GaAs quantum wells. The average population relaxation time of these LO phonons which are responsible for hot-phonon effects in GaAs quantum wells is directly measured to be $\sim 8 \pm 1$ ps at $T \sim 10$ K. These experimental results should help determine quantitatively and accurately the role hot phonons play in the hot-carrier dynamics of MQWS.

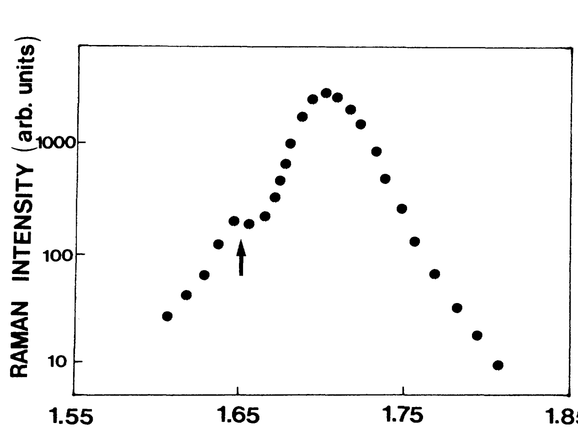

FIG. 3. Resonant Raman profile of the nonequilibrium LO phonons measured at the maximum of their intensity. The arrow indicates the calculated energy of the $n=1$ heavy-hole and first-conduction-subband exciton. See text for discussions.

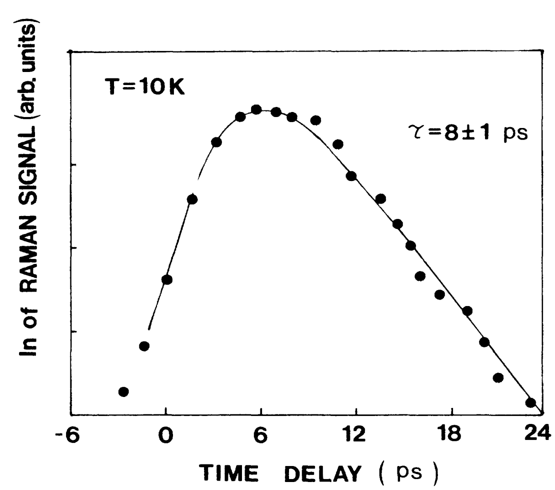

FIG. 4. The logarithm of the integrated anti-Stokes Raman signal of the GaAs LO phonons with wave vectors almost parallel to the layers of GaAs-$Al_xGa_{1-x}As$ MQWS is plotted against the time delay Δt. The solid circles correspond to experimental data. The deduced average population relaxation time of the LO phonons is given by 8 ± 1 ps. The solid curve represents theoretical calculation based on an analytic formulation for the quantum-well system. See text for discussions.

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[17]Two-photon absorption is another process of generating electron-hole pairs in semiconductors; however, we estimate that this process creates about 5×10^{8} cm^{-2} electron-hole pair density which is much less than 3×10^{11} cm^{-2} generated by one-photon process. Consequently, we believe that the observed nonequilibrium LO phonons cannot come from two-photon absorption process.

[18]For the sample parameters used in our experiments, basic quantum-mechanical calculations show that the broadening into bands (as a result of coupling between quantum wells) of the energy associated with motion perpendicular to the layers is about 0.2 meV, which is much less than the LO phonon energy ($\simeq$36 meV); therefore, the wave vector of LO phonons emitted by the photoexcited electrons undergoing intrasubband transitions is going to have a sizable component ($\simeq10^{6}$ cm^{-1}) parallel to the layers.

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[24]Because the anti-Stokes Raman signal of LO phonons extends up to time delay $\simeq$24 ps, their decay constant can be resolved with 3-ps laser pulse width (see also Ref. 12). The major uncertainty in our analysis comes from the relative time jitter ($\simeq$2 ps) between the pump and the probe pulses. We estimate that this time jitter introduces an uncertainty of about ±1 ps to our measured relaxation time.

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