Electron-Optical-Phonon Interactions in Ultrathin GaAs/AlAs Multiple Quantum Wells

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Nonequilibrium populations of both confined and interface phonons generated by picosecond laser pulses in a series of GaAs/AlAs quantum wells have been studied by time-resolved picosecond Raman scattering as a function of well width. The dependence of the nonequilibrium phonon populations on well width is found to be sensitive to the theoretical model which is used to describe the electron-phonon interaction. Our data disagree with the macroscopic models of electron-phonon interaction, but they are in excellent agreement with the microscopic model proposed recently by Huang and Zhu.

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Electron-phonon interactions are fundamental in determining the electronic properties of semiconductor quantum wells (QW) such as mobility and tunneling. So far several theories [1-5] have been proposed for the interaction of electrons with confined longitudinal-optical (LO) phonons and with interface phonons. These theoretical models differ significantly in the way they treat the vibrational modes of the QW and the electron-phonon scattering rates calculated from them can differ by as much as an order of magnitude [6]. It is, therefore, important to test experimentally the validity of these theoretical models. So far there have been a few attempts to differentiate between these models by directly measuring the intersubband scattering rates [7]. However, due to insufficient time resolution the issue has not yet been definitively resolved. In this Letter we report a conclusive test of these theoretical models for the electron-phonon interaction in QW based on a study of the nonequilibrium phonon population generated by intrasubband relaxation of photoexcited hot electrons in a series of GaAs/AlAs QW with varying well widths. We found that only the microscopic model proposed by Huang and Zhu [4] can explain our results quantitatively.

We will first briefly review the theoretical models proposed so far. Two of these models are macroscopic models in which the QW is approximated by a continuum and, using different boundary conditions, the resulting phonons have been referred to either as "slab modes" [1] or as "guided modes" [2,3]. These models have been labeled as the "dielectric continuum model" (to be referred to as model 1) and as the "mechanical model" (to be referred to as model 2). A microscopic lattice-dynamical calculation has recently been performed on GaAs/AlAs QW by Huang and Zhu (HZ). Based on their numerical results these authors have proposed an analytical approximation of the generally complicated solutions for the ionic displacements and the resultant macroscopic electric fields [4]. This model will be labeled as the HZ model. The electron-phonon interaction in these three models differs significantly because of the different boundary conditions used to calculate the electric potential associated with the LO phonon [6], the only exception being that the interactions between the interface phonons and electrons within the dielectric continuum model and within the HZ model are almost identical [5].

The undoped GaAs/AlAs multiple-QW (MQW) samples used in our experiment were grown by molecular-beam epitaxy on a (001)-oriented GaAs substrate. Each sample consists of 100 periods of a GaAs well surrounded by 7-nm-thick AlAs barriers. The thickness of the GaAs wells (L) in different samples varies from 2 to 6 nm. The experiments were performed at a constant sample temperature of 10 K. The samples were excited and probed by two independently tunable dye lasers of about 100 mW of time-averaged power each [8]. The two dye lasers were pumped synchronously by the second harmonic of a cw mode-locked Nd-doped yttrium aluminum garnet laser operating at a repetition rate of 76 MHz. The pulse width of the dye lasers was typically 3 ps, while the jitter between the two dye lasers was about 2 ps, as determined from cross-correlation measurements. The photon energy of one of the dye lasers was chosen to excite electrons into the first subband with the same excess energy of 200 meV for all the samples. The photon energy of the probe dye laser was tuned always to the vicinity of the lowest-energy exciton for maximum Raman intensity. The excitation and time-delayed probe beams were made to overlap on the sample surface with a spot size of about 500 μm. Based on the absorption coefficient of the GaAs/AlAs MQW, the intensity of the excitation beam was adjusted to excite, in all samples, an electron-hole gas of areal density equal to $(2\pm0.4)\times10^{10}$ cm^{-2}. The backscattered Raman signal from the probe beam was analyzed and detected by a double spectrometer with photon counting electronics. All the Raman spectra were measured in the $z(x',x')\bar{z}$ scattering configuration where $x'=[1\bar{1}0]$ and $z=[001]$.

Figure 1 shows the Stokes and anti-Stokes Raman spectra of a GaAs/AlAs MQW with a well width of 2 nm measured at a time delay of 1.5 ps between the excitation and probe laser pulses. The peaks observed in the Stokes spectra [Fig. 1(a)] are very similar to those reported previously under near-resonance condition in a cw Raman experiment [3]. The sharp peaks below 300 cm^{-1} are from LO phonons confined inside the GaAs layer. Using the notation of Sood *et al.* [3], these modes are labeled as LO_m, according to the component of the phonon wave vector perpendicular to the QW: $q_z=m\pi/(L+a)$, where m is a positive integer and a is the thickness of a monolayer of GaAs. In this scattering configuration only modes with even m are observed [3]. The broader features labeled as IF1 and IF2 have been identified as GaAs- and AlAs-like interface optical phonons. The peak labeled as TO is the transverse-optical phonon inside the AlAs layer. At low temperatures the thermal occupation numbers of these optical phonons are vanishingly small. The anti-Stokes Raman signals in Fig. 1(b) arise, therefore, from a nonequilibrium population of optical phonons generated by the excitation laser via intrasubband relaxation of the photoexcited electron-hole pairs. Another striking feature of these spectra is that not all of the many phonon peaks observed in Stokes scattering appear in the anti-Stokes spectra. This suggests that some optical phonons interact more strongly than others with hot electron-hole pairs.

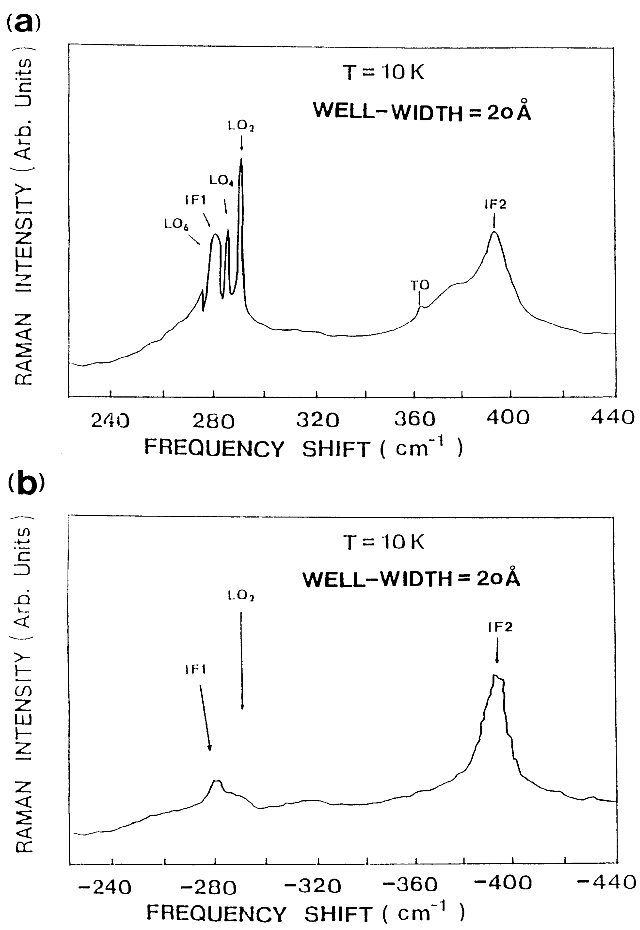

FIG. 1. (a) Stokes and (b) anti-Stokes Raman spectra of a GaAs/AlAs MQW with a 2-nm well width measured at 10 K with a train of 3-ps-long, 1.9-eV probe pulses which have been delayed by 1.5 ps relative to a train of 2.1-eV excitation pulses.

To determine the population N of a phonon mode from the Stokes (I_S) and anti-Stokes (I_{AS}) Raman intensities, we have utilized the following relationships [9]:

$$I_S(\omega_i)=I_0\sigma_S(\omega_i)(N+1)\,, \tag{1}$$

$$I_{AS}(\omega_i)=I_0\sigma_{AS}(\omega_i)N\,. \tag{2}$$

σ_S and σ_{AS} are the Stokes and anti-Stokes Raman cross sections, respectively. I_0 and ω_i stand for the probe-laser intensity and photon frequency. When the probe laser is resonant with excitonic transitions, $\sigma_S(\omega_i)$ and $\sigma_{AS}(\omega_i)$ may no longer be identical due to their different resonant behavior. However, it is known that the following rela-

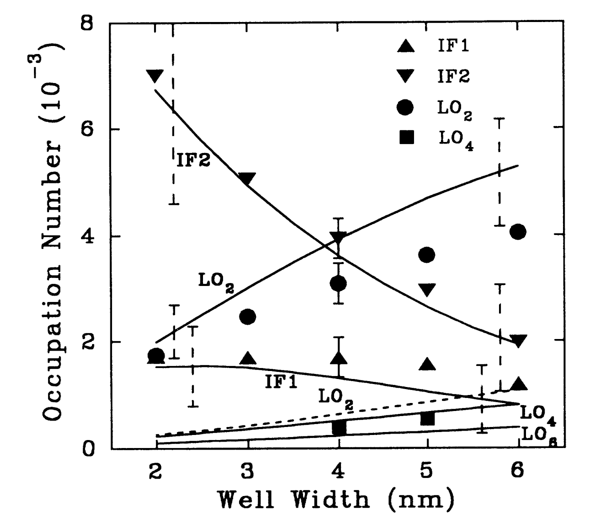

FIG. 2. Nonequilibrium phonon occupation numbers of the IF1,IF2 interface modes and the LO_m ($m=2,4,6$) confined phonon modes as a function of the quantum well width. The symbols are experimental results and the solid vertical lines passing through them are the error bars. The solid curves are calculated with the HZ model assuming a constant Q of 4.6×10^6 cm^{-1}. The dashed vertical bars give the variation in the phonon populations when Q is varied between 4.3×10^6 cm^{-1} and 4.9×10^6 cm^{-1}. The dashed curve labeled LO_2 has been calculated with model 2.

tion is valid for nonresonant Raman scattering [10]:

$$\sigma_S(\omega_i)=\sigma_{AS}(\omega_i-\omega_0)\,, \tag{3}$$

where ω_0 is the optical-phonon frequency. We tested the validity of this relation by requiring that N determined with Eqs. (1)-(3) should be independent of ω_i and found it to be valid for the MQW we have studied even under resonant conditions. Time-resolved pump-and-probe anti-Stokes Raman scattering [8] was used to measure the phonon lifetimes in our MQW samples and we found them to be longer than 5 ps. Since we will concentrate on the phonon occupation numbers at a time delay of 1.5 ps, the influence of phonon decay on the nonequilibrium phonon population may be neglected.

The data points in Fig. 2 represent our results for four modes: IF1, IF2, LO_2, and LO_4. We note that they have quite different dependences on L. The population of confined LO modes decreases, whereas that of interface phonons significantly increases as the well width becomes narrower. To understand our results, we have calculated the nonequilibrium phonon populations generated by the relaxation of hot electrons in QW as a function of L. For samples with small L, only the lowest subband is populated by hot electrons. Nonequilibrium optical phonons are therefore generated only by intrasubband scattering processes. We will assume that the hot-electron distribution function $f_{\mathbf{K}}$ ($\mathbf{K}$ being the electron wave vector parallel to the QW layers) is a Maxwell-Boltzmann distribution, otherwise the calculation would be much more complex. This approximation is reasonable since the picosecond time delay of the probe pulse is long enough for the electrons to reach quasithermal equilibrium. The phonon distributions are obtained by solving a set of rate equations for the population $N_{i\mathbf{Q}}$ of the phonon mode i and wave vector (parallel to the layers) $\mathbf{Q}$ and the electron population N_e:

$$\frac{\partial N_{i\mathbf{Q}}}{\partial t}=\frac{2\pi}{\hbar}\sum_{\mathbf{K}}[\,|\langle N_{i\mathbf{Q}}+1,\mathbf{K}-\mathbf{Q}|\mathcal{H}_{e\text{-ph}}(i)|N_{i\mathbf{Q}},\mathbf{K}\rangle|^2 f_{\mathbf{K}}(1-f_{\mathbf{K}-\mathbf{Q}})\delta(E_{\mathbf{K}-\mathbf{Q}}-E_{\mathbf{K}}+\hbar\omega_{i\mathbf{Q}})$$
$$-|\langle N_{i\mathbf{Q}}-1,\mathbf{K}+\mathbf{Q}|\mathcal{H}_{e\text{-ph}}(i)|N_{i\mathbf{Q}},\mathbf{K}\rangle|^2 f_{\mathbf{K}}(1-f_{\mathbf{K}+\mathbf{Q}})\delta(E_{\mathbf{K}+\mathbf{Q}}-E_{\mathbf{K}}-\hbar\omega_{i\mathbf{Q}})]\,, \tag{4}$$

$$\frac{\partial}{\partial t}(N_e\langle E\rangle)=-\sum_{i,\mathbf{Q}}\hbar\omega_{i\mathbf{Q}}\frac{\partial N_{i\mathbf{Q}}}{\partial t}+\left.\frac{E_0\,\partial N_e}{\partial t}\right|_{\text{laser}}. \tag{5}$$

In Eqs. (4) $H_{e\text{-ph}}(i)$ is the appropriate electron-phonon interaction for phonon mode i and $E_{\mathbf{K}}$ is the electron energy. In Eq. (5) $\hbar$ is Planck's constant, $\langle E\rangle$ is the average electron energy defined by

$$\langle E\rangle=\sum f_{\mathbf{K}}E_{\mathbf{K}}\Big/\sum f_{\mathbf{K}}\,, \tag{6}$$

and E_0 is the initial electron kinetic energy, which is equal to 200 meV in all our measurements.

To integrate Eqs. (4) and (5) we have assumed that an electron gas of density equal to 2×10^{10} cm^{-2} is excited by laser pulses of 3 ps full width at half maximum. $H_{e\text{-ph}}$ have been obtained either from the two macroscopic models or the microscopic HZ model as in Ref. [6]. Model 1 predicts that only the interface modes and confined LO phonons with m equal to an odd integer can participate in intrasubband scattering. Model 2 has already been shown to fail to account for the interface phonons observed in cw Raman scattering [2]. In addition, it predicts that only the LO_2 mode can mediate intrasubband scattering [6]. The microscopic HZ model predicts that the interface phonons and all confined LO phonons with m equal to an even integer can be involved in intrasubband scattering. For the interface modes, model 1 and the HZ model give similar results as pointed out by HZ. As a result, we have calculated the interface mode populations using the Hamiltonian derived from model 1 by Mori and Ando [5] rather than the microscopic HZ model. From the data points in Fig. 2 we note that *of these three models, only the microscopic HZ model explains qualitatively the presence of all the observed nonequilibrium phonons.*

We have used the microscopic model of HZ to calculate the phonon distributions (at 1.5 ps after excitation) for the confined LO modes as a function of L. The distributions for interface phonons were calculated with the Hamiltonian of Ref. [5] as explained earlier. The results for the $L=2$ nm sample are shown in Fig. 3. Before comparing the calculated nonequilibrium phonon populations quantitatively with the experimental results we notice that all the phonon populations in Fig. 3 drop to zero for Q below about 2×10^5 cm^{-1}. From the scattering geometry, the value of Q probed by our Raman experiment is less than 10^5 cm^{-1} due to the conservation of inplane crystal momentum. Thus we should not expect to observe any nonequilibrium phonons if Q were conserved strictly. However, the probe laser was resonant with the lowest-energy exciton. Resonant Raman scattering in MQW has been studied with cw lasers by several authors [11]. To explain the observed resonance profiles, these authors have proposed that Q need not be restricted to $<10^5$ cm^{-1} under resonance conditions due to elastic scattering between resonantly excited excitons and defects. The resonant Raman profile of the LO_2 mode for the $L=2$ nm sample is shown in the inset of Fig. 3. This profile is very similar to those reported in the literature as being due to impurity-assisted resonant Raman processes in that the maximum intensity occurs at the outgoing resonance [11]. In our quantum wells elastic scattering of the lowest-energy exciton near the outgoing resonance

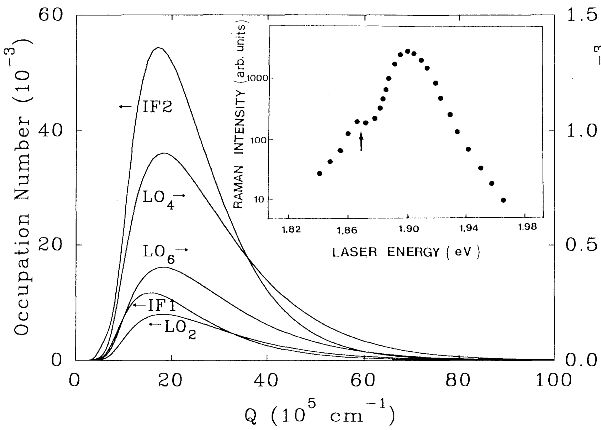

FIG. 3. Calculated nonequilibrium populations of optical phonons as a function of Q for a 2-nm GaAs/AlAs QW using the HZ model. Inset: Resonant Raman profile of the LO_2 confined phonon in the same QW measured at 10 K. The arrow indicates the energy of the lowest-energy exciton.

[11,12] should favor phonons with Q around 4×10^6 cm^{-1} [13]. The defects responsible for this elastic scattering are not definitively known at present. They are most likely carbon or interface roughness.

Assuming that the same defect is responsible for the relaxation in Q conservation for the optical phonons, we have attempted to fit all the modes in Fig. 2 with the value of *Q as the only adjustable parameter.* The solid curves in Fig. 2 have been calculated with the HZ model and $Q=4.6\times10^6$ cm^{-1}. The range of acceptable values of Q is $(4.3\times4.9)\times10^6$ cm^{-1}. For this range of values of Q the variations in the calculated phonon populations are indicated by the dashed vertical bars in Fig. 2. The theoretical curves reproduce well the L dependence of the phonon populations for the IF1, IF2, LO_2, and LO_4 modes. The magnitude of the calculated phonon populations are also in very good agreement with experiment for all the modes. This excellent agreement in the magnitude of the phonon populations is somewhat fortuitous, since the absolute magnitude of the calculated phonon population scales with the electron density and there is an uncertainty of about 20% in the experimental electron density. The two macroscopic models do not explain the data as well. While model 1 explains quantitatively the population of the interface phonon modes as well as the HZ model, it predicts that the LO_2 phonon is *not* allowed in intrasubband scattering. Model 2 predicts correctly that intrasubband scattering with LO_2 phonons is allowed but the calculated populations (shown as the dashed curve in Fig. 2) are much lower than the experimental values. In addition, model 2 cannot explain the large interface phonon population we observed in the narrow wells.

In conclusion, we have found that only the microscopic model of electron-phonon interaction in quantum wells proposed by Huang and Zhu is capable of explaining both qualitatively and quantitatively the nonequilibrium phonons produced by the intrasubband relaxation of photoexcited hot electrons in GaAs/AlAs quantum wells.

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