Time-Resolved Raman Measurements of Intersubband Relaxation in GaAs Quantum Wells

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We have measured the electron intersubband relaxation time due to the emission of longitudinal-optical (LO) phonons in GaAs-AlGaAs quantum wells using time-resolved anti-Stokes Raman scattering. When the energy separation of the subbands is greater than the LO phonon energy we find the lifetime of electrons in the $n=2$ level to be ≤ 1 ps, which is in excellent agreement with theoretical calculations of two-dimensional electron-phonon interactions.

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There has been considerable attention given recently to the subject of electronic intersubband relaxation in semiconductor quantum wells. In these structures electrons are confined spatially in the direction normal to the semiconductor layer, and the energy spectrum displays subbands whose spacing increases with increasing confinement. For weakly confined carriers, where the subband splitting is small, only slow intersubband relaxation by acoustic phonon emission is possible, whereas for highly confined carriers, where the splitting is greater than the longitudinal-optical (LO) phonon energy $\hbar\omega_L$, rapid relaxation by LO phonon emission is allowed. Experimental measurements of the latter process in GaAs quantum wells have given values of the C_2-C_1 interconduction subband relaxation time, τ_{21}, ≥ 10 ps.[1,2] On the other hand, recent theoretical calculations of two-dimensional electron-LO phonon scattering rates[3,4] find that $\tau_{21} \leq 1$ ps for wells with infinite potential barriers, more than an order of magnitude smaller than the experimental values. This discrepancy between experiment and theory has been attributed variously to poor confinement of electrons in the C_2 subband,[4] or to the combined effects of screening, intervalley scattering, and nonequilibrium (NE) phonons.[5] In this Letter we present the first direct measurement of τ_{21} by time-resolved anti-Stokes Raman scattering from a multiple-quantum-well (MQW) structure. This technique probes simultaneously the electrons and the LO phonons emitted by the electrons. The measured electron relaxation time of ≤ 1 ps is in excellent agreement with calculated intersubband scattering via two-dimensional confined phonons.

The sample we used was a 60-period undoped GaAs MQW grown by molecular-beam epitaxy on a (100) undoped GaAs substrate, and it consisted of GaAs layers of thickness $L=14.6$ nm and $Al_{0.36}Ga_{0.64}As$ barriers of thickness 15.7 nm. The energy levels of confined states were measured by photoluminescence excitation (PLE) spectroscopy and gave excellent agreement with the resonance Raman profiles for the GaAs LO phonons. Effective-mass calculations of the subband energies give a C_2-C_1 splitting of 52 meV, in excellent agreement with the PLE data, which is significantly greater than $\hbar\omega_L$. The sample temperature was 30 K for all measurements presented here.

The Stokes Raman spectrum measured in backscattering geometry $z(x'x')\bar{z}$ [where z and $\bar{z}$ are the directions of propagation of the incident and scattered laser beams, respectively, normal to the layers, and x' is the corresponding polarization vector along (110) in the plane of the layers] detects the LO phonon modes of the system: The GaAs mode is at 36.7 meV, and the GaAs-like and AlAs-like modes of the barriers are at 34.9 and 47 meV, respectively. The well width is too large to be able to resolve the splitting of the GaAs LO phonon into confined modes, and there is no evidence of scattering from interface phonons of the type reported for narrow wells.[6,7] An additional peak is observed at 51 meV in the polarized spectrum which shows strong resonant enhancement at both in- and out-going photon resonances of the allowed conduction—heavy-hole optical transitions, C_n-HH_n. Figure 1 shows the Stokes spectra obtained at the $n=4$ resonance for a range of incident laser powers, the scattering having been excited by a pulsed laser with 5-ps pulsewidth. As the photoexcited carrier density is increased, the 51-meV peak increases markedly in intensity and shifts to higher energy, which indicate that the scattering arises from photoexcited carriers. These observations are in close quantitative agreement with the expected behavior of the coupled C_1-C_2 intersubband plasmon-LO phonon mode.[8,9]

In the anti-Stokes spectrum we observe scattering from GaAs LO phonons and from C_2-C_1 intersubband transitions. Since at low temperature there is vanishingly small thermal occupation of either the LO phonon modes or the excited electronic subbands, any Raman scattering observed in the anti-Stokes spectrum must arise from nonequilibrium occupation of these states. The time dependence of the anti-Stokes intersubband scattering gives a direct measure of the electron popula-

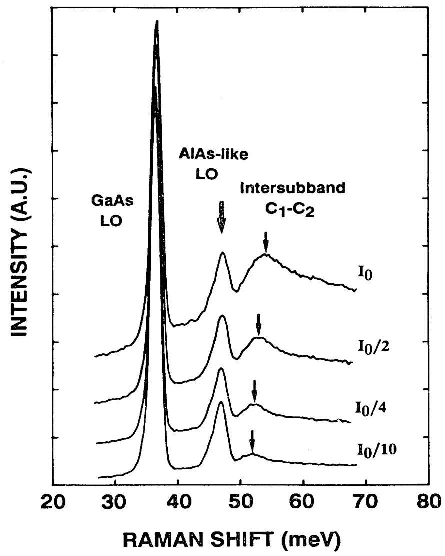

FIG. 1. Time-integrated Stokes Raman spectra obtained as a function of laser intensity from a 14.6-nm GaAs MQW in polarized back-scattering geometry at the C_4-HH_4 resonance. The two sharp peaks are the LO phonon mode of the well and the AlAs-like LO mode of the barrier. The broader peak near 51 meV arises from C_1-C_2 intersubband transitions.

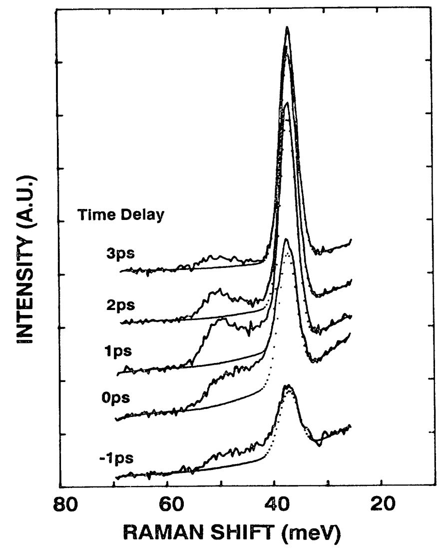

FIG. 2. Time-resolved anti-Stokes resonant Raman spectra for various time delays. The peaks at ~36 and ~51 meV are from nonequilibrium GaAs LO phonons and C_2-C_1 intersubband excitations, respectively. The dotted lines are fits to the phonon and luminescence components of the spectra.

tion in the C_2 subband.

To measure the time dependence of this anti-Stokes intersubband scattering, we used a synchronously pumped mode-locked dye laser with intracavity saturable absorber to generate wavelength-tunable pulses of duration 1 ps. In separate experiments the laser was tuned to the C_4-HH_4 (1.803 eV) and the C_3-HH_3 (1.691 eV) ingoing photon resonances. At both of these laser energies, photoexcited electrons have insufficient energy to allow them to be scattered into the conduction-band L or X valleys, processes which would seriously distort the intersubband dynamics under investigation. The laser pulses were divided into a pump beam and a probe beam with orthogonal polarizations and with a ratio of 5:1 in absorbed power. The Raman spectra were detected using a triple spectrometer and a liquid-nitrogen–cooled charge-coupled device multichannel detector. The polarized spectrum from the probe pulse was measured as a function of time delay between pump and probe pulses; the self-spectrum arising from the pulses in isolation from each other was subtracted to yield the spectrum of electronic excitations produced by the pump pulse. The photoexcited carrier density was kept very low $\leq 3 \times 10^{10}$ cm^{-2} to avoid the effects of intersubband carrier-carrier scattering: Theoretical calculations predict that intersubband carrier-carrier scattering should be considerably weaker than intrasubband scattering at this density.[10] In addition, the effects of final-state blocking of the C_2-C_1 transition and also phonon reabsorption are minimized at these carrier densities due to the negligible occupancy of the lowest subband at the energy $E = E_2 - E_1 - \hbar\omega_L$.

Figure 2 shows time-resolved anti-Stokes spectra obtained at the C_4-HH_4 resonance taken at various time delays between pump and probe pulses. The strong scattering at 36.7 meV is from nonequilibrium GaAs LO phonons, and the weaker scattering at ~51 meV is from C_2-C_1 transitions. The weak background signal arises from hot (anti-Stokes) luminescence and indicates that intraband carrier-carrier scattering does in fact occur. The electronic Raman scattering is very short lived, rising to a peak within 1 ps of the peak of the laser pulse (which coincides with the maximum of the luminescence background) and decaying within a few picoseconds. The phonon Raman scattering reaches a maximum after 2 ps and then decays relatively slowly over 20 ps or so. In contrast to the Stokes spectra presented in Fig. 1, there is no evidence of scattering from AlAs-like phonons in the anti-Stokes spectra of Fig. 2, which indicates a negligibly small nonequilibrium population of such modes. This result provides direct configuration that energy relaxation of confined carriers is by the emission of phonons with large amplitudes of ionic displacements within the wells: The emission of phonons confined to the barriers by remote interactions does not occur in the sample studied here.

Figure 3 shows the time dependence of the integrated anti-Stokes intensities of the phonon and electronic peaks. The autocorrelation profile of the laser pulse is also included for comparison. The electronic Raman

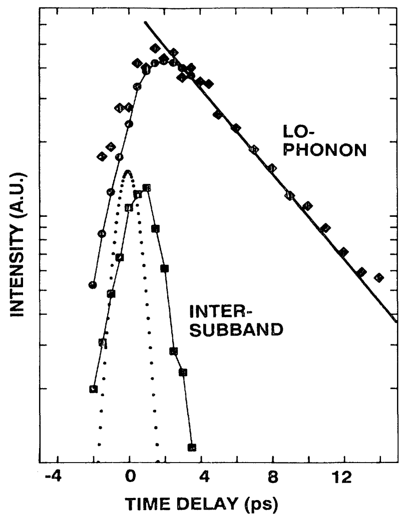

FIG. 3. Time-resolved anti-Stokes scattering intensities from LO phonons and C_2-C_1 intersubband transitions, both obtained at the C_4-HH_4 resonance. The dotted curve is the autocorrelation profile of the laser pulse.

scattering decays exponentially with a lifetime of 1 ps, which places an upper limit on τ_{21}. However, given that four electron subbands are excited, the observed decay time of the C_2 population is likely to be greater than τ_{21} due to scattering of electrons into the C_2 level from the higher states. The LO phonon signal decays exponentially with a lifetime of 5 ps; this value compares with the LO phonon lifetime of bulk GaAs of 7 ps at 77 K,[11] and 6 ps for a 20-nm GaAs MQW [12] also at 77 K.

The electron-phonon scattering rate in this structure can be estimated using the standard Golden rule method:

$$W=\frac{2\pi}{\hbar}\int |\langle K'|e\phi|K\rangle|^2\delta(E'-E)dN\,, \qquad (1)$$

where K, K' are the initial and final electronic states, N being the density of final states, and ϕ is the phonon-induced potential. In the limit of infinite potential barriers the electron states are given in the effective-mass approximation by

$$|K\rangle=(2/V)^{1/2}A(r,z)e^{i\mathbf{k}\cdot\mathbf{r}}\sin(k_z z)\,, \qquad (2)$$

where z is the direction normal to the layers, $0\le z\le L$, $k_z=n\pi/L$, $\mathbf{r}$ is the position vector within the plane, and $\mathbf{k}$ is the wave-vector component within the plane. $A(r,z)$ is the periodic Bloch amplitude, and V is the normalization volume. The potential ϕ depends on the representation of the two-dimensional phonon modes. The issue of what are the most appropriate boundary conditions for two-dimensional phonons in quantum-well structures has not yet been resolved:[3] In the slab-mode approach the ionic displacements in the z direction have antinodes at the interfaces and $\phi_s\sim\sin(q_z z)$, where $q_z=m\pi/L$; in the continuum model guided modes have nodes at the interfaces and $\phi_c\sim\cos(q_z z)$. It follows from general symmetry considerations that intersubband transitions, $n\rightarrow n-1$, must be accompanied by the emission of antisymmetric phonons, i.e., phonons which have in-plane ionic displacements which are antisymmetric with respect to reflection about the center of the well. For ϕ_s this condition requires m even, whereas for ϕ_c m is odd. Following Ridley[3] we obtain for the C_2-C_1 transition rates:

$$\phi_s:\ \frac{1}{\tau_{21}^s}=\frac{1}{2}W_0\left(\frac{\hbar\omega_L}{E_1}\right)^{1/2}\left[\left(\frac{(64/15\pi)^2}{7-\hbar\omega_L/E_1}\right)+\left(\frac{(128/105\pi)^2}{19-\hbar\omega_L/E_1}\right)+\cdots\right], \qquad (3a)$$

$$\phi_c:\ \frac{1}{\tau_{21}^c}=\frac{1}{2}W_0\left(\frac{\hbar\omega_L}{E_1}\right)^{1/2}\left[\left(\frac{1}{4-\hbar\omega_L/E_1}\right)+\left(\frac{1}{12-\hbar\omega_L/E_1}\right)\right]. \qquad (3b)$$

W_0 is the rate constant for the Frölich interaction:

$$W_0=(e^2/4\pi\hbar\epsilon_p)\{2m^*\omega_L/\hbar\}^{1/2},$$

where $\epsilon_p^{-1}=\epsilon_0^{-1}[\epsilon(\infty)^{-1}-\epsilon(0)^{-1}]$. Using values of $m^*=0.067m_e$, $\epsilon(\infty)=12.5$, and $\epsilon(0)=10.9$ for GaAs yields $W_0=6.4\times10^{12}\ s^{-1}$. We obtain values $\tau_{21}^c=360$ fs and $\tau_{21}^s=630$ fs. The scattering rate is smaller in the case of slab modes because electrons do not couple to the lowest-order phonon modes. Transition rates between higher subbands are systematically slower as the number of nodes in the wave functions increases.

Under our experimental conditions the C_2 population depends not only on τ_{21} but also on the transition rates from the higher excited subbands. In order to obtain a more accurate value of τ_{21}, we compare the measurements at the C_3-HH_3 resonance with those at the C_4-HH_4 resonance. The main difference between the two measurements is a delay ~1 ps in the signal risetime at the C_4-HH_4 resonance (see Fig. 4); the decay rates, however, are very similar. In analyzing the data we allow for the effects of population dynamics involving the higher levels by using a simple rate-equation model for a three- or four-level system with a $\mathrm{sech}^2(1.76t)$ pumping term (t in ps). We assume equal excitation of all the subbands, and that electrons relax instantaneously to the subband minima following photoexcitation. Intersubband rate constants are obtained as described above. In the guided-mode representation the overall lifetimes of the C_3 and C_4 subbands are predicted to be 475 and 575 fs, respectively, when the C_4 level is excited; in the slab-mode representation these times are longer by about

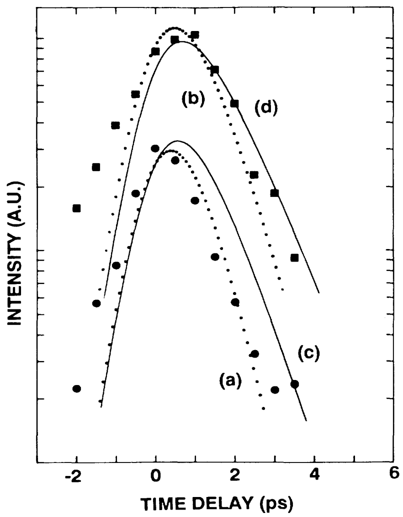

FIG. 4. Time-resolved anti-Stokes scattering from C_2-C_1 transitions at the C_3-HH_3 (lower data set) and C_4-HH_4 (upper data set) resonances. The dotted curve shows the C_2 population calculated using the guided-mode model with (*a*) three and (*b*) four photoexcited subbands. The solid curves show the C_2 population calculated using the slab-mode model with (*c*) three and (*d*) four photoexcited subbands.

50%. The convolution of the C_2 subband population, as a function of time, with the probe laser pulse gives the predicted scattering intensities as shown in Fig. 4. It can be seen that the guided-mode model predicts a scattering rate slightly faster than that observed, whereas the slab-mode model gives excellent agreement with the data. This agreement, however, may be partly fortuitous given the omission of any intrasubband relaxation in our model.

We have already mentioned that for this sample the well width is too large to permit spectral resolution of the confined phonons, and it is therefore not possible to assign the observed NE phonons to either m even or odd. However, it is now well established that for narrow GaAs quantum wells the polarized resonance Raman spectrum detects even m modes.[6,13] If this condition extends to the present case of wide wells, it is likely that the NE phonons observed here are even m modes. In a subsequent paper[14] we will show that for narrow GaAs quantum wells, in which only the C_1 subband is excited, the predominant NE phonons are $m=2$ modes and interface modes. These observations lend support to the idea that the NE measured in the present experiment are in fact produced by intrasubband relaxation, and therefore that ϕ_c is the appropriate phonon potential.

In conclusion, we have measured the C_2-C_1 intersubband relaxation time for LO phonon emission in a 14.6-nm GaAs MQW using an experimental method which avoids many of the effects which have previously prevented direct measurement of τ_{21}: (i) It measures carriers at low density so that carrier-carrier intersubband scattering, many-body effects, and final-state blocking of the C_2-C_1 scattering are negligible; (ii) intervalley Γ-L and Γ-X scattering are not allowed; and (iii) poor confinement of the upper subband is avoided. Our results place an upper limit of 1 ps on τ_{21}; furthermore, we infer a value of ~ 500 fs when allowance is made for relaxation within the multilevel system, which is in excellent agreement with theoretical predictions.

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