Time-Resolved Raman Scattering in GaAs Quantum Wells

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We employ a picosecond–time-resolved Raman-scattering technique to investigate the dynamics of two-dimensional electron-hole plasma confined to a multiple–quantum-well structure composed of layers of GaAs and (GaAl)As. The lifetime and the intersubband scattering time of the electrons photoexcited on the lowest electronic subbands are determined separately. Calculation of the rate of intersubband scattering by longitudinal-acoustical phonons successfully accounts for the observation of relatively long-lived electrons on the second subband of a 215-Å multiple–quantum-well structure.

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The photoexcitation of an electron-hole plasma in semiconductors is a powerful tool to probe the interactions of the carriers between themselves and with the lattice. Previous studies[1-5] of the dynamics of photoexcited carriers have shown that on a very short time scale (less than one picosecond), carrier-carrier scattering effectively randomizes the carrier momentum, and establishes a quasiequilibrium electron-hole plasma which subsequently cools towards the lattice temperature through the emission of phonons. While the mechanisms involved in the cooling of hot carriers are relatively well understood in bulk semiconductors,[6] some aspects of the relaxation of carriers in two-dimensional systems remain unclear. In semiconductor quantum-well structures, the motion of the carriers is confined in the direction normal to the layers by the higher potential of the barriers effecting a ladder of energy subbands. Earlier work has examined the effect of carrier confinement on the interaction of electrons and holes with phonons[7,8]: Time-resolved absorption[9,10] and photoluminescence[11-13] have been the major optical techniques to study the dynamics of the carriers following photoexcitation, and to deduce the cooling rates and the temperature of the plasma. However, these experiments do not directly determine the carrier densities of the plasma, and they have not distinguished between the intersubband and intrasubband acoustic-phonon scattering of carriers. Acoustic-phonon scattering is expected to be the dominant mechanism for intersubband electronic transitions if the electronic energy loss is less than the energy of a longitudinal optical (LO) phonon.

In this Letter, we examine the mechanism of intersubband scattering of electrons by longitudinal phonons within a GaAs quantum well. An electron from a nonequilibrium distribution on an upper energy subband will scatter to a lower subband through the emission of a longitudinal phonon while making an intersubband transition, but if its kinetic and confinement energy is less than one LO-phonon energy, one expects it to emit an LA phonon instead of an LO phonon. In order to determine the dynamics of the photoexcited carriers, we have developed a resonant–Raman-scattering version of the pump and probe technique. Previous studies[14,15] have concentrated on the nonequilibrium LO-phonon population generated as the carriers relax to the band edge, but the selection rules of inelastic light scattering impose the probing of a subset of the phonons that might be generated in the relaxation: those near–zone-center phonons whose momenta are typically $(6-7)\times10^5$ cm^{-1}. In our approach, we probe the inelastic light scattering of the electronic intersubband excitations of a two-dimensional plasma to look directly at the electrons.[16,17] We have observed both single-particle (SP) and collective intersubband excitations of the 2D plasma. The collective excitations provide an independent and absolute calibration of the excitation density, but are otherwise beyond the purview of this Letter.[18] Each SP intersubband excitation is characterized by a peak in the Raman spectrum whose energy shift from the laser frequency is equal to the subband energy separation. The peak originates from a probe photon's inelastically scattering an electron from the lower to the upper energy subband. The intensity of the SP peak is proportional to the difference of the carrier densities on the two subbands.

These experiments are performed with two independently tunable dye lasers synchronously pumped by a mode-locked argon-ion laser at a repetition rate of 76 MHz. Although the duration of the optical pulses is less than 5 ps, the temporal resolution is limited by the relative time jitter of the dye lasers: approximately 8 ps from a cross-correlation measurement. The probe-pulse energy is tuned in resonance to the optical gap separating one of the conduction subbands and the spin-orbit-split-off valence band (the $E_0+\Delta_0$ gap). Both beams are sent collinearly onto the sample at Brewster's angle, and the backscattered light is collected normal to the sample and analyzed in a Spex Triplemate spectrograph. We measure both polarized $z(x',x')\bar{z}$ and depolarized $z(x',y')\bar{z}$ spectra. In this notation, $x'=[110]$ refers to the polarization of the incident light in first position, while x' or y' refers to the polarization of the scattered light in second position, and $z=[001]$ ($\bar{z}$) is the direction of propagation of the incident (scattered) light, normal to the GaAs layers. A key element of our technique is the large sensitivity and parallel detection provided by a charge-coupled-device detector.[19] The sample is held at 5 K in a cryostat by a flow of cold helium gas. Pump fluence is typically 2 μJ-cm^{-2} per pulse, and the ratio of pump to probe laser fluences is kept close to 10. Pump and probe laser beams are respectively focused to spot sizes of 50 and 25 μm and carefully centered with pinholes.

The GaAs-$Al_{0.3}Ga_{0.7}As$ multiple-quantum-well structures (MQWS) were grown by molecular-beam epitaxy on a ⟨100⟩ GaAs substrate, and consist of thirty periods of 100 Å of $Al_{0.7}Ga_{0.3}As$ and either 215 or 116 Å of GaAs. The electron-hole plasma is generated by a near-infrared laser pulse whose penetration depth varies from 1 to 2 μm. The absorption length of the probe at a wavelength of 6510 Å is much shorter (5000 Å); thus, the plasma density in the quantum wells within this length is nearly uniform.

Figure 1 shows how tuning of the energy of the probe photon affects the spectrum of the electronic SP-intersubband excitations. At a laser energy of 1.905 eV, the incident photon is in resonance with a transition between the split-off valence band and the second conduction subband. One observes one peak at 26.8 meV (216 cm^{-1}) in the depolarized spectrum which is attributed to SP-intersubband scattering involving vertical transitions of an electron between the first and the second subbands. In the second spectrum, an additional intersubband excitation, whose energy shift is 41.4 meV (334 cm^{-1}), is seen when the incident photon energy is tuned in resonance with the third electronic subband. Thus by selecting the resonance, one may preferentially enhance the sensitivity to transitions associated with particular subbands. A Kronig-Penney calculation based on the growth parameters allowed us to estimate the energy separation of the lowest electronic subbands. These results are shown in Table I for the two samples studied here.

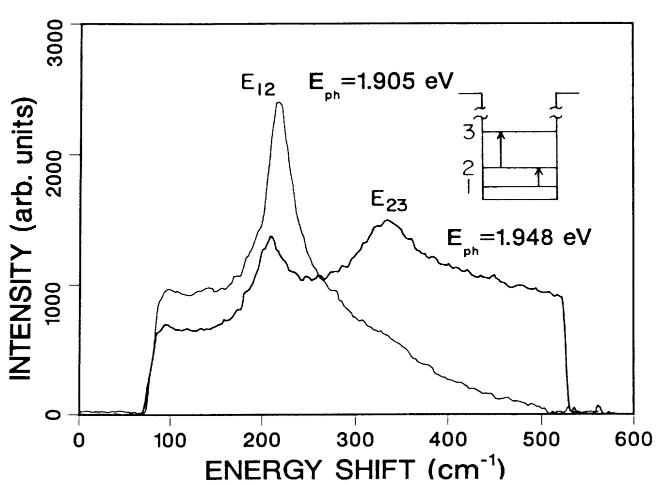

FIG. 1. Raman scattering spectra for two resonant photon energies at a delay time of 50 ps. The scattering geometry is $z(x',y')\bar{z}$.

The 41.4-meV spectral band is present only if the photon energy of the pump laser exceeds the threshold for excitation of electrons directly into the second subband. This indicates that the electronic distribution probed by the second pulse is a nonequilibrium one photoexcited by the pump pulse.

We now show how the intersubband electronic transitions can reveal the presence of excited carriers on the second subband. Figure 2 presents the time evolution of the $n=1$ to $n=2$ intersubband peak following excitation of electrons onto the lowest two subbands as we change the time delay between the pump and probe pulses. Solid points correspond to the excitation of electrons to the bottom of the first subband. After an initial fast rise, the intensity decreases exponentially with a time constant of 750 ps for the 215-Å MQWS. This decay time is the result of radiative and nonradiative recombination of the electron-hole plasma. Any role of the plasma expansion is believed to be minimized by the choice of a ratio of the focal-spot areas of the pump and probe pulses equal to 4. When electrons are initially excited on both the first and the second subbands, the overall intensity is significantly reduced during the first 300 ps (open squares). We attribute this reduction to the blocking effect of the electronic distribution on the second subband. This indicates a relatively long lifetime for the electrons on the second subband. As the electrons scatter down from the second to the first subband, the signal continues to rise. In a 215-Å quantum well, the subband energy separation E_{12} (26.8 meV) is far less than the energy of an LO phonon (36.7 meV). For this reason, we do not expect the thermalized electrons of the second subband to scatter down to the first subband with

TABLE I. Comparison between experimental and calculated values of the energy separations of the lowest electronic subbands ($n=1$ refers to the ground subband; energies are in millielectronvolts).

Width of GaAs layer	E_{12} (calc.)	E_{23} (calc.)	E_{12} (expt.)	E_{23} (expt.)	E_{13} (expt.)
215 Å, $x=0.3$	26.9	44.4	26.8	41.4	70.7
116 Å, $x=0.3$	72.3	107.9	64.2		

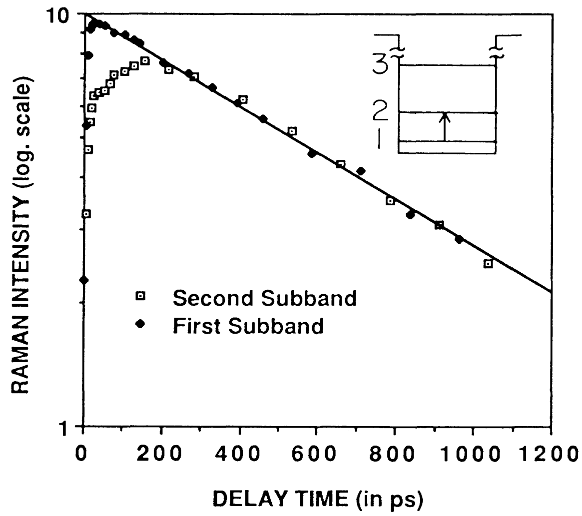

FIG. 2. Raman intensity of the SP-intersubband excitation at various times for two exciting conditions (first subband: energy of the pump photon is 1.530 eV; second subband: $E_{ph}=1.577$ eV). The least-squares fit results in a lifetime of 750 ± 25 ps in this 215-Å MQWS.

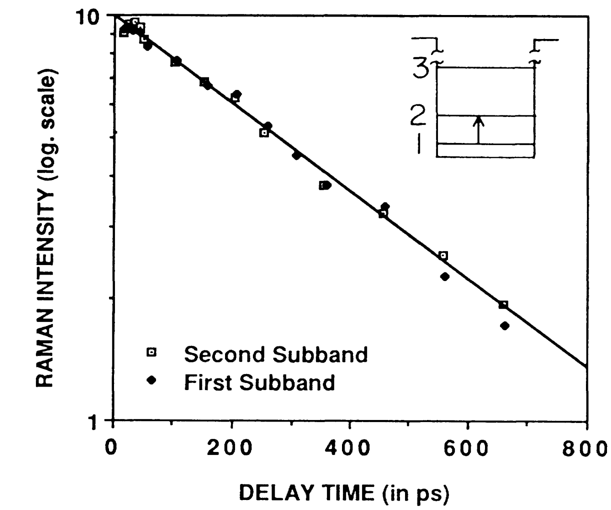

FIG. 3. Raman intensity of the SP-intersubband excitation at various times for two exciting conditions (first subband: energy of the pump photon is 1.556 eV; second subband: $E_{ph}=1.656$ eV). The least squares fit results in a lifetime of 400 ± 10 ps in this 116-Å MQWS.

the emission of LO phonons. However, this would not be the case for a narrower quantum well. We carried out a similar experiment in a 116-Å MQWS, and tuned the probe laser photon into resonance with the second electronic subband. In this narrow well, the intensity of the SP-intersubband excitation is not reduced when the pump-photon energy is raised above the excitation threshold of the second subband (see Fig. 3). We conclude that the electrons on the second subband initially excited by the pump pulse are rapidly scattered to the lower subband by the emission of an LO phonon. The spontaneous lifetime of the electrons is the 116-Å MQWS is also shorter: 400 ps. This supports the idea of an enhancement of the radiative lifetime caused by the increased overlap of the electron and hole wave functions when the size of the well is decreased.[12]

To test further our interpretation of the 300-ps time constant, we have probed the electron resonance at $E_{photon}=1.948$ eV in the 215-Å well sample (see Fig. 1). The intersubband excitation peak E_{23} is observable at this resonance if the energy of the pump photon is tuned to excite above the second subband. Since no electrons are excited onto the third subband, the intensity of this peak is simply proportional to the carrier density of the second subband. Figure 4 indicates the time dependence of the peak intensity. A least-squares fit to these data confirms a lifetime of 325 ps for the lifetime of the electron plasma on the second subband.

Could the electrons observed in the second subband have been scattered up from the first subband? The experimental conditions indicate that this is not the origin of the relatively long lifetime. From the power density of the pump pulse, 2.3 μJ-cm^{-2}, we estimate an average excitation density of $\simeq 4.0\times 10^{11}$-cm^{-2} electrons per layer distributed on the first and second subbands (the absorption coefficient is 10^4 cm^{-1} at $E_{photon}=1.577$ eV, which corresponds to a Fermi energy of 15 meV. From the energy of the pump photon, we estimate the excess kinetic energy of the electrons photoexcited in the first subband to be 40 meV. However, intraband scattering of the electrons by the LO phonons rapidly reduces the electronic temperature to about 30–40 K.[5] Furthermore, time-resolved photoluminescence studies[20] have

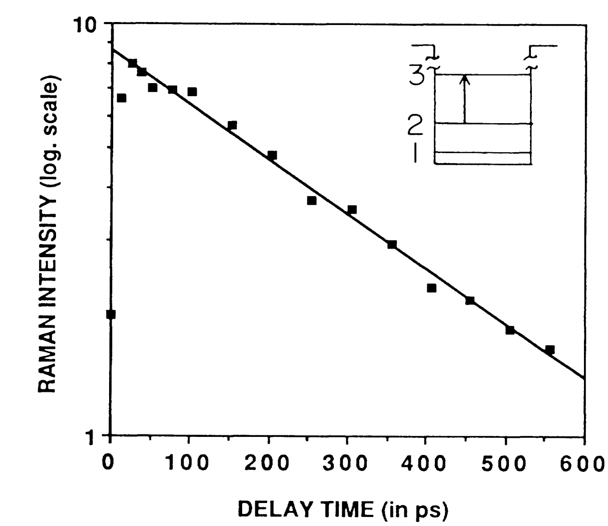

FIG. 4. Raman intensity of the SP-intersubband excitation E_{23} at various delay times. The least-squares fit results in a lifetime of 325 ± 25 ps for the electron gas excited on the second subband of this 215-Å MQWS.

determined the electronic temperature following the photoexcitation of a denser and hotter plasma to be below 60 K after a 50-ps delay time. For these reasons, we conclude that the electrons observed on the second subband within the first 300 ps were generated by the pump pulse, and not subsequently scattered there from the first subband. Thus, the decay rate found in the plot of Fig. 4 is interpreted as the true lifetime of the electrons excited on the second subband of the 215-Å quantum well. This lifetime is related to the average intersubband scattering time τ_{12} as follows:

$$\tau^{-1} = \tau_{r2}^{-1} + \tau_{12}^{-1},$$

where τ_{r2} is the radiative recombination time on the second subband. Because the effective mass of the holes is larger than that of the electrons, we expect the relaxation time of the holes on the second subband to be much shorter. This will effectively increase τ_{r2} and bring τ_{12} closer to 325 ps. However, if we assume that the radiative recombination lifetime is identical on both subbands, we deduce a value of τ_{12} equal to 570 ± 70 ps.

The transition rates for both electron and phonon scattering have been calculated with Fermi's "golden rule" and the appropriate interaction matrix elements.[21] Because of the large momentum transfer associated with intersubband scattering, the strength of the electron-electron scattering is at least one order of magnitude smaller than the deformation-potential scattering by the longitudinal acoustic phonons.[22] Of course, electron-electron scattering effectively thermalizes the electron distribution within each subband. The acoustic-phonon contribution to the intersubband scattering rate is inversely proportional to the square of the well width and, in first approximation, independent of the initial momentum of the electron (except for electrons whose phase velocity is less than the sound velocity of the LA phonons). Taking the deformation-potential energy to be 7 eV,[23] we calculated a value of τ_{12} equal to 490 ps. Considering the experimental uncertainty of the measured lifetime, the agreement with this value is excellent. This determination of the intersubband scattering time is unique, since it cannot be derived from transport measurements. Mobility measurements are known to be dominated by impurity scattering at low temperatures,[24] and longitudinal-optical–phonon scattering at room temperature.[25]

In conclusion, we have demonstrated that the carrier density of an electron plasma can be directly determined for arbitrary delay times following its excitation by use of a Raman-scattering version of the pump and probe technique. These measurements have revealed a relatively long lifetime of the electrons that are photoexcited on the second subband of a 215-Å MQWS. Intersubband scattering by the deformation potential of the acoustic phonons substantiates this result in the wide-well limit, while intersubband scattering by longitudinal optical phonons becomes dominant in the narrow-well limit.

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