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Subpicosecond time-resolved Raman spectroscopy of LO phonons in GaAs-$Al_xGa_{1-x}As$ multiple-quantum-well structures

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

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

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Time-resolved Raman spectra of GaAs-$Al_xGa_{1-x}As$ multiple-quantum-well structures have been measured with subpicosecond time resolution. The average electron–LO-phonon scattering time for the electrons occupying the unbound states is resolved and found to be about 170 fs and is not affected appreciably by the well thickness.

Time-resolved Raman scattering has been used extensively[1–4] to study the relaxation of optically excited carriers in polar semiconductors and semiconductor multiple quantum well structures (MQWS's). The initial relaxation of these hot carriers is believed (at least at very low excitations) to be dominated by the emission of LO phonons through the Frolich interaction. Our previous results on GaAs-$Al_xGa_{1-x}As$ MQWS's (Ref. 4) have shown that the lifetime of the optically generated nonequilibrium LO phonons having wave vectors perpendicular to the layers is about 6±1 ps at $T = 77$ K; however, the average electron-LO-phonon scattering time was not resolved. In this paper, we report the time-resolved Raman scattering results on the average electron-LO-phonon scattering time for electrons occupying the continuum states in GaAs-$Al_xGa_{1-x}As$ MQWS's with subpiosecond time resolution. The LO phonons emitted in the thermalization process is directly detected by measuring the time dependence of their population in the presence of a nonequilibrium carrier population. For GaAs-$Al_xGa_{1-x}As$ MQWS with aluminum concentration $x \simeq 0.05$ and well thickness about 200 Å, the experimental results have demonstrated that the average electron-LO-phonon scattering time for electrons occupying the continuum states is given by 170±40 fs. Similar measurements have been made on samples having well width ranging from 50 to 300 Å, and aluminum concentration $x \simeq 0.1$; within the accuracy of our experiment, the average electron-LO-phonon scattering time is observed to be independent of the well width of GaAs quantum wells.

The ultrashort pulses used in the experiment were generated from a DCM double-jet dye laser synchronously pumped by the second harmonic of a cw mode-locked yttrium-aluminum-garnet (YAG) laser. The pulses had an autocorrelation full width at half maximum of $\simeq 800$ fs and average power of $\simeq 50$ mW at a repetition rate of 76 MHz, In our pump-probe configuration, these ultrashort pulses were split into two beams of equal intensity but different polarization. An appropriate analyzer was placed in front of the entrance slit of the double monochromator so that the scattered light from the pump pulse was effectively eliminated while the scattering from the probe pulse was allowed to be detected. The dye laser was chosen to operate at $\hbar\omega \simeq 1.81$ eV very close to the $E_0 + \Delta_0$ energy gap of GaAs in order to take advantage of the resonance enhancement. The anti–Stokes-Raman signal was collected and analyzed by a computer-controlled Raman system. The experiments were carried out in a backscattering geometry.

The undoped GaAs-$Al_xGa_{1-x}As$ MQWS's investigated in this work were grown by molecular-beam epitaxy on a (001)-oriented undoped GaAs substrate. They consist of ~30 periods of 100-Å-thick $Al_xGa_{1-x}As$ ($x \simeq 0.05$ and 0.1) and different thicknesses of GaAs layers (which range from 50 to 300 Å). The samples were mounted on the cold finger tip of a liquid-nitrogen dewar. The effective temperature of the laser-irradiated area was about 80 K. From the power density and the absorption coefficient of the pump pulse, the average photoexcited carrier density was estimated to be about 2×10^{15} cm^{-3} and the occupation number of the excited nonequilibrium LO phonons is $\simeq 0.005$.

Figure 1 shows the anti–Stokes-Raman signal as a function of the time delay Δt between the pump and the probe pulses for the GaAs-$Al_xGa_{1-x}As$ MQWS's having $x \simeq 0.05$ and well width $\simeq 200$ Å. Here, the time delay is taken to be the time interval between the peak of the pump and that of the probe pulses. The zero delay at the sample was determined to within ±0.2 ps by the observance of the interference effect which occurred when the pump and the probe pulse were spatially and temporally overlapped. The Raman signal is monitored at 295 cm^{-1}. Because the growth of the anti–Stokes-Raman signal reflects the emission of Raman-active LO phonons by the thermalizing carriers, these experimental results enable us to estimate the average electron-LO-phonon scattering time for electrons occupying the continuum states. The number of Raman-active LO phonons emitted by the carriers is estimated to be about five after considering the fact that once the carriers are trapped inside the wells, they are no longer able to emit LO phonons that are active under our experimental conditions. The

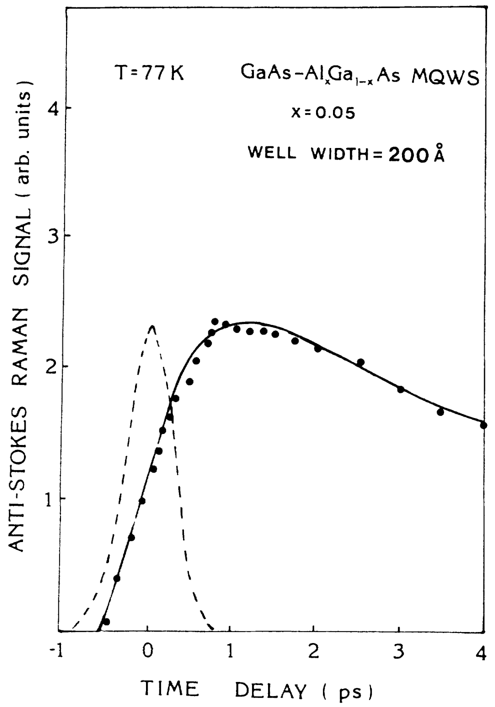

FIG. 1. Spontaneous anti–Stokes-Raman signal of LO phonons having wave vectors perpendicular to the layers is plotted against the time delay between the pump and the probe pulses for GaAs-Al$_x$Ga$_{1-x}$As MQWS's with $x \simeq 0.05$ and well thickness $\simeq 200$ Å. The solid circles correspond to experimental data. The solid curve represents the best fit to the data when the rate equations of electrons and phonons are used with $\tau_{e\text{-LO}} \simeq 170$fs and $\tau_{\text{LO}} \simeq 6$ ps. The dashed curve is the measured cross correlation trace of the pump and the probe pulses.

analysis based on rate equations taking the convolution of the pump and the probe pulses with the electron-LO-phonon generation rate and population relaxation time of LO phonons as parameters shows that the average electron-LO-phonon scattering time for electrons occupying the continuum states is about 170±40 fs. We have also carried out similar experiments on samples with well thickness from 50 to 300 Å and aluminum concentration $x \simeq 0.1$. The experimental results have indicated that the average electron-LO-phonon scattering time for electrons occupying the unbound states is rather insensitive to the well width of GaAs quantum wells.

Recently, it has been shown by Shah *et al.*[5] that the simple LO-phonon cascade model is not adequate to describe the thermalization of photoexcited carriers in GaAs under the circumstances that (1) carrier density is $\geq 5 \times 10^{16}$ cm^{-3}, and/or (2) excitation laser is such that it excites electrons which have excess energy to scatter into *L* or *X* valleys. In order to justify our application of the simple LO-phonon cascade model to deduce the average electron-LO-phonon scattering time in our experiment, the photon energy of the laser has been chosen to operate at $\simeq 1.81$ eV, so that there is no appreciable intervalley scattering of electrons. In addition, the density of the photoexcited electron-hole pairs has been chosen to be $\simeq 2 \times 10^{15}$ cm^{-3}. It is believed that[6] at such low photoexcited density, the carrier-carrier scattering will become less important and carrier-LO-phonon scattering will be dominant in the thermalization process.

We note that the spectral width of the exciting laser is $\simeq 30$ cm^{-1}, for the samples investigated in our experiments ($x \simeq 0.05$), it seems possible that the anti–Stokes-Raman signal would reflect the nonequilibrium population of GaAs LO phonons as well as that of GaAs-like LO phonons from the barriers. However, this is unlikely because our cw Raman measurements have shown that Raman signal of GaAs LO phonons is enhanced much more (by a factor of about 6) than that of GaAs-like LO phonons in the barriers. Therefore, the anti–Stokes-Raman signal observed in our experiments primarily comes from nonequilibrium GaAs LO phonons.

In conclusion, the average electron-LO-phonon scattering time for electrons occupying the continuum states in GaAs-Al$_x$Ga$_{1-x}$As MQWS's has been measured with subpicosecond time resolution. The deduced scattering time is found to be 170±40 fs and is rather insensitive to the well width of GaAs quantum wells.

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