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Raman and hot electron–neutral acceptor luminescence studies of electron–optical phonon interactions in $GaAs/Al_xGa_{1-x}As$ quantum wells

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Abstract

Using two optical techniques, we have studied the hot electron–optical phonon interactions in $GaAs/Al_xGa_{1-x}As$ multiple-quantum wells. Raman scattering measurements at 15 K are presented for the Al composition of $x = 0.3$, 0.5, 0.7 and 1.0. The GaAs-like and AlAs-like phonon frequencies of the first-order modes are also measured as a function of Al composition. The optical phonon energies emitted by the photoexcited electrons in quantum wells are determined by using hot electron–neutral acceptor luminescence techniques. It is shown that the relaxation of hot electrons in the quantum wells is dominated by the GaAs LO phonon emission for small x, but by AlAs-like LO phonons for larger Al composition.

Keywords: A. Quantum wells; D. Electron–phonon interactions; E. Inelastic scattering; E. Luminescence

1. Introduction

Techniques such as molecular-beam epitaxy have stimulated much research on semiconductor heterostructures. Of the semiconductor multiple-quantum wells, those from GaAs/AlGaAs have been the most studied. In order to learn about the characteristic properties of such thin crystalline films, many different methods are employed. Optical methods like reflection, transmission, and luminescence experiments are employed to characterize single and multiple-quantum layers, to learn about recombination mechanisms and the role of interfaces on these mechanisms. There have been a great number of experimental and theoretical studies focused on optical phonons in quantum wells and their interactions with electrons. Among those experimental techniques, Raman scattering has been proven as a versatile and efficient tool for probing long-wavelength and short-wavelength lattice dynamics of ternary alloys [1–5]. The electron–phonon interactions in semiconductor alloys have also been studied by using time-resolved Raman spectroscopy [7–9]. Kash et al. [8] have used time domain pump-probe Raman techniques to measure directly the relative strengths of the Fröhlich coupling for "AlAs-like" and "GaAs-like" LO phonons in the two-mode $Al_xGa_{1-x}As$ system. It was shown that the relative interaction strength with electrons of each mode is a strong function of alloy composition. Their results show that for small values of x, the coupling of electrons to the AlAs-like mode is much weaker than the coupling to the GaAs-like mode, and also much smaller than the coupling expected in pure AlAs. However, there is no available data for samples with Al composition larger than 0.24.

In addition to Raman scattering techniques, it is well known that the radiative recombination of photoexcited carriers with the neutral acceptors can be used to study the hot carrier relaxation processes. The relaxation of hot electrons through optical phonon emission in bulk GaAs [10–12] and heterostructures [13–15] has been extensively studied using the above techniques. Sapega et al. [16] have demonstrated that, for quantum wells with large barrier widths, the energy relaxation mechanism for hot electrons is dominated by the AlAs phonons. For smaller barriers, emission via GaAs phonons is more important. By using conventional hot electron luminescence techniques, Ozturk

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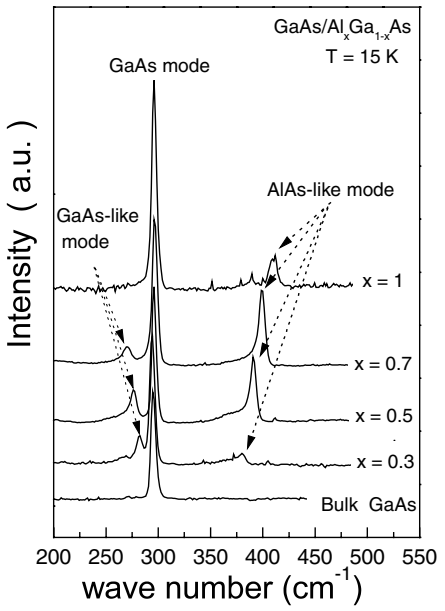

Fig. 1. Raman spectra of four GaAs/Al_xGa_{1-x}As multiple-quantum wells and bulk GaAs samples at 15 K in the back-scattering geometry for incident wavelength of 514.5 nm. The peak labeled GaAs mode is the LO phonon arising from the GaAs wells. The other two peaks labeled GaAs-like and AlAs-like modes are related to the Al_xGa_{1-x}As barrier layers.

et al. [17] have demonstrated that, in GaAs/AlAs quantum wells, the AlAs-like mode has fairly substantial influence on the hot electron relaxation mechanism. Recently, Mirlin et al. [18] have studied electron relaxation in GaAs/AlAs quantum wells with fixed barrier width of 10 nm and well width varying from 4 to 13 nm. It was shown that, for larger wells, the electron relaxation is dominated by GaAs LO phonons. But in the smallest well width sample, it is dominated by AlAs optical phonons. In Ref. [9], it was also shown that the emission of phonons in the barriers by remote interactions does not occur in samples with wider well widths. To our knowledge, there are no investigations of the influence of the Al composition on the electron–LO phonon interactions in GaAs/Al_xGa_{1-x}As quantum well structures.

In this work, we report dependence of the electron–LO phonon interactions on the Al composition in Al_xGa_{1-x}As barrier layers. First, we use Raman spectroscopy to determine the optical phonon energies in GaAs/Al_xGa_{1-x}As quantum well samples with Al composition of $x = 0.3$, 0.5, 0.7 and 1.0, respectively. With the measurements of the energy separation of peaks in the hot electron–neutral acceptor luminescence spectra and the LO phonon energies retrieved via Raman experiments, we then analyze the type of optical phonon emitted by hot electrons during relaxation processes in quantum wells.

2. Experimental techniques

The samples investigated were grown by molecular-beam epitaxy on (100)-oriented undoped semi-insulating GaAs substrate. The four MQW samples studied here were 5 nm GaAs wells, with $x = 0.3$, 0.5, 0.7 and 1.0 Al_xGa_{1-x}As barrier of 12 nm thickness. The central regions of 1 nm of the GaAs layer were doped with Be to 10^{18} cm^{-3}. Two exciting lines were used for the Raman experiments: an Ar^+ laser operated at 514.5 nm and a dye (DCM) laser operated on 655 nm. About 150 mW of the laser power

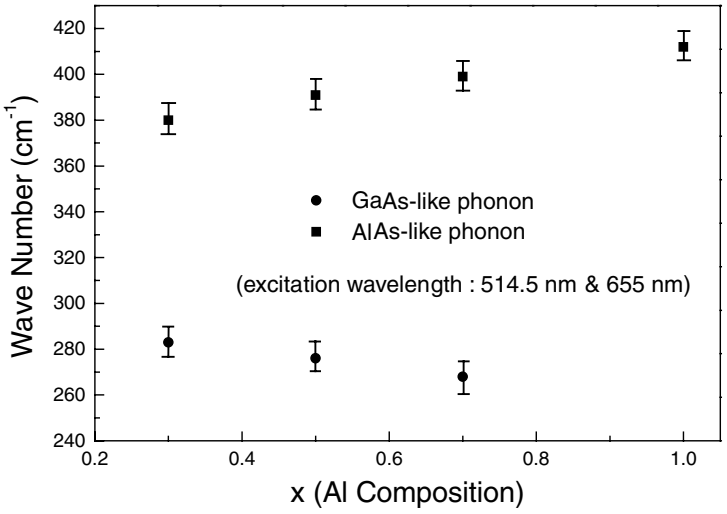

Fig. 2. The AlAs-like LO phonon frequency (square) and GaAs-like LO phonon frequency (circle) as a function of the Al composition for $0 < x \leq 1$ at incident wavelengths of 514.5 and 655 nm.

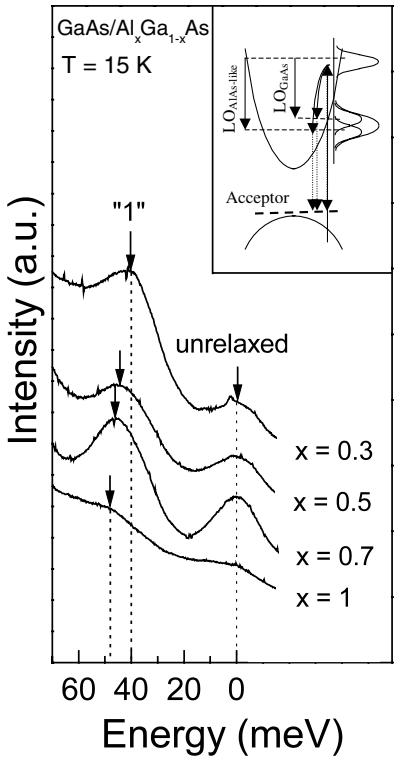

Fig. 3. Hot electron luminescence spectra for four GaAs/$Al_xGa_{1-x}As$ multiple-quantum well samples plotted as a function of the electron energy above the ground state of the quantum wells. The vertical line labeled "unrelaxed" is the energy peak corresponding to recombination at the energy of creation. The peak labeled "1" represents the electron distribution after the emission of one LO phonon. The inset shows schematically the principles of the hot electron–neutral acceptor luminescence technique.

was directed on the samples, which were kept in a closed-cycled refrigerator at 15 K. Raman spectra were obtained in back-scattering geometry and the scattered light was collected by a camera lens and passed through a notch filter before entering the spectrometer. The spectra were recorded with a combination of a SPEX 0.6 m triplemate spectrometer equipped with a liquid nitrogen cooled CCD detector. For the excitation of hot electron–neutral acceptor luminescence, a dye laser (DCM) pumped by an Ar^+ laser was used. The dye laser was operated at appropriate photon energies to excite all four samples in order to give the same amount of excess kinetic energies to electrons. The hot electron luminescence was analyzed with the same spectrometer and detector in the Raman experiments.

3. Results and discussion

The Stokes Raman spectrum measured in back-scattering 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 samples. Fig. 1 shows the Raman spectra for the (50/120) quantum wells of four different Al compositions excited with an Ar^+ laser. On the bottom of the spectra we have placed the Raman spectrum of the bulk GaAs sample for comparison. As the quantum well structures were (001)-oriented, only the LO phonon modes were allowed. The GaAs LO phonon mode is at 36.7 meV and, for the Al_xGa_{1-x} layers, the optical phonons display a two-mode behavior: the GaAs-like (whose energy is below the GaAs LO phonon energy) and AlAs-like modes (whose energy is below the AlAs LO phonon energy). Our detection system is not capable of resolving the splitting of the GaAs LO phonon into confined modes and there is also no evidence of scattering from interface phonons [9]. Note the broadening and asymmetry natures of the peaks which are due to the alloy potential fluctuations [19].

In Fig. 2 we have plotted the AlAs-like and GaAs-like phonon frequencies as a function of Al composition at two excitation wavelengths. The AlAs-like phonon frequencies approach those of the phonons in AlAs as x approaches 1. On the other hand, the GaAs-like phonons have frequencies approaching those of the phonons in GaAs as x approaches zero. We found no dependence of the phonon frequencies on the excitation wavelength. We have also measured the anti-Stokes Raman spectrum, but find no evidence related to the phonon absorption by photons. We attribute this to the vanishingly small thermal occupation of the LO phonon modes at very low temperature.

In Fig. 3 we have shown the hot electron–neutral acceptor luminescence spectra of four samples. The principles of this technique are shown in the inset of Fig. 3 [6]. The peak labeled "unrelaxed peak" in each spectrum corresponds to recombination of electrons, from the state at which they were created, with a neutral acceptor. The peak labeled "1" represents electrons recombining with neutral acceptors after emitting one LO phonon. The width of the peaks is determined by the electron energy distribution at the point of generation, which is related to heavy hole subband warping, as well as the energy distribution of acceptors, the final state of recombination for the hot luminescence process. The power density of the laser used for the excitation was low enough so the main mechanism of energy relaxation in the sample studied is the emission of optical phonons and the phonon–plasmon coupling can be ignored. In order to demonstrate the change of the luminescence spectra with different Al compositions, we have centered the first unrelaxed peaks in the spectra for all four samples. The separation of the "unrelaxed peak" and "1" peaks in the spectra should allow one to determine the energies of the phonons emitted by hot electrons during the relaxation processes. In order to determine the energy separation more accurately, we first subtract the background (which was originated from the band-to-band recombination) from the

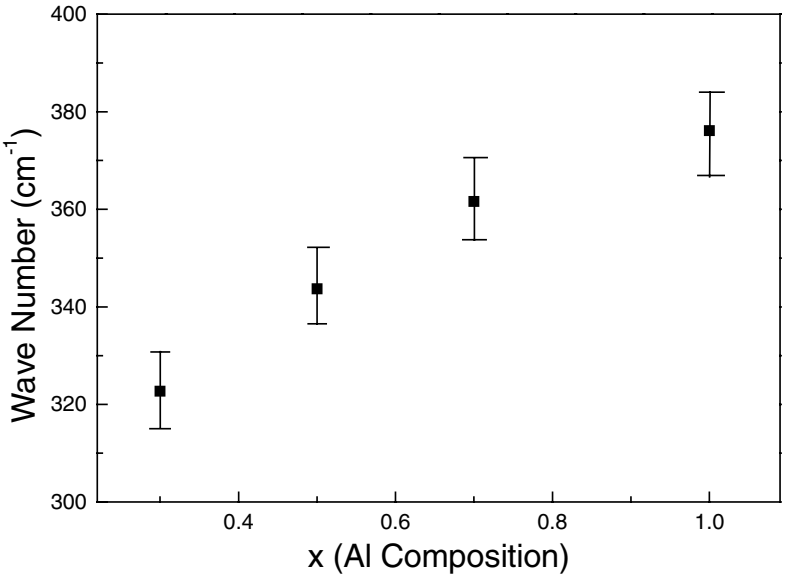

Fig. 4. Measured energy separation between the "unrelaxed" and "1" phonon peaks in the hot electron luminescence spectra as a function of Al composition.

spectra and the energy spectra of the two remaining peaks are then fitted by Gaussian distributions. The energy difference between the two peaks is plotted for all the four samples as a function of Al mole fraction as shown in Fig. 4. For the samples with larger x, the energy separation in the spectrum approaches 400 cm^{-1}, a value in the AlAs phonon regime.

Since GaAs and AlGaAs are polar materials, the phonon modes have a scalar potential Φ associated with them. It is this scalar potential, or equivalently the electric field $E = -\nabla\Phi$, that couples to the electrons by the Fröhlich interactions. In Refs. [4,16], the dispersion curves of the GaAs and AlAs optical phonons have been calculated for GaAs/AlAs multiple-quantum-well structures with different barrier widths. In their calculations, the odd phonon modes have an odd number of anti-nodes in the scalar potential across a particular well layer resulting in an overall macroscopic electrical field, which has a finite value at a distance far from the individual well layers. They argued that it was due to the interactions between this electrical field and the electrical field of the interface modes that the anti-crossings were produced in the phonon dispersions. They also found that, at smaller barrier widths, the upper GaAs interface mode has the largest phonon potential, whereas for large barrier widths, the largest phonon potential is that of the upper AlAs interface mode. In their measurements, the increase of the energy difference between phonon peaks in the hot luminescence spectra as the barrier width is increased was attributed to the increasing scalar potential of the AlAs phonon modes.

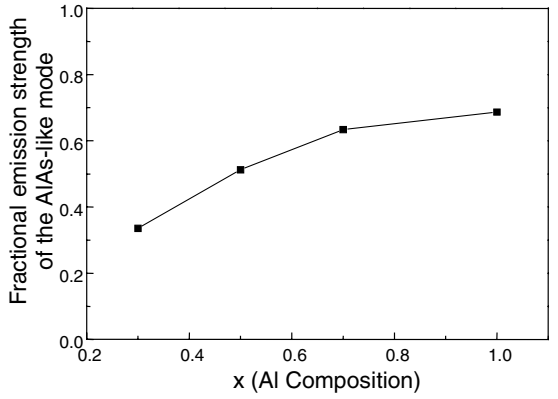

Fig. 5. The estimate relative emission strength of AlAs-like LO phonon mode with electrons as a function of the Al composition.

Even though the well widths and barrier widths of the samples are fixed in our experiments, we have changed the Al composition in the barriers instead. We speculate that the scalar potential of the AlAs-like phonon has been increased as the Al composition in the barriers is increased. Therefore, the Fröhlich interactions between the hot electrons and AlAs-like phonons become stronger and the phonon energy emitted by electrons move toward the AlAs phonon energy (which is about 52 meV in the bulk AlAs). The results have led to the monotonic increase of the energy separation between the phonon peaks in the hot electron luminescence spectra (as shown in Fig. 4). A thorough calculation of the phonon dispersion in quantum wells on the dependence on Al composition in the barriers is currently under investigation.

Nevertheless, if we assume that the emitted phonon

energies by hot electrons (or the energy separations in the hot electron luminescence spectra) are partitioned by the AlAs-like and GaAs LO phonons, whose energies are accurately determined in our Raman scattering measurements, we can estimate the emission strength of AlAs-like LO phonons relative to the GaAs LO phonons for electrons in the wells. In Fig. 5, we have plotted the fractional emission strength of the AlAs-like optical phonon relative to the GaAs LO phonon as a function of barrier width for all samples. In the case of $x = 0.3$, the energy separation of the peaks is about 29 cm^{-1} larger than the energy of the GaAs LO phonons. This indicates that although interaction with the GaAs LO phonon is strong, there is still a significant contribution from the AlAs-like LO phonon. However, for $x = 1.0$, the spectra are dominated by AlAs-like LO phonons and the energy separations are very close to the AlAs LO phonon mode.

Investigations of the GaAs/AlAs multiple-quantum wells by Ozturk et al. [17] have also demonstrated the substantial influence of the AlAs-like LO phonon modes on the hot electron relaxation processes. On the other hand, the GaAs phonons provide the energy relaxation in a similar GaAs/$Al_{0.24}Ga_{0.76}As$ structure. In their works, the predominance of the AlAs-like phonon modes is also attributed to the stronger scattering strength and to their shorter lifetime compared to the GaAs modes. Our results have also indicated that, for quantum wells whose barriers have large Al molar fractions, the hot electrons relax mostly via the AlAs-like optical phonon emission.

4. Conclusions

We have observed phonons in the Raman scattering and hot electron–neutral acceptor luminescence of GaAs/$Al_xGa_{1-x}As$ multiple-quantum wells. In the Raman scattering experiments, the dependence of the mode frequency on the Al composition is the important factor in distinguishing the phonon modes from the bulk optical phonons. We have also demonstrated that, even though the electrons are confined in the wells, they still interact remotely with phonons in the barriers. The interaction strength is a function of Al composition in the barriers. For smaller x in the barrier, the emission of the GaAs optical phonon mode is stronger. But for the largest x investigated, the energy relaxation of hot electrons is dominated by the AlAs-like phonon.

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