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Hot-electron relaxation via optical phonon emissions in GaAs/Al_xGa_{1-x}As quantum well structures: dependence upon the alloy composition and barrier width

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Abstract. We present a systematic investigation of the dependence of the hot-electron–optical-phonon interactions on Al composition and barrier width in GaAs/Al_xGa_{1-x}As MQW structures. Raman scattering measurements at 15 K are presented for samples with different barrier widths and Al composition. The optical phonon energies emitted by the photoexcited electrons in quantum wells were also 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. For samples with larger barriers, the electrons in the GaAs layer relax mostly through the AlAs-like optical phonon emission. However, in samples with smaller barriers, the relaxation of hot electrons is dominated by the GaAs optical phonon emission.

1. Introduction

There has been considerable interest in the problem of electron–optical-phonon interaction in heterostructures. The interaction of electrons with optical phonon modes of layered polar semiconductors (for example, GaAs/AlGaAs) has been the subject of a great deal of both experimental and theoretical work for many years as these modes dominate the energy and momentum relaxation at high fields and temperatures. Theoretically, the controversy over the correct boundary conditions for the long-wavelength confined optical vibrations of these systems has now been clarified by both *ab initio* microscopic calculations [1] and more involved macroscopic approaches [2–4]. The relative importance of interface modes and bulk-like confined modes in single and double heterostructures composed of diatomic polar semiconductor has been studied by Mori and Ando [5].

Experimentally, optical methods such as 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. Among the experimental techniques, Raman scattering has proved to be a versatile and efficient tool

for probing long- and short-wavelength lattice dynamics of ternary alloys [6–10]. The continuous spectrum of acoustical phonons has been reported for single [11] and multiple two-dimensional quantum wells [12–14]. The electron–phonon interactions in semiconductor alloys have also been studied by using time-resolved Raman spectroscopy [16–18]. Kash *et al* [17] 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 are no available data for samples with Al composition larger than 0.24. It is also worth mentioning that, in the time-resolved Raman studies of GaAs/AlAs quantum well systems, Tsen *et al* [19] have demonstrated that, in addition to the GaAs confined mode, interface optical phonon modes are also able to scatter electrons, and in particular that the AlAs-like interface modes dominate the scattering processes for small well widths.

In addition to Raman scattering techniques, it is well known that the radiative recombination of photoexcited

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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 [20–22] and heterostructures [23–25] has been extensively studied using the above techniques. Sapega *et al* [26] demonstrated that, for quantum wells with large barrier widths, the energy relaxation mechanism for hot electrons is dominated by the AIAs phonons. For smaller barriers, emission via GaAs phonons is more important. By using conventional hot-electron luminescence techniques, Ozturk *et al* [27] have demonstrated that, in GaAs/AIAs quantum wells, the AIAs-like mode has a fairly substantial influence on the hot-electron relaxation mechanism. Recently, Mirlin *et al* [28] have studied electron relaxation in GaAs/AIAs quantum wells with a 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 that in the smallest well width sample, it is dominated by AIAs optical phonons. In [18], 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 has been no investigation of the influence of the Al composition on the electron–LO-phonon interactions in GaAs/AI_xGa_{1-x}As quantum well structures.

In this paper, we report on the dependence of the electron–LO-phonon interactions on the Al composition in Al_xGa_{1-x}As barrier layers and barrier width. First, we use Raman spectroscopy to determine the optical phonon energies in GaAs/AI_xGa_{1-x}As quantum well samples. 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 analyse 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 a (100)-oriented undoped semi-insulating GaAs substrate. The multiple quantum well (MQW) samples used in the experiments for investigating the effect of the Al composition on phonon emissions 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 samples used in the studies of the barrier width dependence were 50 Å GaAs wells, with an Al_{0.7}Ga_{0.3}As or AIAs barrier of 5, 25, 50, 120 Å in thickness, respectively. 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 Raman experiments: an Ar⁺ laser was used at 514.5 nm and a dye (DCM) laser operated at 655 nm. About 150 mW of the laser power was directed on the samples which were kept in a closed-cycled refrigerator at 15 K. Raman spectra were obtained in backscattering 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 about 1.893 eV

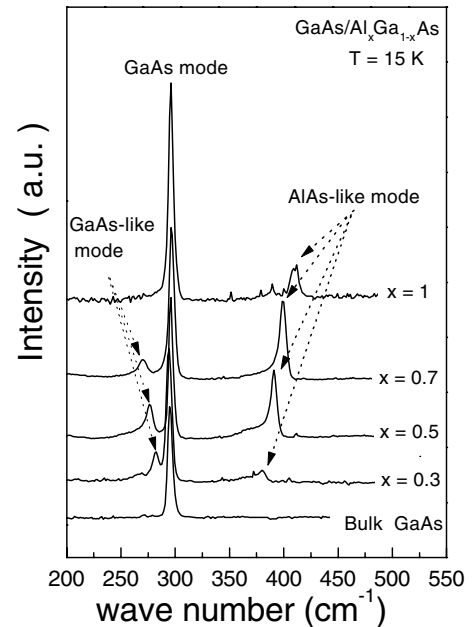


Figure 1. Raman spectra of four GaAs/AI_xGa_{1-x}As MQWs and bulk GaAs samples at 15 K in the backscattering geometry for an incident wavelength of 514.5 nm. The peak labelled GaAs mode is the LO phonon arising from the GaAs wells. The other two peaks labelled GaAs-like and AIAs-like modes are related to the Al_xGa_{1-x}As barrier layers.

with output power of about 100 mW. The hot-electron luminescence was analysed with the same spectrometer and detector in the Raman experiments.

3. Results and discussions

3.1. Dependence on Al composition

In this experiment, 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 samples. Figure 1 shows the Raman spectra for the (50/120) Å quantum wells of four different Al compositions excited with the 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}As layers, the optical phonons display a two-mode behaviour: the GaAs-like (whose energy is below the GaAs LO phonon energy) and AIAs-like modes (whose energy is below the AIAs 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 [18]. Please note the broadening and asymmetric nature of the peaks which is due to the alloy potential fluctuations [29].

In figure 2 we have plotted the AIAs-like and GaAs-like phonon frequencies as a function of Al composition at two

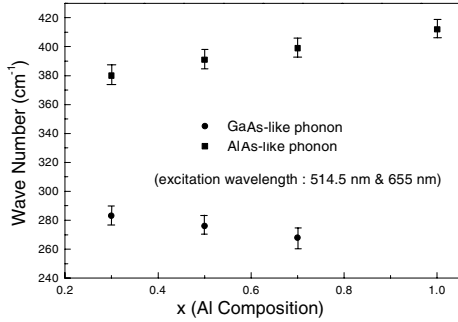


Figure 2. The AlAs-like LO phonon frequency (square) and GaAs-like LO phonon frequency (circle) as a function of the Al composition for $0 < x \leq 1$ at incident wavelengths of 514.5 and 655 nm.

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 that approach those of the phonons in GaAs as x approaches zero. We found no dependence of the phonon frequencies with 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 figure 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 figure 3 [15]. The peak labelled ‘unrelaxed peak’ in each spectrum corresponds to recombination of electrons, from the state in which they were created, with a neutral acceptor. The peak labelled ‘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 such that 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 centred the first unrelaxed peaks in the spectra for four samples. The separation of the ‘unrelaxed’ and ‘1’ peaks in the spectra should allow one to determine the energy 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 originated from the band-to-band luminescence) from the spectra and the energy spectra of the two remaining peaks were then fitted by Gaussian distributions. The energy difference between the two peaks is plotted for all four samples as a function of Al mole fraction as shown in figure 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

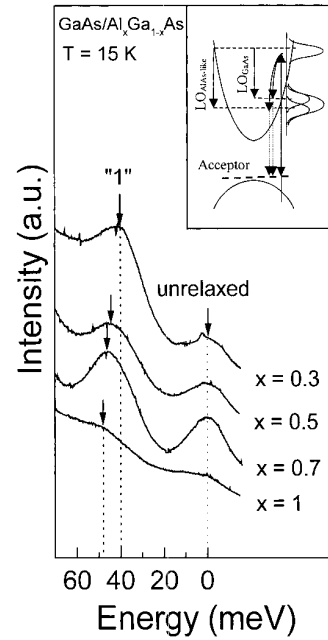


Figure 3. Hot-electron luminescence spectra for four GaAs/Al_xGa_{1-x}As MQW samples plotted as a function of the electron energy above the ground state of the quantum wells. The vertical line labelled ‘unrelaxed’ is the energy peak corresponding to recombination at the energy of creation. The peak labelled ‘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.

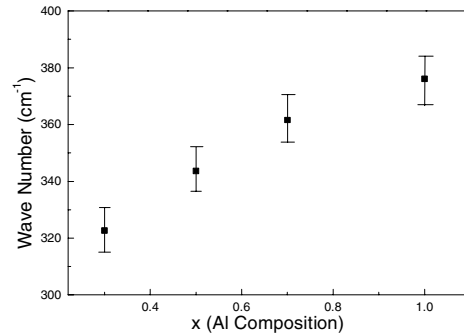


Figure 4. Measured energy separation between the ‘unrelaxed’ and ‘1’ phonon peaks in the hot-electron luminescence spectra as a function of Al composition.

$E = -\nabla\Phi$, that couples to the electrons by the Fröhlich interactions. In [9,26], the dispersion curves of the GaAs and AlAs optical phonons have been calculated for GaAs/AlAs MQW structures with different barrier widths. In their calculations, the odd-phonon modes have an odd number of antinodes 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 this was due to the interactions between this electrical field and the electrical field of the interface modes that have produced the anticrossings 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

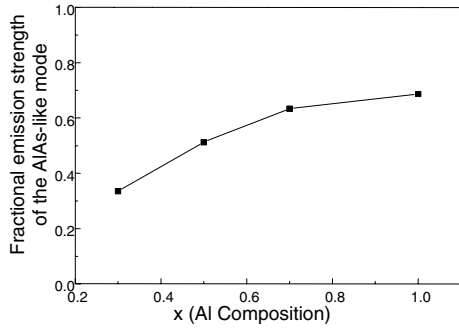


Figure 5. The estimated relative emission strength of the AlAs-like LO phonon mode with electrons as a function of the Al composition.

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.

Although 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 increases 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 moves toward the AlAs phonon energy (which is about 52 meV in the bulk AlAs). The results lead to the monotonic increase of the energy separation between the phonon peaks in the hot-electron luminescence spectra (as shown in figure 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 figure 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 separation is very close to the AlAs LO phonon mode.

Investigations of the GaAs/AlAs MQWs by Ozturk *et al* [27] 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 with 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.

3.2. Dependence on barrier width

In this experiment, the Stokes Raman spectra were also measured in the same backscattering geometry to detect the LO phonon modes of the samples as in the previous experiments. Figures 6(a) and (b) show the Raman spectra for GaAs/Al_{0.7}Ga_{0.3}As and GaAs/AlAs quantum wells samples with different barrier widths excited with an Ar⁺ laser. At the bottom of each figure we have also placed the Raman spectrum of the bulk GaAs sample for comparison. In figure 6(a), the GaAs LO phonon mode is at 36.7 meV and, for the Al_{0.7}Ga_{0.3}As layers, the optical phonons also display a two-mode behaviour. However, in figure 6(b), due to different compositions of the barriers only the AlAs-like and GaAs phonon mode were observed. In figures 7(a) and (b), we have plotted the AlAs-like and/or GaAs-like phonon frequencies as a function of barrier width at two different excitation wavelengths. In figure 7(a), we found that both the GaAs- and AlAs-like phonon frequencies kept relatively constant throughout the whole range of the barrier width. However, for samples with AlAs barrier, the AlAs-like phonon frequencies approach those of the phonons in AlAs with increasing barrier widths as shown in figure 7(b). We also found no dependence of the phonon frequencies with the two different excitation wavelengths, as in the previous experiments.

In figures 8(a) and (b), we have shown the hot electron-neutral-acceptor luminescence spectra from GaAs/AlAs and GaAs/Al_{0.7}Ga_{0.3}As quantum well samples and the first unrelaxed peaks for all four spectra in figures 8(a) and (b) are also centred. The energy difference between the two peaks is determined by following the same procedure used in the previous experiments and is plotted as a function of barrier width as shown in figure 9. For samples with the largest barrier, the phonon energies emitted by the electrons in the GaAs/Al_{0.7}Ga_{0.3}As and the GaAs/AlAs quantum wells approach 360 and 380 cm^{-1} , respectively. In the cases with the smallest barriers, the emitted phonon energies measured from both samples approach 300 cm^{-1} , which is still higher than the GaAs LO phonon energy (293 cm^{-1}).

The experimental results from the GaAs/AlAs samples are in good agreement with earlier works by Sapega *et al* [26]. In their studies, the emission strength of a particular phonon mode is proportional to the square of the overlap integral of the phonon scalar potential with the initial and final electron states in the GaAs layer. The different relaxation paths were weighted by $(\varphi_{\text{Ga}}/\varphi_{\text{Al}})^2$, where φ_{Ga} is the sum for the scalar potentials for all the calculated GaAs modes and φ_{Al} the sum of the AlAs modes at a particular barrier width. In our experiments, the increase of the energy separation between the peaks in the hot-electron luminescence spectra does suggest that the coupling strength between hot electrons and AlAs-like phonon is becoming stronger as the barrier width is increased. In figure 10 we have also plotted the estimated fractional emission strength of the AlAs-like optical phonon relative to the GaAs LO phonon as a function of barrier width for both samples by assuming that the emitted phonon energy

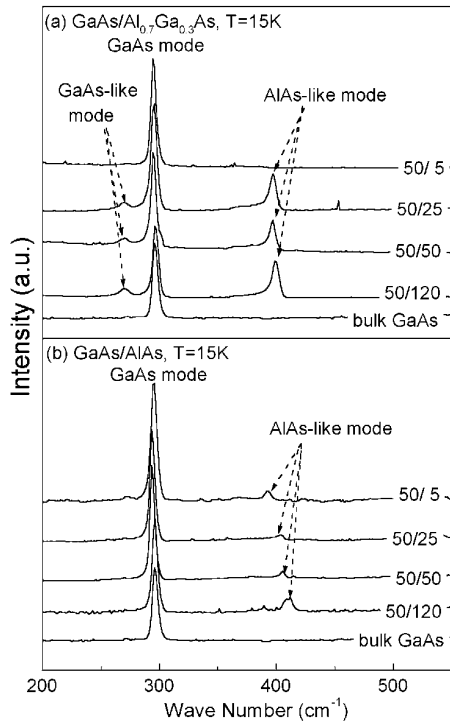


Figure 6. Raman spectra of (a) GaAs/Al_{0.7}Ga_{0.3}As MQWs and bulk GaAs samples and (b) GaAs/AlAs MQWs and bulk GaAs samples at 15 K in the backscattering geometry for an incident wavelength of 514.5 nm. The peak labelled GaAs mode in (a) is the LO phonon arising from the GaAs wells. The other two peaks labelled GaAs-like and AlAs-like modes are related to the Al_{0.7}Ga_{0.3}As barrier layers. Only the AlAs-like LO phonon mode was observed from the barriers in (b). The notations (50/5, 50/25, 50/50, 50/120) in the figure represent samples with fixed well widths of 50 Å and barrier widths varied from 5 to 120 Å.

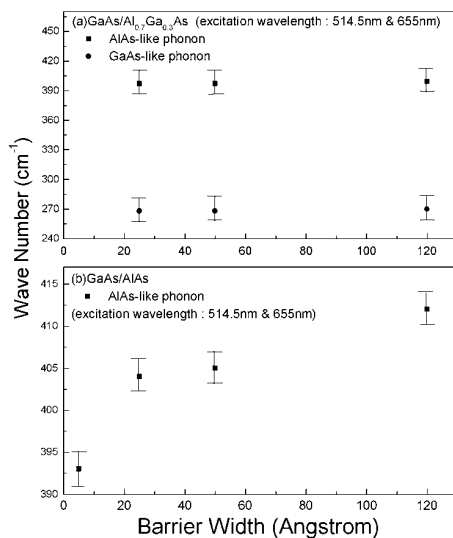


Figure 7. The AlAs-like LO phonon frequencies (squares) and GaAs-like LO phonon frequencies (circles) from (a) GaAs/Al_{0.7}Ga_{0.3}As MQWs and (b) GaAs/AlAs MQWs were plotted as a function of barrier width at incident wavelengths of 514.5 and 655 nm.

was partitioned by AlAs-like and GaAs LO phonons. In the case for the smallest barrier width, the energy separation of

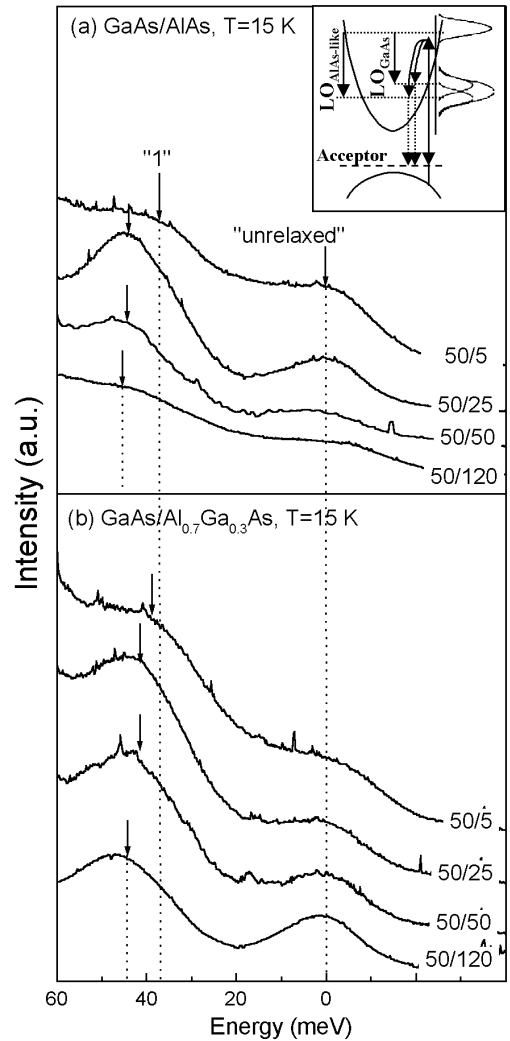


Figure 8. Hot-electron luminescence spectra of (a) GaAs/AlAs MQWs and (b) GaAs/Al_{0.7}Ga_{0.3}As MQWs plotted as a function of the electron energy above the ground state of the quantum wells. The notations (50/5, 50/25, 50/50, 50/120) in the figure represent samples with fixed well widths of 50 Å and barrier widths varied from 5 to 120 Å.

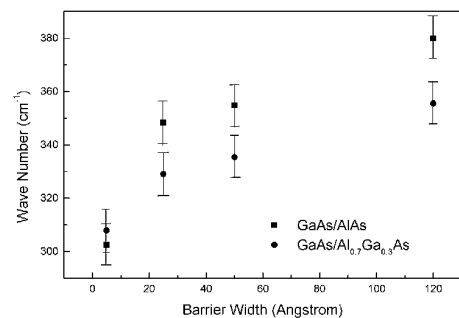


Figure 9. Measured energy separations between the 'unrelaxed' and '1' phonon peaks in the hot-electron luminescence spectra from GaAs/Al_{0.7}Ga_{0.3}As (circles) and GaAs/AlAs (squares) quantum wells as a function of barrier width.

the peaks is about 13 cm⁻¹ larger than the energy of the GaAs LO phonons which indicates that although interaction with

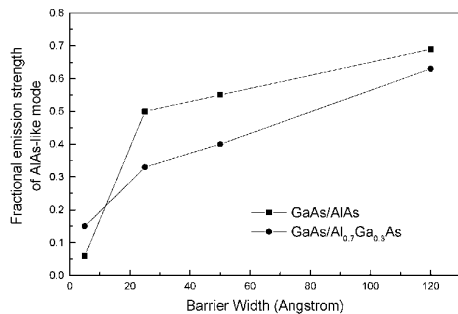


Figure 10. The estimated emission strength of the AlAs-like LO phonon mode relative to the GaAs LO phonon as a function of the barrier width from both samples.

the GaAs LO phonon is strong, there is still a significant contribution from the AlAs-like LO phonon. However, for the larger barrier, the spectra are dominated by AlAs-like LO phonons and the energy separations are very close to the AlAs LO phonon mode.

We have also found that the fraction of the AlAs-like mode increases more rapidly in samples with AlAs barriers as the barrier width is increased in comparison with the GaAs/Al_{0.7}Ga_{0.3}As quantum wells. Our results indicate that, for quantum wells with a fixed well width of 50 Å, the hot electrons relax mostly via the AlAs-like optical phonon emission when the AlAs barrier has a width larger than 25 Å. However, for samples with Al_{0.7}Ga_{0.3}As barriers, the AlAs-like mode emission dominates the relaxation processes only when the barrier width is over 100 Å.

4. Conclusion

In conclusion, we have observed phonons in the present Raman scattering and hot-electron-neutral-acceptor luminescence investigation of the GaAs/Al_xGa_{1-x}As MQWs. In the Raman scattering experiments, the dependence of the mode frequency on the Al composition and barrier width 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 was measured as a function of Al composition and barrier width. 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. On the other hand, for a smaller barrier, the emission of the GaAs optical phonon mode is stronger. But for the largest barrier investigated, the energy relaxation of hot electrons is dominated by the AlAs-like phonon.

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