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Optical phonon emission in GaAs/AlAs and $GaAs/Al_{0.7}Ga_{0.3}As$ multiple quantum well structures

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Abstract

We have performed Raman scattering measurements and hot electron–neutral acceptor (hot(e, Å)) luminescence experiments on Be-doped GaAs/AlAs and $GaAs/Al_{0.7}Ga_{0.3}As$ multiple quantum well structures, with fixed well width of 50 Å and barrier thickness of 5, 25, 50,120 Å, to determine the optical phonon energy emitted by the hot electrons excited in the quantum wells. It was shown that the relaxation of electrons in the GaAs layer is dominated by the AlAs-like optical phonon emission for samples with larger barriers, but by GaAs optical phonons for smaller barriers. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Semiconductor quantum well structures such as GaAs/AlGaAs systems not only have important device applications, but also provide a challenge to our understanding of the physics of hot carrier dynamics in semiconductors. 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. Raman scattering has been proven as a versatile and efficient tool for probing long- 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 [6–8]. It is well known that the $Al_xGa_{1-x}As$ barrier in the quantum wells is a two-mode alloy, in that two longitudinal–optical (LO) phonon modes of lattice vibration exist, even though the material is assumed to be a smooth alloy with average Al/Ga concentration on a single sublattice. More recently, Kash et al. [7] were able to evaluate the relative interaction strength of these two LO modes, with the electrons, using picosecond Raman spectroscopy.

In addition to the Raman scattering technique, it is well known that the radiative recombination of photoexcited carriers with the neutral acceptors

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can be used to the study the hot carrier relaxation processes. The relaxation of hot electrons through optical phonon emission in bulk GaAs [9–11] and heterostructures [12–14] has been extensively studied using above techniques. Sapega [15] has demonstrated that, for GaAs/AlAs 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 phonon is more important. By using conventional hot electron luminescence techniques, Ozturk et al. [16] have demonstrated that in GaAs/AlAs the dominant electron relaxation mechanism is via the interaction with the AlAs interface mode for a device having a well width of 80 Å. But for a similar GaAs/$Al_{0.24}Ga_{0.76}As$ structure, the GaAs phonons provide the energy relaxation. Recently, Mirlin et al. [17] 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.

2. Experimental techniques

In this work, we first use Raman spectroscopy to determine the optical phonon energies in GaAs/$Al_{0.7}Ga_{0.3}As$ and GaAs/AlAs quantum well samples of different barrier widths. 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 the quantum wells.

The samples investigated were grown by molecular-beam epitaxy on (1 0 0)-oriented undoped semi-insulating GaAs substrate. The MQW samples studied here were 50 Å GaAs wells, with $Al_{0.7}Ga_{0.3}As$ or AlAs 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 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 was directed on the samples which were kept in a closed-cycle 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 samples in order to give 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 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 (1 1 0) in the plane of the layers) detects the LO phonon modes of the samples. Fig. 1(a) and (b) show the Raman spectra for GaAs/$Al_{0.7}Ga_{0.3}As$ and GaAs/AlAs quantum well samples with different barrier widths excited with Ar^+ laser. At the bottom of each figure we have placed the Raman spectrum of the bulk GaAs sample for comparison. In Fig. 1(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 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). However, in Fig. 1(b), due to different compositions of the barriers only the AlAs-like and the GaAs phonon mode were observed. 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 [8]. Please

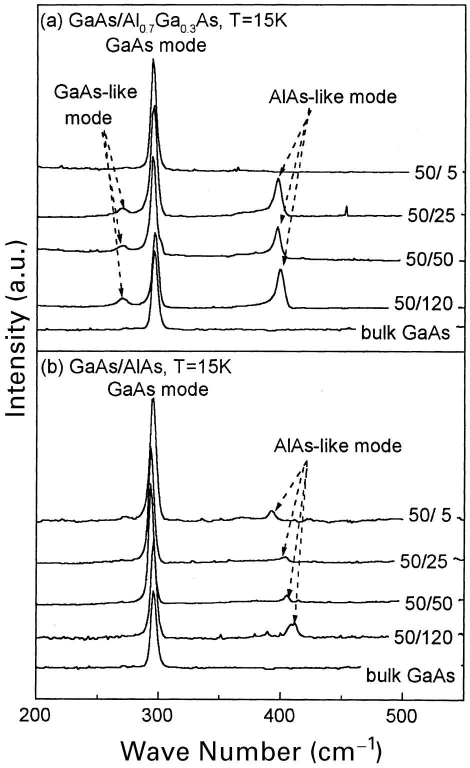

Fig. 1. Raman spectra of (a) GaAs/$Al_{0.7}Ga_{0.3}As$ multiple quantum wells and bulk GaAs samples and (b) GaAs/AlAs multiple quantum wells and bulk GaAs samples at 15 K in the back scattering geometry for incident wavelength of 514.5 nm. The peak labeled GaAs mode in (a) is the LO phonon arising from the GaAs wells. The other two peaks labeled GaAs-like and AlAs-like modes are related to the $Al_{0.7}Ga_{0.3}As$ barrier layers. Only the AlAs-like LO phonon mode was observed from the barriers in (b).

note the broadening and asymmetry nature of the peaks which is due to the alloy potential fluctuation [18].

In Fig. 2(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 Fig. 2(a), we found that both the GaAs-like and AlAs-like phonon frequencies kept relatively constant throughout the whole range of the barrier width. However, for samples with AlAs barriers, the AlAs-like phonon frequencies approach those of the phonons in AlAs with increasing barrier widths as shown in Fig. 2(b). No dependence of the phonon frequencies was found with the two different excitation wavelengths. We have also measured the anti-Stokes Raman spectra, but found 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.

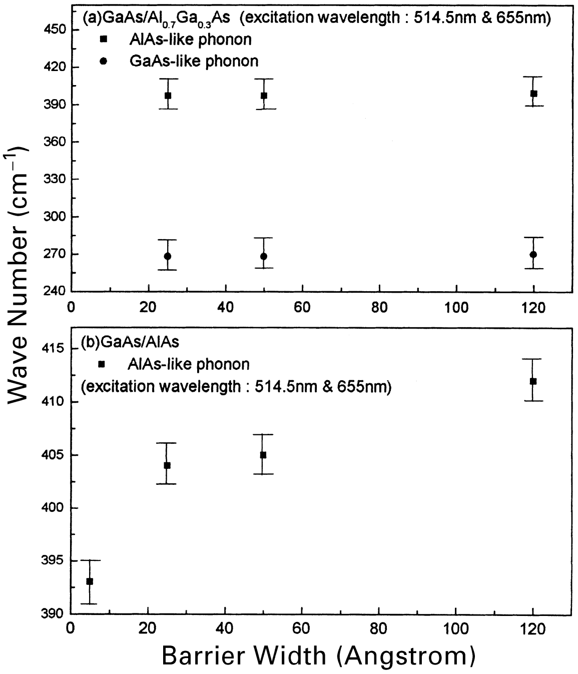

Fig. 2. The AlAs-like LO phonon frequencies (square) and GaAs-like LO phonon frequencies (circle) from (a) GaAs/$Al_{0.7}Ga_{0.3}As$ multiple quantum wells and (b) GaAs/AlAs multiple quantum wells were plotted as a function of barrier width at incident wavelengths of 514.5 and 655 nm.

In Figs. 3 (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. The principles of this technique were given in Ref. [19]. The schematic of this technique is also shown in the inset of Fig. 3(a). 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,

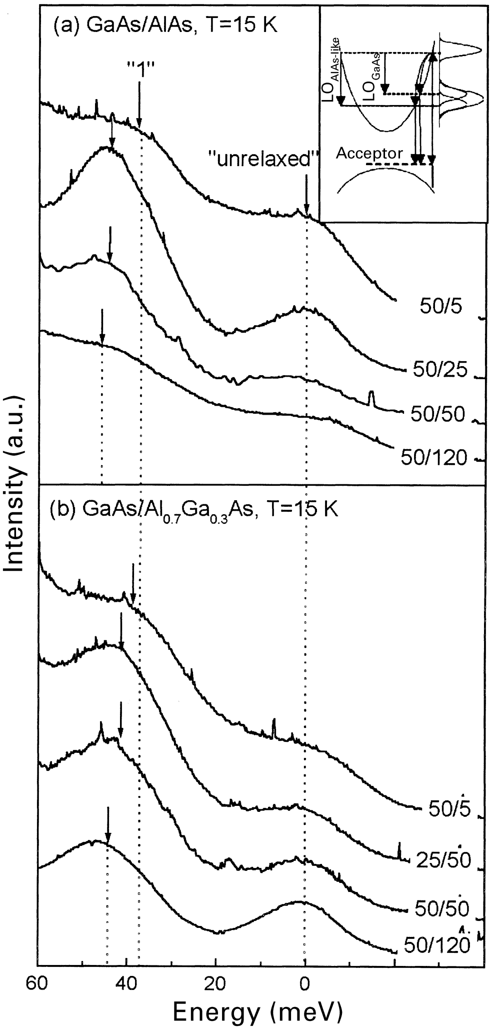

Fig. 3. Hot electron luminescence spectra of (a) GaAs/AlAs multiple quantum wells and (b) GaAs/$Al_{0.7}Ga_{0.3}As$ multiple quantum wells plotted as a function of the electron energy above the ground state of the quantum wells. The vertical line labeled "unrelaxed" is the energy peak corresponding to 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.

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 photoexcited carrier densities are determined from the laser spot size on the sample and the absorption coefficient of the QW samples at the excitation wavelength. The power density of the laser used for the excitation was about 10 W cm^{-2}, which resulted in a 2D carrier density of about $10^9 cm^{-2}$. Our photoexcited carrier densities are low enough so 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 barrier widths and 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 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 was originated from the band-to-band recombination) 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 the samples as a function of barrier width as shown in Fig. 4. 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 smallest barriers, the emitted phonon

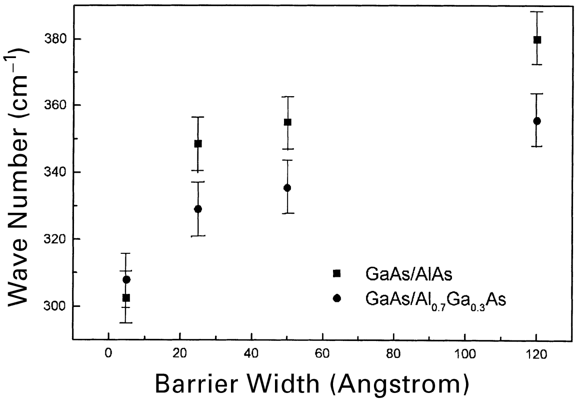

Fig. 4. Measured energy separations between the "unrelaxed" and "1" phonon peaks in the hot electron luminescence spectra from GaAs/$Al_{0.7}Ga_{0.3}As$ (dot) and GaAs/AlAs (square) quantum wells as a function of barrier width.

energies measured from both samples approach $300\,\mathrm{cm}^{-1}$, which is still higher than the GaAs LO phonon energy – $293\,\mathrm{cm}^{-1}$.

In according to Ref. [15], 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 $(\varphi_{\mathrm{Ga}}/\varphi_{\mathrm{Al}})^2$, 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 particular barrier width. In their works, for samples with smaller barrier widths, relaxation through the emission of GaAs modes is more important. But, for the 8 nm barrier widths, the energy relaxation was dominated by the AlAs phonons.

In our experiments, the monotonic increase of the energy separation between the peaks (the phonon energies emitted by hot electrons) in the hot electron luminescence spectra suggests that the coupling strength between hot electrons and AlAs-like phonons is becoming stronger as the barrier width is increased. Therefore, we can estimate the emission strength of AlAs-like LO phonons relative to the GaAs LO phonons by taking into account the optical phonon energies measured in the Raman experiments and the energy separations in the hot electron luminescence spectra. 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 both samples by assuming that the emitted phonon energy was partitioned by AlAs-like and GaAs LO phonons. In the case for the smallest barrier width, the energy separation of the peaks is still about $13\,\mathrm{cm}^{-1}$ 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 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 comparing to the GaAs/$Al_{0.7}Ga_{0.3}As$ quantum wells.

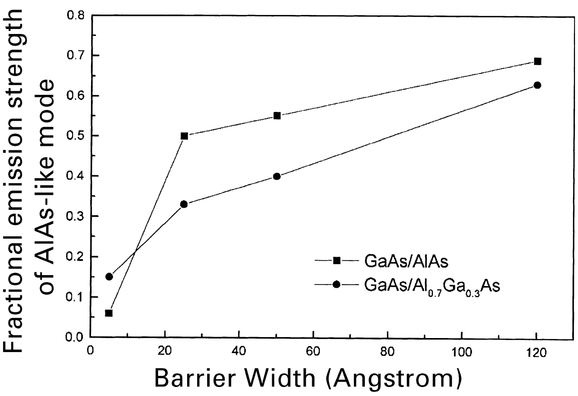

Fig. 5. The estimate emission strength of the AlAs-like LO phonon mode relative to the GaAs LO phonon as a function of the barrier width from both samples.

Investigations of the GaAs/AlAs multiple-quantum wells via high-temperature mobility measurements [16] have also demonstrated the substantial influence of the AlAs-like LO phonon modes on the hot electron relaxation processes. On the contrary, the GaAs phonons provide the energy relaxation in a similar GaAs/$Al_{0.24}Ga_{0.76}As$ structure. The predominance of the AlAs-like phonon modes is attributed to the stronger scattering strength and to their shorter lifetime compared to the GaAs modes. Our results have indicated that, for quantum wells with AlAs barriers, the hot electrons relax mostly via the AlAs-like optical phonon emission with barrier width larger than 25 Å. However, in similar 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 larger than 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/$Al_{0.7}Ga_{0.3}As$ and GaAs/AlAs multiple-quantum wells. In the Raman scattering experiments, we have observed two-mode behavior from $Al_{0.7}Ga_{0.3}As$ barriers, but only one LO phonon mode is observed from AlAs barriers. The AlAs- and GaAs-like phonon frequencies were measured as a function of barrier width. We have also demonstrated that, for 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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