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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 compositions. 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.

Introduction

There has been a great number of experimental and theoretical studies focused on optical phonons in quantum wells and their interactions with electrons. Raman scattering has been proven as a versatile and efficient tool for probing long-wavelength and short-wavelength lattice dynamics of ternary alloys. The electron-phonon interactions in semiconductor allovs have also been studied by using time-resolved Raman spectroscopy. 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 the study the hot carrier relaxation processes. The relaxation of hot electrons through optical phonon emission in bulk GaAs [1, 2] and heterostructures [3, 4] has been extensively studied using above techniques. Sapega [5] has 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 phonon is more important. By using conventional hot electron luminescence techniques, E. Ozturk et al. [6] 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.

1. Experimental techniques

In this work, we first use Raman spectroscopy to determine the optical phonon energies in GaAs/Al_xGa_{1-x}As quantum wells samples with Al composition of x = 0.3, 0.5, 0.7 and 1.0. 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



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 peak labeled GaAs like and AlAs-like mode are related to the $Al_xGa_{1-x}As$ barrier layers.



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

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 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. 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 same amount of excess kinetic energies to electrons.

2. Results and discussion

Figure 1 shows the Raman spectra for the (50/120) Å quantum wells of four different Al composition excited with Ar⁺ laser. On the bottom of the spectra we have placed the Raman spectrum of the bulk GaAs sample for comparison. 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 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.

In Fig. 2 we have plotted the AlAs-like and GaAs-like phonon frequencies as a func-



Fig. 3. Hot electron luminescence spectra for four $GaAs/Al_xGa_{1-x}As$ multiple quantum wells samples plotted as a function of the electron energy above the ground state of the quantum wells. The inset shows schematically the principles of the hot electron neutral acceptor luminescence technique.

tion of Al composition at two excitation wavelengths. The AlAs-like phonon frequencies approach those of the phonons in AlAs as *x* approach 1. On the other hand, the GaAs-like phonons have frequencies approach those of the phonons in GaAs as *x* approach 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 Fig. 3 we have shown the hot electron-neutral acceptor luminescence spectra of four samples. The principles of this technique [6] are shown in the inset of Fig. 3. 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 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 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 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.

The monotonic increase of the energy separation between the peaks (the phonon energies emitted by hot electrons) in the hot electron luminescence spectra (as shown in Fig. 4) suggests that the coupling strength between hot electrons and AlAs-like phonons is becoming stronger as the Al composition is increased. Therefore, we can estimate the emission strength of AlAs-like LO phonons relative to the GaAs LO phonons by taking into



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

account the optical phonon energies measured in the Raman experiments and the energy separations in the hot electron luminescence spectra. In the case for 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 are very close to the AlAs LO phonon mode.

3. Conclusion

In conclusion, we have observed phonons in the present Raman scattering and hot electronneutral acceptor luminescence investigation of the 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, 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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