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Raman spectrum study of δ-doped GaAs/AlAs multiple-quantum wells*

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Three samples of GaAs/AlAs multiple-quantum wells with different quantum well widths and δ-doped with Be acceptors at the well center were grown on (100) GaAs substrates by molecular beam epitaxy. Polarized Raman spectra were recorded on the three samples at temperatures in a range of 4–50 K in a backscattering configuration. The two branches of coupled modes due to the interaction of the hole intersubband transitions and the quantum-well longitudinal optical (LO) phonon were observed clearly. The evaluation formalism of the Green function was employed and each lineshape of the Raman spectrum of the coupled modes was simulated. The dependence of the peak position of Raman shifts of the two coupled modes as well as the quantum-well LO phonon on the quantum-well size and measured temperature were given, and the coupling interaction mechanism between the hole subband transitions and the quantum-well LO phonon was researched.

Keywords: coupled mode, Raman spectrum, δ-doped GaAs/AlAs multiple quantum wells

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1. Introduction

Semiconductor quantum wells and superlattices have attracted a great deal of interest due to their size tunable electronic structures and optical properties, which make them ideal building blocks for a wide variety of optoelectronic devices.[1–5] For example, the resonant-phonon teraherz quantum cascade laser of GaAs–GaAlAs quantum wells utilizes the optical phonon of the adjacent well to achieve electron depopulation for obtaining the population inversion.[6] Therefore, it is important to study the phonons as well as their coupling to the electronic (or hole) intersubband transitions in the GaAs–GaAlAs quantum wells or superlattices. The confined longitudinal optical (LO) and transverse optical (TO) phonon modes in GaAs–GaAlAs multiple-quantum wells have been studied extensively by Raman scattering spectroscopy, and the properties of these phonon modes have been discussed theoretically.[7–10] Some new interface (IF) phonon modes have also been observed in GaAs–GaAlAs quantum wells and superlattices.[11–13] In addition, the coupling of the electronic (or hole) intersubband transitions and the quantum-well LO in n-type modulation-doped GaAs–GaAlAs multiple-quantum wells were also investigated by resonant Raman spectroscopy and more related papers have been published.[14–16] The dependence of two branches of coupled modes, I_+ and I_-, on the incident laser power density and an external magnetic field has been researched.[17–19] However, for the case of GaAs–GaAlAs multiple quantum wells with p-type doping at the quantum-well centers, very few studies have been carried out because of the complicated valence-band four-fold degenerate of the bulk GaAs.

In this paper, we measure polarized Raman spectra at temperatures in a range from 4 K to 50 K in backscattering geometry for three samples of GaAs–GaAlAs multiple quantum wells with different quantum-well widths and δ-doped with Be acceptors at the quantum-well centers. The two branches of coupling modes I_+ and I_- due to the interaction of the hole intersubband transitions and quantum-well LO phonons as well as the quantum-well LO phonon are observed clearly. The evaluation formalism of the Green function is employed and each individual Raman spectra is simulated. The peak position of the Raman shifts of the two coupled modes I_+ and I_- is studied in more detail as a function of the quantum-well size and the measured temperature. Our work is significant for analyzing and improving the performance of opto-electronic devices based on GaAs–GaAlAs multiple quantum wells or superlattices.

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2. Experimental details

Three GaAs/AlAs multiple-quantum well samples with different quantum-well widths used in the experiments, namely, samples A, B, and C, were grown on (100)-oriented semi-insulating GaAs substrates by molecular-beam epitaxy with Be acceptors δ-doped at the quantum-well center. The growth of the layers was performed under the exact stoichiometric condition using the technique of low-temperature growth, which ensured high-quality optical materials even at relatively low growth temperatures. Under these conditions, the quantum-well structures were grown at 550 °C or 540 °C without interruptions at the quantum well interfaces, which ensured negligible diffusion of the Be δ-doping layers. Prior to the growth of the multiple-quantum wells, a GaAs buffer layer of 300 nm was grown. Each of the multiple-quantum well structures investigated contained an identical 5-nm-wide AlAs barrier, while each GaAs well layer was δ-doped at the well center with Be acceptor atoms. The Be δ-doped level is 5×10^{10} cm^{-2} for the three samples investigated. The quantum-well width is $L_{\rm w} = 10$ nm with 200 periods for sample A, $L_{\rm w} = 15$ nm with 50 periods for sample B, and $L_{\rm w} = 20$ nm with 40 periods for sample C.

Our Raman scattering experiments were performed using a Renishaw Raman imaging microscope system. The slit width used here gave a resolution of about 0.5 cm^{-1}. The samples investigated were excited by an argon ionic laser with a wavelength of 514.5 nm. The laser power of about 20 mW was focused onto the sample, which was mounted on a cold finger of a continuous flow helium cryostat, and the light returning back from the sample was collected and passed into a spectrograph for analysis. The polarized Raman spectra were recorded at the temperatures in the range of 4–50 K in the $z(x,x)\bar{z}$ backscattering configuration, where the light polarization x corresponds to the (1 0 0) axis.

3. Experimental results and discussion

3.1. Quantum-well width dependence of Raman spectra

In Fig. 1, we show the experimental results of the polarized Raman scattering from samples A, B, and C with quantum-well widths of 10 nm, 15 nm, and 20 nm, respectively. The three samples studied were excited by the 514.5 nm line of an Ar-ion laser with a power of about 20 mW. The spectral data were taken in the frequency range between 50 cm^{-1} and 1000 cm^{-1} at temperature 4 K in the backscattering $z(x,x)\bar{z}$ configuration and averaged over 20-time runs. For sample A, the dashed line crossing the picture indicates the hot-luminescence background, which comes from the transitions between the conduction and valence subbands, and is always observed in the n-type doped bulk GaAs (20). On the top of this hot-luminescence background, four distinct peaks labeled by LO, I_+, I_-, and H are superimposed. The sharp line at 293 cm^{-1} denoted LO arises from the GaAs-like LO phonon, namely, the quantum-well LO phonon, which is shifted to lower frquency by -2 cm^{-1} as compared with the frequency (295 cm^{-1}) of un-doped bulk GaAs. This value of 293 cm^{-1} presented here is in good agreement with that observed by Kraus *et al.* in p-type modulation-doped GaAs–GaAlAs multiple quantum wells with the well width of $L_{\rm w} = 10.0$ nm.[19] However, the GaAs-like TO-phonon mode is not observed because of the forbidden selection rule of Raman scattering from the (100) surface in the backscattering geometry. The AlAs-like LO-phonon mode from the AlAs barrier layers is completely absent. In addition, a weaker broad peak around 415 cm^{-1} labeled I_+ appears on the high-frequency side of the quantum-well LO phonon, while another broadband peak denoted I_- around 265 cm^{-1} appears on the low-frequency side of the quantum-well LO phonon, which is slightly below the TO-phonon frequency (268 cm^{-1}) of the bulk GaAs. Referring to Raman studies for the many-particle interactions in the two-dimensional electron gases in n-doped GaAs–GaAlAs quantum-well structures,[14–16] both I_+ and I_- peaks are assigned to the upper and lower longitudinal branches of the coupled modes of the hole intersubband transitions and quantum-well LO phonons, respectively, because the behaviors of both modes coincide with the results, which are well understood for the two coupled modes in heavily doped n-type GaAs–GaAlAs multiple quantum wells. The frequency of the upper branch is much higher as usual than that of the LO-phonon of bulk GaAs, while that of the lower branch is closely equal to the bulk GaAs TO phonon frequency. Furthermore, the band peak around 586 cm^{-1} labelled H originated from the group of second-order optical phonons.

As shown in Fig. 1, the quantum-well LO phonon is also observed for the three samples investigated and its position is not found to shift with increasing quantum-well width except for its intensity increasing. Compared to the LO phonon line, the upper branch I_+ of the coupled modes shifts to the lower frequency from 415 cm^{-1} to 354 cm^{-1} as the quantum-well size increases from 10 nm to 20 nm, while its intensity is enhanced and its shape grows into a broadband from a broad peak. However, the lower branch I_- separates gradually from the LO phonon line and also shifts to the lower frequency with increasing quantum-well width, while its shape is changed into a shoulder peak from the initial broadband. These phenomena are interpreted as follows: a macroscopic electrical field is simultaneously attached to the LO phonon. This field will give rise to a strong coupling between the hole intersubband transitions and the quantum-well LO phonons. A mutual renormalization of the coupled modes in the quantum wells will

result, which causes the two branches of the coupled modes to shift to the lower frequency relative to the quantum-well LO phonon line.[21] For the p-type doped GaAs–GaAlAs quantum wells, the hole subbands are quantized in the z direction oriented parallel to the growth direction of the well layers, but the dispersion in the x–y plane is continuous and mixed strongly at a finite in-plane wavevector.[22] As a result, this results in a broadband (or broad peak) of the coupled modes in the polarized Raman spectra, superimposed by hot-luminescence transitions from the higher conduction-subband states. For sample A with the smallest quantum-well size among the three samples, the hole subbands are separated more clearly from each other. This brings about the relative narrower peaks of the coupled modes than those of the other two samples. With the increase of the quantum-well size, the quantum-confined potential for the hole carriers becomes gradually weak, and there are more hole subbands appearing in a quantum well. Therefore, the probability with the hole subband transition in resonance to the quantum-well LO phonon grows and results in the intensities of the two branches of coupled modes to strengthen with increasing quantum-well width. The Lorentzian-type symmetry of the LO phonon profile changes into an asymmetry as the quantum-well width increases, which has been investigated for binary semiconductors.[23] The Raman spectra also indicate the excitations from the second-order optical-phonon groups labelled by H for the three samples. It can be seen obviously that the H-band-peak position does not vary as the quantum-well width increases, but its intensity is enhanced.

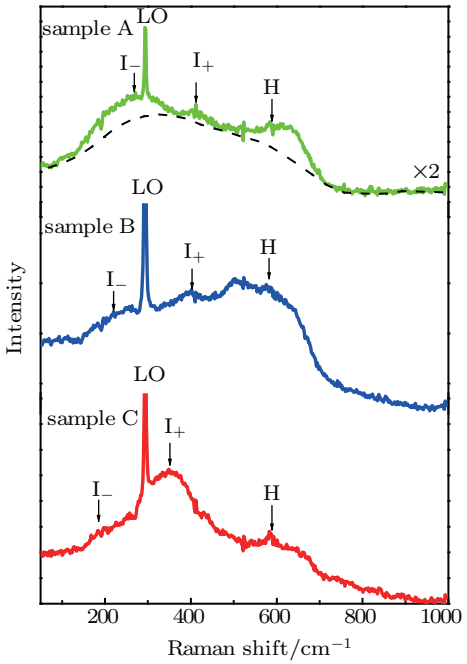


Fig. 1. (color online) Raman spectra taken from samples A, B, and C with the quantum-well widths of 10 nm, 15 nm, and 20 nm, respectively, in the $z(x,x)\bar{z}$ backscattering configuration at 4 K. The samples were excited using the 514.5 nm line of an Ar-ion laser with the power of 20 mW.

3.2. Temperature dependence of Raman spectra

In order to further investigate the behavior of the two coupled modes as well as the quantum-well LO phonon, the Raman spectra at different temperatures were taken for the three studied samples. Figure 2 displays the polarized Raman spectra averaged over 20-time runs from sample C as a function of the measured temperature in the $z(x,x)\bar{z}$ backscattering configuration. The sample was still excited using the 514.5-nm line of an Ar-ion laser with the power of 20 mW, whose photon energy (2.41 eV) is above the band gap of bulk AlAs. It is noted that the frequency of the quantum-well LO phonons is not found to shift obviously when the sample temperature rises from 4 K to 50 K, while its intensity decreases significantly. In general, the lattice constant and dielectric function of the materials constructing the GaAs–AlAs multiple quantum wells are dependent on the temperature, for example, the lattice parameters and thermal expansion coefficients of GaAs and AlAs are 0.5654 nm and 0.5661 nm at room temperature, and 5.8×10^{-6} °C^{-1} and 5.28×10^{-6} °C^{-1}, respectively. A shift of the quantum-well LO phonon frequency with temperature should be clearly observed, but in fact it is on the contrary. According to the investigations of Hu *at al.* on the temperature dependence of the frequency of the GaAs-like LO phonon in chirped GaAs–AlAs superlattices, it can be noted that the frequency of the LO-phonon line shifts to the lower frequency as the measured temperature increases. When the measured temperature is 50 K, the shift of frequency relative to the Raman phonon frequency at 0 K is about -0.01 cm^{-1}.[12] This shift is so small that it cannot be resolved by our Raman spectrometer with a resolution of about 0.5 cm^{-1}. Furthermore, the upper

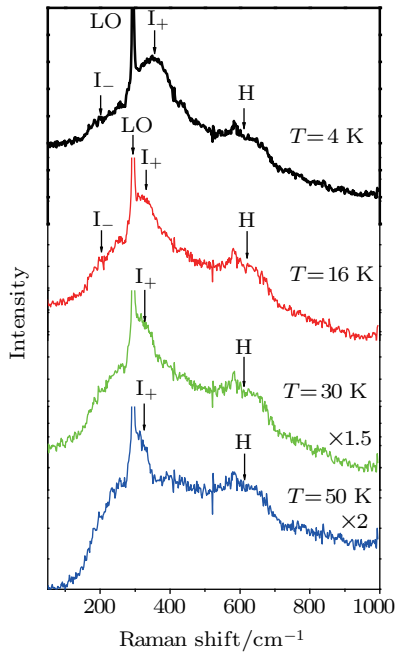


Fig. 2. (color online) Raman spectra recorded from sample C in the $z(x,x)\bar{z}$ backscattering configuration at different temperatures from 4 K to 50 K. The samples were excited using the 514.5 nm line of an Ar-ion laser with the power of 20 mW.

branch I_+ of the coupled modes moves gradually towards the LO phonon line with increasing measured temperature, while its intensity decreases. The lower branch I_- of the coupled modes also decreases in its intensity, and becomes more and more difficult to resolve as the sample temperature increases. The interpretation of these behaviors will be given in the following Subsection 3.3. The H position of the second-order phonon group is independent of the temperature variation, but its intensity also becomes weak with increasing temperature.

3.3. Theoretical analysis of Raman spectra with two coupled modes

For a coupled two excited-level system in which higher-order phonon oscillator states are ordinarily neglected, it has a ground state $|g\rangle$, an excited electronic (or hole) state $|e(n)\rangle$ with excitation energy E_e, and a one-phonon excited state $|p\rangle$ with energy E_p. If the transitions to states $|p\rangle$ and $|e(n)\rangle$ from the ground state $|g\rangle$ are Raman-active, the electron–phonon coupled interaction will produce a mutual repulsion of the levels E_e and E_p to positions E_+ and E_-, respectively. Then, the energies of the two coupled modes, E_+ and E_-, are the roots of the following secular equation:

$$(E_p - E)(E_e - E) - V^2 = 0, \tag{1}$$

where V is the matrix element of the electron–phonon interaction and real. The Raman spectrum of two coupling excitations is proportional to[24]

$$I(\hbar\omega) \propto \mathrm{Im}\left(\sum_{i,j=e,p} T_i \langle i|G(\hbar\omega)|j\rangle T_j\right), \tag{2}$$

with the Green function operator

$$G(E) = \begin{pmatrix} \tilde{E}_p - \hbar\omega & V \\ V^* & \tilde{E}_e - \hbar\omega \end{pmatrix}^{-1}.$$

The energy $\tilde{E}_i$ $(i = p, e)$ is assumed as being complex to describe the effects of an overlap of the interacting lines, $\tilde{E}_i = E_i + \mathrm{i}\Gamma_i$, with the energy E_i and linewidth Γ_i of the fictively uncoupled excitations. T_i is the scattering amplitude of the coupling excitations, which is connected to the ground state $|g\rangle$. The difference in the phase of T_e and T_p and the argument of the complex quantity V^* add up to just one parameter of the fit. In addition, on the theoretical calculation side, the valence-subband structure in two-dimensional GaAs/$Ga_{1-x}Al_xAs$ quantum-well systems is known to be complicated because of the four-fold degeneracy of the bulk GaAs valence band. Although the use of the effective-mass theory in homogeneous bulk semiconductors is very well established, its application to heterostructures has still been the subject of considerable debate, especially with regard to the boundary conditions connecting the envelope functions across an abrupt heterojunction.[25,26] The development of multiband envelope-function-approximation theory has successfully settled the controversy and been applied well to semiconductor multiple quantum well structures, through which the theoretical results for valence subbands are in good agreement with the experimental measurements.[27,28]

In order to find the parameters $E_p, |V|$, and E_e from the evaluation of our Raman spectra measured, the information stored in the lineshape of each individual spectrum is used: the upper I_+ and lower I_- branch coupling modes are simulated using Eq. (2); the luminescence background and the sharp bare LO-phonon line are fitted by sums of Gauss and Lorentz functions. Figure 3(a) shows the Raman shifts of the two coupling modes I_+ and I_- observed and the fitting results of both parameters E_p and E_e versus the quantum-well size for the three samples investigated. It is noted that the coupling quantum-well phonon frequency $E_p = 293.4 \pm 0.5$ cm^{-1} is identical for the three studied samples and independent of the quantum-well width, which is close to the LO-phonon frequency (295 cm^{-1}) of un-doped bulk GaAs. Due to the lattice mismatch between bulk GaAs and AlAs, a tensile strain always exists in the GaAs layers, therefore, the LO-phonon frequency of the bulk GaAs is usually higher than that of the GaAs–GaAlAs quantum wells. In addition, the reduction of the quantum-well LO-phonon frequency should also be related to the acceptor-impurity doping in the quantum wells. The energy of coupling hole intersubband transitions E_e also decreases as the quantum-well width increases, which is consistent with the previous theoretical calculation as well as experimental results.[29–32] For sample A, the parameter $E_e = 322.6 \pm 0.5$ cm^{-1} $(40.0 \pm 0.06$ meV) should be assigned to the transition between the ground and the second excited state of the heavy hole subband, $h_0 \to h_2$, in terms of the calculated results of Kraus *et al.*[19] Accordingly, it is deduced that the two branches of coupled modes I_+ and I_- for sample A as displayed in Fig. 1 should originate from the excitation of the coupling interaction between the $h_0 \to h_2$ hole intersubband transition and a quantum-well phonon. Although the parameter $E_e = 278.2 \pm 0.5$ cm^{-1} $(34.49 \pm 0.06$ meV) of sample C is smaller than that of sample A, it is even nearer to the energy in resonance with the quantum-well LO phonon, such that the intensities of the I_+ and I_- are stronger than those of sample A. Figure 3(b) and the inset show the parameters E_+, E_p, E_e and the absolute value of the coupling strength $|V|$ as a function of the measured temperature in the range of 4–50 K for sample C. The energy of the quantum-well LO phonon does not shift as the measured temperature varies, which has been interpreted in Subsection 3.2. The energy of the coupling hole intersubband transition falls monotonously and is gradually away from that of the coupled LO phonon with increasing measured temperature, which coincides with that reported by Allmen *et al.*[33] In

addition, as the measured temperature rises, the upper branch of the coupled modes E_+ shifts to the lower frequency and is close to the LO phonon, while its intensity is reduced as shown in Fig. 2. There are the following two aspect reasons resulting in the peak position shift and the reduction in the intensity of the coupled modes. On one hand, as the sample temperature increases, even more acceptors δ-doped in the quantum wells are ionized with the hole carrier density being increased, such that the polarized electrical field of the LO phonons is screened and the coupling strength $|V|$ between the intersubband hole transitions and LO phonons is weakened, which is demonstrated in the inset of Fig. 3(b). On the other hand, it leads to the coupling strength $|V|$ being weakened that the energy of the hole intersubband transitions falls and gradually goes away from that in resonance with the quantum-well LO phonon with raising measured temperature.

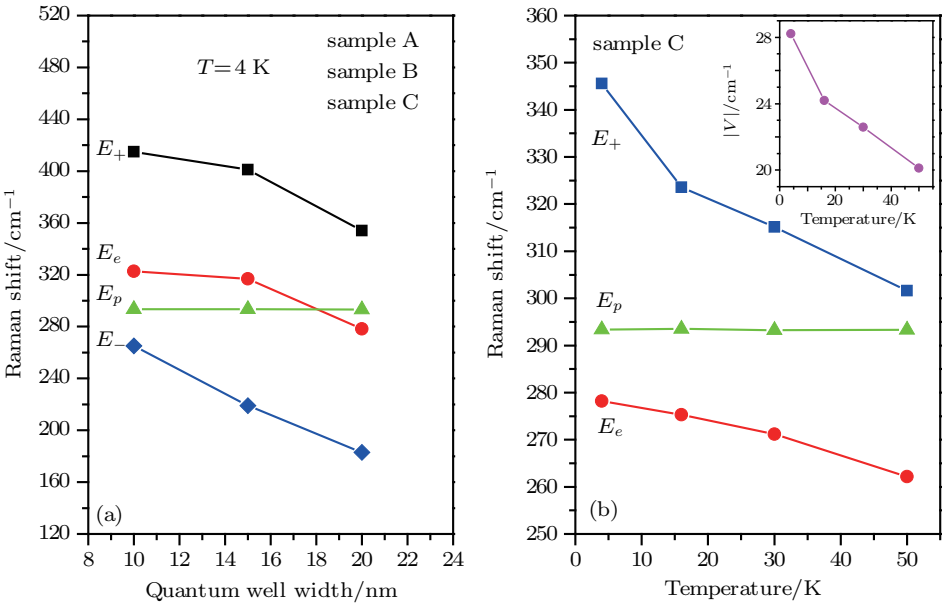


Fig. 3. (color online) (a) Peak positions of Raman shifts versus the different quantum-well widths at 4 K for samples A, B, and C. (b) Peak positions of Raman shifts for sample C as a function of the measured temperature. The inset displays the absolute value of coupling strength $|V|$ versus the temperature for sample C. The values of E_p, E_e and $|V|$ are obtained by a fit of Eq. (1) to each individual spectrum.

4. Conclusion

We have investigated experimentally Raman spectra from Be δ-doped GaAs/AlAs multiple quantum wells with different well widths at the temperatures in a range of 4–50 K. The two coupled modes of the interaction between the hole intersubband transitions and quantum-well LO phonons are observed clearly. The evaluation formalism of the Greem function is employed and the Raman spectrum lineshape of each of the coupled modes is simulated. The dependence of the peak position of Raman shifts of the two coupled modes as well as the quantum-well LO phonon on the quantum-well size and measured temperatures are given and the coupling interaction mechanism between the hole subband transitions and the quantum-well LO phonon is researched.

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