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Near-resonance Raman scattering of longitudinal optical phonon modes and interface modes in GaAs/AlAs superlattices

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Abstract. The near-resonance Raman scattering of GaAs/AlAs superlattices is investigated at room temperature. Owing to the resonance enhancement of Frohlich interaction, the scattering intensity of even LO confined modes with A_1 symmetry becomes much stronger than that of odd modes with B_2 symmetry. The even modes were observed in the polarized spectra, while the odd modes appear in the depolarized spectra as in the off-resonance case. The second-order Raman spectra show that the polarized spectra are composed of the overtone and combinations of even modes, while the depolarized spectra are composed of the combinations of one odd mode and one even mode. The results agree well with the selection rules predicted by the microscopic theory of Raman scattering in superlattices, developed recently by Huang and co-workers. In addition, the interface modes and the combinations of interface modes and confined modes are also observed in the two configurations.

1. Introduction

In GaAs/AlAs superlattices (SLs), the optical phonon modes cannot propagate through the whole structure. They are the confined modes localized in the GaAs and AlAs layers, respectively, because the dispersion curves of optical phonons for bulk GaAs and AlAs are distributed in different energy regions. Usually, the GaAs/AlAs SLs are denoted as $(GaAs)_{n_1}(AlAs)_{n_2}$, in which n_1 and n_2 are the numbers of monolayers representing the thicknesses of GaAs and AlAs constituent layers. $(GaAs)_{n_1}(AlAs)_{n_2}$ SLs have D_{2d} point group symmetry. There are, in total, n_1 GaAs and n_2 AlAs confined LO_m modes where $m = 1, 2, \ldots, n_1$ (or n_2). These modes belong to the B_2 (m odd) and A_1 (m even) representations and are Raman active in depolarized and polarized configurations, respectively [1–3].

Raman scattering measurements are extensively employed in the investigations of phonon modes in SLs. The resonance Raman scattering technique is especially efficient for obtaining more information, such as the second-order Raman modes, through the resonance enhancement effect. It is also used to investigate the electronic structure and electron–phonon interaction in SLs. Double or triple resonances have been realized by

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choosing a suitable layer thickness in order to make the splitting between light- and heavy-hole subbands equal to the energy of one or two LO phonons [4]. Details of recent progress have been reviewed in several papers [5–8]. For resonance Raman scattering, one needs a tunable laser and the experiments become more complicated. However, near-resonance Raman scattering is realizable for most GaAs/AlAs SL samples by using 6471 Å (1.916 eV) or 6764 Å (1.833 eV) lines of a krypton ion laser. Sood *et al* have carried out near-resonance scattering measurements at 10 K by using the excitation of the 6471 Å line and observed the confined LO even models [1], interface modes [9] and second-order phonon modes [10] which were not well resolved under off-resonance conditions. However, they observed only even modes with A_1 representation in both polarized and depolarized configurations [1]. In the triple-resonance scattering measurements, a similar result has been reported [4]. As to the second-order Raman scattering in both the polarized and the depolarized configuration, only overtone and combinations of even modes have been reported. Gant *et al* [11] have also reported similar results in resonance scattering. All of them explained the appearance of even modes in the depolarized spectra by means of impurity- or interface-defect-induced Frohlich interaction. In a recent report, Mowbray *et al* [12] presented observations of the LO_1 odd mode in the depolarized configuration in addition to the LO even modes.

In this paper we present the measurements of near-resonance scattering from GaAs/AlAs SLs at room temperature and in back-scattering geometry. In depolarized first- and second-order Raman scattering measurements, we observed confined LO odd modes and combinations of one odd mode and one even mode, respectively.

2. Samples and experiments

The $(GaAs)_{n_1}(AlAs)_{n_2}$ SL samples were grown on [001]-oriented semi-insulating GaAs substrates by molecular beam epitaxy. A GaAs buffer layer was grown first, and then GaAs and AlAs layers alternatively with the specified thicknesses. The top layer was the GaAs constituent layer with the same specified thickness. The sample growth was controlled by a computer and monitored by RHEED. The structure parameters of SL samples were tested by the double-crystal x-ray diffraction method. The designed layer thicknesses are in agreement with the measured values. The details of the sample preparation and double-crystal x-ray diffraction measurement have been described elsewhere [13]. The structure parameters (n_1, n_2) and the repeating period numbers of the two samples studied in this paper are (10, 3), 150 periods and (6, 6), 200 periods, respectively. Since the thicknesses of the superlattice structures in our sample are significant, no signals either from the buffer layers or from the substrates were observed.

The Raman spectra were measured with a Jobin–Yvon JY-T800 laser Raman spectrometer equipped with a triple monochromator. The measurements were carried out in the back-scattering geometry and in both polarized $z(xx)\bar{z}$ and depolarized $z(xy)\bar{z}$ configurations where $x\|(100)$, $y\|(010)$ and $z\|(001)$. The excitation light sources were the 6471 Å and 6764 Å lines of a coherent CR-I-100-K3 krypton ion laser. A premonochromator was used to filter the output laser beam. The excitation power focused on the sample surfaces was about 300–500 mW. A cylindrical lens was used to avoid heating effects on the sample. The output laser beam was vertically polarized. The polarization analysis of the scattered light was carried out through a polarization analyser in order to accomplish the specified scattering configurations. The scattered light was dispersed through the spectrometer and then detected with a cooled RCA C31034A

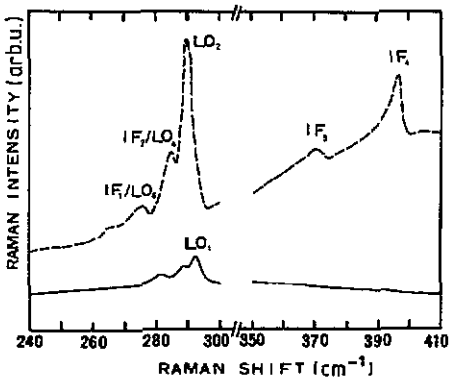

Figure 1. The room-temperature Raman spectra of a $(GaAs)_{10}(AlAs)_3$ SL excited by the 6764 Å line: – – – –, $z(xx)\bar{z}$ configuration; ———, $z(xy)\bar{z}$ configuration.

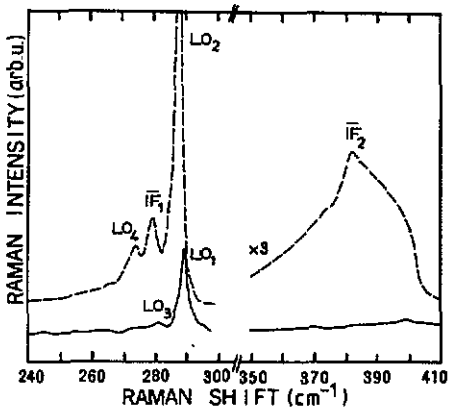

Figure 2. The room-temperature Raman spectra of a $(GaAs)_6(AlAs)_6$ SL excited by the 6764 Å line: – – – –, $z(xx)\bar{z}$ configuration; ———, $z(xy)\bar{z}$ configuration.

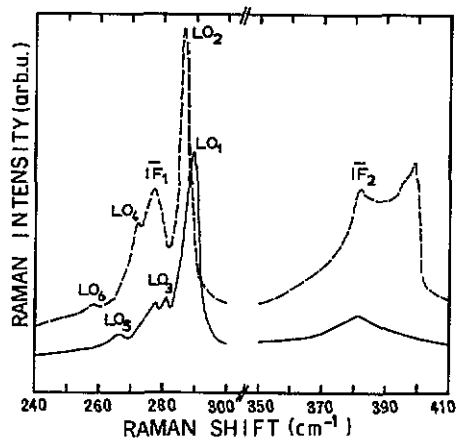

Figure 3. The room-temperature Raman spectra of a $(GaAs)_6(AlAs)_6$ SL excited by the 6471 Å line: – – – –, $z(xx)\bar{z}$ configuration; ———, $z(xy)\bar{z}$ configuration.

photomultiplier and a photon counter. The experimental curves have been treated with a smoothing procedure. All the measurements were made at the room temperature.

3. Results and discussion

In figures 1 and 2 are shown the Raman spectra of two SL samples, (10, 3) and (6, 6), respectively, in the $z(xx)\bar{z}$ and $z(xy)\bar{z}$ configurations. The wavelength of the excitation light was 6764 Å. Under off-resonance scattering, the intensity of the LO odd modes is much stronger than that of the LO even modes [1–3]. Under near-resonance scattering, however, the intensity of GaAs LO even modes is much stronger than that of GaAs odd modes (for the depolarized configuration, appropriately enlarged spectra are depicted in figures 1–3). The signals of the odd modes were weaker than those obtained under off-resonance scattering. Quite similar to the earlier report by other workers, GaAs LO

even modes were observed in the $z(xx)\bar{z}$ configuration [1, 4, 11]. As distinct from the earlier presentation, what we observed in the $z(xy)\bar{z}$ configuration, however, was LO odd modes with B_2 representation instead of LO even modes with A_1 representation. As shown in figure 1, a peak at 291.3 cm^{-1} was observed in the $z(xy)\bar{z}$ configuration. This value is in accordance with the frequency of the confined LO_1 mode and is 2 cm^{-1} away from that of the LO_2 mode observed in the $z(xx)\bar{z}$ configuration. In figure 2, a peak at 289.5 cm^{-1} can be clearly resolved in the depolarized spectra and is 3 cm^{-1} away from the frequency of the LO_2 mode observed in the polarized spectra. From the peak position and polarization analyses of the Raman spectra, these two peaks are reasonably assigned to LO_1 modes. Unfortunately, the high-order modes are too weak to be clearly resolved. However, owing to the stronger resonance enhancement in Raman scattering from sample (6, 6) excited by the 6471 Å laser line, as shown in figure 3, three GaAs LO odd modes and three GaAs LO even modes were clearly observed in depolarized and polarized configurations, respectively. Their frequencies are consistent with our results of the earlier off-resonance experiments [3]. Our near-resonance measurements indicate that the A_1 modes are Raman active in the polarized spectra, while the B_2 modes are Raman active in the depolarized spectra. In other words, the selection rule in the near-resonance scattering is the same as in the off-resonance scattering.

Recently, Huang and co-workers [14, 15] have developed a microscopic theory on Raman scattering in SLS, giving the expressions for Raman scattering efficiency related to the confined modes and interface modes, and derived the selection rules of these processes. According to their results, the LO odd modes are induced by deformation potential scattering and are Raman active in the $z(xy)\bar{z}$ configuration, while the LO even modes are induced by Frohlich scattering and are Raman active in the $z(xx)\bar{z}$ configuration. Although the Frohlich interaction is dipole forbidden in bulk GaAs, it is allowed in SLS. Its contribution may come from both intrasubband and intersubband scattering processes. However, when the splitting between heavy- and light-hole subbands is larger than the LO phonon energy, the contribution from the intersubband process can be neglected. On the other hand, the intrasubband scattering process is weak for the deformation potential interaction. Thus, under near-resonance conditions, only LO even modes are resonantly enhanced, the LO odd modes remaining relatively weak. Our experimental results agree well with their theoretical prediction.

In the depolarized spectra, as well as the LO odd modes, we also observed even modes (not depicted in this paper) when there was a deviation from the exact $z(xy)\bar{z}$ configuration. As has been mentioned above, the Raman scattering intensity of LO even modes is much stronger than those of odd modes in the near-resonance scattering; thus it is easier to activate LO even modes due to a deviation from the exact $z(xy)\bar{z}$ scattering configuration.

In figures 1–3, the Raman spectra in the frequency range of AlAs LO phonons are also shown. The results are very different from what we observed in off-resonance scattering [3]. Some broad and strong peaks appear in the polarized spectra. From their peak frequency, line shape and the behaviour of the resonance enhancement, they should not be assigned to the LO confined modes, but rather to the interface modes. The results of theoretical analysis on the interface modes in the microscopic model [14, 16] are consistent with the dielectric continuum model in the zero-dispersion limit. According to the dielectric continuum mode [9, 11], there are two GaAs interface modes, IF_1, and IF_2, and two AlAs interface modes, IF_3 and IF_4, for each wavevector. In the strict back-scattering geometry, the frequencies of the wavevector-allowed interface modes are very close to that of LO_1 and TO_1 confined modes and are difficult to distinguish experimentally

from each other. However, owing to the disorder induced by impurities or interface defects, scattering by the interface modes may be allowed for in all wavevectors. The interface mode scattering will produce two wide bands corresponding to the density of state. As shown in figure 1, there are two interface modes for both GaAs and AlAs regions. Now, considering the confined LO modes, only GaAs confined modes appear owing to the resonance enhancement, while AlAs confined modes are covered up by the resonance-enhanced interface modes. When $n_1 = n_2$, the gap between two interface mode bands disappear, they merge into one band, $\overline{\text{IF}}_1$ in the GaAs region and $\overline{\text{IF}}_2$ in the AlAs region as shown in figure 2. In fact, the polarized spectra in figure 2 are quite similar to those shown in [9] for a similar sample.

In figure 3 the Raman spectra of the (6, 6) sample excited by the 6471 Å line are depicted. The Raman spectrum in the AlAs region is different from that shown in figure 2 and is rather similar to the earlier result [17]. The spectrum is composed of an AlAs interface mode $\overline{\text{IF}}_2$ and a sharp peak at 398 cm^{-1}. The latter should not be induced by a confined LO_1 mode although the frequency seems to be the same, because the LO odd modes are only observed in the $z(xy)\bar{z}$ configuration. AlAs LO confined modes are very well localized within AlAs layers. They will not couple with the excitons which are localized in GaAs well regions and will not be resonance enhanced; thus it does not seem to be the LO_2 mode either. In the back-scattering geometry, although the frequency of the wavevector-allowed AlAs interface mode is close to that of the LO_1 confined mode, and its electrostatic potential has a near-constant term which extends to neighbouring GaAs regions, the contribution to Raman scattering is ineffective [14, 16]. Thus, further investigation is needed for the assignment of this peak.

A significant resonance enhancement of Raman signal was obtained for the (6, 6) sample by using the excitation of the 6471 Å line. In addition to the first-order Raman peaks, the second-order Raman peaks are clearly detected. The first- and second-order Raman scattering spectra in the $z(xx)\bar{z}$ and $z(xy)\bar{z}$ configurations are shown in figures 4 and 5. It can be seen from figures 4 and 5 that the phonon peaks are superimposed on the background of a broad luminescence peak centred at 1.856 eV, which is quite close to the $E^{\Gamma}_{11\text{h}}$, the transition energy between the first conduction subband and the first heavy-hole subband, calculated by the Kronig–Penney model and also to data from the photoluminescence at room temperature [18]. This broad luminescence band is nearly completely polarized; the intensity of the depolarized component is reduced by at least one order of magnitude. The insets in figures 4 and 5 show the details of the second-order Raman peaks. Similar to the earlier report by other workers [4, 10], polarized second-order Raman spectra are composed of overtones and combinations of LO even modes or interface modes.

As distinct from the earlier report [4, 10], however, neither overtones nor combinations of LO even modes were observed in depolarized second-order Raman spectra. From the analysis of the frequencies of these peaks in depolarized second-order Raman spectra, we assigned them to combinations of a LO odd mode and a LO even mode or an interface mode. The frequency values and these assignments of the peaks in the polarized and depolarized first- and second-order Raman spectra are listed in table 1. The data in table 1 show that the frequencies of the second-order Raman peaks can be correlated well to the overtones and combinations of the confined LO modes and interface modes in the first-order Raman spectra. The assignments are also in agreement with polarization selection rules on the second-order Raman spectra [19]. The peak and shoulder appearing in the low-wavenumber region of figures 4 and 5 are assigned to the disorder-activated transverse acoustic phonon mode and longitudinal acoustic phonon mode.

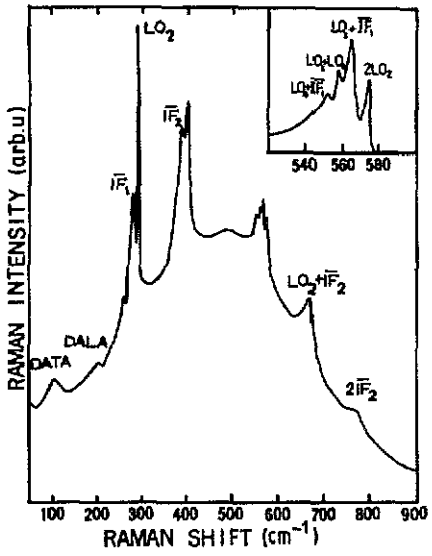

Figure 4. The first- and second-order polarized Raman spectra of a $(GaAs)_6(AlAs)_6$ SL excited by the 6471 Å line at room temperature.

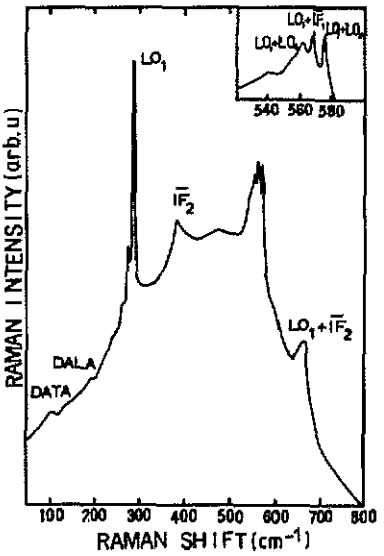

Figure 5. The first- and second-order depolarized Raman spectra of a $(GaAs)_6(AlAs)_6$ SL excited by the 6471 Å line at room temperature.

Table 1. Frequencies and their identifications of the LO confined modes and interface modes in $(GaAs)_6(AlAs)_6$ SLs.

$z(xx)\bar{z}$		$z(xy)\bar{z}$	
Mode assignment	Mode frequency (cm^{-1})	Mode assignment	Mode frequency (cm^{-1})
LO_2	286.5	LO_1	289.5
LO_4	272.5	LO_3	281
LO_6	259	LO_5	266
$\overline{IF}_1$	277.5	$\overline{IF}_1$	277.5
$\overline{IF}_2$	382.5	$\overline{IF}_2$	382
$LO_4 + \overline{IF}_1$	551		
$LO_2 + LO_4$	558	$LO_1 + LO_4$	562
$LO_2 + \overline{IF}_1$	564	$LO_1 + \overline{IF}_1$	566.5
$2LO_2$	573	$LO_1 + LO_2$	575.5
$LO_2 + \overline{IF}_2$	667	$LO_1 + \overline{IF}_2$	668
$2\overline{IF}_2$	765		

4. Conclusion

In conclusion, the Raman scattering spectra of GaAs/AlAs SLs are measured at room temperature and in the back-scattering geometry. The near-resonance excitation is realized by using 6471 Å and 6764 Å lines of a krypton ion laser. The experimental results show that, in the first-order Raman spectra, GaAs LO even modes are Raman

active in the polarized configuration, while GaAs LO odd modes are Raman active in the depolarized configuration. Interface modes are observed in both configurations. In the second-order Raman spectra, overtones and combinations of A_1 modes and interface modes were observed in the polarized configuration, and in the depolarized configuration the combinations of either B_2 and A_1 mode, or B_2 and interface modes were seen.

Acknowledgments

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