Study of Asymmetric Broadening of Raman Scattering in $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{InP}$ and $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ Epilayers

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Raman scattering of $\text{In}_x\text{Ga}_{1-x}\text{As}$ epitaxial layers on InP and GaAs substrates have been investigated with a wide range of composition. It is found that the asymmetric broadening of LO phonon modes varies with composition as well as substrate. With the one-parameter spatial correlation model, the Raman lineshapes have been analyzed, but the fits showed some deviation to the experiments. Comparing the Raman spectra with the measurements of double-crystal x-ray diffraction (DXRD), we found that, besides the alloy disorder, the Raman lineshapes are also influenced by the structural dislocation. Our results provide a framework for the use of the two-parameter spatial correlation model in the study of the Raman spectra in epilayer structures.

Introduction

Raman scattering is a powerful tool to investigate the microscopic nature of the disordered alloy since the compositional potential fluctuation will produce the broadening and the asymmetry of LO Raman lineshape. Parayanthal and Pollak successfully used a spatial correlation (SC) model to interpret the asymmetric and broad Raman lineshapes of $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ alloy semiconductors(1). Nevertheless, some experiments in $\text{GaAs}_{1-x}\text{Sb}_x/\text{InP}$ and $\text{Cd}_{1-x}\text{Zn}_x\text{Te}/\text{GaAs}$ alloys showed that the SC model can not totally explain the Raman lineshapes(2,3). Olego et. al. laterly suggested the Fano-like model may get better fittings than the SC model(3). While Ksenzov et. al. demonstrated the SC model with two parameters is also equivalent to the Fano model as far as fitting procedures are concerned(4). However, the physical meaning for the fitting parameters is not clear.

In this report, we study the Raman scattering of $\text{In}_x\text{Ga}_{1-x}\text{As}$ epilayers grown on InP and GaAs substrates. The theoretical calculation based on the one-parameter spatial correlation model and the correlation between the Raman spectra and the measurements of double-crystal x-ray diffraction (DXRD) have been investigated.

Experimental procedure

The used samples were grown on Fe-doped (100) InP and undoped (100) GaAs substrates by molecular-beam epitaxy. The $\text{In}_x\text{Ga}_{1-x}\text{As}$ epilayers are 2- μm -thick with $0 \leq x \leq 1$ laterly. The Raman scattering measurements were performed at room temperature in the backscattering geometry with the 514.5 nm line of an Ar ion laser. The scattering light was analyzed by a Spex 1403 0.85 m double monochromator and detected by a charge coupled device (CCD) using a Si diode array.

Results and Discussion

Figure 1(a) shows the Raman spectra of $\text{In}_x\text{Ga}_{1-x}\text{As}$ on InP substrate with the indium molar fraction $x=0.31, 0.48, 0.59, 0.88$, and 0.93 . The peaks labeled LO_1 and LO_2 are attributed to GaAs-like LO phonon mode and InAs-like LO phonon mode, respectively. Similarly, figure 1(b) displays the spectra of $\text{In}_x\text{Ga}_{1-x}\text{As}$ on GaAs substrate with the composition $x=0.16, 0.20, 0.50, 0.88$, and 0.93 . The GaAs-like LO mode and InAs-like LO mode are

Figure 1: Raman spectra of $\text{In}_x\text{Ga}_{1-x}\text{As}$ with various x on the a) InP b) GaAs substrate

labeled by LO_3 and LO_4 separately in figure 1(b).

Full width at half maximum (FWHM) of the LO Raman peaks versus x is plotted in the figure 2. We found that the FWHM of LO_1 mode basically increases with x , along with two local maximums occur at the $x=0.3$ and $x=0.85$. The FWHM of LO_3 mode also increases with x but the maximum occurs at 0.5. As for the InAs-like mode, the linewidths of LO_2 and LO_4 both decrease as x approaches to 1 but the FWHM of LO_2 is larger than that of LO_4 .

From this figure, we can see that, the FWHM of Raman signal not only depends on the alloy composition, but also depends on the substrate. For the alloy semiconductor, the Raman lineshape displays an asymmetric profile in phonon band. This phenomenon can be seen in the figure 1 especially as the x value is large. The asymmetric ratio of LO_1 peak and LO_3 peak versus x were shown in figure 3. The asymmetric ratio of LO_1 peak in general increases with x for $0.1 \leq x \leq 0.88$, but at the $x=0.31$ and $0.52 \leq x \leq 0.69$ the ratios are reduced. The evolution of LO_3 is similar, except the reduced ratio occurs at $x=0.5$.

Figure 2: FWHM of Raman peaks vs. x in $\text{In}_x\text{Ga}_{1-x}\text{As}$ epilayers on the InP and GaAs. The solid lines are guided for the eye.

Figure 3: Asymmetric ratios of the GaAs-like mode of $\text{In}_x\text{Ga}_{1-x}\text{As}$ as a function of x . The solid lines are guided for the eye.

It has been reported that the linewidth and profile of Raman peak will depend on the residual stress(5), the structural alloy disorder(1). However, the thickness of epilayers in our measured samples is so thick ($2\mu\text{m}$) that the stress due to lattice mismatch will be released and hence the stress affects the Raman lineshape is impossible. It is well known that the spatial correlation length of the phonon in an alloy crystal is finite owing to the potential fluctuation of the alloy disorder, which gives rise to a relaxation of the $q=0$ selection rule and the asymmetric broaden lineshape in Raman scattering(1). With the SC model(1), we can evaluate the asymmetric broadening by theoretical calculation. In the

Figure 4: Measured and calculated lineshapes of LO_1 with $x=0.31, 0.48, 0.73$ and 0.88 .

SC model the Raman intensity $I(w)$ can be written as:

$$I(w) \sim \int_0^1 \exp(-q^2 L^2/4) \times d^3 q / \{ [w - w(q)]^2 + (\Gamma_0/2)^2 \}, \quad (1)$$

where a is the lattice constant, Γ_0 ($= 3 \text{ cm}^{-1}$ for GaAs) is the intrinsic linewidth, and L is the correlation length. For the dispersion $w(q)$ of the LO phonon, we take the analytical relationship based on the linear-chain model(1):

$$w^2(q) = A + \{ A^2 - B[1 - \cos(\pi q)] \}^{1/2}. \quad (2)$$

For the GaAs-like phonon mode we take $A=4.26 \times 10^4 \text{ cm}^{-2}$ and $B=7.11 \times 10^8 \text{ cm}^{-4}$. By setting L as an adjustable parameter, one can describe the experimental data by the SC model. Figure 4(a),(b),(c), and (d) show the fitted results for $x=0.31, 0.48, 0.73$, and 0.88 , respectively. The fittings are good for the $x=0.88, 0.48$, and 0.73 but deviate much for the $x=0.31$.

If we want to get better fits for all x , however, the fitting parameters must include L as well as Γ_0 . This unsatisfactory result of one-parameter SC

Figure 5: FWHM of DXRD signals from $\text{In}_x\text{Ga}_{1-x}\text{As}$ epilayers on the a) InP and b) GaAs.

model also have been reported in previous studies(2,3). Olego et. al. hence proposed a Fano-like effect produced by an anharmonic decay of the LO phonons into a continuum of other phonon to explain the asymmetric broadening(3). Since the Fano-like model always involves two parameters. Ksendzov et. al. laterly demonstrated that the two-parameter SC model also produces good fit and suggested another anharmonic interactions to account for the change of another fitting parameter Γ_0 (4). Nevertheless, there is no clear evidence to support their suggestion. These studies indicate that the one-parameter SC model can not explain the Raman lineshapes and imply a new mechanism available.

To search for the possible mechanisms of the deviation of the one-parameter SC model, the samples also have been measured with the DXRD (6,7). Figures 5(a) and 5(b) display the FWHM of the DXRD signals in $\text{In}_x\text{Ga}_{1-x}\text{As}$ epilayers with $0 \leq x \leq 1$ on InP and GaAs. Comparing figure 5 with figure 2 and figure 3, it is interesting to see that there is a correlation between the linewidth of the DXRD signals and the Raman lineshapes: [1] At

the $x=0.5$, the Raman FWHM of LO_3 is larger than that of LO_1 as shown in figure 3. And figures 5(a) and 5(b) show that the FWHM of DXRD in $In_{0.5}Ga_{0.5}As/GaAs$ is larger than that in $In_{0.5}Ga_{0.5}As/InP$. [2] The Raman peaks of LO_2 is broader than that of LO_4 in $0.75 \leq x \leq 0.9$ in figure 2. This trend is also consistent with the figure 5 which shows the FWHM of DXRD (≥ 900 arcsec) in $In_xGa_{1-x}As/InP$ is broader than that (≤ 900 arcsec) in $In_xGa_{1-x}As/GaAs$. [3] The asymmetric ratio of the LO_1 is larger than that of the LO_3 at $x=0.5$ in the figure 3. This result is opposite to the linewidth of DXRD in figure 5 which show that the FWHM of $In_xGa_{1-x}As/GaAs$ is larger than that of $In_xGa_{1-x}As/InP$ at $x=0.5$.

Since the FWHM of the DXRD signals is sensitive to the misfit and threading dislocation in heteroepitaxial system, we suggest that, in the mismatched heterostructure, the LO Raman lineshape not only depends on the alloy disorder but also the dislocation. The samples we used were $In_xGa_{1-x}As$ grown on the mismatched InP and $GaAs$. The formation of dislocations in these samples is inevitable as the stress due to lattice mismatch will be released in such a thick film. If the dislocation is so high such that it can change the Γ_0 , the deviation from the one-parameter SC model will occur. This can be used to explain the experimental fact that the values of x where the fit is worse (figure 4(a)) just correspond to the high dislocation densities (figure 5(a)). Besides, the larger FWHM of DXRD in figure 5 corresponds to the larger Raman FWHM in figure 2, it means the more dislocations will increase the Γ_0 . The variation of the Γ_0 also leads to change the asymmetric ratio. This phenomenon can be observed in the figure 3: at $x=0.5$, the asymmetric ratio of LO_3 mode in $In_xGa_{1-x}As/GaAs$ which correspond to high dislocations is less than that of LO_1 mode in $In_xGa_{1-x}As/InP$ which corresponds to low dislocations. Now, if we consider the effect of dislocation in the Raman lineshape, the reason why the SC model succeed in $Al_xGa_{1-x}As/GaAs$ (1) system but failed in $GaAs_{1-x}Sb_x/InP$ (2) and $Cd_{1-x}Zn_xTe/GaAs$ (3) systems in previous studies become clear as the former is a lattice matched system but the others are lattice mismatched systems which contain more dislocations.

Conclusion

In summary, Raman scattering of $In_xGa_{1-x}As$ on InP and $GaAs$ substrates over a wide range of x have been performed. We found that the compositional dependence of FWHM (and the asymmetric ratio) were different in different substrates. The Raman lineshapes have been fitted by the one-parameter SC model and found some deviation. According to the measurements of the DXRD, we proposed that the dislocation may contribute to the Raman lineshape in mismatch heterostructure. Our results provide a framework for using the two-parameter SC model to analyze the compositional dependence of the Raman profiles of epilayer structures.

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