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LINE SHAPE AND PROFILE OF THE LO PHONON MODES IN MIXED ALLOY SYSTEM $In_{1-x-y}Ga_xAl_yAs$ EPITAXIAL LAYERS GROWN ON InP

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Abstract: In this work, linewidth variations versus compositions of the first-order longitudinal optic (LO) phonon modes in the mixed alloy system are studied. The asymmetry and broadening of the LO modes is explained using a "spatial correlation" model with a Gaussian correlation function. The system investigated here were $In_{1-x-y}Ga_xAl_yAs$ quaternary alloys lattice-matched to InP(with $x+y \approx 47\%$). The quaternary alloy samples were grown as epilayers on (001)InP substrates by molecular beam epitaxy(MBE).

1. INTRODUCTION

The quaternary semiconductor system $In_{1-x-y}Ga_xAl_yAs$ is potentially of importance for optical communication devices such as optical emitters, waveguides, and detectors. The material is also very suitable for use in heterojunction bipolar transistors[1]. It can be grown lattice matched to InP for $x+y \cong 0.47$[2], in which case the band-gap is adjustable between those of $In_{0.53}Ga_{0.47}As$ (0.76 eV, 1.63 μm)[3] and $In_{0.52}Al_{0.48}As$ (1.46 eV, 0.85 μm)[4]. The fact that Al has a large distribution coefficient[5] makes it difficult to grow layers with uniform composition by liquid phase epitaxy(LPE) whereas the virtual unity incorporation coefficients of the group III atoms in the molecular beam epitaxy(MBE) growth process allow great flexibility and control over alloy composition since only one group V element is used, and the group III sticking coefficients are easily kept to unity.

To investigate the compositional dependence in mixed alloys, Raman scattering technique provides a simple and accurate way for measurement and calculation of lineshapes and profiles of the phonon modes.

2. EXPERIMENT

The $In_{1-x-y}Ga_xAl_yAs$ crystals investigated here were grown by MBE with nominal lattice matching to semi-insulating InP substrates with (001) surfaces. All layers have lateral compositional homogeneity and an average In content of about 0.53 ± 1. The total layer thickness of the samples is about 1.5 μm.

The Raman experiments were carried out at room temperature usually using the 488 nm line of an Ar^+ laser operated at a power of approximately 200 mW. The spectra were recorded with a spectral resolution between 1 $cm^{-1}$and 3 cm^{-1}. Scan speed of the spectrometer was usually about one minute per cm^{-1}. The scattered light was analysed by a Spex double monochromator spectrometer and detected by a standard photon counting technique. Spectra were mainly

recorded in the, $z(x,x+y)\bar{z}$ backscattering configuration where z is normal to the layers and xy are along the (100]) and (010) direction [This notation $q_i(a_i,a_s)q_s$ refers to an incoming beam along the direction q_i with polarisation a_i, and scattering beam along q_s with polarisation a_s]. In this geometry, the selection rules for Raman scattering permit observation of LO modes with wave vector parallel to (001)[6].

3. LINE SHAPE AND PROFILE OF THE LO PHONON MODES

Although there has been a number of Raman studies of alloy semiconductors, Barker and Sievers[7] give a review of the early work, only a few authors have reported the relation between alloy disorder and the line shape of phonon modes [8,9,10]. Investigations of linewidth and profile have been carried out on only a few pure semiconductors such as GaAs[11] and on GaP[9] Beserman et al.[12] performed this kind of study on the mixed crystal, $In_{1-x}Ga_xP$, where the variation of the asymmetry of the lineshape as a function of composition was studied. In the case of a pure crystal, such as a GaAs, the LO and TO phonon modes display a symmetric profile, which become asymmetric in the alloys.

The asymmetry in the profiles of the GaAs-like mode in our representative $In_xGa_yAl_{1-x-y}As$ sample can clearly be seen in Figure 1. The asymmetry can be characterised by measuring linewidth parameters Γ_a and Γ_b, defined as the frequency separations between the peak frequency and the half-intensity points below and above the central maximum respectively. Figure 2 shows data for Γ_a and Γ_b versus concentration for the AlAs- and GaAs-like LO modes. The asymmetry takes the form of a low energy tail on the phonon profile, so that $\Gamma_a > \Gamma_b$, and can be characterised by an asymmetry parameter $r = \Gamma_a/\Gamma_b$. Note the narrowing of the overall linewidth $\Gamma=\Gamma_a+\Gamma_b$ of the GaAs-like phonon mode when the sample composition is rich in Ga, and the narrowing of the AlAs-like phonon mode when the sample composition is high for Al, and the corresponding approach of the asymmetry parameter r to the symmetric value r = 1.

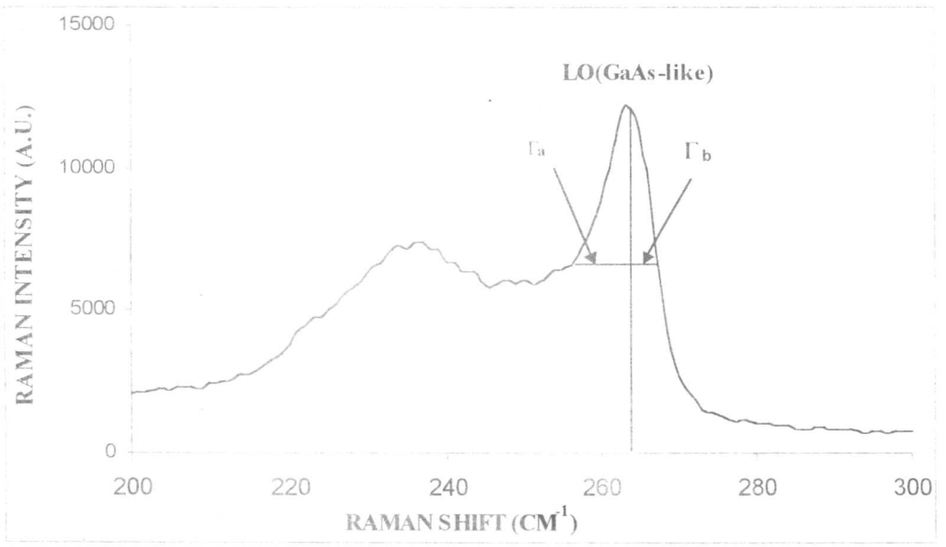

FIGURE 1. At room temperature Raman spectrum from a representative sample MBE539, $In_{0.53}Ga_{0.32}Al_{0.15}As$, at $z(x,x+y)\bar{z}$ geometry.

Two kinds of interpretation can be given for the asymmetry broadening of a Raman line: either an anharmonic decay into a degenerate continuum of two acoustic phonons (with equal and opposite wavevector), as in GaP[13,9] or a disorder induced effect[14]. Jusserand and Sapriel[10] showed that the asymmetry in $Ga_{1-X}Al_XAs$ does not vary substantially with the temperature. Thus a disorder-induced effect seems to prevail over the first one. Here, we present a model which adequately explains the details of the first-order Raman line shape (broadening and broadening asymmetry) due to the disorder-induced effect. We have analysed the longitudinal-optic (LO) phonon line shape in the representative alloy semiconductor using a "spatial correlation" model, based on finite phonon mode correlation related to q-vector relaxation induced by the microscopic nature of the alloy disorder. In a "perfect crystal" (i.e., a crystal with translational symmetry) the spatial correlation length of the phonon in extent is infinite and accordingly the phonon eigenstates are plane wave, leading to the usual $\Delta\mathbf{q}=0$ momentum selection rules of Raman scattering[15]. However, if the crystal is disordered in any way, such as by substitutional

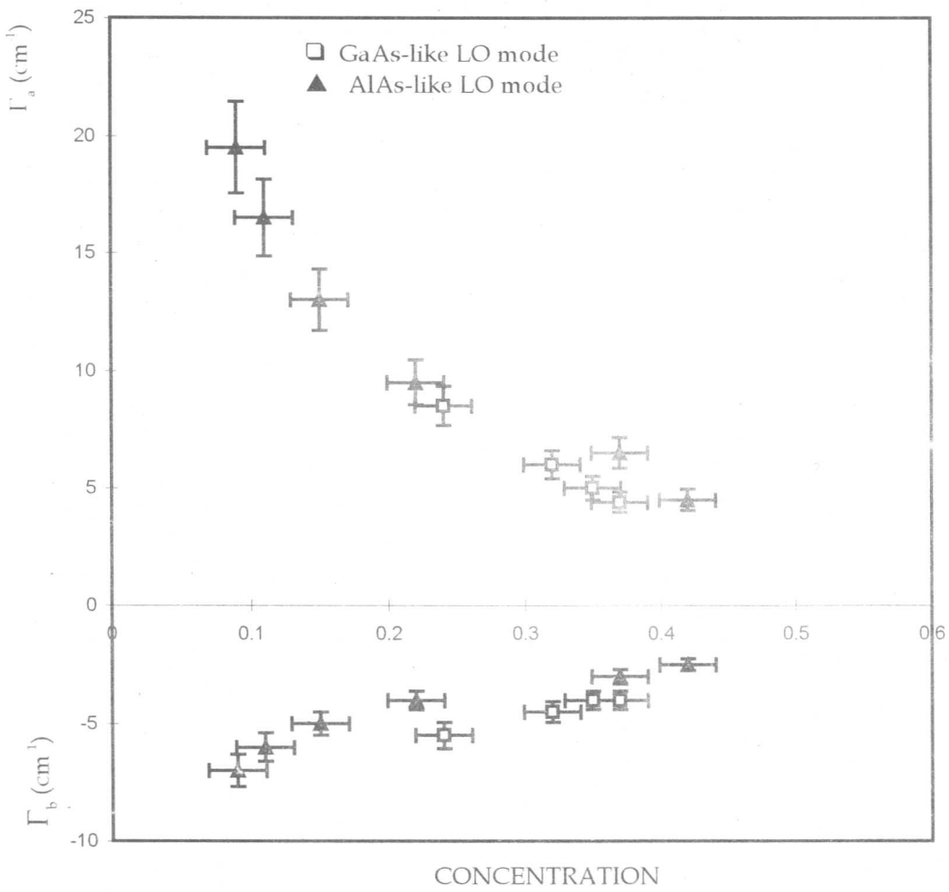

FIGURE 2. Composition variations versus the two half-widths Γ_a and Γ_b for the GaAs-like LO lines. Γ_a is associated with the low frequency side (LFS). The results are uncorrected for the instrumental resolution (1.4 cm^{-1} half-width).

disarrangement of the group III atom, the correlation length becomes finite[16] and there is a relaxation of the $\Delta\mathbf{q}=0$ selection rule. A Gaussian spatial correlation function $\exp(-2r^2/L^2)$ has been successfully used to explain q-vector relaxation related to finite-size effects[15] and structural disorder (ion-damaged materials)[17]. Here, we follow Parayanthal and Pollak[18]. The Raman intensity, $I(\omega)$, at a frequency ω can be written as the convolution

$$I(\omega) = N\int_{o}^{1} q^2 dq \frac{e^{-q^2L^2/4}}{[\omega - \omega(q)]^2 + \left(\frac{\Gamma_o}{2}\right)^2} \qquad (1)$$

Here, q is the wavevector (in units of $2\pi/a$ where a is the lattice constant), $\omega(q)$ is the frequency of the phonon mode with wavevector q, assumed to have natural linewidth Γ_0 in the absence of defect-induced scattering. L is the correlation length (in units of a), and N is a normalising constant. For simplicity, we assume a spherical Brillouin zone, with $\omega(q)$ given by the one-dimensional linear-chain expression[19]

$$\omega(q)^2 = 1/2\left[\omega_o^2 + \sqrt{\omega_o^4 - 2\omega_z^2[\omega_o^2 - \omega_z^2(1-\cos(\pi q))]}\right]^{1/2} \qquad (2)$$

ω_0 is the zone-centre frequency (q=0) and ω_Z the zone-boundary frequency (q=1). Figure 3 shows the type of asymmetry produced by the convolution Equation (1), compared with the GaAs LO profile for the sample $In_{0.53}Ga_{0.32}Al_{0.15}As$.

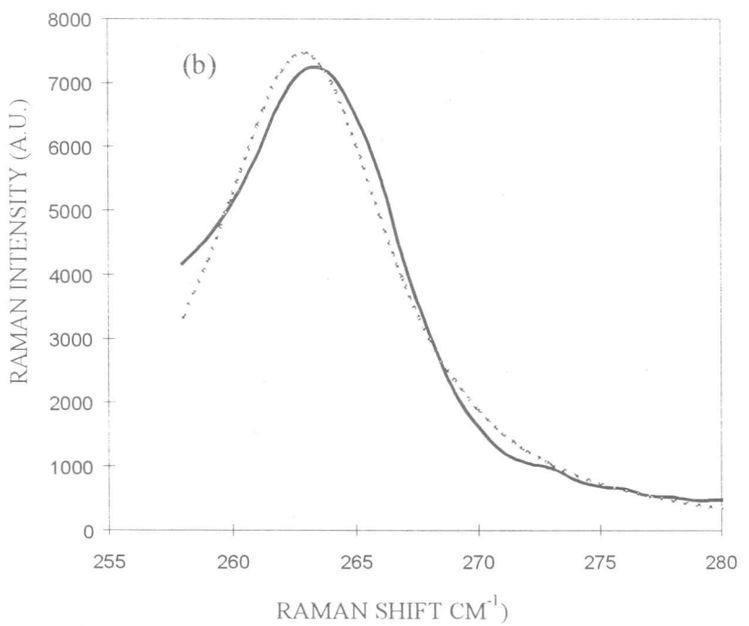

FIGURE 3. Raman Spectrum of phonon mode profile in the GaAs-like LO from $In_{1-x-y}Ga_xAl_yAs$ with y=0.15: solid curve-experimental values and dashed curve-theoretical line shape obtained from equation (1).

Figure 4 plots the relation between overall linewidth Γ and asymmetry parameter $r = \Gamma_a/\Gamma_b$ for the AlAs-like LO phonon modes, comparing the experimental measurements with a theoretical curve generated by varying the correlation length L in Equation (2). Here, we use $\omega_0 = 405$ cm^{-1} and $\omega_Z = 320$ cm^{-1}. The values of L are shown on the right hand side of the figure; L increases

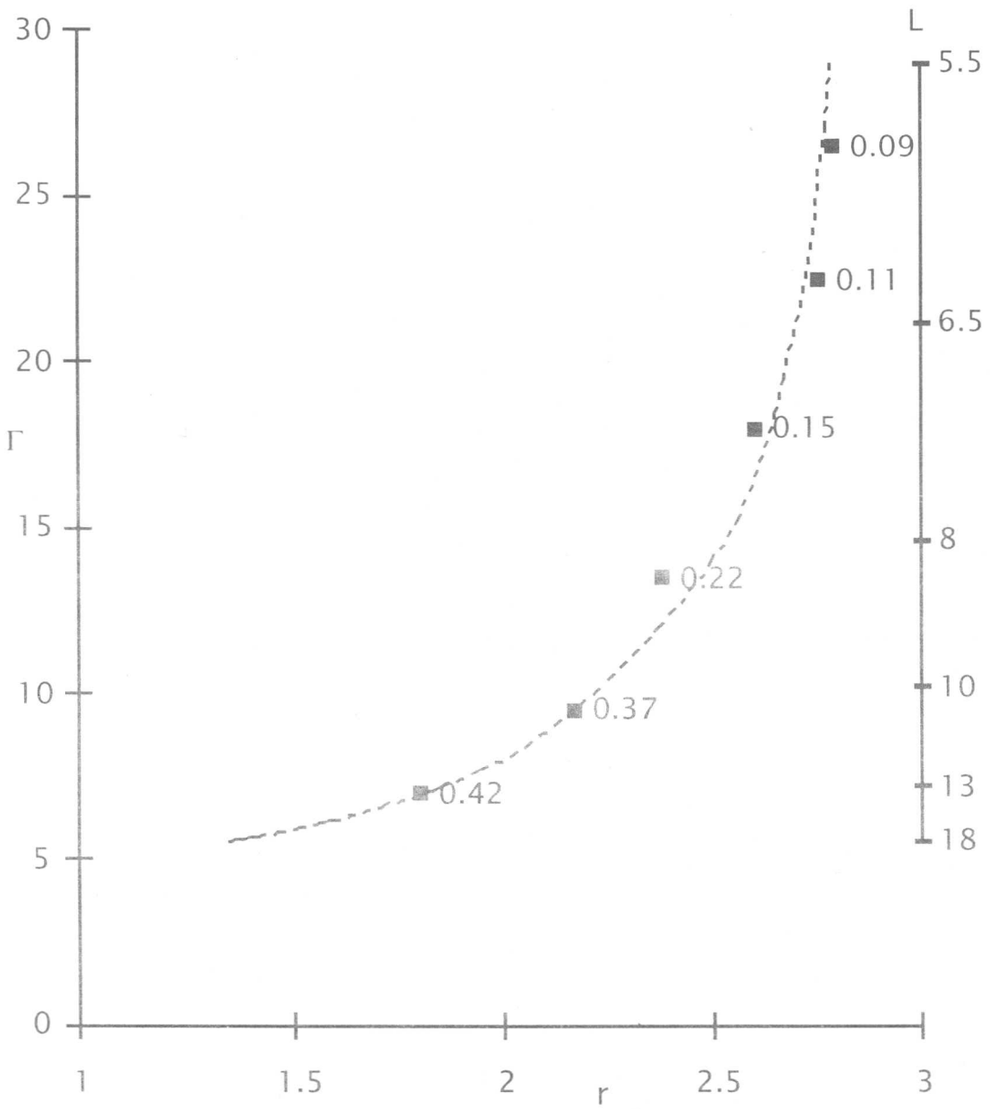

Figure 4. Total linewidth Γ (in cm^{-1}) vs. asymmetry parameter $r = \Gamma_a/\Gamma_b$ for the AlAs-like LO modes in $In_{0.53}Ga_{0.47-y}Al_yAs$. The experimental points are shown as squares, labelled with their y-values. The dashed curve is theory generated using Equation (1). The correlation length L associated with the theoretical curve is shown by the scale on the right of the figure.

roughly proportionately to the Al concentration y. The agreement between theory and experiment is not so good at the shorter correlation lengths unless the intrinsic linewidth parameter Γ_0 is allowed to vary with alloy concentration. It appears that Γ_0 reduces with decreasing Al content y. To include this, we assume in plotting Figure 4 that

$$\Gamma_0 = (0.9 + 0.15\ L)\ cm^{-1} \qquad (3)$$

Whilst the precise values obtained by this procedure should not be taken too literally, the good agreement between theory and experiment shown in Figure 3 indicates the basic validity of the explanation for the origin of the asymmetry in the linewidth. A similar analysis for the GaAs-like LO phonon modes indicates a somewhat different correlation length for GaAs, perhaps due to the proximity of the InAs-like modes. Parayanthal and Pollak[18] have demonstrated that the microscopic alloy disorder depends, in general, on the growth parameters as well as the alloy composition, and that the spatial correlation length in principle gives information about the distribution of constituent atoms in a disordered alloy

4. CONCLUSIONS

In conclusion we show that in $In_{1-x-y}Ga_xAl_yAs$ mixed alloys the narrowing of the overall linewidth $\Gamma=\Gamma_a+\Gamma_b$ of the GaAs-like phonon mode when the sample composition is rich in Ga, and the narrowing of the AlAs-like phonon mode when the sample composition is high for Al, and the corresponding approach of the asymmetry parameter r to the symmetric value r = 1. We also demonstrates that the asymmetric broadening in the Raman line shapes of the LO phonon mode due to the disorder-activations can adequately be explained by a spatial correlation model.

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