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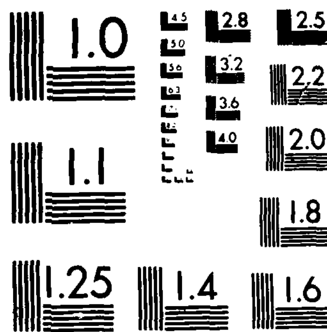

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by

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PHONON SHIFTS AND STRAINS IN STRAIN-LAYERED $(Ga_{1-x}In_x)As$

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Abstract

We have measured the phonon frequencies (Raman technique) and the strains (x-ray rocking curve technique) in $(Ga_{1-x}In_x)As$ films on GaAs (100) substraites. Films with various x-values and various thicknesses were studied. The films range from perfect epitaxial ones to those that have relaxed by different amounts. Using both of these measurements, all of the films gives internal agreement, which indicates that thte Raman technique can be used for in situ monitering of the growth process.

There is a great deal of interest in non-lattice matched heterostructures[1] where, for example, a film of one cubic semiconductor of a given lattice constant is grown on a substrate of another cubic semiconductor with a different lattice constant[2]. For most device applications it is important that the non-lattice matched epi-layers are commensurate (or pseudomorphic) with respect to substrate and to avoid any lattice relaxation which leads to either interface misfit dislocations or a high density of threading dislocations[3]. Lattice relaxation[4] during epitaxy will occur when the layer thickness exceeds some critical value h_c due to excessive strain energy[1]. At present RHEED is the only practical in-situ method for characterizing lattice constants during growth under high vacuum conditions[3]. Unfortunately the e-beam energies used in RHEED adversely affects the surface properties and hence subsequent interface properties of the grown layers. Therefore, it is desirable to have an alternative method for in-situ characterization which operates at low energy and power densities, and which can be used under any pressure conditions. Raman spectroscopy is one possible candidate which meets these criteria.

We report measurements in thin $(Ga_{1-x}In_x)As$ films grown on GaAs substrates. Principally we have looked at samples in the range $0 \leq x \leq 0.2$ and measure the optic phonons (by the Raman technique) and the parallel and perpendicular strains in the film (by the x-ray rocking curve technique)[6]. For a given value of x, we measure films of various thickness, so that they are strained by various amounts.

The films, similar to those reported previously[2], are grown by MBE on (100) oriented GaAs faces. The x-ray rocking curve measurement were carried out using the Fe $K\alpha$, radiation in a non-dispersive double crystal geometry[4]. To measure the lattice constants of the film, the 400 reflection was used to obtain the lattice constant normal to the sample surface, and the 422 reflection, with a large angle of incidence, to obtain the in-plane constant. The angular separation of the rocking curve peaks due to the misorientation of the film has been cancelled out by taking an average of the two rocking curves differing by 180 degrees in angle with respect to the sample normal. The Raman measurements were performed using a 5145 A laser with a standard double monochrometer.

Because the GaAs substrate is massive compared to the film, the substrate is assumed to retain its normal internuclear distances, with a lattice constant given as a_s. For the film we use the term parallel to denote lattice constants in the film parallel to the (100) plane of the substrate; thus $a_{f\parallel}$ is the parallel lattice constant of the film. Similarly, $a_{f\perp}$ is the perpendicular lattice constant of the film.

The seven single crystal films of $(Ga_{1-x}In_x)As$ on (100) GaAs substrates that were studied are listed in Table 1, where the value of x and nominal film thickness is given. Consider the three films of composition X=0.10 as an example. For this composition we have grown one film with thickness less than the critical thickness, h_c and two with thickness thicker than h_c. The critical thickness is defined as the maximum thickness the film can be grown in perfect epitaxy for a given substrate and composition of the film. For each film we measure the relative unit cell size of the film with respect to the substrate and obtain what is often called x-ray strain. For example, $\epsilon_{\parallel}^{xr} = (a_{f\parallel} - a_s)/a_s$. These results are listed in the table. For perfect epitaxy $\epsilon_{\parallel}^{xr} = 0$, and this is found for the 1500 Å film. We also list the measured position of the LO phonon of this film; as the film become thicker this value decreases. The

measured frequencies are plotted in Fig. 1 as solid circles. For example, for x=0.1 there are are solid circles corresponding to the frequency measured from each of these three films. These phonon frequency values make physical sense; the film with perfect epitaxy is under the largest compressive stresses. Since we know that compressive stresses shift the phonons to high energies, these films will have the largest frequency shift, as observed.

In order to calculate the phonon frequency shifts due to the strains in the film, we must determine the elastic strains. Clearly these are related to the measured x-ray strains. Physically we know that the largest negative in-plane elastic strain (ϵ_{xx}) will be required to produce a perfect epitaxial film (i.e., a film with $\epsilon_{\parallel}^{xr} = 0$). Similarly, a perfect epitaxial film will have the largest elastic strain perpendicular to the plane (ϵ_{zz}). From the x-ray strain, and with the help of elasticity theory, ϵ_{xx} and ϵ_{zz} can be calculated. For the case discussed here, for a two dimensional stress in the xy-plane, from elasticity theory $\epsilon_{zz}/\epsilon_{xx} = 2\nu/(1-\nu)$, where ν is Poisson's ratio. Since $\nu \approx 1/3$, we have $\epsilon_{zz} \approx -\epsilon_{xx}$, and the deviations from this result shown in Table 1 are due to the fact that Poisson's ratio is not 1/3 exactly and varies slightly with x.

Given the elastic strains it remains for us to calculate the frequencies of the optic modes in the presence of the strains. Assuming the threefold degeneracy of the k=0 optical phonons in a material with the diamond structure, in the presence of strain the frequencies of the optic modes are given by the following secular equation[7,8].

$$\begin{vmatrix} p\,\epsilon_{xx} + q(\epsilon_{yy} + \epsilon_{zz}) - \lambda & 2r\,\epsilon_{xy} & 2r\,\epsilon_{xz} \\ 2r\,\epsilon_{xy} & p\,\epsilon_{yy} + q(\epsilon_{zz} + \epsilon_{xx}) - \lambda & 2r\,\epsilon_{yz} \\ 2r\,\epsilon_{xz} & 2r\,\epsilon_{yz} & p\,\epsilon_{zz} + q(\epsilon_{xx} + \epsilon_{yy}) - \lambda \end{vmatrix}$$

The eigenvalue $\lambda = \Omega^2 - \omega_0^2$ where ω_0 is frequency with zero strain and Ω is the strain dependent frequency. The phonon deformation constants, p, q and r describe the change of the spring constants with strain[9]. Since the frequency shift due to strain is small, $\Omega \approx \omega_0 + \lambda/2\omega_0$. In III-V semiconductors with the zinc blend structure, the long range coulomb forces split the LO and TO modes at small k. This splitting is small and is essentially independent of strain; so the strain dependent frequency shifts are obtained by adding the LO-TO splitting to the frequencies obtained from Eq. 1.

The phonon deformation constants have been measured by applying uniaxial stresses to single crystal samples with various orientations[7,8,10]. Hence, in subsequent experimental papers[11] the shifts are expressed in terms of stress. However, since here we directly measure the strains, we can ignore the stress (which is applied to the film by the substrate). Then the eigenvalue of Eq. 1 for the LO phonon that we measure from (100) oriented faces is given by

:f.lambda $= p\epsilon_{zz} + q(\epsilon_{xx} + \epsilon_{yy})$:ef.

where $\epsilon_{xx} = \epsilon_{yy}$ for our geometry. Given the phonon deformation constants p and q, and using our measured strain (listed in Table 1), the frequency shift Ω-ω_0, is obtained and given in the last column in Table 1.

We measure Ω, calculate λ (using the measured x-ray strain), hence can determine ω_0, which is the equivalent bulk frequency for the phonon. These results are plotted in Fig. 1 as crosses. For each data point (solid circles) we obtain a cross; as can be seen, these bulk equivalent frequencies are in good agreement with each other. For ;f.x nearly 0.15, we find that h_c is between 1500 A and 2500 A; this is in reasonable agreement with published data[12]

We have shown that Raman spectroscopy appears to be a suitable method for characterizing strain in strained layer epitaxial systems. Since it is an optical technique of lower power density than RHEED, it should be ideal as a damage-free, insitu tool to characterize the state of an epitaxial layer during growth by MBE, MOCVD,, of other techniques.

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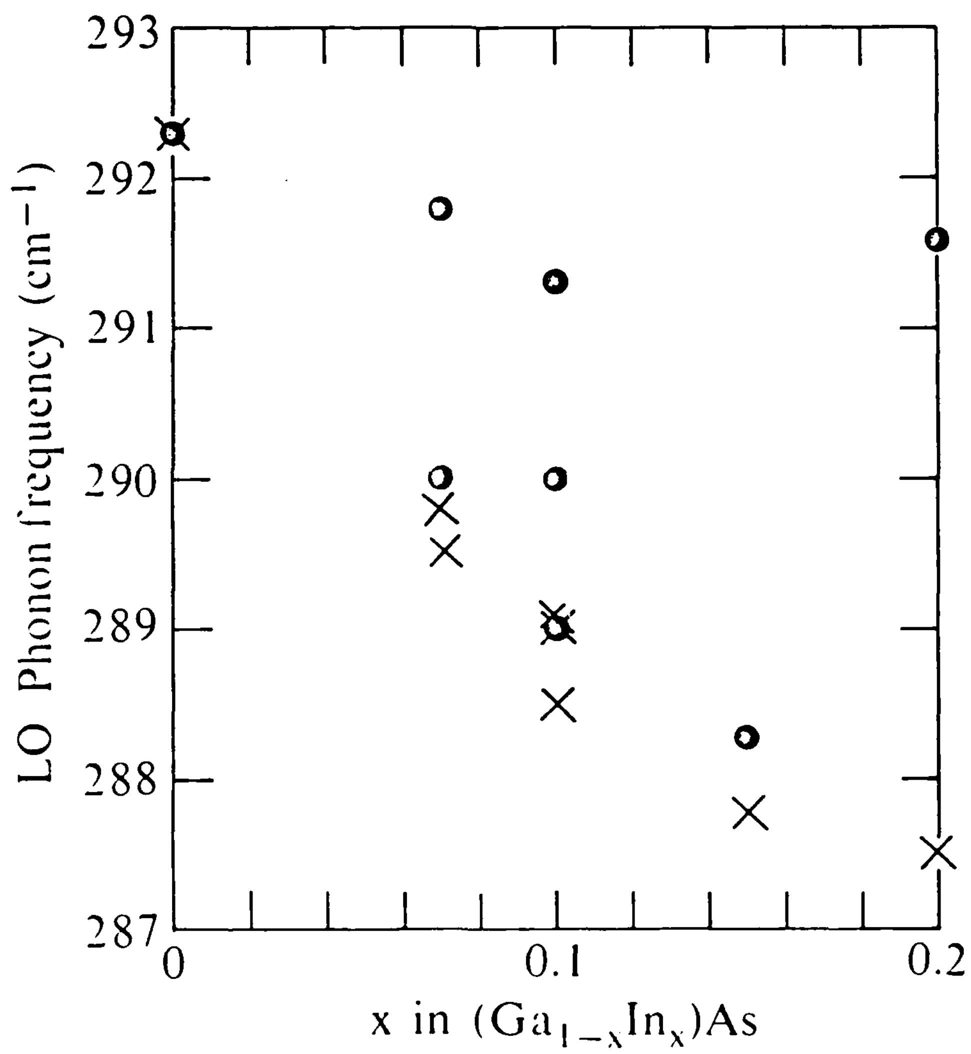
Figure Captions

Fig. 1. The solid circles are the measured frequencies for the samples listed in Table 1. For each sample, a bulk equivalent frequency (cross) is calculated from the Raman measured frequency and the x-ray measured strain, as described in the text.

Table 1 Various properties of our films. The first three columns list the sample number, the composition in $(Ga_{1-x}In_x)As$, and h is the film thickness. The next three columns are the measured x-ray strains and LO is the measured frequency of the LO mode in the film. The last three columns are calculated quantities: the elastic strain parallel (ε_{xx}) , and perpendicular (ε_{zz}) to the sample surface, and the frequency shift.

Sample	x	h	$\varepsilon_{\parallel}^{xr}$	ε_{0}^{xr}	LO	ε_{xx}	ε_{zz}	Freq. Shift
#1	0.07	1000A	0.00	1.21%	291.8cm^{-1}	-0.63%	0.58%	2.03cm^{-1}
#2	0.07	1μm	0.42%	0.71%	289.9cm^{-1}	-0.15%	0.14%	0.49cm^{-1}
#3	0.15	1μm	0.61%	0.91%	288.3cm^{-1}	-0.16%	0.14%	0.50cm^{-1}
387	0.10	1500A	0.00	1.30%	291.3cm^{-1}	-0.67%	0.63%	2.17cm^{-1}
389	0.10	2500A	0.32%	0.98%	290.0cm^{-1}	-0.34%	0.32%	1.03cm^{-1}
388	0.10	1μm	0.46%	0.79%	288.9cm^{-1}	-0.17%	0.16%	0.55cm^{-1}
390	0.20	400A	0.00	2.46%	291.6cm^{-1}	-1.25%	1.21%	4.06cm^{-1}



293
292
291
290
289
288
287
LO Phonon frequency (cm^{-1})
0
0.1
0.2
x in ($Ga_{1-x}In_x$)As

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