Theory of the anomalously low band-gap pressure coefficients in strained-layer semiconductor alloys

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The band-gap pressure coefficients of III-V ternary semiconductor alloys within strained layers are significantly lower than the bulk binary values and the drop in pressure coefficient, of about $6x$ meV/kbar for $In_xGa_{1-x}As$ grown on GaAs remains unexplained. Linear elasticity has been used to predict first-order (linear) effects of pressure, but for strained layers, this procedure fails to predict the pressure coefficients. We show that the nonlinear elasticity theory is necessary, and when evaluated with a consistent level of approximation throughout, it accounts for the pressure coefficients, largely through an approximately linear increase of Poisson's ratio with pressure. Earlier experimental data and some photoabsorption results for $In_xGa_{1-x}As$ on InP are reviewed and they agree well with values predicted using our analysis.

I. INTRODUCTION

Coherency strain arises from the pseudomorphic growth of layers of materials with different lattice constants in the same crystal. Such strained layers have become most well known for their use in the active region of semiconductor lasers. There has been a great deal of basic research carried out on their physical properties. In this paper, we present work on the fundamental elasticity theory that can explain the unusually-low-pressure coefficients of the heavy-hole and light-hole band gaps in strained layers.

As Paul and Warschauer noted, the pressure coefficients of the zone-center band gaps of the III-V semiconductors are all of the order 10 meV/kbar.[1] It is surprising therefore that the presence of only a few percent of indium in an $In_{0.1}Ga_{0.9}As$ strained layer can reduce the pressure coefficient by about 1 meV/kbar. Many speculative ideas have been put forward to account for these low-pressure coefficients. Electronic effects, alloy ordering, misfit-dislocation generation, and nonlinear elasticity have all been suggested.[2,3] However, structural studies have shown that these reduced coefficients are observed for perfect two-dimensional strained layers with abrupt interfaces, so unusual interface effects like the nucleation of quantum dots or dislocation generation can be discounted.

We show that the nonlinear elasticity theory is able to explain the phenomenon. Initially, in Sec. II, we use the nonlinear elasticity theory to derive expressions for the heavy-hole and light-hole band gaps under pressure. In Sec. III, we go on to present the experimental data currently available in the literature and, in Sec. IV, we show how this is consistent with the analysis presented in Sec. II.

II. THEORY

Apart from *ab initio* calculations,[4] to our knowledge, there is currently no complete theory of nonlinear elasticity that can treat nonhydrostatic finite strains, which means that treatments of strained layers under hydrostatic pressure have to be approximations.[2,5–7] Murnaghan's equation of state for a solid under high pressure is the most appropriate theory to use in the analysis of band-gap data from bulk material.[8] Several groups have used this in their work on strained layers under pressure,[5–7] however, the Murnaghan equation of state takes no account of the variation of the individual elastic constants under pressure. Only the bulk modulus is considered, giving rise to the equation of state,

$$1+\frac{\Delta V}{V_0}=\left(1+\frac{B'}{B_0}P\right)^{1/B}, \tag{1}$$

where B_0 and B' are the ambient pressure bulk modulus and its pressure derivative, respectively, V_0 is the initial volume of the solid, and compressive pressure is taken as negative. For a material in a general state of strain, knowledge of the elastic constants is important.

The stress and strain tensors for a pseudomorphic strained layer in cubic symmetry under external pressure P are related. We have

$$\begin{pmatrix} \sigma_0+\sigma & 0 & 0 \\ 0 & \sigma_0+\sigma & 0 \\ 0 & 0 & P \end{pmatrix} = C_{ijkl}\begin{pmatrix} \varepsilon_0+\varepsilon & 0 & 0 \\ 0 & \varepsilon_0+\varepsilon & 0 \\ 0 & 0 & -\nu_{2D}(\varepsilon_0+\varepsilon) \end{pmatrix}, \tag{2}$$

where ε_0 and σ_0 are the stresses and strains in the layer at ambient pressure and can be found by setting ε, σ, and P equal to zero. The terms σ, ε, and P arise from the applied hydrostatic pressure and are to be determined. C_{ijkl} is the elastic stiffness tensor and here it is only the elements C_{11} and C_{12} (in reduced Voigt notation) that are relevant. In general, they will depend upon the applied pressure. The factor ν_{2D} is the biaxial Poisson ratio, equal to $2C_{12}/C_{11}$.

The heavy-hole and light-hole band gaps, for layers grown in the usual $\langle 001\rangle$ orientation, are generally accepted to depend on strain as[9,10]

$$E_g^{hh}=E_g+(a_{CB}-a_{VB})\varepsilon_{vol}-b_{VB}\varepsilon_{ax},$$

$$E_g^{lh}=E_g+(a_{CB}-a_{VB})\varepsilon_{vol}+b_{VB}\varepsilon_{ax}, \tag{3}$$

where E_g is the ambient pressure band gap of the bulk material, a_{CB} and a_{VB} are the hydrostatic deformation potentials of the conduction and valence bands, respectively, and b_{VB} is the axial deformation potential of the valence band. $\varepsilon_{\mathrm{vol}}$ is the volumetric strain, $\varepsilon_{\mathrm{ax}}$ is the axial strain equal to $\frac{1}{2}(\varepsilon_{xx}+\varepsilon_{yy}-2\varepsilon_{zz})$ and ε_{xx}, ε_{yy}, and ε_{zz} are the compressive cartesian strains. Expressions (3) combine the pressure dependences of the conduction-band minimum and the average valence-band maximum in the second term, which describes the variation of the average band gap. The third term describes the light-hole/heavy-hole splitting.

The pressure coefficients measured in experiments are usually quoted for ambient pressure. The band-gap versus pressure data are usually sublinear, but has a well-defined slope at zero pressure from which the pressure coefficient is determined. For this reason we are concerned with the pressure coefficient at ambient pressure, and that means pressure only needs to be considered to first order. At low pressure, $\varepsilon_{\mathrm{vol}}$ is equal to P/B and the second term of Eq. (3) can then be written as $(a_{\mathrm{CB}}-a_{\mathrm{VB}})P/B$. Thus, $(a_{\mathrm{CB}}-a_{\mathrm{VB}})/B$ is, for bulk material, equal to the pressure coefficient we are concerned with, dE_g/dP as $P\rightarrow 0$, and so we shall use experimental values of the pressure coefficient for the second term of Eq. (3).

Within expressions (3), there are a number of terms that can be expected to vary with strain or pressure. The hydrostatic strain varies nonlinearly with hydrostatic pressure. The Poisson ratio can be expected to vary with pressure or strain. It is not unreasonable to expect the deformation potentials themselves to vary with strain. In fact, earlier work by Frogley, Sly, and Dunstan has shown that a better linear fit for the bulk band gap is obtained if one uses density instead of strain or pressure as the independent variable.[11] If the band gap depends linearly on density rather than pressure, then the nonlinear relationship between density and pressure described by the Murnaghan equation would imply a pressure-dependent volume deformation potential. Similarly, one might also expect the axial strain deformation potential to vary with pressure. In this work, we do not include any deformation potential dependence on pressure to avoid confusing the presentation of the nonlinear elastic effects. It would be necessary to invoke these pressure dependences in order to explain the pressure coefficients over a wide range of pressure.

Remarkably, working to first order in pressure requires that we use the nonlinear elasticity theory. This is because we are investigating the pressure coefficient of the band gap and not the band gap itself. The strains appearing in Eq. (3) depend on the elastic constants. The elastic constants vary with pressure and so this must be accounted for when calculating the strains. Essentially, because there are several parameters that will vary with pressure, a consistent level of approximation must be maintained for all terms.

As the pressure is applied, the lattice constants of the substrate and layer vary and so, too, does the misfit. Using Murnaghan's equation, the misfit is

$$\varepsilon_0(P)=1-\left(1+\frac{B_s'}{B_s}P\right)^{-1/3B_s'}\left(1+\frac{B_l'}{B_l}P\right)^{1/3B_l'}\frac{a_s}{a_l}, \quad (4)$$

where the subscripts l and s refer to the layer and substrate, respectively. Equation (4) can be expanded, to first order in pressure, to the well-known expression

$$\varepsilon_0(P)\approx\frac{a_l-a_s}{a_l}+\frac{1}{3}\frac{a_s}{a_l}\left(\frac{1}{B_s}-\frac{1}{B_l}\right)P=\varepsilon_0+\varepsilon_0'P.$$

Then,

$$\varepsilon_{\mathrm{vol}}=[2-\nu_{2\mathrm{D}}(P)](\varepsilon_0+\varepsilon_0'P), \quad (5)$$

$$\varepsilon_{\mathrm{ax}}=2[1-\nu_{2\mathrm{D}}(P)](\varepsilon_0+\varepsilon_0'P). \quad (6)$$

Similarly, the Poisson ratio varies with pressure. In order to calculate the variation, we need to know how the elastic constants vary with pressure. Third-order elasticity theory is available, but it is expressed in terms of strains and does not give a simple method of treating the variation of the elastic constants with pressure.[12] Furthermore, it does not take into account finite deformation[12] and has previously failed to account for the low pressure coefficients.[2] Rather than using the third-order theory, we choose to use the experimentally determined pressure derivatives of the elastic constants C_{ij}'. Thus, the elastic constants depend linearly on pressure and with the C_{ij}' as the coefficients. This automatically accounts for finite deformation and is entirely consistent with the way bulk modulus depends on pressure in the Murnaghan equation. In this way, the two-dimensional Poisson ratio varies with pressure as

$$\nu_{2\mathrm{D}}(P)=+2\frac{C_{12}+C_{12}'P}{C_{11}+C_{11}'P}\approx+2\frac{C_{12}}{C_{11}}-2\frac{C_{12}C_{11}'-C_{11}C_{12}'}{C_{11}^2}P$$
$$=\nu_{2\mathrm{D}}+\nu_{2\mathrm{D}}'P. \quad (7)$$

The expressions (5) and (7) can now be used in Eq. (3) to obtain the pressure dependence of the band gaps. The axial strain varies with pressure because both Poisson's ratio, which is used to calculate the growth direction strain from the misfit, and the misfit itself vary with pressure. The heavy-hole band gap is

$$E_g^{\mathrm{hh}}(P)=E_g+(a_{\mathrm{CB}}-a_{\mathrm{VB}})[2-\nu_{2\mathrm{D}}(P)]\varepsilon_0(P)$$
$$-b_{\mathrm{VB}}[1+\nu_{2\mathrm{D}}(P)]\varepsilon_0(P). \quad (8)$$

This can be expanded to first order in pressure as

$$E_g^{\mathrm{hh}}(P)\approx E_g+(a_{\mathrm{CB}}-a_{\mathrm{VB}})(2-\nu_{2\mathrm{D}})\varepsilon_0-b_{\mathrm{VB}}(1+\nu_{2\mathrm{D}})\varepsilon_0$$
$$+\frac{a_{\mathrm{CB}}-a_{\mathrm{VB}}}{B_l}P+\{(a_{\mathrm{CB}}-a_{\mathrm{VB}})[(2-\nu_{2\mathrm{D}})\varepsilon_0'$$
$$-\varepsilon_0\nu_{2\mathrm{D}}']\}P-b_{\mathrm{VB}}[(1+\nu_{2\mathrm{D}})\varepsilon_0'+\varepsilon_0\nu_{2\mathrm{D}}']P, \quad (9)$$

from which it is possible to read off an expression for the pressure coefficients of the layers at low pressures as

$$\left.\frac{\partial E_g^{\mathrm{hh\{lh\}}}}{\partial P}\right|_{P\rightarrow 0}\approx\frac{a_{\mathrm{CB}}-a_{\mathrm{VB}}}{B_l}+\{(a_{\mathrm{CB}}-a_{\mathrm{VB}})[(2-\nu_{2\mathrm{D}})\varepsilon_0'$$
$$-\varepsilon_0\nu_{2\mathrm{D}}']\}-\{+\}b_{\mathrm{VB}}[(1+\nu_{2\mathrm{D}})\varepsilon_0'+\varepsilon_0\nu_{2\mathrm{D}}']. \quad (10)$$

TABLE I. Room-temperature values for parameters in Eqs. (5), (7), and (10), used to obtain the predictions shown in Fig. 2. The values in parentheses are those taken from the literature. The bulk-pressure coefficients are taken from Frogley *et al.*, and Tsay, Mitra, and Bendow. Elastic and lattice constants and their pressure dependencies are taken from the INSPEC-EMIS Datareview Series (Refs. 25-27). The axial deformation potentials are taken from Krijn (Ref. 9). N/A indicates that these values were not required for the calculation.

Material	$\frac{a_{CB}-a_{VB}}{B}$ (meV/kbar)	b_{VB} (eV)	C_{11} (kbar)	C_{12} (kbar)	C'_{11}	C'_{12}	a (Å)
GaAs	11.6	1.7	1184	537	3.9	4.8	5.6536
	(11.6)	(1.7)	(1184)	(537)	(4.6)	(4.4)	(5.6536)
InAs	9.6	1.8	832.9	453	3.9	4.8	6.0583
	(9.6–10.8)	(1.8)	(832.9)	(453)	(4.5)	(4.9)	(6.0583)
InP	N/A	N/A	1022	573	N/A	N/A	5.8687
			(1022)	(573)			(5.8687)

The terms of Eq. (10) can be interpreted as follows. The term $(a_{CB}-a_{VB})/B_l$ is the usual band-gap pressure coefficient that includes the shift of both the conduction band and the average valence-band energies. The second term represents the shift in average band gap of the heavy and light holes from the change in misfit, and Poisson's ratio of the layer and substrate under pressure. The change in Poisson's ratio is important because for mismatched layers there is hydrostatic strain even at ambient pressure. The third term represents the heavy-hole/light-hole splitting and again arises from the change in misfit and Poisson's ratio of the layer and substrate.

It is primarily the second term that accounts for the low-pressure coefficients and that has previously been overlooked. While a typical value for $1/B_l$ is 1.33 Mbar^{-1}, for the values given in Table I $(2-\nu_{2D})\varepsilon'_0$ is insignificant, being of order 0.004 Mbar^{-1}, but $\varepsilon_0\nu'_{2D}$ is of order 0.08 Mbar^{-1} for 1.5% misfit strain, which has a considerable effect.

III. EXPERIMENTAL REVIEW AND RESULTS

Several groups have studied the pressure dependence of the band gap in quantum wells made of strained semiconductor alloys.[2,3,5,13–18] The most complete work has been carried out on the $In_xGa_{1-x}As/GaAs$ system. Wilkinson *et al.*[2] investigated both the effects of composition and well width of the pressure coefficients. As well as measuring alloy pressure coefficients lower than expected from a linear interpolation between bulk values, they showed that the pressure coefficient depends on well width and converges asymptotically for very wide wells. This has been independently corroborated by Hou *et al.*[5] and we will make use of this result when comparing data from experiments on samples with different widths. The results of Wilkinson *et al.*[2] and Shen *et al.*[3] are particularly important because the pressure coefficients of the layers were determined by measuring the energies of several different strained layers relative to the bulk GaAs substrate. In this way the error in pressure calibration is not important as pressure becomes a dummy variable. The relative pressure coefficients of the layers are accurately determined.

There are also data concerning the light-hole/heavy-hole splitting available, although this often relies on comparing measurements of the pressure coefficients of the light hole and heavy hole from separate analyses and so the accuracy is often poor. With the substrate for comparison, the data of Shen *et al.*[3] are most reliable and indicate a pressure coefficient for the light-hole/heavy-hole splitting of not more than 0.1 meV/kbar for $In_xGa_{1-x}As$ layers on GaAs with up to 20% indium content. It is important to know, too, how the splitting behaves at larger indium content and on a different substrate. We studied $In_xGa_{1-x}As$ superlattices grown on InP, with twenty-five 50 nm compressive layers ($x=0.615$, $\epsilon_0=0.6\%$) alternating with 50-nm tensile layers ($x=0.50$, $\epsilon_0=-0.2\%$) to prevent strain relaxation. The heavy-hole and light-hole band gaps of the compressive layers were clearly visible in absorption spectra measured at room temperature. Loaded in a diamond-anvil cell with argon as the hydrostatic pressure medium, the splitting was found to vary, up to 50 kbar, at 0.05 ± 0.1 meV/kbar (Ref. 19)—again, essentially independent of pressure.

The experimental data available in the literature are presented in Fig. 1. The data as a whole show considerable scatter. However, when considering individual sets of data, one can see a linear decrease in pressure coefficient with indium content or strain that is consistent between data sets from GaAs and InP substrates. The solid lines in the figure are guides to the eye for three of the sets of data and serve to highlight the decrease of pressure coefficient with compressive strain.

The scatter in the data can be reduced by making corrections. Where the pressure coefficients were measured relative to a GaAs substrate, the coefficient given for GaAs by Frogley, Sly, and Dunstan, is used.[11] They showed that the most appropriate value for the pressure coefficient of bulk GaAs is not 10.7 meV/kbar,[20–24] but 11.6 meV/kbar. Therefore, the data of Wilkinson *et al.*[2] and Shen *et al.*[3] are rescaled to this higher value for the GaAs bulk coefficient. The other data are not related to a bulk substrate and cannot be rescaled in the same way.

The width of the wells affects the pressure coefficients. The results of Wilkinson *et al.* and Hou *et al.* can be used to

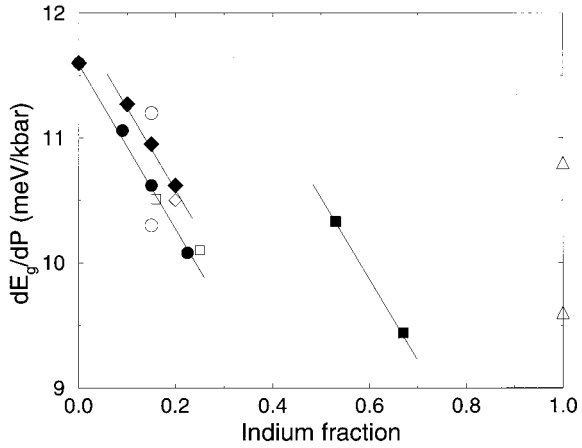

FIG. 1. Values given in the literature for the pressure coefficients of the heavy-hole band gap. Corrections are made for layer thickness effects. The solid circles are from Wilkinson *et al.*, solid diamonds from Shen *et al.*, solid squares from Sly and Dunstan, open squares from Hou *et al.*, open circles from Shan *et al.*, the open diamonds from Li *et al.*, and the upwards-open triangles from Tsay, Mitra and Bendow. The straight lines through the three sets of solid points indicate the results on which we place emphasis in this paper. The three data points at $x=0.0$, 0.53, and 1.00 are for unstrained material; all others are for pseudomorphic strained layers.

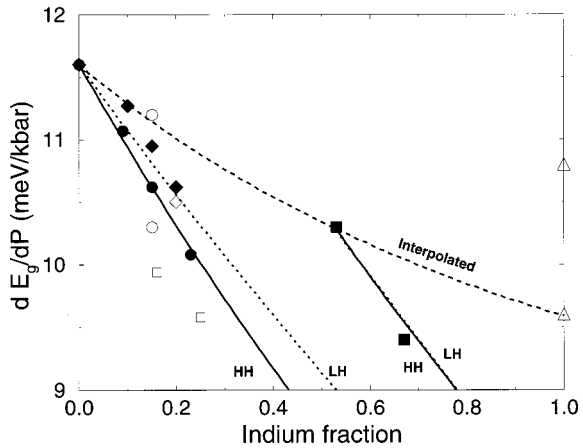

FIG. 2. Predictions for pressure coefficients from Eq. (9) and the experimental values given in the literature. The experimental data have been corrected for well-width effects, and for the GaAs bulk-pressure coefficient. The dashed line is an interpolation between GaAs and InAs for unstrained $In_xGa_{1-x}As$. The two solid lines are the predictions from Eq. (10) for the heavy-hole pressure coefficient for $In_xGa_{1-x}As$ strained layers on both GaAs and InP substrates. The dotted lines are the predictions for the light-hole pressure coefficients. The solid circles are from Wilkinson *et al.*, solid diamonds from Shen *et al.*, solid squares from Sly and Dunstan, open squares from Hou *et al.*, open circles from Shan *et al.*, open diamonds from Li *et al.*, and upwards-open triangles from Tsay, Mitra and Bendow.

correct for this as follows. The single point of Li *et al.* should be shifted down by 0.1 meV/kbar to account for the thin well of 80 Å. Shan *et al.* gives two values for a 15% In composition that are different by 0.9 meV/kbar. The difference in layer thickness, 80 Å and 150 Å, cannot account for the discrepancy. The value of 11.2 meV/kbar is corrected down to 11.1 meV/kbar to account for the thin well. Hou *et al.* gives values for compositions of 16% and 25% In. For the 16% In composition, they studied a variety of well widths and showed convergence to about 9.9 meV/kbar for wide wells. For the 25% In samples, they only looked at thin wells and the values of 10.1 and 10.0 meV/kbar should be corrected downwards to 9.6 meV/kbar.

The data of Fig. 1 are shown again in Fig. 2 following these corrections. Even after correction the data are not all entirely consistent. This is not surprising because it is known that the band-gap pressure dependence is sublinear. The different experimental groups measure over different pressure ranges and often use linear fits to their data, so the pressure coefficients they find are likely to differ.[11] For the purposes of comparison to the predictions of Eq. (10), most emphasis should be placed on obtaining the slopes of the data sets of Wilkinson *et al.*,[2] Shen *et al.*,[3] and Sly and Dunstan[16] (shown as solid symbols in Figs. 1 and 2) because, for these experiments, problems with pressure calibration and finite pressure range are avoided by measuring coefficients relative to the substrate.

IV. DISCUSSION

All the parameters in Eq. (10) are available in the literature,[26–28] and are given in Table I. The literature values of the elastic constants typically have errors of a few percent and their pressure derivatives have a considerably larger error.[26–28] Although there is a large error associated with the individual pressure derivatives, they must combine to give a bulk-modulus pressure derivative of about 4.5, because this value has been well tested.[29] There is no appreciable error associated with the lattice constants. There are no measurements for the elastic constants for $In_xGa_{1-x}As$ alloys and there is no standard interpolation scheme for calculating them from the bulk values. The elastic constants were chosen so that they followed Keyes's empirical rule for the scaling of elastic constants with the lattice constant. This meant choosing elastic constants such that the bulk modulus of an $In_xGa_{1-x}As$ layer lattice matched to InP was equal to that of the InP substrate. To achieve this, a positive bowing parameter of 0.25 Mbar was used to interpolate the elastic constants of $In_xGa_{1-x}As$ alloys. The constants were given by

$$C_{In_xGa_{1-x}As}=xC_{InAs}+(1-x)C_{GaAs}+x(1-x)\ \ 0.25\ \ \text{Mbar}. \tag{11}$$

The band-gap pressure coefficient for unstrained bulk $In_xGa_{1-x}As$ was interpolated between the well-established GaAs (Ref. 11) value and the less certain InAs (Ref. 25) value. A bowing parameter of 1.5 meV/kbar was used instead of a linear interpolation for agreement with the experimental value of the coefficient for lattice-matched (unstrained) $In_xGa_{1-x}As$ grown on InP.

The pressure dependence of the elastic constants can also be derived from the third-order elastic constants of nonlinear elasticity.[12] Using experimental values for the third-order elastic constants and the expressions of Sklar *et al.*,[12] one finds C'_{11} and C'_{12} to be about 5.5 and 3.3, respectively. These values are consistent with the values of 4.5 for B', but

compared with the experimentally observed values of about 4.5 for both C_{11}' and C_{12}',[26–30] indicate the range of uncertainty in these quantities.

The predictions of Eq. (10) are shown alongside the experimental data (corrected as described in Sec. III) in Fig. 2. Save for the data of Shan *et al.* and Hou *et al.*, the results agree remarkably. It should be noted that these two data sets are less reliable than the data that were referenced to the substrate. After adjustment for well thickness, the two values given by Shan *et al.* lie either side of that predicted by Eq. (10). The two values given by Hou *et al.* do not agree with our predictions; however, they also disagree with most of the other experimental data. The coefficient for a 20% In layer given by Li *et al.* is in good agreement and the data of Sly and Dunstan fits with the predictions.

In agreement with our recent results and with Shen *et al.*,[3] the pressure dependence of the light-hole/heavy-hole splitting is predicted to be small for the $In_xGa_{1-x}As$ on GaAs and $In_xGa_{1-x}As$ on InP. This is a consequence of the bulk moduli for the lattice-matched material being equal to that of the substrate in our interpolation scheme. This adds weight to the earlier observation of Prins and Dunstan[31] that lattice-matched layers have bulk moduli equal to their substrates.[31]

The fit is obtained using the set of parameters shown in Table I. Good agreement between Eq. (10) and the experiment could be obtained by varying the pressure derivative of the elastic constants within their uncertainty.

V. CONCLUSIONS

A treatment to first order in pressure of nonlinear elastic effects is able to explain the large decrease in pressure coefficient for strained III-V layers relative to their unstrained bulk values. This is achieved by including the rate of change of both the misfit strain and the Poisson's ratio with pressure. Predictions calculated using parameters from the literature agree very well with experimental data.

The theory also explains the very low-pressure coefficient of the light-hole/heavy-hole splitting for the $In_xGa_{1-x}As$ strained layers. By following Keyes's rule and choosing a bulk modulus for a lattice-matched layer equal to that of its substrate, the theory predicts very little variation of the light-hole/heavy-hole splitting with pressure.

In short, we have shown that the low band-gap pressure coefficients of $In_xGa_{1-x}As$ alloys within strained layers can be understood in terms of nonlinear elasticity.

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