



Is there an elastic anomaly for a (001) monolayer of InAs embedded in GaAs?

>Ua Yg'9''6YfbUfX'UbX'5'YI 'Ni b[Yf'

7]Ujcb.'5dd]YX'D\ng]Mg'@YHYfg'65z%') f%- (L/Xc]. '%\$"%\$*' #/%%&* * \$

J]Yk'cb'jbY.'\hhd.##XI "Xc]"cf[#/%\$"%\$*' #/%%&* * \$

J]Yk' HUVY'cZ7 cbHYbng.\hhd.##gV]Ujcb"Ujd"cf[#/%\$"%\$*' #/%%&* * \$) #&3j Yf1dXZVj

Di V]g\YX'VmH\Y'5-D' Di V]g\]b[

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H\Yfa c!d]YncWYa]gfmZ-b5g'cb'; U5gf\$%\$%

5dd""D\ng"@YH'80z'(+(+ f&\$&\$&L/%\$"%\$*' #/%%(, - +\$(

8YYd'Yj Y'fUbg]YbhgdYVfcgVcdmUbX'dgYi XcdchYbh]U'gi dYfUHQW'WU'W'Ujcb'Zcf'Ub'5g'a cbc'UnYf

Ya VYXXYX'jb'; U5g'

>"5dd""D\ng"88z-(, f&\$&\$&L/%\$"%\$*' #/" +' +*%

GdYVfcgVcd]WV`]dgca Yhf]Wgh X]Yg'cZ-b5g'a cbc'UnYfg'Ya VYXXYX'jb'; U5g'

5dd""D\ng"@YH'70z%' * f%- +L/%\$"%\$*' #/%%&* ,)'

GfU]b'UbX'fY'UI Ujcb'jb'5g'UbX'5g'; U5g'Z'a g:[fck b'cb'; U5gf\$%\$%

5dd""D\ng"@YH'68z' '\$%\$ f%- * L/%\$"%\$*' #/%%&* , \$'

C dh]W'V\UfUWVf]nUjcb'cZ-b5g'a cbc'UnYfg'fi Vh fYg:[fck b'cb'f%\$ t5'UbX'f\$%\$%; U5g'gi VghUfYg'

5dd""D\ng"@YH'62z'%'\$%\$ f%- -' L/%\$"%\$*' #/%%\$,) *%

Is there an elastic anomaly for a (001) monolayer of InAs embedded in GaAs?

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When a coherently grown (001)-oriented layer of InAs is embedded in a GaAs host, the coherency strain induces a perpendicular distortion of the embedded layer, predicted by continuum elasticity theory to be $\epsilon_{\perp} = 7.3\%$. Brandt, Ploog, Bierwolf, and Hohenstein, [Phys. Rev. Lett. **68**, 1339 (1992)] have described a high resolution electron microscopic analysis of such buried layers that appears to

report on an investigation into whether a first-principles local-density total energy minimization shows such an elastic anomaly in the monolayer limit. We find that it does not.

When a layer of material with cubic lattice parameter a_0 is grown coherently in a host with cubic lattice parameter $a_h \neq a_0$, the buried layer undergoes a distortion of its lattice parameter c perpendicular to the plane of the layer in response to the distortion of its lattice parameter parallel to the plane. Since surface effects are absent, the amount of the distortion is determined by minimizing the bulk energy as a function of all the structural degrees of freedom. The simplest way to approximate this is to use harmonic continuum elasticity theory (see, e.g., Ref. 1). For example, for cubic materials with a layer orientation parallel to (001), the perpendicular strain $\epsilon_{\perp} = (c - a_0)/a_0$ is related¹ to the parallel strain $\epsilon_{\parallel} = (a_{\parallel} - a_0)/a_0$ by

$$\epsilon_{\perp} = -2 \frac{C_{12}}{C_{11}} \epsilon_{\parallel}, \quad (1)$$

where the C_{ij} are elastic constants of the embedded material. For example, with the measured² $C_{11} = 0.8329$ and $C_{12} = 0.4526$ (Tdyn/cm²) for InAs, Eq. (1) predicts $\epsilon_{\perp} = 7.27\%$ for a layer of InAs buried in GaAs.

Recently, Brandt *et al.*³ examined the strain of buried (001) layers of InAs in GaAs experimentally, via a high-resolution transmission electron microscope (HRTEM) technique.

Responding to $\epsilon_{\parallel} = 12.46\%$, much greater than the prediction of Eq. (1), Brandt *et al.* concluded that the harmonic elasticity theory applied to a three monolayer film of InAs revealed a lattice distortion corresponding to $\epsilon_{\perp} = 6.06\%$ in good agreement with Eq. (1). Thus, they concluded that the widely used harmonic elasticity theory breaks down in the extreme limit of a single monolayer.

Equation (1) involves several approximations: (1) the strain is assumed to be uniform in the monolayer; (2) the strain energy is minimized as a function of the lattice parameter of the embedded layer. Regarding (1), we can expect the strains, however, the lattice mismatch between GaAs and InAs ($\approx 7\%$) is not sufficient to cause a substantial departure

from harmonicity. Regarding (2), it was previously shown⁴ that for certain interface orientations nonuniform atomic relaxations near an interface can be large. But for (001)-oriented interfaces the allowed relaxations all lie parallel to [001], and the nonuniformity of these interplanar relaxations does not substantially alter the applicability of the simple picture provided by continuum elasticity theory. Regarding (3), the question of anisotropy dependence has, until recently, been untested in semiconductor layers.⁵ One might imagine, however, that the anisotropy of the elastic constants, which is small, interaction between them might substantially alter the theory.

In view of the unprecedented result of Brandt *et al.*,³ we tested the validity of Eq. (1) via an atomistic, nonharmonic, first-principles theory of the relaxations of such a system to determine whether it would reveal the breakdown ostensibly demonstrated in the experiment. Our first-principles results in no way depend upon Eq. (1) or the approximations on which it is based. We found good agreement with Eq. (1) and no theoretical evidence for such a breakdown, in spite of using state-of-the-art techniques for relaxing the total energy of the system.

Our tests improve on harmonic continuum elasticity theory in two steps. First, we employ the same discrete atomistic approach, still limited to purely elastic energies. This is done with the Keating valence-force-field (VFF) model, which uses a description in terms of microscopic quantities: the two-body bond stretching and three-body bond bending elastic energies. The strain energy is minimized as a function of $\{R_i\}$, the set of bond lengths and bond angles.

Second, we employ a first-principles method for relaxing the total energy of the system. This is done with the local-density approximation (LDA). With the total energy represented as a functional of the electron density of the system, all contributions

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to it are treated on the same quantum-mechanical footing. This avoids all the potential pitfalls discussed above, and can be regarded as giving the best available theoretical estimate

In the first step, we applied the VFF model, with the bond lengths, to the calculation of the relaxed geometry of a structure containing an embedded (001)-oriented layer of InAs in a GaAs host, all confined to the GaAs lattice constant in the (001) plane. We found values ranging from $\epsilon_1 = 7.09\%$ with 1 monolayer (ML) of InAs, to $\epsilon_1 = 7.17\%$ with 10 ML.⁹ This demonstrates good agreement with the prediction of Eq. (1) and shows no evidence of anomalously large distortion in the ultrathin limit.

We note that in a recent paper Massies and Grandjean¹⁰ used a one-dimensional variant of the VFF model to investigate the behavior of monolayer-height InAs surface islands on GaAs, finding a relaxation away from the surface larger

ever, the clear agreement of our bulk three-dimensional VFF calculations with continuum elasticity theory demonstrates that this comparison of a one-dimensional model of surface relaxation with bulk strain is inappropriate.

The inability of the VFF model to reproduce the

method¹¹ pseudopotentials were generated by the method of

equivalent of 10 special points¹² in the irreducible zinc-

5%, far less than the more than 70% difference between the

prediction of Eq. (2). The theoretical equilibrium lattice constants of zinc-blende GaAs and InAs were determined by fitting to a Birch equation of state¹⁵ the total energies from seven calculations with differing lattice constants. The fit gives the values (\AA) 5.60 ± 0.005 (5.65) for GaAs and 6.02 ± 0.005 (6.06) for InAs, where experimental values² are

constants induces only about a 4% (0.3 percentage points) overestimation in our calculated ϵ_1 .

For the first-principles calculations of the properties of this layer of InAs buried in a GaAs host we used a model

to obtain the structural effect of a thick GaAs substrate. Tests

more than adequate to converge the structural features of the interface and the GaAs layer to match those with much thicker layers. We minimized the SL total energy with respect to all structural degrees of freedom, subject to the coherency constraint. Quantum-mechanical forces¹¹ were used to relax the atomic positions, and a fit of total energy vs c/a

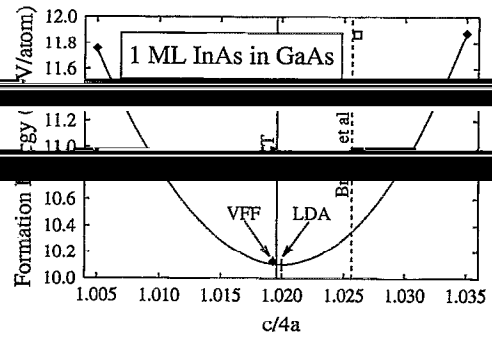


FIG. 1. Formation energy of the atomically relaxed eight-layer structure containing 1 monolayer of InAs vs its $c/4a$ ratio. Filled diamonds denote direct first-principles LDA results; the curve is a cubic fit to those points. The minimum of the fit curve is marked by a vertical tick, labeled "LDA." The diamond symbol labeled "VFF" lies at the $c/4a$ value predicted by the VFF model. The solid vertical line, labeled "CET," marks the prediction of

the InAs layer has $\epsilon_1 = 12.5\%$, illustrating the large energy penalty incurred if the strain is concentrated in the InAs layer.

was used to find the equilibrium c/a_h ratio. While the symmetry of the structure permits additional relaxations beyond the relaxation of merely the InAs bond length, the effect of these was found to be rather small.

Our results are illustrated for the single-monolayer case also shown. The equilibrium c/a_h ratios for the three meth-

lattice constants). We estimate the error in the LDA value

ratio to be about 1%. Similarly good agreement among the

recent pseudopotential calculation by Shiraishi and Yamaguchi.¹⁶ However, correspondence with one of the authors¹⁷ has clarified that their results do not, in fact, show evidence of an elastic anomaly in the monolayer limit.

One situation that could increase the apparent measured

so because the determination of ϵ_1 is based on the overall measured shift in the lattice.⁹ Because the VFF model provides a sufficiently accurate reproduction of the structural parameters obtained from the LDA calculations, we used it

respectively can raise ϵ_1 to 13.05%, comparable to the

refinement of the analysis of the HRTEM data of Ref. 3, permitting more precise determination of the individual spacings, suggests that they should be able to detect excess In, at least in the two larger amounts we have suggested.²⁰

It is interesting that the effect of substituting In in the first subsequently grown GaAs layer would be to change the

ratio of the two expanded spot-row spacings in different [110] or along [110]—in one case increasing the ratio, and in the other case decreasing it. Thus it would be possible to compare such images as another test for excess In. To our knowledge, this has not been done.

In a recent paper, Giannini *et al.*²¹ have studied the composition of InAs to be distributed among three layers, with 75% in the first layer, 25% in the second, and 15% in the third. Unfortunately, the samples used for this study were not characterized by HREM, so it is not clear whether they would exhibit the expected lattice expansion.

principles total-energy methods to the determination of the equilibrium lattice constants of the layers, in order to determine whether such a layer would exhibit the expected lattice expansion.

ment to help determine whether the experimental results of Brandt *et al.*³ could be a consequence of excess In in the GaAs layer.

mental findings, and further work to attempt to resolve the discrepancy is warranted.

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InAs in GaAs (2.55 Å). However, Dr. Shiraishi has informed us that the lattice constant of the InAs layer is 2.49 Å, corresponding to $\epsilon_1 = 4.6\%$.

the calculated bond length to their calculated zinc blende GaAs bond length. Their calculated bond length for the embedded layer is 2.49 Å, corresponding to $\epsilon_1 = 4.6\%$.

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