Phase stability and the arsenic vacancy defect in $\text{In}_x\text{Ga}_{1-x}\text{As}$

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(Received 13 September 2011; revised manuscript received 19 October 2011; published 17 November 2011; corrected 18 November 2011)

The introduction of defects, such as vacancies, into $\text{In}_x\text{Ga}_{1-x}\text{As}$ can have a dramatic impact on the physical and electronic properties of the material. Here we employ ab initio simulations of quasirandom supercells to investigate the structure of $\text{In}_x\text{Ga}_{1-x}\text{As}$ and then examine the energy and volume changes associated with the introduction of an arsenic vacancy defect. We predict that both defect energies and volumes for intermediate compositions of $\text{In}_x\text{Ga}_{1-x}\text{As}$ differ significantly from what would be expected by assuming a simple linear interpolation of the end member defect energies/volumes.

DOI: 10.1103/PhysRevB.84.184108 PACS number(s): 71.55.Eq

I. INTRODUCTION

In recent years the $\text{In}_x\text{Ga}_{1-x}\text{As}$ system has drawn significant interest due to a myriad of potentially useful applications. These include use as a material for $n$-channel metal-oxide field-effect transistors ($n$-MOSFETS)\textsuperscript{1,2} and as a bottom cell material in multijunction solar cells.\textsuperscript{3} Additionally, the efficient manipulation of quantum dots (QD) using strain-driven epitaxy has opened up the possibility of applications in optoelectronic devices such as high-efficiency QD lasers.\textsuperscript{4}

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employ DFT simulations to investigate the thermodynamics of the \( \text{In}_x\text{Ga}_{1-x}\text{As} \) system and compare our results with the thermodynamic data from the literature before moving on to examine the formation energy of the arsenic vacancy defect as a function of \( x \).

II. METHODOLOGY

A. Density-functional theory simulations

All DFT simulations presented here were conducted using the CASTEP 5.5 simulation package.\(^{18,19}\) CASTEP is a plane-wave pseudopotential code that describes a crystal using supercells and periodic boundary conditions with special point integration over the Brillouin zone. Exchange correlation was described using the generalized gradient approximation of Perdew, Burke, and Ernzerhof (GGA-PBE).\(^{20}\) A Monkhorst-Pack\(^{21}\) scheme is used to sample the Brillouin zone with a density of \( 0.03 \text{Å}^{-1} \) on each axis, corresponding to \( 3 \times 3 \times 3 \) and \( 2 \times 2 \times 2 \) \( k \)-point grids for the 64 and 216 atom supercells, respectively. Pseudopotentials were developed using the “on-the-fly” pseudopotential generating feature in CASTEP and the smoothing parameters were optimized following Murphy et al.\(^{22}\) Semicore 3d and 4d electrons are treated as valence for Ga and In, respectively, and the plane-wave cutoff energy was set to \( 380 \text{eV} \), offering a precision of \( 10^{-2} \text{eV} \) per formula unit. Finally, the Fourier transform grid for the electron density was larger than that of the wave functions by a scaling factor of 2.0 (represented in CASTEP using the grid_scale parameter).

To ensure that the optimum precision was obtained, we used a convergence criterion for the self-consistent simulations of \( 5 \times 10^{-6} \text{eV} \). Similarly, robust criteria for the geometry optimization were employed as follows: energy \( = 2 \times 10^{-3} \text{eV \text{atom}^{-1}} \), forces \( = 0.05 \text{eV/Å} \), stress \( = 0.1 \text{GPa} \), and displacement \( = 2 \times 10^{-3} \text{Å} \).

Five compositions of \( \text{In}_x\text{Ga}_{1-x}\text{As} \) have been studied corresponding to \( x = 0.0, 0.25, 0.5, 0.75, \) and 1.0, where \( x = 0.0 \) and 1.0 correspond to the end members GaAs and InAs, respectively. The arsenic vacancy defect was selected for investigation here due to it being the only vacancy defect that can exist across the entire compositional range.

B. Special quasirandom structures

In order to model a disordered system, such as the 4a sublattice in \( \text{In}_x\text{Ga}_{1-x}\text{As} \), while still retaining full atomic detail and, including all local relaxation effects, Zunger et al.\(^{23}\) have developed a series of special quasirandom supercells (SQS). These small unit cell structures closely mimic the most relevant near-neighbor pair and multisite correlation functions of random alloys. The efficacy of DFT simulations employing SQS cells has been established by Haas et al.\(^{24}\) The simulations presented here employed 64 atom supercells based on the SQS methodology and are the same as those used in our previous work\(^{25}\) (more details, including pictorial representations of the supercells, can be found in this previous work). This combination of DFT simulations and SQS supercells has been used previously to examine the stability of \( E \) centers in \( \text{Si}_{1-x}\text{Ge}_x \).\(^{26}\)

C. Defect formation formalism

Within the supercell methodology, the formation of a defect can be considered as exchange between the host material and some atomic and/or electronic reservoir. Following the formalism of Zhang and Northup\(^{27}\) the formation energy of a defect is given by

\[
E_F = E_{\text{DFT}}(D) - E_{\text{DFT}}(\text{In}_x\text{Ga}_{1-x}\text{As}) + \sum_i n_i \mu_i + q_D E_f,
\]

where \( E_{\text{DFT}}(D) \) and \( E_{\text{DFT}}(\text{In}_x\text{Ga}_{1-x}\text{As}) \) are the DFT total energies of \( \text{In}_x\text{Ga}_{1-x}\text{As} \) with and without the defect, \( D \); \( n_i \) is the number of atoms removed/added (in this convention addition of an atom would result in a negative value for \( n_i \)); \( \mu_i \) is the chemical potential of the atoms removed; \( q \) is the charge on the defect; and \( E_f \) is the Fermi energy. As this study is limited to the investigation of charge-neutral defects, the final term in Eq. (2) can be neglected. Furthermore, it is not necessary to employ any post-simulation corrections normally associated with the calculation of defect formation energies of charged defects in semiconductors.\(^{28}\)

III. RESULTS AND DISCUSSION

A. Elemental solids

Single unitcells of the elemental In, Ga, and As were relaxed to the criteria described in Sec. II.A. Table I shows that there is excellent agreement between the lattice parameters predicted by our simulations and the experimental values. The DFT values are slightly larger than the experimental values, which is common for simulations employing the GGA exchange correlation functional.

Atomization energies were determined using Eq. (3) as follows:

\[
E_a = E(\text{In}_x\text{Ga}_{1-x}\text{As}) - [i E(\text{In}) + j E(\text{Ga}) + k E(\text{As})],
\]

where \( E(\text{In}_x\text{Ga}_{1-x}\text{As}) \) is the energy per formula unit of \( \text{In}_x\text{Ga}_{1-x}\text{As} \) (for example, if \( j = k = 0 \), this corresponds to elemental In). \( E(\text{In}), E(\text{Ga}), \) and \( E(\text{As}) \) are the reference

<table>
<thead>
<tr>
<th>Element</th>
<th>Property</th>
<th>DFT</th>
<th>Experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>In</td>
<td>a /Å</td>
<td>3.23</td>
<td>3.25(^{30})</td>
</tr>
<tr>
<td>I4/MMM</td>
<td>c /Å</td>
<td>5.28</td>
<td>4.95(^{30})</td>
</tr>
<tr>
<td></td>
<td>Volume /Å(^3)</td>
<td>54.91</td>
<td>52.26(^{30})</td>
</tr>
<tr>
<td></td>
<td>( E_a ) /eV</td>
<td>2.32</td>
<td>2.26(^{30})</td>
</tr>
<tr>
<td>Ga</td>
<td>a /Å</td>
<td>2.90</td>
<td>2.81(^{31})</td>
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<tr>
<td>I4/MMM</td>
<td>c /Å</td>
<td>4.57</td>
<td>4.46(^{31})</td>
</tr>
<tr>
<td></td>
<td>Volume /Å(^3)</td>
<td>38.34</td>
<td>35.15(^{31})</td>
</tr>
<tr>
<td></td>
<td>( E_a ) /eV</td>
<td>2.61</td>
<td>2.81(^{32})</td>
</tr>
<tr>
<td>As</td>
<td>a /Å</td>
<td>3.81</td>
<td>3.76(^{31})</td>
</tr>
<tr>
<td>R3-MH</td>
<td>c /Å</td>
<td>10.62</td>
<td>10.55(^{31})</td>
</tr>
<tr>
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<td>Volume /Å(^3)</td>
<td>133.18</td>
<td>129.12(^{31})</td>
</tr>
<tr>
<td></td>
<td>( E_a ) /eV</td>
<td>3.01</td>
<td>3.12(^{31})</td>
</tr>
</tbody>
</table>
states of the constituent atoms modeled as single atoms in a monoclinic cell with \( a = 15.79 \text{ Å}, b = 15.50 \text{ Å}, c = 15.00 \text{ Å}, \) and \( \beta = 98.75^\circ \) with periodic boundary conditions. Spin-polarized calculations used 1, 1, and 3 unpaired electrons for In, Ga, and As atoms, respectively. The atomization energies for the elemental solids are in good agreement with the experimental data measured at 298.15 K to derive a value of 6.62 eV for GaAs of 6.20 eV using the B3LYP functional. The dashed line in Fig. 2 shows a small positive enthalpy of mixing across the entire composition range, in agreement with the experimental data of Rugg et al., hinting at the presence of a miscibility gap in the GaAs-InAs phase diagram.

If the free energy, \( 
\Delta G = \Delta H_m - T \Delta S,
\)

where \( T \) is the temperature and \( \Delta S \) is the entropy associated with reaction (1) is positive, then the In\(_{x}\)Ga\(_{1-x}\)As will undergo phase separation into distinct regions of InAs and GaAs, a process that is limited by the diffusion rate of the atoms. The Gibb’s free energy can be calculated using

\[
\Delta G = \Delta H_m - T \Delta S,
\]

where \( k_B \) is the Boltzmann constant and \( \Omega \) is the number of ways of arranging \( x \) moles of In and \( 1-x \) moles of Ga. (For an alloy, such as In\(_{x}\)Ga\(_{1-x}\)As, where the In and As atoms are randomly distributed on the 4\( a \) Wyckoff sites, the configurational entropy can be determined by

\[
\Delta S = k_B \ln \Omega,
\]
from the liquid-solid equilibria of Shen et al. showing the low-temperature miscibility gap. Our data are compared to the DFT-B3LYP simulations of Pentin et al. and data extrapolated from the liquid-solid equilibria of Shen et al. for $0 \leq x \leq 1$ of Ga on a mole of 4$a$ lattice sites. Setting $\Delta G$ to zero and solving for $T$ from Eq. (5), it is possible to determine the critical temperature defining the limit of the In$_x$Ga$_{1-x}$As phase. The low-temperature phase diagram showing the miscibility gap in In$_x$Ga$_{1-x}$As is given in Fig. 4 where our data are compared with similar data from the literature. The results suggest that In$_x$Ga$_{1-x}$As will be stable across the entire compositional range at temperatures greater than 550 K. Figure 4 shows that our results fall between the previous DFT-B3LYP simulations of Pentin et al. and the experimental data of Shen et al. that were extrapolated from thermodynamic functions obtained close to the liquid-solid interface. Pentin et al. argue that such extrapolations are frequently unreliable while acknowledging that their DFT simulations ignore any vibrational contribution to the enthalpy.

The observation that $E(\text{In}_x\text{Ga}_{1-x}\text{As}) > E(\text{InAs}) + E(\text{GaAs})$ has implications for the determination of the chemical potential of the atoms in the alloy since the chemical potentials of InAs or GaAs in In$_x$Ga$_{1-x}$As should not exceed the chemical potentials of the bulk binary alloys. Consequently, the chemical potentials of the atoms in In$_x$Ga$_{1-x}$As are determined with reference to the elemental states only by assuming $\mu_{\text{InGa$_{1-x}$As}} = x\mu_{\text{In}} + (1-x)\mu_{\text{Ga}} + \mu_{\text{As}}$. The chemical potentials of the atoms may not exceed that of the bulk element; therefore, taking As as an example, $\mu_{\text{InAs}} = \mu_{\text{As(bulk)}}$ represents the upper bound of the chemical potential. Continuing with As as our example, the lower bound (corresponding to As-poor growth conditions) can be determined by assuming the growth conditions were rich in both In and Ga, i.e., $\mu_{\text{As}}^{\text{poor}} = \mu_{\text{InGa$_{1-x}$As}} - [x\mu_{\text{In(bulk)}} + (1-x)\mu_{\text{Ga(bulk)}}]$.

C. As vacancy defects in In$_x$Ga$_{1-x}$As

1. Size effects

For intermediate compositions of In$_x$Ga$_{1-x}$As and InAs the arsenic vacancy formation energies are determined using the 64-atom supercells only. The local distortions of the arsenic sublattice due to the random arrangement of the In and Ga atoms on the 4$a$ Wyckoff sites means that each of the As sites in the simulation supercell is potentially unique. For In$_{0.25}$Ga$_{0.75}$As and In$_{0.75}$Ga$_{0.25}$As, all 32 sites are unique but for the In$_{0.5}$Ga$_{0.5}$As only 16 As sites are unique as it is constructed from a 32-atom supercell. Consequently, it is necessary to perform a total of 80 spin-polarized defect simulations. Employing a larger SQS supercell would increase the computational size on the formation energy and, in particular, on the configuration of the arsenic vacancy in GaAs can sometimes be significant. El-Mellouhi and Mousseau observe a Jahn-Teller distortion around the $V_{\text{As}}$ defect in their 216 atom supercell, however, in their 64-atom supercell they found a $C_{2v}$ symmetry. Clearly, it is important that the magnitude of any possible errors relating to the use of a 64-atom supercell are understood. Therefore, an arsenic vacancy defect was introduced into both 64 and 216 atom GaAs supercells and the supercells were minimised under constant pressure conditions (i.e., atoms positions and lattice vectors were allowed to change) and the formation energies and the symmetry of the surrounding Ga atoms were determined using MATERIALS STUDIO.

In the nondefective GaAs structure the separation between two nearest-neighbor Ga atoms is predicted to be 4.06 Å, in good agreement with the experimental value of 4.00 Å. Two simulations were performed for the charge-neutral defect in the 64-atom supercell, one in which all of the atoms in the supercell are left on their perfect lattice positions and another where each atom is slightly displaced in a random direction. For the simulation where the atoms are left on their perfect lattice sites the separations between the four Ga atoms surrounding the arsenic vacancy are all reduced to 3.73 Å, maintaining a perfect tetrahedral site symmetry. Conversely, when the atoms initial positions are distorted the final symmetry of the surrounding Ga atoms can be best approximated to a $D_{2d}$ point group. The maximum deviation of the atoms from a perfect $D_{2d}$ structure was 0.04 Å; however, the deviation from a perfect $T_d$ symmetry was only 0.044 Å. This shows that while the symmetry has changed from $T_d$ to $D_{2d}$ the distortion is not large.

For the simulation of the $V_{\text{As}}^0$ defect in a 216 atom supercell the atoms were randomly displaced from their perfect lattice sites. After minimization two of the separations between Ga atoms were reduced to 3.93 Å and the remaining four were reduced to 4.04 Å, representing a shift to a Jahn-Teller $D_{2d}$ symmetry with a maximum deviation of 0.019 Å. Clearly, the move to a larger supercell allows the Ga atoms surrounding the vacant 4$c$ site to move closer to a perfect $D_{2d}$ structure.

The $V_{\text{As}}^0$ defect formation energies in the As-rich limit were 3.22, 3.23, and 3.33 eV for the 64-atom undistorted, 64-atom distorted, and 216-atom supercells, respectively, compared to a value of 3.25 eV from El-Mellouhi and Mousseau. These results show that for the charge-neutral defect the 64-atom supercell gives formation energies in good agreement with the larger supercell and is also able predict the same symmetry around the defect. We note that for intermediate In$_x$Ga$_{1-x}$As compositions the perfect $T_d$ symmetry of each lattice site is broken due to the quasirandom distribution of the In and Ga atoms on the 4$a$ sublattice; therefore, the presence of a Jahn-Teller distortion surrounding the vacancy.
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FIG. 5. (Color online) Plots of the defect formation energy of a $V_{\text{As}}^0$ defect in In$_x$Ga$_{1-x}$As under As-rich (a) and As-poor (b) conditions. Red diamonds represent configurationally averaged formation energies at 1000 K.

would be swamped by the perturbation due to the differing local environments.

2. Defect formation energies

Figures 5(a) and 5(b) show plots of the formation energies of charge-neutral As vacancy defects in As-rich and -poor growth conditions, respectively. The formation energy of the charge-neutral As vacancy, $V_{\text{As}}^0$, in GaAs has already been discussed in Sec. III C 1. The formation energy for this defect in InAs was found to be 2.53 and 2.06 eV under As-rich and -poor growth conditions, respectively. Van Vechten$^5$ predicted a value of 2.07 eV in a stoichiometric sample and Hoglund et al.$^16$ report a value of $\geq 2.6$ eV under As-rich conditions.

As discussed previously, the adoption of SQS supercells allows a sampling of the different local environments on the disordered sublattice. This quasirandom distribution of ions allows for local relaxation effects to be modeled and gives rise to the distribution in the values of the defect formation energies shown in Figs. 5(a) and 5(b) for intermediate compositions of In$_x$Ga$_{1-x}$As. Figures 5(a) and 5(b) clearly illustrate the significant variation ($\leq 1.2$ eV) in defect formation energies due to different local environments in the random alloys. A typical formation energy for a neutral As vacancy defect was then determined by configurational averaging using Eq. (7)$^{45}$ as follows:

$$\langle A \rangle = \frac{\sum_c C_c A_c \exp \left(-\frac{E_{\text{cell}}^{c}}{k_B T} \right)}{\sum_c \exp \left(-\frac{E_{\text{cell}}^{c}}{k_B T} \right)},$$

(7)

where $\langle A \rangle$ is the configurationally average, $A_c$ is the property being averaged for configuration $c$, $E_{\text{cell}}^{c}$ is the defect formation energy for configuration $c$, and $T$ is the temperature (for this study a temperature of 1000 K was adopted). The configurationally averaged defect formation energies are included as the (red) diamonds in Figs. 5(a) and 5(b).

As a first approximation the formation energy of $V_{\text{As}}^0$ in an intermediate In$_x$Ga$_{1-x}$As compound can be estimated from a linear interpolation of the vacancy formation energies determined for the end member binary alloys; this is represented by the dashed line in Figs. 5(a) and 5(b). Clearly, the configurationally averaged formation energies are significantly lower than would be expected from such an interpolation and, indeed, are lower than for the end-member binary alloys. By considering the detailed local relaxations it is possible to understand why this behavior occurs. At intermediate compositions one of the four surrounding atoms undergoes relaxation toward the now-vacant site, creating a quasi-$V_{\text{As}}^-\text{In/Ga}_i-V_{\text{In/Ga}}^-\text{As}$ defect cluster. In the end-member binary alloys none of the surrounding atoms undergo this relaxation and the four atoms remain approximately equidistant from the vacancy. It is this difference in response to the introduction of the vacancy that allows the intermediate In$_x$Ga$_{1-x}$As compounds to have lower defect formation energies than present in the end members. The atom that undergoes the greatest relaxation toward the vacancy was always found to be an In atom except when there were no In atoms immediately surrounding the vacancy. Configurations in which there are no In atoms surrounding the defect were found to have the highest formation energies, as illustrated in Fig. 6 where the defect formation energy is plotted as a function of the number of In nearest neighbors. Thus, as the number of In atoms surrounding the vacancy increases the formation energy of the $V_{\text{As}}^0$ defect decreases until there are three In atoms surrounding the vacancy. The defect formation energy when there are four In atoms surrounding the vacancy is then similar to the value obtained for three In atoms. As the concentration of InAs in In$_x$Ga$_{1-x}$As increases the average number of indium atoms surrounding any 4c lattice site will increase and, hence, the configurationally averaged formation

FIG. 6. Plot showing the formation energy of a $V_{\text{As}}^0$ defect in In$_x$Ga$_{1-x}$As as a function of the number of In atoms located on nearest-neighbor 4a lattice sites surrounding the vacancy.
parameters of the intermediate \( \text{In}_x\text{Ga}_{1-x}\text{As} \) compositions are lower than would be expected by taking a linear interpolation of the lattice parameters of the end members, i.e., there is a small negative deviation from Vegard’s law in agreement with previous simulations.\(^\text{25}\) The atomization energies of the solid \( \text{In}, \text{Ga}, \text{As}, \) and \( \text{GaAs} \) are in excellent agreement with the literature values, indicating that the GGA-PBE exchange correlation functional is adequate for determining energetic properties. We have determined the 0 K atomization energies across the compositional range of \( \text{In}_x\text{Ga}_{1-x}\text{As} \) and observed a negative deviation from a linear interpolation of the atomization energies of \( \text{GaAs} \) and \( \text{InAs} \). Consequently, we predict a positive enthalpy of mixing for \( \text{In}_x\text{Ga}_{1-x}\text{As} \) from \( \text{InAs} \) and \( \text{GaAs} \), and, therefore, the presence of a miscibility gap, that would correspond to the low-temperature region of the \( \text{GaAs-InAs} \) phase diagram. \( \text{In}_x\text{Ga}_{1-x}\text{As} \) is, however, expected to be thermodynamically stable at temperatures greater than 550 K. The relatively low temperature range at which \( \text{In}_x\text{Ga}_{1-x}\text{As} \) is expected to undergo spinoidal decomposition may be the reason this has not been observed experimentally. Miscibility gaps are, however, common in other III-V materials,\(^\text{46}\) such as \( \text{Ga}_x\text{In}_{1-x}\text{P} \),\(^\text{47}\) \( \text{GaP}_x\text{Sb}_{1-x} \),\(^\text{47}\) \( \text{InP}_x\text{Sb}_{1-x} \),\(^\text{47}\) and \( \text{InAs}_x\text{Sb}_{1-x} \).\(^\text{47,48}\)

The formation energies for the introduction of a \( V_{\text{As}}^0 \) defect under As-rich conditions in \( \text{GaAs} \) and \( \text{InAs} \) were predicted to be 3.31 and 2.53 eV, respectively. These values are in excellent agreement with previously determined values of 3.25 eV for \( \text{GaAs} \)\(^\text{25}\) and >2.6 eV for \( \text{InAs} \).\(^\text{16}\) For intermediate values of \( x \) there is a large variation in the formation energies and defect volumes of the charge-neutral arsenic vacancy defect: This arises due to the random arrangement of \( \text{In} \) and \( \text{Ga} \) atoms on the 4\( a \) sublattice. As a consequence of the random arrangement of \( \text{In} \) and \( \text{Ga} \) atoms an As vacancy may be surrounded by between zero and four \( \text{In} \) atoms. As the number of \( \text{In} \) atoms surrounding an arsenic vacancy increases, the formation energy of the vacancy decreases (see Fig. 6). The formation energy of a \( V_{\text{As}}^0 \) defect in \( \text{In}_{0.25}\text{Ga}_{0.75}\text{As} \) surrounded by three atoms was found to be particularly low due to the propensity for one of the \( \text{In} \) atoms to move closer to the vacant arsenic site than was observed in \( \text{In}_{0.5}\text{Ga}_{0.5}\text{As} \) and \( \text{In}_{0.75}\text{Ga}_{0.25}\text{As} \). It is these low-energy configurations that cause the apparent distortion in the configurationally averaged formation energies shown in Figs. 5(a) and 5(b) at \( x = 0.25 \). Across the entire compositional range the configurationally averaged formation energies for the \( V_{\text{As}}^0 \) defect were found to be significantly lower than would be estimated from a linear interpolation of the end-member binary alloys, thereby demonstrating the necessity of performing a rigorous study of the different defect configurations in random alloys such as \( \text{In}_x\text{Ga}_{1-x}\text{As} \).

**ACKNOWLEDGMENTS**

This paper was based on work supported in part by King Abdullah University of Science and Technology (KAUST). Computational resources were provided by the Imperial College High Performance Computing Service (http://www.imperial.ac.uk/ict/services/teachingandresearchservices/highperformancecomputing).
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