Molecular dynamics study of structural, mechanical, and vibrational properties of crystalline and amorphous Ga$_{1-x}$In$_x$As alloys

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Using an interaction potential scheme, molecular dynamics (MD) simulations are performed to investigate structural, mechanical, and vibrational properties of Ga$_{1-x}$In$_x$As alloys in the crystalline and amorphous phases. For the crystalline phase we find that: (i) Ga–As and In–As bond lengths vary only slightly for different compositions; (ii) the nearest-neighbor cation–cation distribution has a broad peak; and (iii) there are two nearest-neighbor As–As distances in the As (anion) sublattice. These MD results are in excellent agreement with extended x-ray absorption fine structure and high-energy x-ray diffraction data and also with ab initio MD simulation results. The calculated lattice constant deviates less than 0.18% from Vegard’s law. The calculated phonon density of states exhibits a two-mode behavior for high-frequency optical phonons with peaks close to those in binary alloys (GaAs and InAs), which agrees well with a recent Raman study. Calculated elastic constants show a significant nonlinear dependence on the composition. For the amorphous phase, MD results show that: (i) the nearest-neighbor cation–anion distribution splits into well-defined As–Ga and As–In peaks as in the crystal phase; (ii) the cation–cation distribution is similar to that in the crystal phase; and (iii) the As–As distribution is quite different from that in the crystal, having only one nearest-neighbor distance. © 2003 American Institute of Physics.

[I. INTRODUCTION]

Semiconductor pseudobinary alloys with chemical formula $A_{1-x}B_xC(0 \leq x \leq 1)$ have drawn much attention for their interesting properties and their possible applications in optoelectronic devices. These alloys form a zinc-blende structure in the entire composition range, $0 \leq x \leq 1$, where component $C$ is arranged in a fcc sublattice (anion sublattice), and components $A$ and $B$ are distributed over another fcc sublattice (cation sublattice) displaced from the first one by $(111)_C$. For bulk materials synthesized at high temperatures, $A$ and $B$ components are randomly distributed within the cation sublattice, although they can be ordered under specific growth conditions.\footnote{Electronic mail: priyav@usc.edu} When a completely miscible solution is obtained, for random cation distributions, physical properties may be varied continuously across the composition range. For example, the wavelength of a solid-state laser made of Ga$_{1-x}$Al$_x$As can be tuned by changing $x$.

A particularly interesting pseudobinary alloy is Ga$_{1-x}$In$_x$As, which has a simple zinc-blende structure at low pressures. This alloy closely follows Vegard’s Law,\footnote{Electronic mail: priyav@usc.edu} which states that the average lattice parameter, $d(x)$, of the alloy is a linear function of the composition $x$, i.e., $d(x) = (1 - x) d_{AB} + x d_{AC}$, ($d_{AB}$ and $d_{AC}$ are the lattice parameters of the pure binary alloys $AC$ and $BC$). Consequently, the weighted average of the nearest-neighbor atomic distances is $\frac{\sqrt{3}a}{4}$ ($a$ is the lattice parameter of the alloy). The bond lengths in the binary alloys vary significantly from 2.448 Å in GaAs to 2.623 Å in InAs. Extended x-ray absorption fine structure (EXAFS) and x-ray measurements\footnote{Electronic mail: priyav@usc.edu} indicate that this large lattice mismatch ($\approx 7\%$) is accommodated by local bond distortions while the zinc-blende structure is preserved. Recent studies on other $A_{1-x}B_xC$ semiconductors alloys found that the cation–anion bond lengths remain al-
most the same as those in the pure binary compounds due to local bond distortions. This apparently contradicts the virtual crystal approximation, which explains the structural disorder of the alloys assuming an average crystal, where all the atoms are distributed over one zinc-blende lattice with the lattice parameter calculated from x-ray diffraction data.\(^5\)

Vibrational properties of Ga\(_1-x\)In\(_x\)As alloys are complex as well and not well understood. Infrared measurements by Brodsky et al.\(^6\) indicated a mixed-mode behavior, in which the high-frequency optical phonons are characterized by a single broad peak. However, in a recent Raman-scattering study, Groenen et al.\(^7\) observed a two-mode behavior, in which the high-frequency optical modes have two distinct peaks corresponding to GaAs and InAs optical phonon modes. The vibrational properties of similar alloys, e.g., Ga\(_{1-x}\)Al\(_x\)As, were also studied computationally based on lattice dynamics, where supercells were used to study the effects of disorder on the spectra.\(^8\) It was shown that the phonon dispersions are well defined, despite lattice distortion and the absence of periodicity.

In this article, we report classical and \textit{ab initio} molecular dynamics (MD) simulations of Ga\(_1-x\)In\(_x\)As alloys. We have investigated the structural, mechanical, and dynamical properties of these alloys in the composition ranging from \(x = 0\) (GaAs) to 1 (InAs) for both the crystalline and amorphous phases. This article is organized as follows: In Sec. II we discuss the interatomic potentials used in the classical MD study of the alloys, and in Sec. III we summarize the MD method. The preparation of the crystalline and amorphous states and details of the simulations are described in Sec. IV, and the \textit{ab initio} simulations are explained in Sec. V. In Sec. VI we discuss the MD results and compare them with available experiment data, and finally, conclusions are given in Sec. VII. A short letter discussing some of these results was published in the journal of Applied Physics Letters.\(^9\)

### II. INTERATOMIC POTENTIAL SCHEME

The interatomic potential model encodes interactions among all the atoms, and thus is the essential ingredient of molecular dynamics simulations. Our interatomic potential for Ga\(_1-x\)In\(_x\)As alloys is an interpolation of those for binary alloys,\(^10\) which consist of two- and three-body terms,\(^11\)

\[
V \approx \sum_{i<j}^N V_{ij}^{(2)}(r_{ij}) + \sum_{i<j<k}^N V_{ijk}^{(3)}(r_{ij}, r_{ik}),
\]

where \(N\) is the number of atoms, \(r_{ij} = |\mathbf{r}_{ij}|\), \(r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|\), and \(\mathbf{r}_i\) is the position of the \(i\)th atom.

The two-body term represents steric repulsion, Coulomb interactions due to charge transfer, induced charge–dipole interaction due to large electronic polarizability of the anions, and van der Waals (dipole–dipole) interaction:

\[
V_{ij}^{(2)}(r) = \frac{H_{ij}}{r^{\eta_{ij}}} + \frac{Z_i Z_j}{r} e^{-r r_{1s}} - \frac{1}{2} (\alpha_i Z_i^2 + \alpha_j Z_j^2) \frac{1}{r^4} e^{-r r_{4s}} - \frac{w_{ij}}{r^6},
\]

where \(H_{ij}\) and \(\eta_{ij}\) are the strength and exponents of the steric repulsion, \(Z_i\) the effective charge, \(\alpha_i\) the electronic polarizability, \(r_{1s}\) and \(r_{4s}\) the screening constants for the Coulomb and charge–dipole interactions, and \(w_{ij}\) the strength of the van der Waals interaction. The three-body term represents covalent bond bending and stretching:

\[
V_{ijk}^{(3)}(r_{ij}, r_{ik}) = B_{ijk} \exp \left( -\frac{\xi}{r_{ij} - r_0} + \frac{\xi}{r_{ik} - r_0} \right) \times \frac{(\cos \theta_{ijk} - \cos \theta_0)^2}{1 + C_{ijk} (\cos \theta_{ijk} - \cos \theta_0)^2 (r_{ij} r_{ik} r_0)}
\]

where \(B_{ijk}\) is the strength of the three-body interaction, \(r_0\) the cutoff radius, \(\xi\) and \(C_{ijk}\) constants to adjust the stretching and bending terms, and \(\theta_0\) the angle formed by \(r_{ij}\) and \(r_{ik}\), \(\theta_0\) in the case, of GaAs and InAs is the tetrahedral angle 109.5°, with \(\cos \theta_0 = -\frac{1}{3}\).

This interatomic potential model has been successfully applied to a number of ceramics such as SiO\(_2\),\(^1\) Si\(_3\)N\(_4\),\(^12\)–\(^16\) SiC,\(^17\) Al\(_2\)O\(_3\), and AIN, and semiconductors such as GaAs,\(^18\) InAs,\(^19\) CdSe, SiS\(_2\),\(^20\)\(^21\) and GeSe\(_2\).\(^22\) In the present work, we use this model with the parameters optimized to reproduce selected experimental data for GaAs and InAs.\(^23\) Our fitting database includes crystalline lattice constants, cohesive energies, elastic constants, melting temperature as well as structural transition pressures, phonon density of states, and neutron-scattering data for crystalline and amorphous structures. The potential parameters for the binary alloys, GaAs and InAs, are given in Tables I and II.

For Ga\(_1-x\)In\(_x\)As the potentials for GaAs and InAs are interpolated in such a way that the resulting potential depends on the local chemical environment.\(^24\)\(^25\) This is achieved by applying the following rules:

(i) The two-body potential between different cations is the average of the cation–cation interatomic potentials in the pure alloys,

<table>
<thead>
<tr>
<th>(Z)</th>
<th>(\alpha)</th>
</tr>
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<tbody>
<tr>
<td>Ga</td>
<td>0.9418</td>
</tr>
<tr>
<td>As</td>
<td>0.9418</td>
</tr>
<tr>
<td>H</td>
<td>\eta</td>
</tr>
<tr>
<td>Ga-Ga</td>
<td>1.4747\times 10^{-16}</td>
</tr>
<tr>
<td>Ga-As</td>
<td>6.5111\times 10^{-16}</td>
</tr>
<tr>
<td>As-As</td>
<td>4.4683\times 10^{-16}</td>
</tr>
</tbody>
</table>

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**TABLE I.** Constants in the interaction potential for GaAs. Unit of length/energy is Å/Joule. \(Z\) is the effective charge in \(e^2\) units, and \(\alpha\) the electronic polarizability, which has dimension of volume, Å\(^3\). \(H\) is the repulsive strength, \(\eta\) parameters are repulsive exponents, \(w\) the van der Waals strength, and \(r_{1s}\) and \(r_{4s}\) the screening constants. The others constants pertain to the three-body part of the interaction potential, where \(B\) is the strength, and \(\xi\) and \(r_0\) are defined by the three-body potential equation.
TABLE II. Constants in the interaction potential for InAs. Units and constants are defined in Table I.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th>Z</th>
<th>α</th>
</tr>
</thead>
<tbody>
<tr>
<td>In</td>
<td>1.1276</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>As</td>
<td>1.1276</td>
<td>2.0</td>
<td></td>
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</tbody>
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<p>| | | | |</p>
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<tr>
<th></th>
<th></th>
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</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>η</td>
<td>w</td>
<td>r_{1s}</td>
</tr>
<tr>
<td>In-In</td>
<td>7.8128 × 10^{-17}</td>
<td>7.0</td>
<td>0.0</td>
</tr>
<tr>
<td>In-As</td>
<td>3.9578 × 10^{-16}</td>
<td>9.0</td>
<td>0.0</td>
</tr>
<tr>
<td>As-As</td>
<td>2.3649 × 10^{-16}</td>
<td>7.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

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<tbody>
<tr>
<td>B</td>
<td>C</td>
<td>ξ</td>
<td>r_{0}</td>
</tr>
<tr>
<td>In-As-In</td>
<td>1.3 × 10^{-19}</td>
<td>4.0</td>
<td>1.0</td>
</tr>
<tr>
<td>As-In-As</td>
<td>1.3 × 10^{-19}</td>
<td>4.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>

\[
V_{Ga-As} = \frac{V_{Ga-Ga} + V_{In-In}}{2}. \tag{4}
\]

(ii) The two-body interatomic potential between As atoms is an interpolation of the As–As potentials in the pure alloys. Since the first-neighbor shell of an As atom in the zinc-blende structure consists of 4 – \(m\) Ga and \(m\) In atoms, there are five possible first-neighbor configurations (\(m = 0, 1, 2, 3, 4\)) for the As atom, see Figs. 1(a)–1(c). Accordingly, the potential between two As atoms is a linear combination of the As–As potential, \(V^{(0)}_{AsAs}\), in GaAs and that, \(V^{(1)}_{AsAs}\), in InAs,

\[
V^{(m,n)}_{AsAs} = \left(1 - \frac{m+n}{8}\right)V^{(0)}_{AsAs} + \frac{m+n}{8}V^{(1)}_{AsAs}, \tag{5}
\]

where \(m\) and \(n\) are the number of In neighbor atoms for the two As atoms, respectively.

(iii) The three-body interatomic potential \(V_{Ga-As-In}\) is the average of those in the pure alloys,

\[
V_{Ga-As-In} = \frac{V_{Ga-Ga} + V_{In-In}}{2}. \tag{6}
\]

All the other potentials are identical to the corresponding potentials for the pure GaAs and InAs alloys.

III. MOLECULAR DYNAMICS METHOD

In the MD approach, one obtains the phase–space trajectories, i.e., the time evolution of the set of positions and momenta for all the atoms from the numerical solution of the Newton’s equations of motion,\(^{26}\)

\[
m_i \frac{d^2 r_i}{dt^2} = -\frac{\partial}{\partial r_i} V(i = 1, \ldots, N), \tag{7}
\]

where \(m_i\) is the mass of the \(i^{th}\) atom, and \(N\) is the number of atoms.

Equation (7) is integrated by discretizing time with an interval, \(\Delta t\), and applying a finite-difference integrator that depends on the statistical ensemble. In this study we use both the microcanonical (NVE) ensemble, which conserves the number of atoms, \(N\), the system volume, \(V\), and the total energy, \(E\), and the canonical (NVT) ensemble, which conserves temperature, \(T\), instead of energy. For the NVE ensemble the velocity-Verlet algorithm is implemented, whereas the Nosé–Hoover-chain method is applied to generate the NVT ensemble. For both algorithms we applied the multiple time-scale (MTS) scheme,\(^{27}\) which uses different
time steps for different force components to reduce the number of force evaluations.

**IV. PREPARATION OF THE CRISTALLINE AND AMORPHOUS PHASES**

**A. Preparation and characterization of the crystal phase**

Initial crystalline configurations of the alloy are prepared as follows: (i) a GaAs zinc-blende lattice is assembled; (ii) for any given value of \( x \) a Ga\(_{1-x}\)In\(_x\)As alloy is then obtained by randomly replacing Ga atoms by In atoms with probability \( x \); (iii) As atoms are assigned “types” depending on the number of the nearest-neighbor Ga and In atoms (these “types” are used to calculate the As-As potential during the simulation); and (iv) the lattice parameter is determined by minimizing the potential energy. Calculations are performed for 1000 and 8000 atom systems using periodic-boundary condition, the simulation results depend little on the system size. The dimension of the cubic MD box is between 56.534 and 60.576 Å for the 8000 atom system, depending on the composition \( x \). The equations of motion are integrated using \( \Delta t = 2.177 \) fs. The structural correlations of the first and second nearest neighbors are analyzed by calculating pair distribution functions at the temperature \( T = 10 \) K to reduce thermal broadening of the peaks. Elastic constants are calculated at \( T = 0 \) K, after the system has been quenched over 10 000 \( \Delta t \), during which the velocities are scaled by a factor of 0.001 each 100 time steps to bring the system to a minimum energy configuration. Phonon density of states is calculated at room temperature by calculating pair distribution functions at the temperature \( T = 10 \) K to reduce thermal broadening of the peaks.

**B. Preparation and characterization of the amorphous phase**

We also calculate the structural correlations in the amorphous state, which is obtained as follows: the system is melted and thermalized at \( T = 2000 \) K for 30 000 \( \Delta t \), and subsequently, the system is quenched slowly to \( T = 100 \) K during 50 000 \( \Delta t \). The As “types” are reassigned at every 10 \( \Delta t \) to reflect the change in the local environment. Because of the discrete changes of the As types during the simulation and the resulting discontinuities in the interatomic potential, the temperature of the system increases gradually during the simulation. This necessitates the control of the temperature using the \( NVT \) ensemble. In the crystalline and amorphous phases, atomic diffusions are negligible and the simulation is performed in the \( NVE \) ensemble.

**V. AB INITIO CALCULATION**

To validate the classical MD simulations, \( ab \) \( initio \) MD simulations are performed for a 216 atom crystalline system for \( x = 0.1, 0.5, \) and \( 0.9 \), in which the interatomic potentials are calculated quantum mechanically. The electronic-structure calculations are based on the local density approximation in the framework of the density functional theory. We use the real-space grid based approach with a higher-order finite-difference method and norm-conserving pseudopotentials. The energy functional is minimized using an iterative scheme based on the preconditioned conjugate-gradient method with multigrid accelerations.

The real-space grid is chosen as 0.22 Å, which corresponds to a plane-wave cutoff of 14.3 Ry. The calculations are performed on 128 processors of an SGI Origin 2000 supercomputer, using spatial decomposition and a message passing interface for parallel computation. Also shown are results for a 216-atom system in MD (open symbols) and \( ab \) \( initio \) MD (dotted open symbols) simulations.

**VI. RESULTS AND DISCUSSION**

**A. Crystalline phase**

**1. Nearest-neighbor environment**

The volume of the Ga\(_{1-x}\)In\(_x\)As alloy is an increasing function of \( x \) because of the 7% difference of the lattice constants for GaAs and InAs in the zinc-blende structure. To investigate the effect of volume change on the nearest-neighbor local environment, we have calculated pair distribution functions [Fig. 2(a)]. We have found a well-defined split in the first-neighbor shell, corresponding to Ga–As and In–As bonds. Peak positions plotted in Fig. 2(b) for various \( x \) values indicate that the bond lengths of Ga–As and In–As are almost constant functions of \( x \). This result is in contradiction with the virtual crystal approximation, which states that all the atoms in the alloy occupy average lattice positions defined by the x-ray lattice constant. It also contradicts
Pauling’s assumption that atomic radii are conserved quantities and should be the same in different chemical environments. Figure 2~b! shows the weighted average of the bond length, which closely follows Vegard’s Law and is \( a \sqrt{3}/4 \), where \( a \) is the lattice parameter. These MD results are in good agreement with EXAFS/x-ray data, shown in Fig 2~a!, and valence force field calculations. The two sets of MD results for 1000 and 216 atoms compared in Fig. 2~b! demonstrate that system size effects are negligible. Figure 2~b! also includes ab initio MD results for \( x = 0.1, 0.5, \) and \( 0.9 \), showing that small modeling errors do not change the main conclusions based on the classical MD results.

2. Second-neighbor environment

Pair distribution functions in Figs. 3~a!–3~c! show the structure of the cation–cation sublattice. The results are in good agreement with the virtual crystal approximation, showing a single peak for the cation–cation distance. On the other hand, the first peak in the As–As distribution in Figs. 3~d!–3~f! splits, suggesting the existence of two As–As nearest-neighbor distances. This is in good agreement with EXAFS data by Mikkelsen and Boyce, and demonstrates that the second-neighbor correlations in the alloy are accurately described by MD simulations. The data is obtained from the lattice relaxed over 10,000 time steps for a 8,000-atom system.

3. Violation of Vegard’s law

Although Fig. 2~b! shows that the weighted average of the bond lengths roughly agrees with the Vegard’s law, there is a small but systematic convex deviation. This deviation is highlighted in Figs. 5~a! and 5~b!, which show that this con-
curves in Fig. 6 quantify the stretching and bending forces in GaAs and are larger for InAs than for GaAs. The potential energy per atom associated with the stretching of a GaAs/InAs lattice. Also included is an inverted curve for the InAs curve for easier comparison with the GaAs curve. (b) Energy per bond associated with angle changes between As-Ga-As and As-In-As bonds.

The deviation in Fig. 5b has an asymmetrical shape, being more pronounced at high Ga concentration, which may be understood by considering the difference of the stretching and bending forces for the GaAs and InAs components inside the Ga1-xInxAs crystal. As discussed by Fong et al., for x = 0, the bond angles around the large In ions tend to expand, which in turn induce changes in the bond angles at neighboring Ga ions, see Fig. 1. If the bond-bending forces in the GaAs host lattice are sufficiently large, the lattice will resist angular distortions, and thus prevent the bonds around the In ions from expanding. Therefore, the average lattice parameter of the alloy at x = 0 would be smaller than the Vegard’s value. For x ≈ 1, when a small amount of Ga ions is present in the InAs host lattice, a similar argument suggests a lattice parameter above the Vegard’s value. Therefore, the deviation should have an S shape, i.e., negative at small x and positive at large x. On the contrary, if the bond-stretching forces are dominant, one instead finds either a concave or convex curve of the lattice parameter, depending on whether the stretching force constant of GaAs is larger or smaller than that of InAs. Based on this argument, the convex curve in Fig. 5(b) can be explained if the stretching forces in Ga1-xInxAs are dominant and are larger for InAs than for GaAs. The potential curves in Fig. 6 quantify the stretching and bending forces in Ga1-xInxAs and show that this is, indeed, the case, i.e., the stretching forces (which are larger for InAs than for GaAs) are orders of magnitude larger than the bending forces.

The asymmetry in the deviation in Fig. 5b is due to the increasing difference between the force constants of GaAs and InAs as the InAs bonds are contracted; see Fig. 6(a). Gehrsitz et al. have found a similar deviation (3%) from Vegard’s law for Ga1-xAlxAs through near-infrared Brillouin scattering and high-energy x-ray diffraction experiments.

4. Elastic constants

Figure 7 shows the calculated elastic constants. The constants C_{11}, C_{12}, and C_{44} are obtained by calculating energy changes due to applied strains at T = 0 K.

The Bulk modulus is then calculated as $B = 1/3(C_{11} + 2C_{12})$. We observe a nearly linear decrease of $C_{12}$ and $C_{44}$, and a significant nonlinear decrease of $C_{11}$ as a function of x. Figure 7 also shows experimental data for x = 0 (GaAs) and x = 1 (InAs), which are in good agreement with the calculated values. Although we are not aware of any experimental data on the dependence of the elastic constants with composition for Ga1-xInxAs, Gehrsitz et al. have experimentally observed a similar nonlinear dependence of the Poisson ratio on x for Ga1-xAlxAs.

5. Phonon density of states

Figure 8 shows the calculated phonon density of states of Ga1-xInxAs at T = 300 K.

The data are obtained for simulation times over 20 000 Δt. For x = 0 (GaAs) and 1 (InAs), the MD results agree well with experimental and ab initio electronic-structure calculation data. For intermediate x values, experimental data on optical phonons associated with high-frequency peaks are controversial. Brodsky and Lucovsky showed that the optical modes have a mixed mode spectrum. However, a more recent experimental study supports a two-
mode-type spectrum. In the two-mode-type, two bands of frequencies corresponding to those in the binary systems persist in the mixed crystal. The strength of each band increases from zero to the maximum as the concentration of the corresponding constituent increases. In the one-mode-type spectrum the band varies continuously from one characteristic binary alloy to the other, with the strength of the band remaining almost constant. The results in Fig. 8 unambiguously support a two-mode spectrum: the GaAs and InAs types change strengths across the composition while keeping the high-frequency-peak positions close to those in the pure alloys. This is in agreement with recent experimental studies by Groenen et al. For intermediate \( x \) values the peaks are significantly broadened. This may be explained by the disorder activated modes caused by distortions in the crystal lattice.

B. Amorphous phase

1. Nearest-neighbor environment

Figure 9(a) shows the pair distribution function and coordination number for As–cation pairs for \( x = 0.5 \) at \( T = 100 \text{ K} \) in the amorphous alloy. Here, the coordination number, \( Cn(r) \), is the number of neighbor atoms inside a sphere of radius \( r \). \( Cn_{\text{As–(Ga,In)}}(r) \) is related to \( g_{\text{As–(Ga,In)}}(r) \) by

\[
Cn_{\text{As–(Ga,In)}}(R) = 4\pi\sigma_{\text{(Ga,In)}} \int_0^R g_{\text{As–(Ga,In)}}(r) r^2 dr,
\]

where \( \sigma_{\text{(Ga,In)}} \) is the density of cations.

As in the crystalline phase, the first peak splits into the bond lengths of As–Ga (first peak) and As–In (second peak). The arrow indicates that the coordination number is 4, as expected for the tetrahedral structure of the zinc blende. From the area under the first peak, the nearest-neighbor coordination of As is found to be 2; the same is found for the second peak, as expected for the composition \( x = 0.5 \). Nevertheless, the disorder at the second peak (As–In), defined by the FWHM, is noticeably larger than that at the first peak (As–Ga). In the crystalline alloy, on the contrary, the FWHM for the InAs peak is only slightly larger than that for the GaAs peak, see Fig. 4. This suggests that the difference in disorder between GaAs and InAs correlations is further amplified in the amorphous phase.

2. Second-neighbor environment

The pair distribution function for As–As pairs for \( x = 0.5 \) at \( T = 100 \text{ K} \) in Fig. 9(b) shows a well-defined first peak without splitting, indicating that the slight difference between GaAs- and InAs-like As–As distances (which is present in Fig. 3) is smeared out by the amorphous disorder. As a result, the As–As and cation–cation structures become nearly identical, see the pair distribution function for cation–cation pairs for \( x = 0.5 \) and \( T = 100 \text{ K} \) in Fig. 9(c). In both As–As and cation–cation cases, the second and third peaks of the crystalline function are merged to form a broad second peak. The arrows in Figs. 9(b)–9(c) indicate the coordination number 12, as expected for the first shell of the fcc lattice.

3. Bond-angle distribution

Bond-angle distributions are also calculated from MD trajectories. To calculate the \( Y–X–Y \) angle between \( Y \) and \( X \) components we first construct a list of all nearest-neighbor atoms of type \( Y \) around atoms of type \( X \). The cutoff distance for the \( X–Y \) separation is taken to be the first minimum of the corresponding radial distribution \( g_{XY}(r) \). From the list of all \( Y \) atoms, the angles are calculated for all the \( X–Y–X \) bonds and the results are averaged over all \( X \) atoms to obtain the bond-angle distribution.

Bond-angle distribution in the amorphous phase for \( x = 0.5 \) and \( T = 100 \text{ K} \) is shown in Fig. 10(a). The As–Ga–As bond angle has a well-defined peak at 109°. In Fig. 10(b) the
As–In–As bond angle shows a broad distribution with a peak at \( \sim 100 - 110^\circ \), which is consistent with the disorder shown in the corresponding radial distribution.

Figures 10(c)–10(f) show distributions of bond angles around As atoms. In contrast to Ga and In, which have only As atoms as neighbors, As atoms at \( x = 0.5 \) have different local environments, as discussed before. This is reflected in the angle distribution: (i) The Ga–As–Ga bond-angle distribution in Fig. 10(c) shows two peaks at 85° and 110°. (ii) The In–As–In bond-angle distribution in Fig. 10(d) has a broad and asymmetrical distribution peaked at \( \sim 100^\circ \). The cation–As–cation bond-angle distribution in Fig. 10(e) has a complex shape with a peak at 109° due to various local environments. Similar behavior is observed for the Ga–As–In distribution shown in Fig. 10(f).

VII. CONCLUSION

We have performed molecular-dynamics simulations of the pseudobinary alloy Ga\(_1\)–xIn\(_x\)As using an interaction potential scheme based on the potential for binary alloys. We have studied structural, mechanical, and dynamical properties in the crystalline and amorphous phases. The MD results reveal a local distortion in the crystalline lattice that is characterized by two well-defined bond lengths, Ga–As and In–As. While the cation sublattice exhibits a single broad peak at the first neighbor shell, there are two As–As distances. These results are in excellent agreement with experimental data\(^3,4\) as well as our \textit{ab initio} molecular-dynamics simulation results. We calculate the deviation (\( \sim 0.18\% \)) of the lattice constant from Vegard’s law. The phonon density of states shows a two-mode behavior, with the high-frequency optical modes, close to those in the binary materials. This is in excellent agreement with a recent Raman study.\(^7\) The calculated elastic constants exhibit a significant nonlinear dependence on the composition. For the amorphous phase the MD results show a well-defined split in the nearest-neighbor peak for the As–cation structure, similar to that in the crystalline structure. However, the difference between the disorder at the first (As–Ga) and second (As–In) peaks is amplified in the amorphous phase. For the cation–cation and As–As distributions, similar structure results were obtained, with the crystalline distortion over the As–As sublattice being completely overcome by the amorphous disorder.

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