

Spatial Correlations in GaInAsN Alloys and their Effects on Band-Gap Enhancement and Electron Localization

Kwiseon Kim and Alex Zunger

National Renewable Energy Laboratory, Golden, Colorado 80401

(Received 14 August 2000)

In contrast to pseudobinary alloys, the relative number of bonds in quaternary alloys cannot be determined uniquely from the composition. Indeed, we do not know if the $\text{Ga}_0, \text{In}_0, \text{As}_0, \text{N}_0$ alloy should be thought of as $\text{InAs} + \text{GaN}$ or as $\text{InN} + \text{GaAs}$. We study the distribution of bonds using Monte Carlo simulation and find that the number of In-N and Ga-As bonds increases relative to random alloys. This quaternary-unique short range order affects the band structure: we calculate a blueshift of the band gap and predict the emergence of a broadband tail of localized states around the conduction band minimum.

DOI: 10.1103/PhysRevLett.86.2609

PACS numbers: 71.20.Nr, 71.55.Eq, 78.20.Bh

The need to simultaneously control both the band gap and the lattice constant of semiconductor alloys prompted interest in not only ternary (e.g., $\text{Ga}_{1-x}\text{In}_x\text{As}$) but also in quaternary (e.g., $\text{Ga}_{1-x}\text{In}_x\text{As}_{1-y}\text{P}_y$ [1], $\text{Ga}_{1-x}\text{In}_x\text{As}$

thought (e.g., Ref. [1]) that, in quaternary semiconductor alloys, one can simultaneously tune band gaps and the lattice constant by altering only the composition x, y , we find that the ensuing band gap is, in fact, not unique and an additional thermodynamic variable z controls it. (v) The level repulsion in the conduction band has an important consequence on localization: While the statistical distribution of As-centered As-Ga_mIn_{1-m} tetrahedra in ternary InGaAs causes a small ~ 1 meV broadening of the conduction band minimum (CBM), the distribution of N-centered N-Ga_mIn_{1-m} tetrahedra in GaInAsN leads to a far broader range ~ 0 meV of quasi-N-localized band tail states near the CBM. This wide range is due to the stronger perturbations by the higher energy cluster (L_{1c} -derived) levels in the nitride alloy. Such strong alloy band edge fluctuations can locally capture carriers, affecting carrier dynamics [6].

Our model energy functional depends both on site occupancy variables S_c, S_a and on atomic positions \mathbf{R}_i :

$$E(S_c, S_a, \{\mathbf{R}_i, i = 1, \dots, M\}) = E_{\text{kin}} + E_{\text{pot}}. \quad (2)$$

Here $S_c = 1 - S_a$

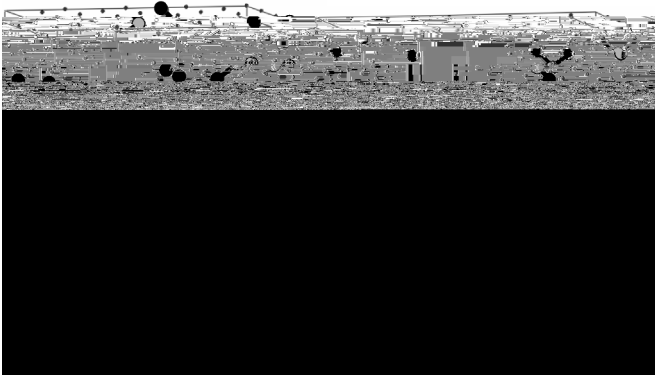


FIG. 2. A visualization of the real space supercell of the alloy $\text{Ga}_{0.4}\text{In}_{0.11}\text{As}_{0.49}\text{N}_{0.0}$ (a) for a random configuration and (b) with SRO determined by MC simulation at $T = 0$ K. Note the preponderance of N-In Ga_1 and N-In clusters. Solid dots denote the positions of Ga and As atoms.

Figure 2 provides a visualization of the real space atomic positions in the supercell of $\text{Ga}_{0.4}\text{In}_{0.11}\text{As}_{0.49}\text{N}_{0.0}$ with SRO obtained with MC simulation at $T = 0$ K. Taking statistics over many final configurations such as that shown in Fig. 2 reveals that *the main effect of SRO is that the concentration of the N-centered In Ga_1 clusters is statistically enhanced significantly (local In enrichment), whereas the In*

