

Impact of random composition fluctuations on electron and hole states in InAlN and InGaN alloys

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Abstract—We overview the impact of random alloy fluctuations on the electronic properties of InAlN and InGaN systems. Both density functional theory and empirical atomistic approaches have been applied. We describe our empirical atomistic approach to describe the electronic structure of III-N alloys and quantum wells (QWs). We show that random alloy fluctuations lead to wave function localization effects in the electronic structure of InAlN and InGaN alloys and QWs.

Keywords—InAlN; InGaN quantum wells; atomistic models; carrier localization.

I. INTRODUCTION

The electronic and optical properties of InAlN and InGaN systems are of interest for a variety of different applications. However, key material parameters are still not well understood. For instance, a large degree of uncertainty in the band bowing parameters has been reported in the literature and even composition dependent bowing parameters have been suggested. Furthermore, experimental studies clearly show that electronic and optical properties of III-N alloys are significantly affected by random alloy fluctuations present in these systems. An example is the unusual defect insensitivity found in (In,Ga,Al)N alloys, which has been imputed to localisation of carriers due to alloy fluctuations [1]. Also, alloy fluctuations have been shown to affect excited states of these systems, as evidenced by the “S-shaped” temperature dependence of the photoluminescence peak energies [2]. However, theoretical studies have focused mainly on continuum based descriptions, which do not capture fully the effects produced by fluctuations on an atomic scale.

We show here the importance of random alloy fluctuations in bulk InAlN and InGaN alloys as well as in InGaN quantum well (QW) systems. Several scales of model are required to describe key details of the electronic structure of III-N materials, and the consequences of random alloy fluctuations for device design and analysis. Many of the electronic properties of III-N structures are dominated not just by the large built-in polarization potentials found in such materials, but also by the impact of random alloy effects on the electronic states. *Ab-initio* techniques based on widely

available density functional theory (DFT) codes provide key III-N material parameters [3,4]. However, the accurate treatment of random alloy effects requires empirical atomistic models that can include of the order of 10^5 atoms in the calculation [4].

Based on DFT calculations in the framework of local density approximation (LDA) and hybrid functional schemes, we have extracted key material parameters, e.g. piezoelectric coefficients, and studied the impact of isolated and clustered In atoms in AlN and GaN bulk systems. With input from DFT we have developed an sp^3 tight-binding (TB) model to describe the electronic structure of InAlN and InGaN alloys. To address the impact of random alloy effects on *local* variations in the built-in potential we have also developed a local polarization theory [4]. This model has been benchmarked against DFT-based Berry phase calculations for ordered and disordered supercells (SCs), showing a very good agreement between the two approaches. Strain field calculations underlying the local polarization theory are performed by means of an in-house developed atomistic valence force field approach, which includes also electrostatic effects to account for the high ionicity of the III-N bonds. Combining these different empirical models, the overall model allows us to achieve an atomistic description of the electronic properties of InAlN and InGaN alloys, and to explicitly include local alloy, strain and built-in potential fluctuations.

We overview here in Section II our DFT calculations which show that In-related localized states are formed both in the conduction and the valence bands of InAlN for low In compositions. These localized states dominate the evolution of the band structure with increasing In composition, explaining the experimentally reported composition dependent band gap bowing parameter. In Section III the impact of alloy and well width fluctuations on the properties of InGaN QWs is discussed. We conclude in Section IV that quantitative modeling of the role of random alloy fluctuations is critical to understand electronic and optical properties of III-N alloys.

II. BAND STRUCTURE OF INALN

First-principles calculations that we carried out show that In-related localized states are formed both in the conduction band (CB) and the valence band (VB) in $\text{In}_x\text{Al}_{1-x}\text{N}$ for low In composition, x , and that these localized states dominate the evolution of the band structure with increasing x . Figure 1 shows the charge densities of (a) the CB edge (CBE) and (b)

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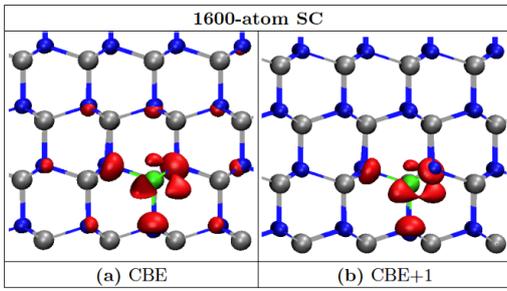


Figure 1: LDA-DFT charge densities in $\text{In}_1\text{Al}_{799}\text{N}_{800}$ for CBE and CBE+1 at $\mathbf{k}=\mathbf{0}$. The dark (red) isosurface of the charge density corresponds to 10% of the maximum value.

the first excited CB state (CBE+1), calculated for a $\text{In}_1\text{Al}_{799}\text{N}_{800}$ SC using LDA-DFT. Figure 1(b) shows that the single In atom introduces a strongly localized state (CBE+1) above the CBE. This localized state hybridizes with the CBE: the wave function of the lowest CB state is a linear combination of the AlN CBE state and of the localized state (CBE+1) (Fig. 1(a)).

Turning to the VB states, we showed through DFT studies that the highest VB states are also strongly localized in InAlN. We first considered ordered SCs, in which each In atom only has Al atoms as second nearest neighbors. Our analysis revealed that the VB edge (VBE) energy has a much weaker variation with composition in such structures compared with that which is observed in structures containing In pairs and clusters, where an In pair is formed when a N atom has two In neighbors. We found that even a single In pair in a SC can strongly shift the VBE energy upwards. It also tends to localize the highest valence state, consistent with In pairs introducing localized states below the VBE in $\text{In}_x\text{Al}_{1-x}\text{N}$. As a consequence, we found the band-edge optical polarization switches from TM- to TE-mode at approximately 15-18% InN in InAlN; from a linear interpolation of the crystal field splitting energies such a switching should occur for ~90% InN. Hence the detailed local atomic environment strongly influence the optical properties of $\text{In}_x\text{Al}_{1-x}\text{N}$ alloys. In addition, our TB results on the composition dependence of the energy gap in InGaN and InAlN alloys [4,5] show very good agreement with experimental and theoretical data.

III. POLAR INGAN QUANTUM WELLS

Having established the importance of localized states in InAlN alloys, we now turn to InGaN QWs. The particular examples considered here are approximately 3.5 nm wide *c*-plane QWs with varying InN contents (10% or 25%) and with well width fluctuations. For each composition several different random atomic configurations are investigated. SCs of around 82,000 atoms with periodic boundaries are used.

The importance of alloy fluctuations is demonstrated quantitatively in Figure 2, where the variation in the electron and hole ground state energies for 10% and 25% InN content is displayed. The ground state variation is given with respect to the ground state energy of configuration 1. It can be seen that random changes at the atomistic level have a clearly discernible effect on the properties of the electron and hole

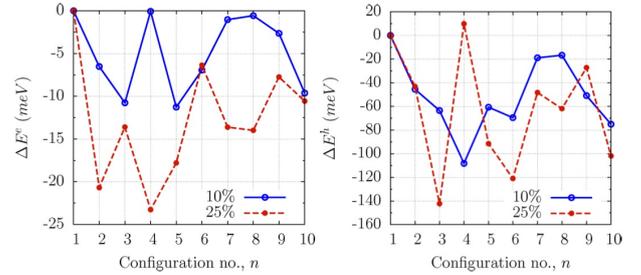


Figure 2: Variation of electron (left) and hole (right) ground state energies in 10 nominally identical InGaN QWs with 10 and 25% InN.

states. For electrons we see that the fluctuations manifest in a variation of ~ 10 meV for 10% InN and ~ 24 meV for 25% InN. For the holes this variation is much more pronounced, with ~ 100 meV for 10% InN and ~ 150 meV for 25% InN. Further calculations show that this increased sensitivity of the holes to local environment is due to very strong hole wave function localisation, whilst the electron wave functions in InGaN are only slightly affected by random alloy fluctuations. However, the electrons are strongly affected by well width fluctuations. These effects can be further investigated by a close examination of the electron and hole wave functions for a large number of excited states, showing that hole states are localized over an increasingly wide energy range, as the InN composition is increased from 10 to 25%.

IV. SUMMARY AND CONCLUSIONS

In summary, we have presented an atomistic analysis of wave function localization effects in $\text{In}_x\text{Al}_{1-x}\text{N}$ alloys and $\text{In}_x\text{Ga}_{1-x}\text{N}$ QWs with random In distributions and well-width fluctuations. The local In configurations strongly affect the CBE wave function in $\text{In}_x\text{Al}_{1-x}\text{N}$ alloys with small *x*, while the VBE state becomes increasingly localised as an increasing number of In atoms share a N atom in $\text{In}_x\text{Al}_{1-x}\text{N}$ alloys and in $\text{In}_x\text{Ga}_{1-x}\text{N}$ QW structures. Our overall analysis using the TB method is in very good agreement with DFT calculations and with experimental data. We conclude that random alloy fluctuations have a critical impact on electronic structure of InAlN and InGaN, and that these fluctuations are best assessed using an atomistic approach such as that described here.

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