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First-Principles Calculation of Bandgap Bowing Parameter for Wurtzite InAlGaN Quaternary Alloy using Large Supercell

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1. Introduction

Solid state ultraviolet (UV) light emitters have drawn a great deal of interests in recent years as the light sources for solid-state lighting, future high density optical storage, photo-catalytic disinfection and so on. At present, GaN-based UV emitters are the only viable choices for such applications. Although most of the demonstrated UV emitters use the AlGaN ternary alloy with high Al content to emit lights with shorter wavelengths, increase of the AlGaN thicknesses and Al contents is limited because of the epitaxial cracks caused by the lattice mismatch between the AlGaN and the underneath GaN. Addition of In into the ternary alloy can relax the material strain and thus formed InAlGaN alloy is reported to increase the UV emission intensities[1]. The quaternary alloy can be lattice-matched with wide variation of the bandgaps by choosing the Al/In composition to be 4.7[2]. Understanding of material parameters of the quaternary InAlGaN is critical as a part of device design including the bandgap bowing parameters. First principle calculation predicted the bowing parameter of zinc-blende InAlGaN to be as high as 2.59 eV, however, it was not on practical wurtzite structure and their calculation is limited to small 16 atoms supercell[3].

In this study, we calculate the bowing parameter of wurtzite InAlGaN using large supercells by first-principles technique. Neighboring effect of the group-III atoms in the 192-atoms supercell is examined. The calculated bowing parameters are over 2 eV regardless of the atomic positions which support the experimental results.

2. Calculation model

We employ 192-atoms ($3a\times3a\times2c$) supercell models for $Ga_{96}N_{96}$, $Al_1Ga_{95}N_{96}$, $In_1Ga_{95}N_{96}$ and $In_1Al_1Ga_{94}N_{96}$ to extract the bowing parameter of InAlGaN. In this supercell, the minimum composition ratio of substitutional elements (Al and In) is 1.04 % (=1/96), which is small enough precise calculation. Neighboring effect between Al and In in GaN host material is investigated using two types of interatomic configurations for the InAlGaN. The Al and In are placed at 2nd nearest neighbor position (abbreviated to 2NN, the distance between In and Al is 3.2 Å) and at the farthest position (FAR; 10.8 Å) .

The Vienna Ab initio Simulation Package[4,5] based on the Kohn-Sham density-functional theory using a plane-wave basis set is used for the first-principle calculations. Ultrasoft Vanderbilt pseudopotentials[6] using the generic gradient approximation[7] are employed for the exchange-correlation functional. The cutoff energy of the basis set is set at 21 Ry.

After completing the structural relaxation for each model, the bandgaps are calculated.

3. Calculation

Fig. 1 shows the calculated bandgaps for the four compositions. The difference of the bandgap between the alloy and $Ga_{96}N_{96}$ is plotted as a function of the (x-y), where the x and y are the number of In and Al atoms in the unit cell, respectively. By simple linear interpolation between the bandgap of $In_1Ga_{95}N_{96}$ and that of $Al_1Ga_{95}N_{96}$, the bandgap of $In_1Al_1Ga_{94}N_{96}$ *without* bowing parameter can be estimated as seen in the cross mark in Fig. 1. On the contrary, the calculated bandgaps of $In_1Al_1Ga_{94}N_{96}$ for the 2nd nearest neighbor and the farthest are far lower than the interporated value, implying a large bowing parameter.

As shown in Fig.1, the difference of the bandgap between the two quaternary models is negligibly small compared with another quaternary alloy, such as GaInNAs. This can be explained by the following two reasons: (1) Al and In atoms are connected only with N atom, (2) the physical properties of Al, Ga and In are very similar. On the contrary, In-N clusters in GaInNAs alloy cause strong neighboring effect[8]. The total energies of the two models are almost same ($\Delta E<k_BT$), which implies Al and In atoms can be randomly placed in GaN host material keeping the almost same value of the bandgap.

The bowing parameter of InAlGaN can be derived from the following equation[9],

$$\begin{aligned} E_g^{\mathrm{InAlGaN}} &= \left[xE_g^{\mathrm{InN}} + (1-x)E_g^{\mathrm{GaN}}\right] \\ &+ \left[yE_g^{\mathrm{AlN}} + (1-y)E_g^{\mathrm{GaN}}\right] \\ &- E_g^{\mathrm{GaN}} - (x+y)(1-x-y)b \end{aligned} \quad , \qquad (1)$$

where b is the bowing parameter, and E_g^{material} denotes the bandgap of each binary material. The first and second terms denote the bandgaps of $In_xGa_{1-x}N$ and $Al_yGa_{1-y}N$, respectively. Using the calculated bandgaps shown in Fig. 1, the bowing parameter of InAlGaN quarternary alloy for the two models is,

$$b = \begin{cases} 2.49 & \text{(2nd nearest neighbor)} \\ 2.19 & \text{(farthest)} \end{cases} . \quad (2)$$

The above two bowing parameters are very close reflecting the close bandgaps shown in Fig. 1.

4. Comparison with Experimental Results

In order to verify the calculated results, lattice-matched InAlGaN films were successfully grown on GaN (2.5 μm thickness) templates by metalorganic chemical vapor deposition (MOCVD). The thickness of InAlGaN films was around 100 nm. The bandgap and the composition ratio of each element were evaluated by cathodoluminescence and electron-probe microanalysis, respectively.

Fig. 2 shows the typical reciprocal space mapping of InAlGaN film by x-ray diffraction. A single peak is observed at the same diffraction angle of GaN, which implies the InAlGaN film is lattice-matched to the underlying GaN layer.

Among the various InAlGaN grown films, closely lattice-matched samples with small strain ($\Delta a/a_{GaN}$ <0.4 %) were picked up and plotted as open circles in Fig. 3. Bandgaps of the lattice-matched InAlGaN using the calculated bowing parameters in Eq. (2) are also shown in the same figure. The calculated bandgap does not much depend on the interatomic configuration explained in the previous section. As shown in Fig. 3, the experimental values well agree with the calculated bandgaps supporting the large bowing parameters predicted in the first-principle calculations.

5. Conclusion

We calculate the bowing parameter of wurtzite InAlGaN using large 192-atoms supercells by first-principles technique. Atomic neighboring effect of the group-III atoms is examined and the calculated bandgaps are not much dependent on the atomic positions. The bandgap bowing parameters are estimated to be 2.19-2.49 eV, which well agree with our experimental results.

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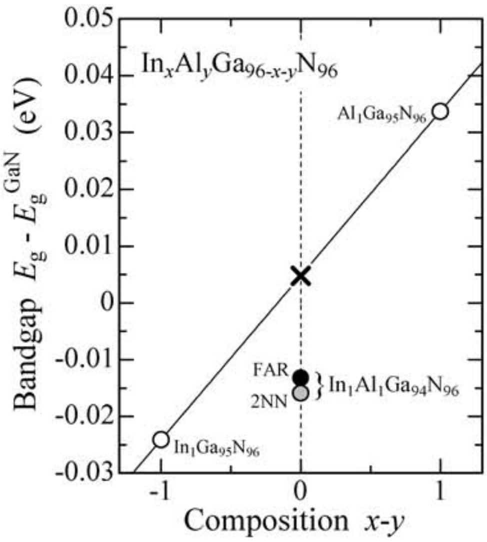

Fig. 1. Calculated bandgaps of AlGaN, InGaN and InAlGaN using first-principles calculation.

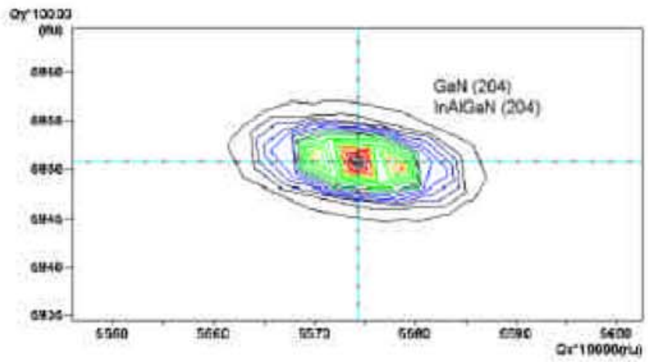

Fig. 2. Reciprocal space mapping of InAlGaN film by x-ray diffraction.

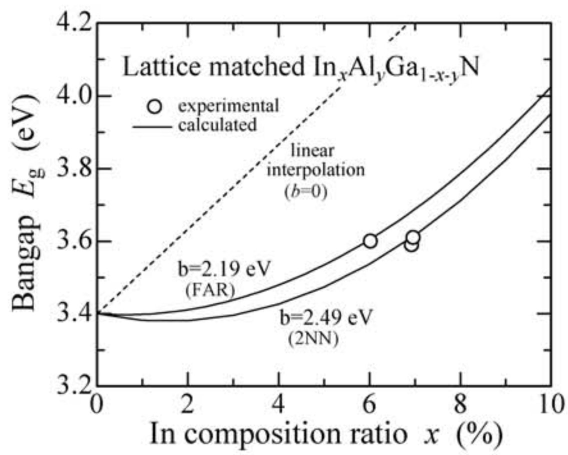

Fig. 3. Comparison of the InAlGaN bandgap between experimental results and calculated bowing curves.