Impact of random alloy fluctuations on the electronic and optical properties of c-plane $Al_xGa_{1-x}N$/AlN quantum wells

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***Abstract*—We present a theoretical study of the electronic and optical properties of c-plane $Al_xGa_{1-x}N$/AlN quantum wells emitting in the ultraviolet-A (UV-A) to UV-C spectral range. Special attention is paid to the impact of alloy fluctuations on the results. We find that random alloy fluctuations in (Al,Ga)N are already sufficient to cause strong carrier localization effects. Moreover, our results suggest that the degree of optical polarization is impacted by alloy induced carrier localization effects. More specifically, in comparison to a virtual crystal approximation, which neglects effects of alloy disorder, the switching from transverse electric to transverse magnetic polarization occurs at higher Al contents. This feature is important for the light extraction efficiencies of (Al,Ga)N-based light emitters operating in the deep UV spectral range.**

I. Introduction

Light emitters using the semiconductor alloy aluminium gallium nitride ((Al,Ga)N) have attracted considerable attention due to their potential for a wide range of applications in the ultraviolet (UV) spectral region, including water purification, medical diagnosis or sterilisation procedures [1]. However, the efficiencies of such (Al,Ga)N-based emitters are very low and understanding their fundamental electronic and optical properties is essential to aid the design of UV devices with improved efficiencies.

Overall, it is important to note that the III-N materials (In,Ga)N or (Al,In)N exhibit very different properties than other more conventional III-V materials. For instance, in (In,Ga)N quantum wells (QWs), random alloy fluctuations lead to very strong carrier localization effects [2]. Recently, experimental studies indicate a similar behavior in (Al,Ga)N/AlN QWs, evidenced for instance by an "S-shaped" temperature dependence of the photoluminescence (PL) peak position [3]. However, theoretical investigations on the impact of random alloy fluctuations on the fundamental electronic and optical properties of these systems are sparse.

Here, we target this question by employing a nearest neighbor tight-binding (TB) model to analyze the electronic and optical properties of (Al,Ga)N/AlN QWs with Al contents ranging from 10% up to 75%. Our calculations show that already random alloy fluctuations are sufficient to lead to strong hole wavefunction localization effects. Moreover, we find that with increasing Al content, *electron* wave functions may also be localized by alloy fluctuations, an effect usually *not* observed in (In,Ga)N QWs. Furthermore, by comparing the results of our atomistic calculations with the outcome of studies utilizing a virtual crystal approximation (VCA), and thus neglecting alloy fluctuations, we find that the switching from transverse electric (TE) to transverse magnetic (TM) optical polarization occurs at higher Al contents in the random alloy case when compared to VCA. Thus, while alloy fluctuations may be detrimental for the electron and hole wave function overlap and thus the radiative recombination rate, these fluctuations may have benefits for the light extraction efficiency in (Al,Ga)N-based LEDs operating in the deep UV spectral range.

II. Method

We employ here a nearest neighbour sp^3 tight-binding (TB) model that includes strain and polarization fields to gain insight into the impact random alloy fluctuations have on the electronic and optical properties of of c-plane $Al_xGa_{1-x}N$/AlN QW systems. Given the microscopic resolution of our framework, our calculations are performed on supercells with periodic boundary conditions that contain 81,920 atoms; this resembles a cell with the dimension of approximately 10 nm $\times$ 9 nm $\times$ 10 nm. We start with a cell of pure AlN, but in a specified sub-region defining the well (between specified z-coordinates of the cell), Al atoms are replaced by Ga atoms, determined by the Al/Ga content in the well. Here, Al atoms are randomly replaced and no preferential positioning or clustering is assumed. The width of the QW is approximately 3 nm. We investigated systems with Al contents of 10%, 25%, 50% and 75%. These chosen Al contents cover the window which is experimentally relevant for developing UV emitters operating in the UV-A to UV-C spectral range. Moreover, to gain insight into the impact of the alloy microstructure on the electronic and optical properties of (Al,Ga)N/AlN QW systems, for each Al content mentioned above, the calculations have been repeated for 20 different random alloy configurations.

III. Results

Before looking at the electronic structure of (Al,Ga)N/AlN QWs, Fig. 1 shows a linescan of the diagonal components of the strain tensor, ϵ_{ii}, along the growth direction (c-axis) in a

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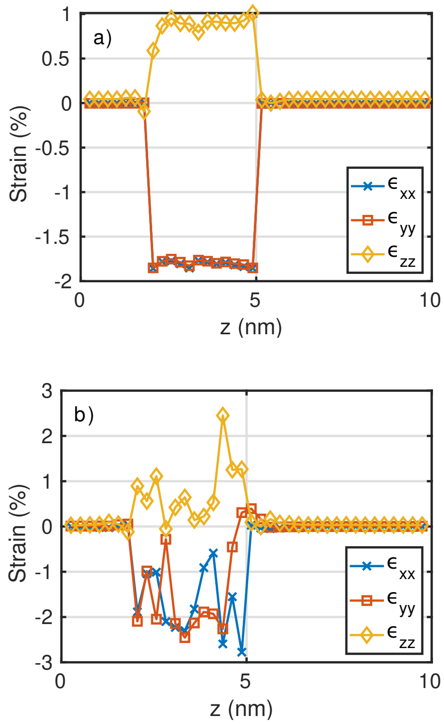

Fig. 1. Diagonal components of the strain tensor, ϵ_{ii}, for a linescan along the c-axis of an $Al_{0.25}Ga_{0.75}N$/AlN QW. a) Strain tensor averaged over the growth plane, b) single linescan along the c-axis.

$Al_{0.25}Ga_{0.75}N$/AlN QW. In the upper part of this figure, Fig. 1 (a), the strain tensor components are averaged over the growth plane. The figure reveals a behavior that could be expected from a continuum-based calculation, namely $\epsilon_{xx} = \epsilon_{yy} < 0$ and $\epsilon_{zz} > 0$. However, when looking at Fig. 1 b), which displays a *single* linescan, it is clear that the alloy fluctuations lead to strong *local* strain field fluctuations. These fluctuations in strain will also result in local built-in field fluctuations; all this can give rise to carrier localization effects.

Equipped with this insight, Fig. 2 shows the electron (red) and hole (blue) ground state charge densities of an $Al_{0.25}Ga_{0.75}N$/AlN QW; the charge densities are shown for a top-view (along the c-axis) and the light (dark) surfaces correspond to 10% (50%) of the maximum charge density. It is clear that even at this relatively low Al content in the well, we observe noticeable hole localization effects. In contrast the electron charge density is far less affected by the alloy fluctuations and presents a more delocalized nature.

For higher Al contents in the well i.e. 50% and 75% Al, the results can change. While we still observe strong hole localization effects, we also find configurations with noticeable electron localization effects. We note this behavior is in contrast to (In,Ga)N systems, where the electrons are mainly localized by well width fluctuations [4]. It needs to be stressed that we have not included any well width fluctuations in these calculations, and that therefore random alloy fluctuations are already sufficient to lead to strong carrier localization effects.

Moreover, our calculations indicate that the alloy fluctuation

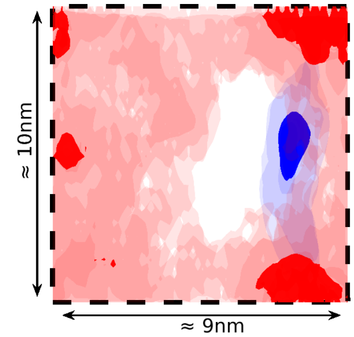

Fig. 2. Isosurface plots of the electron (red) and hole (blue) ground state charge densities for a c-plane $Al_{0.25}Ga_{0.75}N$/AlN QW. The charge densities are shown for a topview (down the c-axis). The dashed lines indicate the supercell boundaries

induced carrier localization effects can be beneficial for the light extraction efficiency in deep UV light emitters. More specifically, when comparing results from VCA and random alloy fluctuation calculations on the degree of optical polarization, we find that the cross-over from TE to TM occurs at higher Al contents and thus shorter wavelengths when accounting for alloy fluctuations. Our investigations show that this originates from differences in the mixing of the atomic-like orbitals in the random alloy case when compared to the 'ideal' VCA system. Thus while alloy fluctuations may have a detrimental effect on the radiative recombination rate, they may be beneficial for light extraction efficiency in UV-C (Al,Ga)N-based LEDs.

IV. Conclusion

In conclusion, we present an atomistic study on the impact of random alloy fluctuations on the electronic and optical properties of (Al,Ga)N/AlN QWs. Our results show that over a wide range of Al contents and thus wavelengths, random alloy fluctuations are already sufficient to facilitate carrier localization effects in these systems. Moreover, our calculations indicate that in addition to hole localization effects, electrons may also be localized by alloy fluctuations, at least at higher Al contents in the well. Overall, although carrier localization may have a detrimental impact on the radiative recombination rate, for the light extraction efficiency in deep UV-C range they may become advantageous.

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