Segregation effects and bandgap engineering in InGaN quantum-well heterostructures

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ABSTRACT

The analysis of In surface segregation and its impact on the composition profile and light emission spectra of the InGaN single quantum well heterostructures grown by Metalorganic Vapor Phase Epitaxy (MOVPE) is carried out by coupled solution of the Poisson and Schrödinger equations. Effective methods of controlling the composition profile, indium predeposition and temperature ramping during the cap layer growth are considered in terms of surface segregation model. General trends in spectra transformation upon the forward bias variation and their correlations with the quantum well electronic structure are discussed.

INTRODUCTION

Recently, In surface segregation has been recognized as a factor critical to control of the composition profiles in III-nitride light emitting diode (LED) quantum well (QW) heterostructures. Much effort was made to find correlations between the growth recipes, segregation effects, and emission characteristics of the grown diodes. Along with variation of growth parameters (temperature, pressure, precursor flow rates, etc.), specific procedures – growth interruption at the QW interfaces [1], indium predeposition [2,3], and temperature ramping during barrier or cap layer growth [4] – were suggested to improve the composition profiles in InGaN QWs, normally serving as active regions in blue and green LEDs. Recent theoretical studies [5,6] considered basic mechanisms of In surface segregation in the multi-layer structures grown by MOVPE and some approaches to control of the composition distributions in the InGaN QW. However, the interrelation between the segregation effects and the characteristics of light emitted from the LED structures still remains poorly understood.

In this paper, we report on the quantum-mechanical study of segregation effects on the composition profiles and emission spectra of MOVPE-grown InGaN single-quantum-well (SQW) structures. Special attention is given to the most effective ways of controlling the front and back QW interfaces – indium predeposition and temperature ramping during the cap layer growth.

CONTROL OF COMPOSITION PROFILE IN InGaN-SQW HETEROSTRUCTURES

Consider a simple heterostructure that consists of an undoped InGaN SQW between n- and p-doped thick GaN layers, grown by MOVPE at 730°C in a vertical rotating-disk reactor under the conditions reported in [1]: pressure of 200 Torr, flow rates of TMGa, NH_3 , and N_2 (carrier gas) of 10.8 μmole/min, 406 mmole/min, and 196 mmole/min, respectively. TMIn flow rate was adjusted

to obtain steady-state indium composition of 0.27. We will distinguish between (i) the ideal structure, which can be projected from the steady-state calibration of the precursor flow rates, (ii) the real structure with profile affected by In surface segregation, and (iii) the structure grown with the temperature jump from 730°C to 800°C at the onset of the GaN cap layer deposition (hereafter referred to as T-ramping). Figure 1a compares the composition profiles of these structures computed by using the model suggested in [5,6]. Because of delayed In incorporation into the crystal at the onset of the InGaN QW growth, the InN fraction does not reach its steady-state value in the real structure and exhibits the presence of indium (In "tail") in the cap layer. The application of T-ramping reduces conside-rably the In incorporation into the cap layer due to intensive In desorption observed at elevated temperatures.

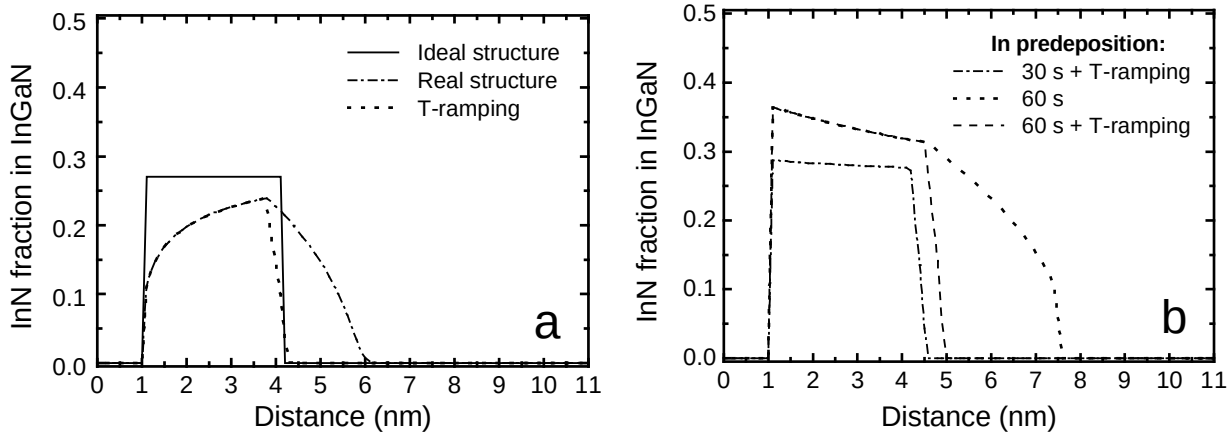

Figure 1 Composition profiles in GaN/InGaN/GaN SQW heterostructures grown under various conditions (see text for more detail).

TMIn deposition on the GaN surface prior to InGaN growth proves to be quite effective for improving the abruptness of the QW front interface. The variation of the TMIn exposure combined with the T-ramping during the cap layer growth allows one to control the entire InGaN QW composition profile (Figure 1b). In particular, the In predeposition for 30 s followed by T-ramping results in the nearly rectangular profile very close to that of the ideal heterostructure.

COMPUTATION OF BAND DIAGRAMS AND LIGHT EMISSION SPECTRA

The band diagrams of the InGaN SQW structures are found from self-consistent solution of the Poisson and Schrödinger equations. To calculate the space/surface charges due to piezoeffect and spontaneous polarization as a function of the QW composition, we use the non-linear approximations suggested in [7]. The donor and acceptor concentrations in the n- and p-GaN cladding layers are assumed to be 3×10^{18} cm^{-3} and 2×10^{19} cm^{-3}, respectively. The ionization energies of donor and magnesium acceptors are chosen to be, correspondingly, of 13 meV and 170 meV. Inside the QW, we take into account only the contribution of confined electrons and holes in the space charge. Outside the well, we use the local three-dimensional statistics relating the carrier concentrations to the separations of their quasi-Fermi levels, F_n and F_p, from the conduction band bottom E_C and the valence band top E_V. The concentrations of the ionised donors and acceptors are calculated via the quasi-Fermi levels in a conventional way. We assume here that

the quasi-Fermi levels are nearly constant throughout the heterostructure, neglecting the effects of carrier injection on band diagram. The quasi-Fermi levels at the edges of the structures are found from the electro-neutrality conditions, while their separation equals the forward bias applied, i.e. $U_b = F_n - F_p$. The band diagrams of the above SQW heterostructures computed for the bias of 3.0 V are shown in Figure 2a.

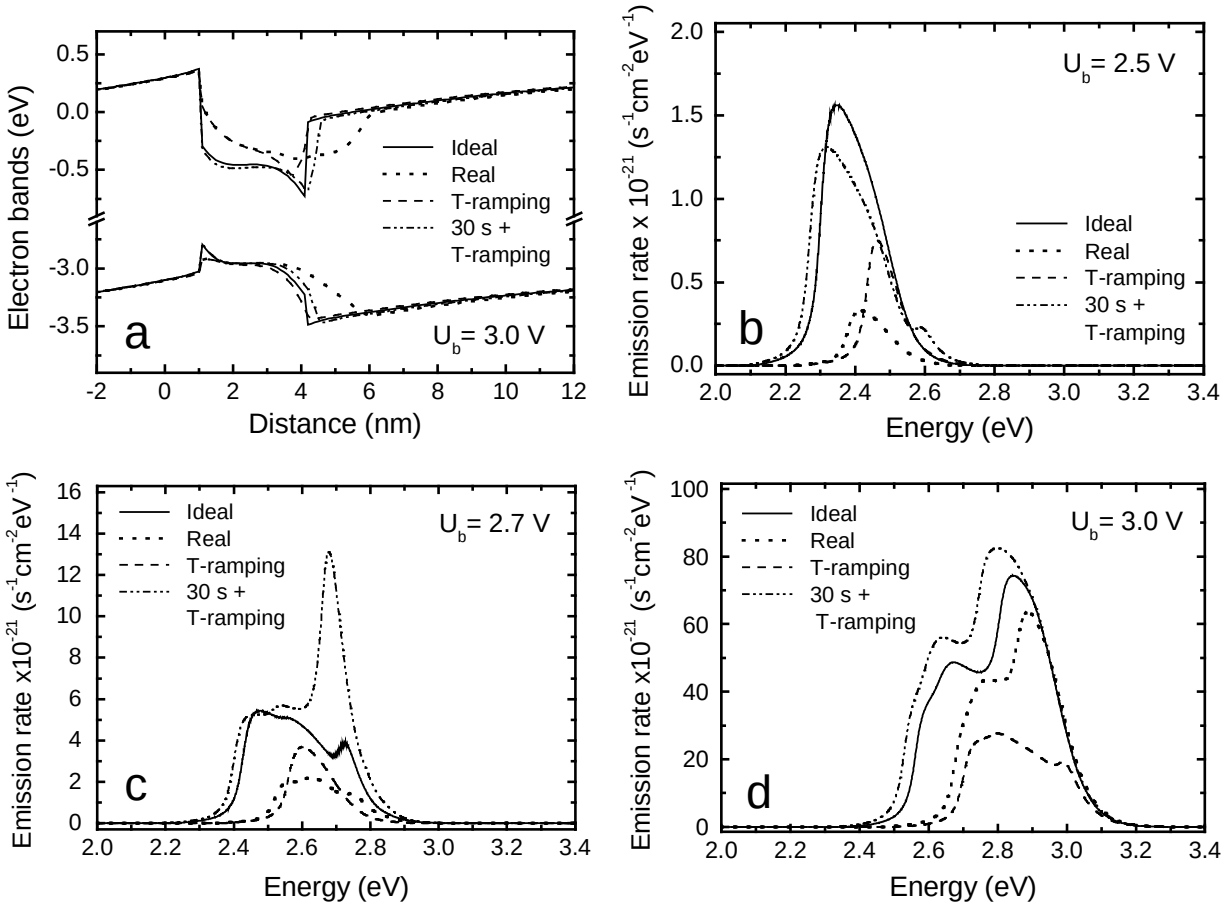

Figure 2 Band diagrams of InGaN SQWs (a) and their electroluminescence spectra (b-d) computed for different forward biases.

The spectra of electroluminescence (EL) are computed by accounting for only the spontaneous recombination between the electrons and holes confined in the QW. We neglect the radiative recombination through the donor and acceptor levels assuming the carrier injection into the QW to be sufficiently high at the biases of interest. The effect of indium composition fluctuations on the luminescence spectra is also neglected in this study. The Luttinger-Kohn approach is used to consider the complex valence band structure of nitrides [8]. The carrier effective masses, spin-orbital and crystal-field splittings are borrowed from [8,9]. The bowing parameter for the energy gap variation with InGaN composition is chosen to be 2.5 eV after [10]. The angle-averaged EL spectra are computed assuming a uniform electron level broadening Γ to be equal to 20 meV, the value typical for other III-V compounds.

RESULTS

Without broadening, an EL spectrum consists of several sharp peaks corresponding to the transitions between various electron and hole levels. The peak intensities depend on the filling of the levels with carriers and on the overlap of the electron and hole wave functions, which are spatially separated within the QW due to high build-in polarization field. The heavy holes (HH) and light holes (LH) in InGaN have rather similar effective masses in the [0001] direction and their spin-orbital splitting is small. Hence, they give rise into the nearly the same range of the EL spectrum. However, the heavy holes provide a higher contribution into the light intensity due to a larger effective mass in the lateral direction and, respectively, a higher density of states. Because of a small effective mass of the crystal-field split-off holes (CH) in the [0001] direction and of a large crystal field splitting, the CH energy levels are shifted down from the HH and LH levels. Thus, the concentration of the crystal-field split-off holes and their contribution to the EL spectra is relatively small.

In our computations, the separation between the hole levels in the QW turns out to be less or comparable with the broadening Γ. Therefore the fine structure of the hole levels is not resolved in the EL spectra. That is not the case for electrons, where the level separation exceeds the broadening remarkably. Therefore, the EL spectra become of a multi-peak character when new electron levels are produced by the QW transformation and filled by the carriers due to the bias increase (see Figure 2c,d).

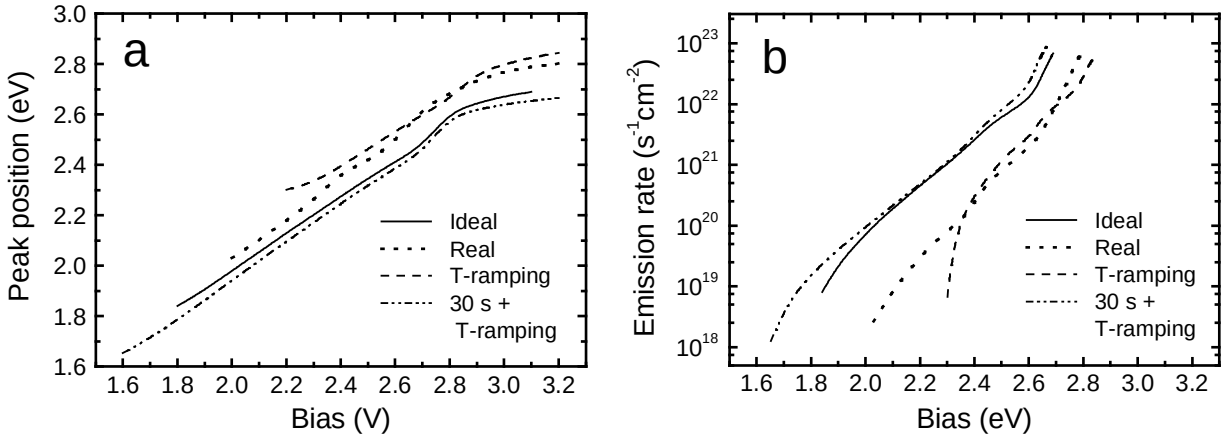

Figure 3 First EL peak spectral position (a) and its intensity (b) as a function of forward bias.

Electroluminescence spectra as a function of bias

At a small bias, there is practically no light emission from these heterostructures due to negligible carrier concentration in the QW. The concentration of electrons and holes gradually rises with bias, resulting in a more intensive light emission. In addition, a higher carrier concentration favours the screening of the polarization field inside the QW. Thus, the overlap of the wave functions corresponding to the ground electron and hole states increases, giving additional contribution to the light emission. First, there is the only peak in the EL spectra, with the energy close to separation of the electron and hole levels. The increasing forward bias produces the

second electron level and, respectively, the second peak in the EL spectra. This results in a blue shift of the whole luminescence spectra. An additional blue shift originates from the screening of the polarization field inside the QW, which produces a higher separation of the electron and hole energy levels (Figure 3a).

Segregation effects on the band diagram and emission spectra

As discussed above, the In surface segregation produces a remarkable difference between the composition profiles in the ideal and real InGaN SQW structures. The real QW has a smaller depth than the ideal one because of a lower peak composition, but it is wider due to the In tail in the cap layer. As a result, the EL spectra of the real structure are systematically blue-shifted by 50-100 meV compared to the ideal SQW (see Figure 3). T-ramping makes the QW narrower that leads to further blue shift in the spectra. As mentioned above, the combination of the 30 s indium predeposition with the T-ramping allows getting the structure very close to the ideal one by its shape. So, it is no wander that the EL spectra of such a structure are quite similar to those of the ideal SWQ. For all the structures considered here the first peak spectral position exhibits a nearly linear dependence with approximately the same slope until the bias of ~2.8 V is reached. This is due to the fact that under low biases, the effect of electron and hole injection into the QW on the space charge in the whole structure is negligible. At higher injection level, (U_b > 2.8 V) the electron and holes screen the build-in electric field, which makes the separation of their energy levels much less dependent on the applied bias.

The intensity of the first emission peak increases exponentially with bias U_b . This correlates well with the tendency reported in literature [11]. In [11], the first maximum in the EL spectra was associated with the “tunnel recombination” between unconfined electrons and holes. Our computations show that this peak may come from the recombination of electron and holes occupying the ground states in the QW.

Discussion

The most critical assumption made in our analysis of the luminescence spectra is neglecting the effects of In composition fluctuations on the light emission from the InGaN QWs. Normally, the fluctuation result in additional broadening of theemission spectra (~50-60 meV as measured in [11]) and in additional red shift of the spectra due to carrier localization in the In-rich regions. The account of the fluctuations, nevertheless, could not change the general trends in the EL spectra behavior predicted by our analysis. In particular, the second peak in the luminescence spectra also should be resolved as the separation between the first and second electron levels in the QW (~120-140 meV) is still larger than the fluctuation-related broadening.

CONCLUSION

In this work, we have studied theoretically the effect of In surface segregation on the composition profiles, band diagrams, and light emission spectra in the InGaN SQWs. The most effective approaches to control of the the abruptness of front and back interfaces in the SQW – indium predeposition and temperature ramping during the cap layer growth – are considered by using the

model [5,6]. It is shown that the combination of the In predeposition with the temperature ramping allows getting the SQWs with the composition profile very close to the desirable one (here, to that of the ideal rectangular QW).

Assuming a uniform broadening of the electron energy levels, we have computed the EL spectra from the SQW structures with different degrees of segregation impact. It is shown that In surface segregation normally results in a systematic blue shift of the EL spectra (~50-100 meV) due to incomplete In incorporation into the crystal in unsteady MOVPE growth. The bias variation produces the transformation of the QW band diagram in such a way as new electron and hole states are formed at high biases. The latter phenomenon results in a multi-peak structure of the EL spectra and, consequently, in additional blue-shift of the emission wavelength. Besides, the secondary peaks provide a higher broadening of the spectra, which is undesirable for practical applications.

Generally, this work demonstrates that the detailed analysis of a device structure growth, accounting for unsteady effects like surface segregation, coupled with the computations of light emission spectra is a powerful tool for bandgap engineering of the nitride LEDs. Further efforts should be made to consider fluctuations of In composition in an InGaN QW as well as the specificity of the active region doping.

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