Session: Photonics/optoelectronics/plasmonics

Multiscale modelling of the impact of intrinsic disorder and localisation effects on the optical and electronic properties of III-N LEDs

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Over the last twenty years, research into nitride-based semiconductor materials (InN, GaN, AlN, and their respective alloys) has gathered pace. This stems from their potential to emit light over a wide spectral range. Despite very high defect densities, blue emitting InGaN-based devices exhibit high quantum efficiencies. The widely accepted explanation for this is that the carriers are spatially localized due to alloy fluctuations and are thus prevented from diffusing to defects. It is important to note that in wurtzite InGaN systems the effect of these fluctuations is much more severe compared to that found, e.g., in zinc-blende InGaAs alloys. This originates from the very different physical properties (e.g., band gap and lattice spacing) of the binary constituents (InN and GaN). A further complication is that InGaN/GaN quantum wells (QWs), compared with InGaAs/GaAs wells, exhibit much stronger electrostatic built-in fields, arising in part from the strain dependent piezoelectric response (Fig. 1). Thus even random alloy fluctuations in InGaN/GaN QWs affect the electronic structure through a complicated interplay of local alloy, strain, and built-in field fluctuations.

To date, most simulation of the optoelectronic properties of III-N heterostructures and devices has been based on using **k.p** and effective mass models to describe the carrier states, as illustrated in Fig. 1. However there is a growing realisation that further device development and optimisation requires accurate understanding, control and exploitation of material properties. We first show here that calculations using density functional theory (DFT) provide clear evidence for the importance of random alloy effects in III-N alloys. Detailed treatment of the electronic structure then requires an empirical atomistic model capable to treat large-scale (100,000 atom) structures. We have developed a *sp*3 tight-binding Hamiltonian which gives an excellent description of the electronic and optical properties of III-N alloys and nanostructures. Preliminary non-equilibrium Green's function (NEGF) calculations show that inclusion of alloy disorder effects are also important for an accurate treatment of carrier transport in III-N heterostructures. There are still however many challenges to apply NEGF calculations to a full LED device. We discuss how modified drift-diffusion approaches can include alloy disorder effects and the consequences of disorder on transport and recombination properties.

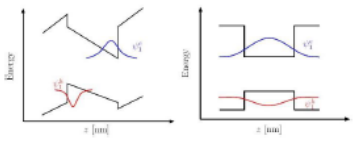

Fig. 1: Idealised variation of band-edge potential along growth direction in a c-axis quantum well structure, with spatial separation of electron (blue) and hole (red) wavefunctions (left), compared with strong overlap in absence of polarization potential (right).

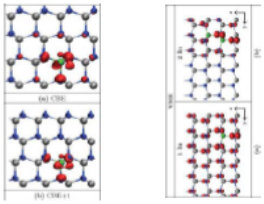

Fig. 2: Left: calculated LDA charge densities for (a) lowest conduction and (b) second conduction state in an $In_1Al_{799}N_{800}$ supercell. Right: Highest valence state in a 1600 atom InAlN supercell with (a) one In (b) In pair.

The left hand panel in Fig. 2 shows the charge density calculated using the local density approximation (LDA) in DFT for the lowest (CBE) and the second (CBE+1) conduction state in an $In1Al_{799}N_{800}$ supercell. It can be seen that the second state in this calculation is a resonant state, localised on the indium (In) site, with the lowest state also showing large probability density at the In site [1]. In the valence band (RHS, Fig. 2), the highest valence state remains delocalised in an $In1Al_{799}N_{800}$ supercell (lower panel), but evidence of

localisation can be observed when a pair of In atoms are considered in the supercell. These results highlight the importance of including detailed atomistic effects in III-N alloy calculations. We have developed a *sp*3 tight-binding model and benchmarked it against DFT calculations and experiment, in order to treat the large supercell structures required for accurate modelling of heterostructures.

Figure 3 shows the ground state electron and hole charge densities in an exemplary $In_{0.25}Ga_{0.75}N/GaN$ *c*-plane QW structure, using the *sp*3 model, and including the effects of well width fluctuation, local alloy composition, strain and built-in field fluctuations as well as Coulomb effects [2]. This and related calculations show that while the electron states are mainly localized by well-width fluctuations, the holes states are already localized by random alloy fluctuations. These localization effects dominate the QW optical properties, leading to strong inhomogeneous broadening of the lowest interband transition energy. Even when including Coulomb interactions, the electron and hole states remain separated along the *c*-axis in Fig. 3, due to the built-in polarisation potential.

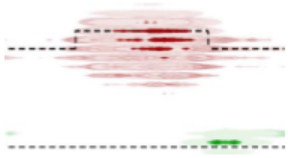

Fig. 3: Ground-state electron (red) and hole (green) charge densities with Coulomb interaction for an exemplar $In_{0.25}Ga_{0.75}N/GaN$ QW. Dashed lines indicate the QW interfaces.

The built-in polarisation potential can be eliminated by growing on non-polar substrates. This allows improved electron-hole overlap and increased exciton binding compared to growth on *c*-plane substrates. We have made a detailed theoretical and experimental comparison of the electronic structure and optical properties of a (non-polar) *m*-plane InGaN/GaN QW structure [3]. Our microscopic theoretical description again reveals strong hole wave function localization effects due to random alloy fluctuations, resulting in strong variations in ground state energies and consequently the corresponding transition energies (Fig. 4). This is consistent with the experimentally observed broad photoluminescence (PL) peak. Likewise, the calculations find strong exciton localization effects which explain the form of the PL decay transients. Additionally, the theoretical results confirm the experimentally observed high degree of optical linear polarization. Overall, the theoretical data highlight the strong impact of the microscopic alloy structure on the optoelectronic properties of these systems.

Given the strong impact of localization effects on the electronic states and optical properties, it can be expected that they will also strongly impact carrier transport in III-N heterostructures. We present preliminary calculations that confirm this expectation and conclude by discussing the importance of linking different scale models to ensure an accurate description and analysis of III-N LEDs and related devices.

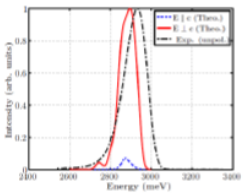

Fig. 4: Calculated excitonic ground state emission spectrum with light polarization vectors parallel and perpendicular to the c axis. The dashed-dotted line shows the experimental (unpolarized) PL spectrum

[1] S. Schulz, M. A. Caro and E. P. O'Reilly, Appl. Phys. Lett. 104, 172102 (2014)
[2] S. Schulz, M. A. Caro, C. Coughlan and E. P. O'Reilly, Phys. Rev. B 91, 035439 (2015)
[3] S. Schulz *et al.*, Phys. Rev. B 92, 235419 (2015).