Nonlinear Polarization and Efficiency Droop in Hexagonal InGaN/GaN Disk-in-Wire LEDs

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Abstract—Recent studies suggest that piezoelectric polarization can play an important role in determining the electronic and optical properties of nanoscale nitride heterostructures. Among a few models available, recent first-principles calculations performed by Prodhomme et al. provide a simple yet accurate description of linear and nonlinear piezoelectric coefficients in reduced dimensionality structures having wurtzite crystal symmetry. In this paper, first, within a fully atomistic VFF- sp\(^3\)\(^*\) tight-binding framework, we employ the model proposed by Prodhomme et al. to evaluate the importance of nonlinear piezoelectricity on the single-particle electronic states and interband optical transitions in a recently reported hexagon shaped In\(_{25}\)Zn\(_{25}\)Ga\(_{25}\)N/GaN disk-in-wire LED. The microscopically determined transition parameters are then incorporated into a TCAD toolkit to investigate how atomicity and the net polarization field affect the internal quantum efficiency of the LED and lead to a degraded efficiency droop characteristic.

Index Terms—nonlinear piezoelectricity, tight-binding, optical anisotropy, disk-in-wire LED, efficiency droop.

I. INTRODUCTION

NANOENGINEERED InGaN heterostructures are promising candidates for efficient, full-spectrum optical emitters for use in both conventional (e.g., lasers, solid-state lighting) and novel (e.g., near-field photolithography, single phonon based quantum cryptography, diagnostic medicine and biological imaging) application domains [1]. Traditionally, most InGaN heterostructures have been demonstrated in two forms: a) quasi 2-D quantum wells (QW) grown using molecular beam epitaxy; and b) quasi 0-D self-assembled quantum dots (QD) [2][3]. QDs, as compared to QWs, benefit from the presence of a relaxed (less-strained) active layer that leads to a reduced quantum confined Stark effect (QCSE). Nevertheless, recent studies show that the electronic and optical properties of these quasi zero-dimensional systems can be adversely affected by an intricate interplay of the quantum confinement effects and the atomicity in the underlying crystal lattice and interfacial discontinuities [4][5].

Very recently, exptially grown InGaN heterostructures realized in disk-in-wire architectures have been hailed as a breakthrough technology for applications in quantum photonics [6]. Various experimental studies demonstrate that these novel structures: a) are highly stable possessing greater reproducibility; b) can be grown free of extended defects, which allows the use of higher indium content and a wider spectral tuning; and c) due to the presence of greater geometrical symmetry, are subject to a lesser degree of internal electro-mechanical fields. In wurtzite heterostructures, compared to zincblende systems, the effects of polarization fields is strong, where, in addition to the off-diagonal strain tensors, biaxial strain induces a piezoelectric field parallel to the C axis ([0001] direction). Additionally, recent studies indicate that nonlinear (2\(^{nd}\)-order) piezoelectric polarization can also influence the electronic and optical properties of nanoscale III-V heterostructures [5][7][8]. For strained InAs/GaAs QDs, it was shown that, for large epilalexial strains, the 1\(^{st}\)-order and the 2\(^{nd}\)-order contributions are of comparable magnitude [9]. In this paper, considering a recently reported hexagon shaped InGaN/GaN disk-in-wire LED [6] as a reference device, we address the following three questions: a) Can the InGaN active disk buried in a GaN wire be completely strain-free?; b) Should one, while designing these devices, take into account the contribution of nonlinear polarization?; and c) How do various internal fields affect the internal quantum efficiency and associated droop (roll-over) characteristics in InGaN LEDs?

II. THEORY AND MODELS USED

Nanostructured InGaN optical emitters, in addition to quantum confinement, are subject to: a) symmetry lowering due to fundamental crystal atomicity and material discontinuity at the interfaces; b) non-homogeneous strain distribution; c) strain-induced piezoelectric polarization, \(P_{\text{piez}}\); and d) spontaneous polarization, \(P_{\text{sp}}\). For an accurate treatment of these internal fields, in this work, the numerical analysis has been carried out using a combination of valence force-field molecular mechanics (VFF-MM), 10-band \(sp^{3}\)\(^*\) atomistic tight-binding (TB), and a TCAD based quantum-corrected drift-diffusion transport framework as available in our in-house QuADS 3-D (quantum atomistic device simulation in three dimensions) simulator [10]. Note that, tight-binding is a local basis representation, which naturally deals with finite device sizes, alloy-disorder and hetero-interfaces and results in computationally tractable sparse matrices. The overall simulation strategy has been described elsewhere [5][11]. After creating the entire structure from a basis set, the atom positions are relaxed and the resulting strain fields are calculated via the valence force-field (VFF) method using the

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Keating potentials. The strain parameters used in this work are taken from Ref. [12] and validated through the calculation of Poisson ratio of the underlying materials. The small thermal strain contribution is neglected. The spontaneous polarization is strain-independent and arises from fundamental asymmetry of the crystal structure. In contrast, the piezoelectric polarization is obtained from the diagonal and shear components of the anisotropic atomistic strain fields [13]. To account for the 2nd-order piezoelectric field, a recent model of Prodhomme et al. [8] has been implemented in this work. In Ref. [8], to obtain the second-order piezoelectric coefficients, the authors have used a finite difference technique in conjunction with density functional perturbation theory (DFPT) within the local density approximation (LDA). The calculations were carried out with the Abinit toolkit using the Troullier-Martins (TM) pseudopotentials. Here, the net piezoelectric polarization (including the 1st-order and the 2nd-order contributions) along the [0001] direction in a wurtzite crystal is given by:

\[
P_{PZ,z}^{net} = e_{31} \left( e_{xx} + e_{yy} \right) + e_{33} e_{zz} + \frac{1}{4} \left( e_{111} + e_{312} \right) \times \left( e_{xx} + e_{yy} \right)^2 + \frac{1}{2} e_{333} e_{zz}^2 + 2 e_{313} e_{xx} e_{zz}.
\]

The polarization constants (in C/m²) used in this study are listed in Table I. Note that: a) the second-order polarization coefficients are almost an order of magnitude higher than the linear counterparts; and b) all of the coefficients, \( e_{33} \), has the largest magnitude and can induce significant second-order effects especially in materials with a rather small linear coefficient \( e_{33} \) and large epitaxial strain. Also, note that, compared to an earlier 2nd-order model [7] proposed by Pal et al. (that was implemented in a recent work [5] of the current authors), the new model proposed by Prodhomme et al. reported (from symmetry considerations) a zero value for the coefficient \( e_{133} \) [14]. Next, the polarization induced potential is obtained by solving the 3-D Poisson equation on an atomistic grid using the open source PETSc toolkit [15]. Next, the single-particle eigenvalues, wave functions, and the interband optical transition rates are calculated using an empirical nearest-neighbor \( sp^3s^* \) tight-binding model as implemented in the open source NEMO 3-D simulator [16]. Finally, the microscopically determined optical transition parameters are incorporated in the Synopsys’ Sentaurus toolkit to obtain the terminal characteristics of the LED and investigate how various internal fields affect the internal quantum efficiency of the device.

### III. Results and Discussion

Fig. 1 shows the schematic of the simulated InGaN disk-in-wire LED. The core atomistic simulation domain assumes a hexagonal geometry for the wurtzite crystal system. The GaN nanowire is oriented in the [0001] direction (c-axis) with longest diagonal, \( d \sim 14 \text{ nm} \) (side, \( s \sim 7 \text{ nm} \)) and height, \( h \sim 100 \text{ nm} \). The In\(_{0.25}\)Ga\(_{0.75}\)N quantum disk is positioned at the center of the GaN nanowire and has a height of \( h_e \sim 3 \text{ nm} \). Looking at the strain profiles, as shown in Fig. 2, both the hydrostatic \( \left( e_{xx} + e_{yy} + e_{zz} \right) \) and the biaxial \( \left( e_{xx} + e_{yy} - 2e_{zz} \right) \) components were found to be compressive within the disk and tensile in the surrounding material matrix.

![Fig. 1. (left) Schematic representation of the simulated InGaN quantum disk-in-wire LED. (right) Atomicity of the active region.](image)

Fig. 3(a) shows the polarization-induced piezoelectric potential distribution (for different model configurations) along the growth direction and through the center of the disk. The salient features one can extract from this Figure are as follows: a) The 2nd-order piezoelectric contribution is significantly smaller than the 1st-order counterpart, magnitude being only \(-18.3 \text{ mV} \) at the InGaN/GaN interfaces and almost negligible in the GaN substrate and cap layers; and b) The net potential peaks at the interfaces at \(-134 \text{ mV} \), tends to be symmetric around the center of the dot (along the growth direction), and, as seen in Fig. 3(b), features strong dipole formation in the \( x-z \) plane.
Fig. 2. (a) Atomistic diagonal, biaxial, and hydrostatic strain profiles along the growth ([0001]) direction through the center of the structure. Strain is seen to penetrate deep (~15 nm) into the GaN matrix. (b) Off-diagonal strain variations along the [0001] direction.

Fig. 4 shows the HOMO ($E_V$) and the LUMO ($E_C$) wavefunctions projected on the $x$-$y$ plane in the InN quantum disk. Here, it is clear that, due to the atomistic and random nature of the distribution of atoms, even in the absence of internal fields (depicted as WO), isotropy is never achieved. Inclusion of atomistic strain and electrostatic fields in the calculation leads to a strong localization in the HOMO wavefunctions, and, therefore, lowers the overall electronic symmetry of the system. The bottom row in this Figure shows the single-particle bandgap, $E_G$, of the LED structure, which, in general, agrees well with the experimentally measured value (~2.73 eV at 300°K) as reported in Ref. [6].

The influence of the internal fields on the single-particle bandgap is summarized in Fig. 5. Looking at this Figure, we can infer that: a) both quantum confinement and strain lead to a large blue-shift in the bandgap, strain being the stronger contributor of the two; b) pyroelectric contribution is negligible; c) the 1st-order piezoelectricity alone introduces a significant redshift and cannot be ignored; d) the bulk or experimental (expt) piezoelectric constant overestimates the degree of redshift, as compared to the same obtained from first-principles calculations for nanostructures; and e) 2nd-order piezoelectricity, while opposing the 1st-order counterpart, is very weak in magnitude and may be ignored in the calculation of the single-particle energy bandgap.

Fig. 6 shows the polar plots of the interband optical transition rates between ground hole (HOMO) and ground electron (LUMO) states. It is found that, all internal fields (as compared to the WO case) break the isotropy in the emission characteristic with varying degrees. Overall, the in-plane polarization anisotropy was found to be ~0.91, due mainly...
Fig. 7. Simulated internal quantum efficiency (IQE) as a function of injected current for the InGaN disk-in-wire LED. Here, Control refers to a device without any structural disorder and $k_0 = 1$.

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Next, Synopsys’ Sentaurus TCAD toolkit is used to simulate the optical characteristics of the disk-in-wire LED. Here, the core active region comprises of an undoped 3 nm thick In$_{0.25}$Ga$_{0.75}$N disk sandwiched between an n-doped GaN buffer region and a p-doped GaN cap layer, both having a base length, $b=14$ nm and height, $h=48.5$ nm. Note that, the effect of internal fields, which is characterized by a strong suppression of interband optical transition rate, is modeled via a scaling factor ($k_0$) for the optical matrix element accessible in the TCAD toolkit. The $k_0$ values used here are the average transition rates obtained from Fig. 6. Fig. 7 shows the variation of internal quantum efficiency (IQE) as a function of injected current for various models used in the calculation. Here, the rollover in efficiency ($dop$), even in the absence of any structural disorder (defects, SRH centers, strain and polarization fields, depicted as Control structure), is due mainly to carrier loss from within the confined disk region. Importantly, it is found that, inclusion of the internal polarization fields in the calculation: a) reduces the IQE drastically; and b) further degrades the dop characteristic by inducing the same at a smaller injected current value.

IV. CONCLUSION

InGaN/GaN disk-in-wire heterostructures, although realized with a greater geometrical symmetry and a more relaxed lattice compared to the self-assembled QD counterparts, are still subject to non-negligible epitaxial strain and compositional and interfacial atomicity. For a recently reported device, full atomistic calculations of the electronic structure reveal a strong dependence of the bandgap on the internal fields and feature large growth-plane optical polarization anisotropy. The 2nd-order piezoelectricity, while minimally affecting the energy bandgap, renders a non-vanishing effect in retaining the symmetry of the system. Overall, the net polarization field was found to degrade the efficiency dop characteristic and, therefore, must be taken into account in modeling these novel LED structures.

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