Influence of strain relaxation in axial $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ nanowire heterostructures on their electronic properties

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Abstract

We present a systematic theoretical study of the influence of elastic strain relaxation on the built-in electrostatic potentials and the electronic properties of axial $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ nanowire (NW) heterostructures. Our simulations reveal that for a sufficiently large ratio between the thickness of the $\text{In}_x\text{Ga}_{1-x}\text{N}$ disk and the diameter of the NW, the elastic relaxation leads to a significant reduction of the built-in electrostatic potential in comparison to a planar system of similar layer thickness and In content. In this case, the ground state transition energies approach constant values with increasing thickness of the disk and only depend on the In content, a behavior usually associated to that of a quantum well free of built-in electrostatic potentials. We show that the structures under consideration are by no means field-free, and the built-in potentials continue to play an important role even for ultrathin NWs. In particular, strain and the resulting polarization potentials induce complex confinement features of electrons and holes, which depend on the In content, shape, and dimensions of the heterostructure.

Keywords: Nanowires, elastic relaxation, piezoelectricity, electronic properties

The ternary alloy $\text{In}_x\text{Ga}_{1-x}\text{N}$ constitutes the semiconductor material of choice for the development of red–green–blue light emitting diodes for display technology, since its emission wavelength can in principle be tuned from the near-infrared to the ultraviolet via the In content $x$ [1–3]. However, due to the large lattice mismatch of about 10% between InN and GaN and the tendency for phase separation, it is difficult to produce $\text{In}_x\text{Ga}_{1-x}\text{N}$ films on GaN with the In content required for red emission while retaining a sufficiently high crystal quality [4].

Additionally, the strong polarization potentials occurring in planar $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ heterostructures induce a spatial separation of electrons and holes and correspondingly low recombination rates.

A possible solution to overcome the limitations of planar heterostructures is the growth of GaN nanowires (NWs) with axial $\text{In}_x\text{Ga}_{1-x}\text{N}$ insertions [5–11]. In contrast to planar structures, the large surface-to-volume ratio facilitates elastic relaxation of the lattice mismatched axial insertions [12–15], thus making the incorporation of larger In contents in the insertions possible without inducing plastic relaxation. Additionally, it is widely believed that elastic relaxation in axial $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ NW heterostructures also leads to a significant reduction of the built-in piezoelectric potential and thus the quantum confined Stark effect (QCSE) [16]. In fact, some researchers even reported evidence for a vanishing QCSE [17–20], similar to findings in other material systems [21, 22].

The influence of elastic relaxation of axial $\text{In}_x\text{Ga}_{1-x}\text{N}$ disks on the piezoelectric potential in GaN NWs has only recently been subject of a systematic study, and it was shown that a total elimination of piezoelectric fields cannot be achieved in axial NW heterostructures [23]. In fact, much research effort has been dedicated to axial...
In$_x$Ga$_{1-x}$N/GaN NW heterostructures in the past. However, the question of how this elastic relaxation and the resulting polarization potential influences carrier confinement and transition energies in axial In$_x$Ga$_{1-x}$N/GaN NW heterostructures has not been addressed so far in a systematic manner.

In the following, we will shed light on the impact of elastic relaxation in axial In$_x$Ga$_{1-x}$N/GaN NW heterostructures on their piezoelectric and electronic properties. We focus particularly on thin NWs, where the ratio between the thickness of the In$_x$Ga$_{1-x}$N insertion and the NW radius approaches unity, such that significant elastic relaxation is expected [24]. We observe a strongly reduced magnitude of the polarization potential in the NW heterostructure in comparison with a planar structure of similar thickness and In content. For larger disk thicknesses, the transition energies are found to approach constant values that depend only on the In content, a behavior which at the first glance reflects that expected for a field-free structure with a vanishing QCSE. At the same time, this phenomenon leads to a limitation of the energy range that can be accessed using axial In$_x$Ga$_{1-x}$N/GaN NW heterostructures. However, we also show that polarization potentials are still present and have a significant influence on both the transition energies and the confinement of charge carriers.

Our model system is a GaN NW with a diameter of 15 nm containing an In$_x$Ga$_{1-x}$N disk of thickness $t$. Such ultrathin GaN NWs have been recently fabricated by either direct growth [25] or post-growth thermal decomposition [26]. The system under consideration comprises about 500 000 atoms in a hexagonal crystal lattice. While it is nowadays possible to compute the elastic and electronic properties of systems of such dimensions using atomistic approaches, the large parameter space spanned by the variations in both In content and disk thickness makes a computationally less expensive approach desirable. For the calculation of the electronic properties, we have thus employed an eight band $k \cdot p$ model for strained wurtzite crystals [27], implemented within the plane-wave framework of the S/PHI/nX library [28, 29]. In fact, an excellent agreement between atomistic tight-binding models and this eight band $k \cdot p$ model was obtained for even smaller III-nitride nanostructures [30].

The elastic properties that enter the above $k \cdot p$ model were computed using continuum elasticity theory [31]. Here, the plane-wave framework of the elasticity theory module of the S/PHI/nX library is not well suited as it is a priori subject to periodic boundary conditions, such that very large supercell dimensions along the (0001) direction would be required to ensure a well-converged description of the elastic properties of the In$_x$Ga$_{1-x}$N disk and its vicinity. Strain and polarization potentials were therefore computed using the finite element method (FEM) implemented within the commercial FEM solver MSCMarc$^{63}$. The material parameters for our simulations were taken from [32]. However, following a previous study [33], we use a negative value for $e_{15} = e_{31}$. The ground state recombination energies as a function of the layer thickness of NW heterostructures are compared with those obtained for planar In$_x$Ga$_{1-x}$N/GaN heterostructures. For the sake of comparability, strain and polarization of the planar layer were transferred to a structure with the same cross section as the NW, such that small energy contributions from in-plane quantization are consistent in both the NW and the planar system. To test the accuracy of our numerical FEM-based approach, we have performed a comparison between the analytical solution of the elastic and piezoelectric problem for axial NW heterostructures presented in [15, 23] and the numerical model for the simplifications that are intrinsic to the analytical model (spatially independent elastic, piezoelectric, and dielectric constants and a cylindrical NW cross section). Deviations of strain and piezoelectric potentials resulting from discretization issues were below 1%. In the following, the more realistic representation of a hexagonal NW cross section and spatially dependent parameters was employed.

In a previous work, we have investigated the influence of surface potentials resulting from unintentional doping and Fermi level pinning [34]. In the present study, we can neglect this contribution for several reasons. Assuming a doping density of $10^{17}$ cm$^{-3}$ as in [34], the maximum of the surface potential would amount to less than 3 meV, a negligible value compared to the potential contributions arising from strain and polarization. In the more realistic picture of individual, randomly distributed dopants, as discussed in [35], this doping density corresponds to an average of 0.3 charges in the NW segment under consideration, meaning that the segment is most likely to be entirely undoped. In all what follows, we therefore consider the NW segment under consideration as undoped and thus free of surface potentials.

To illustrate the elastic relaxation of an axial In$_x$Ga$_{1-x}$N/GaN NW heterostructure, we show in figure 1 the

![Figure 1](image_url)
diagonal components of the elastic strain tensor $\varepsilon_{xx}$ and $\varepsilon_{yy}$ for an In content of 40%. A significant relaxation is seen for thicknesses of 4.5 and 6.5 nm. For thin disks, lateral relaxation occurs only at the side facets of the NW so that the central area of the In$_x$Ga$_{1-x}$N disk is subject to elastic strain that is very similar to the one of a planar layer. In all cases, however, the lattice mismatch between In$_x$Ga$_{1-x}$N and GaN induces significant strains at the interfaces which will modify the polarization potentials and thus the electronic properties of the NW.

Line traces of the polarization potential along the central axis as well as along a parallel line at the surface ($r = R$) are shown in figure 2 for an In$_{0.4}$Ga$_{0.6}$N disk of 4.5 nm thickness. The absolute extrema of the potential are located at the side facets rather than at the center of the NW for this thickness, with the maximum and minimum at the bottom and top interfaces of the In$_x$Ga$_{1-x}$N disk.

Figure 3(a) shows the maximum of the polarization potential along the axis of the NW as well as along a parallel line at the side facet for different In contents as a function of the disk thickness. The maxima along the NW axis are largest for all In contents at a thickness of 2.5 nm and then decrease to a minimum value at 4.5 (x = 20%), 5.5 (x = 40%), and 6.5 nm (x = 60%). After this point, the maximum of the polarization potential along the NW axis increases again due to the spontaneous polarization. At the side facet, the maximum of the potential has its largest value for disk thicknesses of 4.5 nm. This finding is in agreement with data reported in [23], where the maximum of the polarization potential along the side facet was calculated as a function of the disk thickness for different NW diameters based on an analytical model for cylindrical NW heterostructures. The maximum occurs due to progressive strain relaxation for a thickness/diameter ratio of 0.3, and hence has a value of 4.5 nm for a diameter of 15 nm. Note that the precise ratio depends on the elastic and piezoelectric constants and is thus specific for a certain material.

The polarization potential scales almost linearly with the In content of the In$_x$Ga$_{1-x}$N disk. The qualitative behavior of the polarization potential as a function of the disk thickness can therefore be described better after normalizing the potential with the In content. The overall maximum of the normalized polarization potential throughout the whole NW with a diameter of 15 nm is shown in figure 3(b) as a function of the disk thickness. The absolute extrema of the polarization potential have a maximum at a disk thickness of about 4.5 nm and a reduction is seen after this point. This behavior translates into a limitation of the range of electron–hole transition energies, as we will discuss in the following. It is also observed that the absolute maximum of the polarization potential scales with the In content and only small differences
are seen between the curves normalized with the In content in figure 3(b).

The insets in figure 3(b) show that the potential has its extremal at the central axis of the NW for thin disks and at the side facets for thick disks. This behavior is quantitatively depicted in figure 3(c), which shows the ratio $\xi = \max[V_P(0)]/\max[V_P(R)]$ between the maximum potential along the central axis and the one along a parallel axis at the side facets as a function of the disk thickness. Above a thickness of about 2 nm, the extrema of the potential are at the side facets rather than at the central axis of the NW. The splitting of $\xi$ for the different In contents above a thickness of 4.5 nm results from the minima of $\max(V_P)$ along the NW axis as shown in figure 3(a), which occur at different thicknesses for the In contents considered.

Figure 4 shows the energy difference between the electron and hole ground states, $\Psi_{e0,0}$ and $\Psi_{h0,0}$, as a function of the disk thickness $t$ differs drastically between the planar and the NW heterostructures. For the former, the transition energies decrease linearly with layer thickness due to the strong polarization potential. In contrast, we obtain values of $4.06 \times 10^{-3}$ and $1.56 \times 10^{-3}$ for the NW heterostructures with 40% In content and 5.5 nm thickness and 60% In content and 1.5 nm thickness, respectively. For the thin disk with 60% In content, the large overlap results from the negligible spatial separation of electron and hole along the growth direction. However, even for an In content of only 40%, the electron–hole overlap is significantly larger than for a planar layer.

As a further consequence of the saturation of the transition energies, long wavelength emission requires a significantly higher In content than for the corresponding planar heterostructure. For example, red emission at about 1.9 eV is predicted to be achieved with a planar heterostructure containing 20% In at a layer thickness of about 3.5 nm. A NW heterostructure emitting at this wavelength and at the same layer thickness would require an In content of about 45%. Alternatively, one could employ a 5.5 nm thick disk with an In content of 40% or a 1.5 nm thick disk with an In content of 60%.

Since we have assumed that the NW has a diameter of only 15 nm, the strain induced by the In$_{x}$Ga$_{1-x}$N disk should be accommodated elastically for the entire parameter range depicted in figure 4 [36]. This elastic strain relaxation strongly reduces the polarization potential in the NW heterostructure as compared to the equivalent planar heterostructure (see figure 3(b)), but at the same time also reduces the redshift induced by the QCSE, requiring in turn significantly larger amounts of In for obtaining the same emission wavelength. We emphasize that the reduction of the polarization potential requires a large ratio between insertion thickness and NW diameter and is thus specific to the emerging field of ultrathin NWs. The crucial question at this point is whether or not the reduced piezoelectric potential increases the electron–hole overlap and thus constitutes an actual advantage. Let us consider the example given above of structures emitting at 1.9 eV. For the planar heterostructure, the electron–hole overlap $\mathcal{O}$ as defined in [34] is extremely small ($1.17 \times 10^{-9}$) due to the strong polarization potential. In contrast, we obtain values of $4.06 \times 10^{-3}$ and $1.56 \times 10^{-3}$ for the NW heterostructures with 40% In content and 5.5 nm thickness and 60% In content and 1.5 nm thickness, respectively. For the thin disk with 60% In content, the large overlap results from the negligible spatial separation of electron and hole along the growth direction. However, even for an In content of only 40%, the electron–hole overlap is significantly larger than for a planar layer.

In general, however, we cannot predict simple monotonic trends for the electron–hole overlap as a function of disk thickness and In content. The overlap depends on the confinement of electron and hole, which is determined by the strengths of polarization potential and strain in a complicated manner, so that each configuration of thickness, diameter, and In content of a NW heterostructure needs to be studied individually. To illustrate this fact, we show the electron and hole ground state charge densities together with the respective overlap $\mathcal{O}$ for a few selected configurations in figure 5. The electron is typically confined in the center of the NW for both thin disks and low In contents, but tends to be confined in the corner of the NW for thicker disks and larger In contents. For the hole, a confinement in the center of the NW is observed only for an In content of 20%. Otherwise, the hole is confined at the corner (e.g., for disk thicknesses of 4.5 and 6.5 nm for an In content of 60%) or the side facets (for disk thicknesses of 2.5 nm for $x = 40\%$ and 60% and for disk thicknesses of 4.5 and 6.5 nm for an In content of 40%), as also reported in [37]. Note that in all cases the electron is confined at the top

Figure 4. Electron–hole ground state transition energy in the In$_{x}$Ga$_{1-x}$N/N/GaN NW heterostructure under consideration as a function of the disk thickness. In content $x$ is indicated at each curve. For comparison, the transition energies for the limiting case of a planar heterostructure with the same In contents are indicated in shades of gray from light (20%) to dark (100%).

[Figure 4 description]

$E(\Psi_{e0,0}) - E(\Psi_{h0,0})$ (eV)

$t$ (nm)

1 2 3 4 5 6 7

20% 40% 60%

100%

0.5 1 1.5 2 2.5 3

Note that in all cases the electron is confined at the top...
facet of the active In$_x$Ga$_{1-x}$N disk, whereas the hole resides at its bottom, due to the drop of the polarization potential along the NW axis seen in figure 2. This vertical separation leads to a reduction of the overlap for larger disk thicknesses, as seen in figure 5. We note that, while the employed formalisms can be applied to NW heterostructures based on any other direct semiconductor material, the conclusions are highly specific and apply to In$_x$Ga$_{1-x}$N/GaN NWs only. In particular, In$_x$Ga$_{1-x}$N/GaN is characterized by both a very large lattice mismatch and a polarization potential that largely originates from piezoelectricity. For GaN/Al$_x$Ga$_{1-x}$N NWs, for example, the strain is smaller and the polarization potential is dominated by the difference in spontaneous polarization. Strain relaxation is thus expected to have a far less significant influence than in the present case.

To retain an electron–hole overlap higher than that for a planar heterostructure with comparable transition energy, it is imperative to choose a configuration for which no radial separation of electrons and holes occurs. This effect, however, evidently depends on the structural parameters of the NW heterostructure in a highly nontrivial manner. In the present study, we have considered the In$_x$Ga$_{1-x}$N insertion to be represented by a disk, but it is known from experiment that the insertion may assume rather complex shapes that have been found to affect the spatial distribution of the electron and hole charge densities as well [35]. These different shapes will not affect the general behavior found here for simple disks, particularly the trend to approach constant transition energies with increasing thickness of the insertion. However, the absolute transition energies as well as the actual values for the electron–hole overlap will depend sensitively on shape. If experimental and theoretical values for these quantities are to be compared, a complete three-dimensional reconstruction of the insertion’s size, shape, and composition on a nm scale is thus required. Electron and atom-probe tomography are experimental techniques offering the possibility of such a reconstruction. Once the geometry of the structure is known, we can proceed exactly as outlined in the present paper even if the NW and the insertion exhibit irregular shapes.

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