Influence of composition, strain, and electric field anisotropy on different emission colors and recombination dynamics from InGaN nanodisks in pencil-like GaN nanowires

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This work reports an experimental and theoretical insight into phenomena of two-color emission and different electron-hole recombination dynamics in InGaN nanodisks, incorporated into pencil-like GaN nanowires. The studied nanodisks consist of one polar (on c facet) and six (nominally) identical semipolar (on r facets) sections, as confirmed by transmission electron microscopy. The combination of cathodoluminescence with scanning electron microscopy spatially resolves the nanodisk two-color emission, the low-energy emission (\sim 500 nm) originating from the polar section, and the high-energy emission (\sim 400 nm) originating from the semipolar section. This result has been directly linked to a "facet-dependent" nanodisk composition, the In content being significantly higher in the polar ($\sim 20\%$) vs semipolar ($\sim 10\%$) section (as quantified by energy dispersive x-ray spectroscopy), further leading to a strong facet-dependent strain anisotropy. Time-resolved cathodoluminescence reveals significantly different electron-hole recombination times in the two sections, moderately fast (\sim 1.3 ns) vs fast (~0.5 ns) in polar/semipolar sections, respectively, the difference being linked to a strong anisotropy in the nanodisk internal electric fields. To determine the influence of each of the three contributing "facet-related" anisotropies (composition, strain, and electric field) on the two-color emission, a proper simulation [relying on virtual crystal approximation and involving three-dimensional (3D) continuum mechanical modeling, a 3D Poisson equation, and a one-dimensional Schrödinger equation] has been performed. The theoretical simulations allow the three effects to be quantitatively disentangled, revealing a clear hierarchy among their contributing weights, the facet-dependent composition inhomogeneity being identified as the dominant one (and the strain inhomogeneity being identified as the least significant one). As for different recombination times, while it is mainly linked to the internal electric field anisotropy, we also suggest that it is, very likely, influenced by gradually increasing In content along the nanodisk growth direction (lattice-pulling effect); the latter mechanism keeps electrons and holes in (relative) proximity within the polar section, enabling their relatively fast and efficient radiative recombination.

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I. INTRODUCTION

During the last two decades highly efficient wide-band-gap III-nitride emitters have revolutionized the market of solidstate lighting. The achievement of white solid-state light finally led to the Nobel Prize award to its principal inventors [1]. However, typical white LEDs generally combine blue emitters with a yellow phosphor, which drags efficiency (because of unavoidable energy conversion loss) and leads to a poor colorrendering index (CRI) [2,3]. To boost efficiency and improve CRI various approaches toward broadband phosphorus-free light emitters have been suggested; a particularly attractive one relies on InGaN/GaN multi-quantum-wells (MQWs) grown on GaN multifacet microstructures [4–6]. Recent improvements in selective area growth (SAG) of ordered (In)GaN nanowires (NWs) led to a further miniaturization of multicolor InGaN emitters, their size now being controlled in the sub-100-nm range [7,8]. Despite several experimental reports, the exact background mechanisms which drive the multicolor emission phenomenon still remain largely unknown.

The lack of central symmetry of the III-N wurtzite structure leads to a strong anisotropy of its physical properties. First, a strong anisotropy of the GaN crystal facets' surface energy per unit area (γ) favors (In)GaN growth in polar over semipolar and nonpolar crystal directions [9,10]. This intrinsic γ -value anisotropy strongly determines and (constrains) the shape of (In)GaN nanostructures. Second, it is well known that In incorporation strongly depends on crystal facet, being significantly higher on polar with respect to semipolar and nonpolar ones [11,12]. This "facet-related" anisotropic behavior determines the chemical composition and, consequently, the strain distribution across the InGaN nanostructure volume. Finally, the strong anisotropy in polarization fields, which can be orders of magnitude higher on the polar vs semipolar and nonpolar crystal interfaces, determines (and constrains) the electron-hole (e-h) recombination dynamics within the nanostructure [13]. The exploitation of the multicolor emission phenomenon in real devices requires a significant degree of emission wavelength/intensity control, for which a detailed insight into its underlying driving mechanisms is necessary.

This work reports on the fabrication, characterization, and modeling of two-color emitting InGaN nanodisks incorporated into site-controlled pencil-like GaN NWs, consisting of one

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FIG. 1. (a) Top SEM view of a representative sample area revealing ordered hexagonal array of pencil-like NWs. (b) TEM micrograph of a representative single NW (sample B) revealing the structure of its building blocks. (c) Atomic resolution HAADF STEM reveals structural properties of the GaN-capping/InGaN-nanodisk interface: (up) polar *c*-facet and (down) semipolar *s*-facet interface.

polar (on *c* facet) and six (nominally) identical semipolar (on *r* facets) InGaN sections [7,8]. The first part of the article provides a detailed experimental insight into inherent facet-related anisotropies (composition, strain, and electric field) of the III-N materials, which lead to the two-color emission and different *e*-*h* recombination dynamics. In the second part, we perform appropriate theoretical modeling and provide a detailed discussion aiming to quantitatively disentangle and compare the influence that each of the three contributing anisotropies has on the two-color emission. Finally, we provide an extended discussion, in which experimental and theoretical findings are compared, pointing out a good agreement between the two.

II. METHODS

A. Growth

The samples were grown in a RIBER Compact 21 molecular beam epitaxy (MBE) system, equipped with a radio-frequency plasma nitrogen source and standard Knudsen cells, one for In and two for Ga. All growths were performed on ~ 3.3 - μ mthick commercial GaN-on-sapphire (0001) templates grown by metal-organic chemical-vapor deposition and covered with a Ti nanohole mask fabricated by colloidal lithography (for details see the Supplemental Material [14]). The samples under consideration, designated as A, B, and C respectively, were grown following a three-step procedure, each step defining one building block: (i) initial pencil-like GaN NW, (ii) InGaN nanodisks, and (iii) top GaN capping. Building blocks (i) and (iii) were kept constant whereas the InGaN nanodisk was varied in composition and thickness.

(i) Initial GaN NWs are grown selectively on a Ti nanohole mask following a standard SAG process [15,16]. They have

diameters of ~ 180 nm (fixed by the nanohole mask) and heights of ~ 600 nm (controlled by growth rate and time).

(ii) InGaN nanodisks in samples B and C have the same nominal thickness $[d(B) = d(C) \sim 30 \text{ nm}]$, while sample A has a thinner nanodisk [nominal thickness $d(A) \sim 15 \text{ nm}]$ resulting from a shorter growth time. Similarly, the InGaN nanodisks in samples A and B have the same nominal composition, while sample C nanodisk has a higher In content [i.e., $X_{\text{In}}(A) = X_{\text{In}}(B) < X_{\text{In}}(C)$], resulting from lower growth temperatures: $T_{\text{A}} = T_{\text{B}} = 625 \,^{\circ}\text{C}$ and $T_{\text{C}} = 610 \,^{\circ}\text{C}$.

(iii) GaN cappings have a constant thickness of \sim 50 nm. A detailed structure of the samples is shown in Fig. 1, in the following section, whereas further growth details can be found in the Supplemental Material [14].

B. Characterization

The local structural and chemical characterization of the samples has been performed using an aberration-corrected scanning transmission electron microscope (STEM) JEOL-JEMARM200cF, operating at 200 kV with a probe size of \sim 0.8 nm, and enabled for energy dispersive x-ray spectroscopy (EDS) experiments. An initial microstructural analysis was performed using bright-field (BF) STEM imaging. High-angle annular dark field (HAADF) STEM with atomic resolution was performed using a solid semiangle between 68 and 280 mrad. Compositional mapping was achieved through EDS measurements using the Oxford INCA-350 detector. Standard macrophotoluminescence (PL) was performed at ~ 10 K, exciting with the 325-nm HeCd laser line over a range of 0.1-10 mW power and the signal was detected with a Hamamatsu photomultiplier. Direct correlation of the optical and structural properties on a nanometer scale was achieved using low-temperature cathodoluminescence (CL) spectroscopy (T < 20 K) in an STEM-CL system [17]. The CL detection unit is integrated in an FEI STEM Tecnai F20 equipped with a liquid helium stage. The emitted CL is collected by a retractable parabolically shaped aluminum mirror, focused onto the entrance slit of a grating monochromator (MonoCL4, Gatan) and detected by a liquid nitrogen cooled back-illuminated Si-CCD. Simultaneously to the detection of the CL signal at each position, the electrons that are forward scattered are acquired by an annular dark-field (ADF) detector. The STEM acceleration voltage is optimized to 80 kV to minimize sample damage and to prevent luminescence degradation. Time-resolved (TR) CL measurements have been performed in combination with scanning electron microscopy (SEM). The electron beam is switched on and off by an electrostatic beam blanking. An excitation voltage of 5 kV, probe current of 600 pA, and a pulse length of 20 ns at 1 MHz repetition rate were used. The temperature was varied from 4 to 300 K. The collected luminescence is sent to a 27-cm-focal-length grating monochromator equipped with a single-photon-counting system working with delayed coincidence in inverted mode, based on a cooled microchannel plate Hamamatsu photomultiplier.

C. Theoretical modeling

Spatial distribution of strain tensor components ε_{ij} was modeled using a three-dimensional (3D) continuum mechanical model and calculated using a finite element method [18,19]. The effects that arise due to the presence of spontaneous and piezoelectric polarization were modeled assuming that piezoelectric polarization depends linearly on strain tensor components. Electrostatic potential ϕ originating from polarizationinduced charges was derived by solving the corresponding 3D Poisson equation. Input geometry of the structure was obtained from TEM measurements, while In content in InGaN regions was estimated from EDS measurements. Material parameters were taken from Vurgaftman et al. except for the piezoelectric constants e_{15} which have been taken from Tomić et al. [20,21]. The corresponding parameters of InGaN alloy were estimated relying on a virtual crystal approximation, making use of a quadratic Vegard approximation, in the case of energy gap and spontaneous polarization, and a linear Vegard approximation for the other parameters (conduction and valence band deformation potentials, lattice constants, elastic constants, piezoelectric constants, and valence band offset between InGaN and GaN). Finally, to estimate the effects of quantum confinement on electron/hole energies (in the polar InGaN section), a one-dimensional (1D) Schrödinger equation has been solved (further details regarding the theoretical methods and justification of their use are given in Sec. III C and the Supplemental Material [14]).

III. RESULTS

A. Effects of growth-related anisotropies: shape, composition, and strain

Figure 1(a) shows a top SEM view of a representative sample area, whereas Fig. 1(b) shows a BF STEM micrograph of a representative NW (sample B). Figure 1(a) confirms ordered NW growth, with highly uniform diameters and

crystallographic tilts [22], while the TEM micrograph 1(b) reveals the shape and size of each of the three NW building blocks.

An initial GaN NW [Fig. 1(b)], shows a truncated pyramidal top, built out of a polar c facet and six semipolar r facets (c-6r shape). During the second growth step, the InGaN nanodisk grows basically on the polar c and semipolar r facets, with a negligible radial growth (on the nonpolar m facets). When starting the second growth step (InGaN nanodisk) the growth temperature and III/N ratio are both significantly reduced producing a change in the energetically preferential form of the NW top, which changes to c-6s shape [Fig. 1(b)]. During the third step (GaN cap), the growth temperature is kept unchanged whereas the III/N ratio is further slightly reduced. This slight change does not affect the preferential form of the NW top, as seen in Fig. 1(b). HAADF STEM images with atomic resolution reveal that NWs crystallize in a common wurtzite form and that they are free of extended defects [Fig. 1(c)] [22]. Both polar and semipolar intracolumnar interfaces are found to be atomically flat and free of extended defects, their absence indicating the presence of strain.

Figure 2 shows a chemical mapping of the tip of a representative NW (sample B) by EDS. In the EDS experiment, a single lying NW is probed by a high-energy electron beam. The *e* beam penetrates perpendicularly to the growth direction and excites the NW tip at different heights [Fig. 2(a)]. Figure 2(b) shows the spatial distribution of the In- $L\alpha_1$ line intensity. When the NW is probed along the central axis, a strong variation in In- $L\alpha_1$ line intensity is observed [Fig. 2(c)]. The probing at different heights, denominated as I, II, and III, allows retrieving information about In content in each of the building blocks. A moderate intensity of In- $L\alpha_1$ spectral line at height I (the spectrum taken just below the bottom InGaN/GaN polar interface: semipolar InGaN excited and polar InGaN not excited), indicates a moderate In content of the semipolar InGaN, estimated at ~10%. The In- $L\alpha_1$ line intensity rises significantly when polar InGaN gets excited. Probing at height II (the spectrum taken just below the top GaN/InGaN polar interface: semipolar InGaN not excited and polar InGaN excited), estimates the In content of the polar InGaN at ~20%. Finally, the In-L α_1 intensity drops to zero when GaN capping is probed (height III) indicating absence of In (for further details about EDS mapping see the Supplemental Material [14]).

The strong difference of In contents in the semipolar and polar nanodisk sections leads to their different emission colors [7,8]. The scanning electron microscopy combined with cathodoluminescence (SEM CL) probing ~20 representative NWs at room temperature (sample C), reveals that the semipolar and polar InGaN exhibit violet and green luminescence, respectively. Figure 3(b) shows a direct nanoscale correlation of the annular dark-field (ADF) STEM image and the CL wavelength image (CLWI) of a single NW from sample (C) at T = 15 K. The CLWI exhibits a dominant emission peak centered at ~360 nm, attributed to the nearband-edge emission of the GaN NW. The InGaN nanodisk clearly shows two emission bands. The first one, centered at \sim 405 nm, originates from the semipolar section, whereas the second one, centered at \sim 505 nm, originates from the polar one.



FIG. 2. (a) A NW probing is performed along the central axis (red line) with the *e* beam penetrating perpendicularly to it (green arrow). Different probing heights (I, II, and III) have been used to assess different building blocks: (I) GaN-capping/semipolar-InGaN/core-GaN/semipolar-InGaN/GaN-capping/polar-InGaN/GaN-capping, and (III) GaN-capping only. (b) Spatial distribution of the In- $L\alpha_1$ line intensity yields information about In incorporation throughout the NW tip region. (c) EDS spectra taken at three heights, I, II, and III, confirm In presence in the InGaN nanodisk, exclusively, its content being significantly higher in the polar (~20%) vs semipolar (~10%) section.

The STEM, EDS, and CL analyses coherently point to a strong anisotropic character of the nanodisk shape and composition, as well as its (consequent) optical properties. The NW top changes from c-6r to c-6s shape (as a function of growth conditions). The nanodisk sections with different compositions are developed, further implying an inhomogeneous strain distribution. The composition/strain inhomogeneities exhibit a strong facet-related anisotropic character, directly linked

to the facet on which a specific nanodisk section has been grown.

B. Effects of electric-field anisotropy

Due to the lack of central symmetry of the wurtzite unit cell, the III-nitride heterointerfaces may host uncompensated



FIG. 3. (a) SEM image combined with monochromatic room temperature CL measurements, performed at 405 nm (violet) and 505 nm (green) wavelengths. (b) ADF STEM image of a single NW combined with a highly spatially resolved CL mapping (at 15 K). The images reveal that the green luminescence originates from the top-facet InGaN, and violet luminescence originates from side-facet InGaN, whereas the UV luminescence comes from the core GaN (the wavelength is color coded).

surface charges, which can induce internal electric fields [13,23]. More specifically,

(a) Polar heterointerfaces typically host uncompensated charge with a high surface density (σ_p) ,

(b) Semipolar heterointerfaces typically host uncompensated charge with moderate to low surface density (σ_{sp}),

(c) Nonpolar heterointerfaces typically host no charge $(\sigma_{np} = 0 < \sigma_{sp} \ll \sigma_p)$.

The internal electric fields modify the conduction and valence bands' potential profile that (as in this work) may yield a significant e-h spatial separation (i.e., a reduction of their wave functions overlap). This separation reduces the e-h recombination energy (leading to emission redshift) and slows down recombination dynamics (leading to a drop of emission intensity).

The influence of the internal electric field on the eh recombination energy and dynamics was assessed by macro-PL and TR CL measurements. Figure 4(a) shows PL spectra of the three samples under study that exhibit



FIG. 4. (a) Low-temperature PL (10 K) spectra show three intense peaks attributed to GaN, semipolar InGaN, and polar InGaN, respectively. (b) Power-dependent PL measurement shows that when the PL excitation intensity is increased two orders of magnitude, only the polar-InGaN emission blueshifts (around 30 meV). (c) TR CL measurements (\sim 5 K) reveal monoexponential and nonmonoexponential CL decays for semipolar and polar InGaN, respectively. (d) Temperature-dependent TR CL reveals significantly shorter initial decay time of semipolar vs polar InGaN. In addition, the initial decay times of the polar InGaN sections are strongly affected by the nanodisk thickness, being shortest in the case of sample A.

three intense emission peaks attributed to GaN, semipolar InGaN, and polar InGaN respectively. In the case of reference sample B, the semipolar InGaN PL emission is ultraviolet (\sim 384 nm) whereas polar InGaN PL emission is turquoise (\sim 488 nm). When the nominal thickness of the InGaN nanodisk is reduced (sample A), the semipolar InGaN emission blueshifts very slightly, whereas the polar InGaN emission blueshifts significantly, entering the blue range (\sim 465 nm). When the nanodisk In content is increased (sample C), both semipolar and polar InGaN emissions show strong redshift, entering the violet (\sim 405 nm) and green (\sim 505 nm) emission ranges, respectively [see also Fig. 3(a)].

Figure 4(b) shows power-dependent PL measurements of reference sample B. As PL excitation power increases from 0.1 to 10 mW, the PL emission from polar InGaN blueshifts by 30 meV while the PL emission from semipolar InGaN remains unaffected. The observed blueshift points to an electric field screening by excited photocarriers in polar InGaN. These results point to low (high) electric field on the semipolar (polar) intracolumnar interfaces, respectively.

Figure 4(c) summarizes TR CL data from reference sample B. The CL decay from the semipolar InGaN is found monoexponential and fast, as compared to the nonmonoexponential and less fast decay from the polar section (initial time decays are ~ 0.6 vs ~ 1.5 ns, respectively) [24,25]. In addition, as the photocarrier population dies out, the CL decay time progresively increases, in the polar InGaN case. The possible origins of this behavior are addressed in the Discussion section.

Figure 4(d) shows the initial CL decay time dependence on temperature over the 5–300 K range. The recombination dynamics is found barely influenced by the nanodisk thickness in the semipolar InGaN. In polar InGaN the recombination dynamics is found fastest in thiner nanodisks (sample A). This latter result is attributed to a lower *e*-*h* separation in sample A and is addressed in detail in the Discussion section.

Both macro-PL and TR CL measurements evidence the presence of an internal electric field (*e-h* separation) in the polar section of the InGaN nanodisks. In contrast, no significant electric field has been traced in the semipolar sections. Comparing the emission intensity and recombination dynamics in the polar section with those from the semipolar one (at 10 K where nonradiative recombination centers can be considered inactive), the following conclusions can be drawn. The *e-h* recombination which drives the polar InGaN emission is

(i) About an order of magnitude less intense than the reference e-h transition which drives the semipolar InGaN emission [8];

(ii) Around a half order of magnitude slower than the reference *e*-*h* transition in the semipolar InGaN.

C. Theoretical modeling

Theoretical simulations were performed to help the interpretation of experimental results, taking into account the following facet-related anisotropies:

(1) Composition: different In content in the polar (20%) and semipolar (10%) sections (as quantified by EDS);

(2) Strain: different strain in the polar and semipolar sections, a consequence of the preferentially elastic strain

relaxation (due to the lack of extended defects, as observed by HAADF STEM) and different compositions (as quantified by EDS);

(3) Electric field: the uncompensated surface charge at polar c and semipolar r and s interfaces (as evidenced by PL and TR CL).

Before a detailed explanation of the theoretical methods is given, let us note that, due to its complexity, the assumption related to the nanodisk composition issue (1), requires additional comments. The In distribution across the InGaN nanodisk volume can be governed by three different mechanisms, all of which have been widely addressed (both theoretically and experimentally) in the literature. The three mechanisms are

(1.1) Facet-dependent In incorporation, the main driving mechanism of which is different (chemical) properties of dangling bonds on polar vs semipolar NW facets [11,12]. The influence of this mechanism is directly experimentally observed by TEM EDS technique (Figs. 1 and 2) and the effect is fully taken into consideration in theoretical simulations presented in this section.

(1.2) Height-dependent In incorporation (in strained nanostructures), the main driving mechanism of which is the total energy minimization of the growing nanostructure. High compressive strain at the InGaN/GaN interface provokes the In atoms "to be expelled" from the crystal lattice toward the less strained nanostructure regions (i.e., an apparent "upwards lattice pulling") [26,27]. The influence of this effect (on both two-color emission and electron-hole recombination dynamics) is additionally included and elaborated in the Discussion section.

(1.3) Random In fluctuations, the main driving mechanism of which is the spinodal decomposition of the ternary InGaN material [28,29]. Unlike (1.1) and (1.2), this mechanism has no directional preference. The relevance of this mechanism for the phenomena under study is addressed in the Discussion section.

The strain, derived via continuum 3D mechanical modeling (see Supplemental Material [14]), and the electric field distribution, derived from solving the 3D Poisson equation (see Supplemental Material [14]), across the NW volume (sample B) are presented in Figs. 5(a) and 5(b). As expected, the strain in the InGaN nanodisk is found compressive and larger in the polar section, due to a higher In content [Fig. 5(a)]. The intensity of the internal electric field is stronger close to the nanodisk polar interfaces, where the density of uncompensated surface charges is highest [Fig. 5(b)].



FIG. 5. Theoretical simulations for sample B. (a) Hydrostatic strain $\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}$ (left) at the NW cross section and (right) along the NW central axis (green arrow). (b) Electric field intensity (left) at the NW cross section and (right) along the NW central axis (orange arrow). (c) Conduction and valence band edge energies at the NW cross section. (d) Band-gap profiles in the semipolar and polar nanodisk sections, along the trajectories designated in (c).

Information on composition X_{In} , strain $\varepsilon_{ij} = \varepsilon_{ij}(x, y, z)$ and electrostatic potential $\phi = \phi(x, y, z)$ across the InGaN volume allows estimating the spatial distribution of the conduction $[E_c = E_c(x, y, z)]$ and valence $[E_v = E_v(x, y, z)]$ band-edge energies:

$$E_c = E_{c0} + f_c(\varepsilon) - |e|\phi, \qquad (1a)$$

$$E_v = E_{v0} + f_v(\varepsilon) - |e|\phi.$$
(1b)

 E_{c0} and E_{v0} are the unstrained bulk material conduction and valence band energies, $f_c(\varepsilon)$ and $f_v(\varepsilon)$ are strain-induced changes of conduction and valence band edge energies (obtained by diagonalizing the relevant strain-dependent $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian, as described in detail in Supplemental Material [14]), and e is the elementary charge. Results are presented in Figs. 5(c) and 5(d).

Since characteristic dimensions of the nanostructure are of the order of 20-30 nm, i.e., much larger than the exciton Bohr radius in GaN (\sim 3 nm), one might expect that quantum confinement effects are not important in this structure. Note, however, that internal electric fields (effect 2) and heightdependent In distribution (effect 1.2), as well as random In fluctuation (effect 1.3) can create significant potential variation at the scale of exciton Bohr radius or smaller. We emphasize thus that, as for (2), its influence on quantum confinement has been fully considered in simulations presented in this section; to calculate quantum confinement energy for conduction band electrons, a 1D Schrödinger equation (with In_{0.2}Ga_{0.8}N effective mass) has been used (in the Supplemental Material we show that such an approach gives results that are as accurate as results that would be obtained from a three-dimensional Schrödinger equation or by explicitly taking into account the coupling between the conduction and valence band in the eight-band $\mathbf{k} \cdot \mathbf{p}$ model [14]). Concerning (1.2) and (1.3), their influences are not taken into consideration in the simulations presented in this section; however, they are additionally addressed in the Discussion section. Finally, it is also worth noticing that to calculate optical transitions energies, the effect of attractive interaction between an electron and a hole (exciton binding energy) was not considered. This is justified by the fact that exciton binding energy is of the order of 20-30 meV (for bulk GaN and InN) [30], having similar or even smaller values in InGaN quantum wells [31-33]. The value of exciton binding energy in the polar section is similar to its value in quantum wells, while its value in the semipolar section (where electric field is weak) is similar to the bulk value. Since both of these values are small its influence on transition wavelengths, in the present case, is estimated at 3-4 nm at most, and thus not found critical for the present study.

Theoretical calculations reveal strong differences in bandgap profiles across semipolar and polar nanodisk sections [Figs. 5(c) and 5(d)]. First, the local band gap inside the polar section is found significantly reduced mainly as a consequence of enhanced In incorporation on polar *c* facets [11,12]. Second, a strong conduction/valence band bending is found at the edges (at the top and bottom heterointerfaces) of the polar InGaN section. This effect is barely observed in the semipolar InGaN section, where the bands are rather flat. These results are attributed to a high anisotropy in the internal electric field [see Fig. 5(b)], having its origin in a high (low) density of uncompensated surface charge at the polar (semipolar) intracolumnar heterointerfaces [13].

IV. DISCUSSION

A. Identification of dominant transitions

The flat band profile in the semipolar nanodisk section leads to a relatively uniform spatial distribution of generated photocarriers. Consequently this leads to a weak *e*-*h* spatial separation (i.e., strong wave-function overlap), in agreement with experimental data. To calculate the *e*-*h* recombination energy which corresponds to the dominant emission line of the semipolar InGaN section, the value of the local band gap is considered [see Fig. 5(d), left]:

$$E_1 = E_{1c} - E_{1v}.$$
 (2)

On the other hand, the band profile bending in the InGaN polar section leads to a nonuniform carrier distribution and a consequent e-h separation. The e-h photocarriers, generated over the entire polar InGaN volume, may get rapidly separated according to the sign of their charge; electrons drift toward the conduction band minimum located on the upper side of the polar section (top heterointerface), while the holes drift toward the valence band maximum located at the bottom part of this section (bottom heterointerface); therefore, both electrons and holes may eventually get confined in triangularlike potential wells (TPWs) created by the strong electric fields.

The identification of the *e-h* transition corresponding to the dominant emission line is less straightforward in the case of polar InGaN. Both power-dependent PL and TR CL measurements reveal screening and descreening effects, pointing out that carriers (electrons and/or holes) confined by the internal electric fields are involved in the recombination. However, due to the quite high spatial separation (several tens of nanometers), the emission of the polar section cannot be driven by recombination of the electrons and holes confined in TPWs; this transition would yield a very weak emission and a very slow recombination, which is not experimentally observed (this issue is discussed in detail in Sec. IV C).

Due to technical limitations of EDS technique for the geometry under study, the theoretical modeling presented in the previous section did not take into consideration the influence of lattice pulling on In distribution. In the EDS TEM experiment the NW is probed by a beam of high-energy electrons which penetrates the entire structure, yielding the average "weight" of In, Ga, and N atoms across the probed volume (for details see the Supplemental Material [14]). Due to coexistence of polar and semipolar InGaN, across the probing volume, the influence of lattice pulling on In distribution cannot be precisely determined, since due to pencil-like geometry, it is "screened" by the stronger "facetdependent" effect. Note that unlike internal electric field, which draws electrons and holes in opposite directions, the increasing In content toward the nanodisk top draws both types of photocarriers upwards. In this process, due to positive uncompensated charge at the top polar interface, the electrons are getting additionally accelerated until they eventually get confined in the upper TPW. The holes, on the other hand, are getting progressively decelerated until they stop midway between the upper and lower heterointerfaces. The coexistence



FIG. 6. CB and VB profiles along the direction shown in Fig. 5(c). (Solid line) pulling effect considered, assuming linearly graded InGaN in the polar section: $X_{In} = 0\%-20\%$. (Dotted line) pulling effect not considered, assuming uniform InGaN composition in the polar section: $X_{In} = 20\%$.

of strong internal electric field (drawing holes toward the bottom heterointerface) and composition gradient (drawing them in the opposite direction) leads to a formation of an additional (in a first approximation) paraboliclike potential well (PPW). This well, placed in the upper part of the polar section, acts as a reservoir of confined holes, for *e*-*h* recombination. Finally, we attribute the polar InGaN emission to the recombination between electrons confined in the upper TPW with holes confined in the PPW (as depicted in Fig. 6). Due to the lack of precise information about lattice-pulling-related In distribution, the energy of this transition can only be approximated.

Figure 6 shows conduction band (CB) and valence band (VB) profiles, across its central axis at the NW upper part [see also Fig. 5(c) for, clarity]. The comparative profiles are shown for (i) linearly graded polar section: $X_{In} = 0\%-20\%$ (solid lines) and (ii) uniform polar section: $X_{In} = 20\%$ (dotted lines). Note, first, that (even for the most extreme case corresponding to zero initial In content) the graded composition introduces only a very slight perturbation of VB potential in the upper part of the polar section ($\Delta E_{2\nu}$), where the dominant

e-h recombination takes place. Second, the strong triangular potential (created by the uncompensated positive charge at the top heterointerface) is barely affected by the CB perturbation, leading to a virtually unchanged electron confinement (ΔE_e) in the upper TPW. Finally, the parabolic potential (PPW), which confines holes engaging in the dominant *e-h* transition, is much weaker than the triangular potential (TPW) confining electrons. The weaker confining potential, combined with the hole (vs electron) higher effective mass, leads to a significantly weaker hole (vs electron) confinement. It can be shown that the following relation holds: $\Delta E_h < \Delta E_{2v} < \Delta E_e$, allowing the estimation of the corresponding *e-h* recombination energy:

$$E_2 = (E_{2c-\min} - E_{2v}) - \Delta E_{2v} + \Delta E_h + \Delta E_e$$

$$\approx (E_{2c-\min} - E_{2v}) + \Delta E_e.$$
(3)

The exact energy for the dominant transition, taking lattice pulling into consideration, is quantified on the left-hand side of Eq. (3) and depicted in Fig. 6. The approximate energy for this transition, taking no lattice pulling into consideration, is quantified on the right-hand side of Eq. (3) and depicted in Fig. 5(d).

The confinement energy ΔE_e was estimated from an effective mass modeling solving the 1D Schrödinger equation (see Supplemental Material [14]) for the conduction band potential shown in Fig. 5(d), right. Making use of Eqs. (2) and (3) the wavelengths of the emissions from semipolar and polar InGaN sections are estimated. The results, found in good agreement with the experimental values, are summarized in Table I (see calculation 0).

B. Quantitative disentanglement of the three contributing anisotropies

The wavelength differences between observed emission bands are driven by (i) composition, (ii) strain, and (iii) electric-field inhomogeneities, all of which show strong facetrelated anisotropic character. The high separation (\sim 100 nm) measured in the studied case is a joint result of all of them. The three effects are not independent and, consequently, not additive. Nevertheless, to roughly disentangle their influences and estimate relative weights of their contributions, three additional calculations (I–III) have been performed. In each of them, one of the contributing factors was "turned off" in the following way:

TABLE I. Emission wavelengths from the semipolar and polar InGaN nanodisk sections, determined experimentally (by PL) and theoretically (calculation 0). Emission wavelengths obtained from additional calculations (I, II, and III), aimed to quantitatively disentangle contributions of (I) composition, (II) strain, and (III) electric field anisotropies are also shown.

Sample B	Experiment PL (at 10 K)	Simulation			
		Calculation 0. Full calculation	Calculation I. Composition differences neglected: $X_{\text{In}} = 10\%$	Calculation II. Strain neglected: $\varepsilon = 0$	Calculation III. Electric field neglected: $\vec{E} = 0$
Semipolar InGaN	384 nm	385 nm	385 nm	399 nm	397 nm
Polar InGaN	488 nm	474 nm	395 nm	525 nm	427 nm
Wavelength separation	104 nm	89 nm	10 nm	126 nm	30 nm
Contribution to separation		0 nm	+79 nm	-37 nm	+59 nm

(i) Composition inhomogeneity turned off: assuming the same homogeneous In content ($X_{In} = 10\%$) in both polar and semipolar InGaN sections. Mathematically, the same values of unstrained conduction band E_{c0} and valence band E_{v0} edge energies were ascribed to the two sections in the expression for band edge energies [Eq. (1)].

(ii) Strain anisotropy turned off: mathematically, the $f_c(\varepsilon)$ and $f_v(\varepsilon)$ terms were excluded from the expression for band edge energies [Eq. (1)]. Nevertheless, the effect of strain on piezoelectric polarization remains included.

(iii) Electric field anisotropy turned off: setting the electric field to zero ($\vec{E} = 0$) across the entire nanodisk volume. Mathematically, the $|e|\phi$ term was excluded from the expression for band edge energies [Eq. (1)].

Note first that among the three contributing effects, a higher In composition and a higher internal electric field across the polar nanodisk section lead to a redshifted emission. A higher (compressive) strain produces an opposite effect, leading to a blueshifted emission. Second, among the three contributing factors, the contribution from composition inhomogeneity is found significantly higher than those from the other two anisotropies; when the former contribution is neglected, the giant emission redshift from the polar section (originally 89 nm, from Calculation 0) is practically eliminated (10 nm, from Calculation I). This remaining redshift is the joint result of strain and electric field anisotropy contributions that act in opposite directions, being that the influence of electric field significantly stronger (the same qualitative conclusion is obtained by Calculations II and III, which estimate the two contributions as -37 and +59 nm, respectively).

These results introduce a clear hierarchy among the three effects. The multicolor emission is mainly driven by composition inhomogeneity. The influence of electric field anisotropy is the next most significant one. Finally, the influence of strain anisotropy is the least significant one. The strain acts in the opposite direction, leading to a slight compensation of the emission wavelength separation, induced jointly by composition and electric field anisotropies.

C. Final remarks about dominant mechanisms which drive the nanodisk two-color emission and recombination dynamics: Comparison of experimental and theoretical findings

1. Two-color emission

Experimental results obtained by SEM CL, Fig. 3 (and also by micro-PL performed on few NWs; see our separate publication by Chernysheva *et al.*) [29], clearly reveal a "high stability" of nanodisk emission colors across sample surface; there is virtually no shift of the two emission bands (from one NW to another). Concerning nanodisk geometrical parameters, the SAG growth leads to (0) a highly predictable NW shape, which is systematically found as pencil-like [i.e., one polar and six semipolar top facets; see Fig. 1(a)]. However, SEM inspections reveal that the underlying GaN NWs (building block I; for details see Gačević *et al.*) [22] can have significantly varying size of the top polar facet (the NWs can look "more or less sharp"), which inevitably leads to (i) variations in InGaN nanodisk sections' sizes (from one NW to another). Extended (intra-NW) defects, which can lead to (ii) a significant strain

reduction at intracolumnar interfaces (see Urban et al.) can also sporadically appear [34]. Finally, sporadic appearance of (iii) random In fluctuations (see Chernysheva et al.) [29], which can lead to a formation of QD-like states (intradisk localization), can also be expected. Unlike effect 0, the effects i-iii are all far beyond the controllability of the SAG process. High stability of the nanodisk emission colors [from one NW to another; see Fig. 3(a)] thus strongly suggests that the type of involved facet (which directly determines the nanodisk section composition and electric field) has much stronger influence on emission color (effect 0) than quantum confinement due to the varying section size (effect i) and/or conduction/valence band perturbation due to the varying grade of plastic/elastic intracolumnar strain relaxation (effect ii). These experimental findings are in excellent qualitative agreement with theoretical findings presented in Table I.

Despite the well-known InGaN proneness to composition fluctuations (effect iii), leading to an intradot, intradisk and/or intrawell localization [29,35,36], experimental evidence in the present case strongly suggests that these random fluctuations cannot be a determining factor for nanodisk emission colors, either. This result can be qualitatively explained by micro-PL measurements (performed on few NWs, and reported in our separate publication; see Chernysheva et al.) [29]. According to micro-PL evidence, the random intradisk In fluctuations lead to shallow QD-like states (typically depopulated <80 K) characterized by exceptionally low integrated intensity vs integrated intensity of the polar and semipolar emission bands [29], and thus neglected in the present study. These QD-like states are much more commonly observed in polar (vs semipolar) InGaN, the result which strongly suggests significantly stronger potential fluctuation ("potential landscape") in polar vs semipolar InGaN [29]. This result is in good agreement with reports arguing that random potential fluctuations are stronger for polar (vs semipolar) InGaN and for higher (vs lower) In content (20% polar InGaN vs 10% semipolar InGaN, in the present case) [37]. Consequently, we point out that it is very likely that the hereby applied simulation methods relying on virtual crystal approximation are somewhat better suited for semipolar (vs polar) InGaN. To precisely quantify the influence of potential landscape on dominant transitions, full 3D atomistic simulations (taking into account facet-related In incorporation, lattice-pulling In distribution, and random alloy fluctuations) are desired [38-40].

2. Recombination dynamics

The fast recombination in the semipolar InGaN is attributed to a high e-h wave-function overlap, a consequence of low electric field across this section. The slower recombination in the polar InGaN is attributed to the existence of a strong electric field in the proximity of polar intracolumnar interfaces, which consequently leads to a certain e-h separation. As experimentally observed by many authors [26,27], the emission from ternary nanostructures mainly originates from its top part; that is, both electrons and holes migrate upwards. This particular result is attributed to the gradually increasing In content along the growth direction (lattice pulling). While the sole effect of electric field anisotropy cannot fully explain recombination dynamics features in Figs. 4(c) and 4(d), its coexistence with the lattice-pulling effect can give a good qualitative explanation. We identify the lattice pulling as the mechanism which keeps electrons and holes in relative proximity (contrary to electric field which tends to separate them), enabling their relatively fast and efficient recombination. If there was no vertical In gradient across the nanodisk polar section, then nothing would prevent electrons/holes from migrating toward its top/bottom parts, leading to a giant electron-hole separation (of the order of 30 nm in the present case). That would finally lead to extremely slow and weak polar InGaN emission which is not experimentally observed.

Coexistence of electric field anisotropy with lattice pulling can also qualitatively explain results presented in Figs. 4(c) and 4(d). The CL decay is inversely related to e-h separation. This separation, induced by internal electric field (created jointly by uncompensated intracolumnar interface charge and photogenerated carriers) is counterbalanced by the lattice-pulling effect (driven by strain). As the population of photogenerated carriers dies out (upon excitation), the total internal electric field gradually increases, increasing thus the e-h spatial separation. This finally leads to progressively increasing decay time, (only) in the case of polar InGaN [see Fig. 4(c)], as experimentally observed. Similarly, thinner InGaN nanodisk geometry (sample C) leads to a higher strain, that is, stronger lattice-pulling effect. As for e-h separation, this leads to "more efficient counterbalancing" to electric-field-induced separation, leading to faster CL decay, as experimentally observed.

The coexistence of strong internal electric field and lattice pulling can (solely), in a first approximation, explain nonmonoexponential (vs monoexponential) CL decay in the polar (vs semipolar) section. Nevertheless, Morel *et al.* suggested the InGaN potential landscape (the origin of which is random In fluctuation) as the main origin for nonmonoexponential decay, in the case of polar InGaN quantum wells [25]. This interpretation could also explain the different trends observed for polar and semipolar InGaN in the present case. To determine the origin(s) leading to these different trends with certainty, further experimental analyses accompanied with full 3D atomistic simulations is desired.

V. CONCLUSION

A detailed experimental and theoretical insight into the phenomenon of two-color emission from InGaN nanodisks, grown on site-controlled pencil-like GaN NWs, has been performed. The In composition is found significantly higher in the polar (\sim 20%) vs semipolar (\sim 10%) nanodisk sections, as quantified by energy dispersive x-ray spectroscopy. This facet-related inhomogeneity leads to a highly anisotropic strain distribution across the nanodisk volume. While the

spatially resolved CL resolves different emission wavelengths (\sim 500 and \sim 400 nm, respectively), the TR CL reveals significantly different e-h recombination times (~1.3 and ~ 0.5 ns, respectively) in the polar and semipolar sections, respectively. The fast recombination in the semipolar InGaN is attributed to a virtually zero e-h separation, a consequence of low electric field across this section. The slower recombination in the polar InGaN is linked to the existence of strong electric field in the proximity of polar intracolumnar interfaces, which induces a certain e-h separation. To clearly determine the weight of the three contributing facet-related anisotropies (composition, strain, and electric field) on the emission wavelength difference, proper theoretical simulations relying on virtual crystal approximation (involving 3D continuum mechanical modeling, a 3D Poisson equation, and a 1D Schrödinger equation) have been performed. The theoretical simulations allow the effects of the three anisotropies to be quantitatively disentangled. The composition inhomogeneity has been identified as the dominant factor, whereas the strain anisotropy has been identified as the least significant one. Finally, concerning recombination dynamics, we suggest that the lattice-pulling effect very likely partially counterbalances the e-h separation, as induced by internal electric field. The coexistence of two effects keeps electrons and holes in (relative) proximity, providing thus a moderately fast and efficient recombination. All theoretical findings are found in good agreement with vast experimental evidence gathered by numerous experimental techniques. This work establishes a base to understand and interpret cases of multicolor emission from III-N nanostructures, built up of different facets and having different geometries.

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I. METHODS

Colloidal lithography. For nanohole masks preparation a commercial colloidal solution, consisting of polyestirene (PS) nanobeads was used. The nanobeads average diameter is ~270 nm and they are sulfate covered i.e. negatively charged. Previous to the lithographic process, the GaN templates were alternately dipped in poly(sodium4-styr- enesolfonate) (PSS) and poly(diallyldimethylammoniumchloride) (PDDA) solutions (2% w/v, during 1 min each step), which are negatively and positively charged poly-electrolytes, respectively. A tri-layer PSS/PDDA/PSS (5 nm thick) is formed on the substrate surface to achieve a negatively charged hydrophilic surface. The substrate surface is then covered with the colloidal solution through a spin-coating (2000 rpm for 30 s). The result is a monolayer of PS nanobeads, with a close-packaged arrangement in large areas (domains up to $400 \times 400 \ \mu\text{m}^2$). Once the nanobeads were arranged on the surface, reactive ion etching oxygen plasma (dissociation power: 20 W, oxygen pressure: 10 mTorr and etching time: 120 sec) was used to reduce the diameter of the beads to ~180 nm and, consequently, to isolate them. After the plasma etching, 7 nm of Ti was evaporated on the surface. Finally, in the lift-off process the colloidal nanobeads are removed, leaving behind a hexagonal 2D array of nanoholes (diameter ~180 nm) on the Ti layer covering the GaN substrate surface [15].

Growth. The samples were grown in a RIBER Compact 21 Molecular Beam Epitaxy system, equipped with a radio-frequency plasma nitrogen source and standard Knudsen cells, one for In and two for Ga. All growths were performed on ~3.3 µm thick commercial GaN-on-sapphire(0001) templates grown by metal organic chemical vapor deposition. For the growth, the following fluxes (Φ) and growth temperatures (T_g) were used: (step I) $\Phi_N = 4$ nm/min, $\Phi_{Ga} = 15$ nm/min and Tg = 860 °C; (step II) $\Phi_N = 8$ nm/min, $\Phi_{Ga} = 5$ nm/min, $\Phi_{In} = 2.5$ nm/min and Tg = 610 - 625°C; and step (III) $\Phi_N = 8$ nm/min, $\Phi_{Ga} = 5$ nm/min and Tg = 610 - 625°C.

II. INDIUM CONTENT DETERMINATION

The In content of the InGaN nanodisk was estimated through two-dimensional EDS chemical maps obtained at the NW tip. In all measurements the studied NW was laid onto the surface, as shown in Fig. S1. The e-beam, with subnanometric cross-section, penetrates the NW tip along the [10-10] crystal direction, at different points. In the experiment, the e-beam height (h_I or h_{II} in figure S1) and e-beam radial position (from 0 to r3 in figure S1) were varied. The analysis at height I retrieves information about In composition of semipolar nanodisk section, whereas the analysis at height II retrieves information about the polar one. The NW cross-sections at the two designated heights are essentially different. At height I, the NW cross-section contains: hexagonal GaN core, hexagonal InGaN ring (semipolar InGaN), and hexagonal GaN ring (capping) (see figure S1 (I) for clarity). At height II, the NW contains: hexagonal InGaN core (polar InGaN) and hexagonal GaN ring (capping) (see figure S1 (II) for clarity).



FIG. S1. A schematic view of the e-beam probing a single NW. In experiment, the e-beam position is varied in: height (I or II) and radial distance (from 0 to r3) At different heights, the NW shows different cross-sections. (I) At height I, the semipolar InGaN has a form of a regular hexagonal ring. (II) At height II, the polar InGaN has a form of a regular hexagon.

The EDS technique retrieves average atomic In fraction across the probed volume (V_{total}). Consequently, the experimentally measured In content ($c_{In-meas}$) is a function of both content ($c_{In-REAL}$) and volume (V_{InGaN}) of the probed InGaN subsection:

$$c_{In-meas} = c_{In-REAL} \frac{V_{InGaN}}{V_{total}}$$
(S1)

As can be seen in figure S1 (I and II), the volume of the probed InGaN subsection varies as a function of e-beam spatial position.

The atomic In fraction, measured at the two heights (I and II) over the full radial range, is summarized in figure S2. The In content shows symmetrical variation along the radial direction in both cases, as expected.



FIG. S2. Measured In contents at two different heights (I: semipolar InGaN, and II: polar InGaN) along the full radial range. The observed variation in the In content is due to the hexagonal geometry of the system. Real In content of the nanodisk sections is determined after applying the corresponding geometrical correction coefficients.

In order to determine the real composition of side and middle nanodisk sections, equation S1 has been used. The geometrical corrections $\left(\frac{V_{InGaN}}{V_{total}}\right)$ are estimated from the TEM measurements. The two values, calculated at the central axis (zero radial distance) at heights I and II, are shown in table S1. The real In contents of side- and middle-nanodisk sections are finally estimated at: 9.95% and 19.68%, respectively.

Table S1. Correction coefficients, measured In content and real In content of side- and middle nanodisk sections. The three radii featured in figures S1 and S2 have respective values of: r1 = 25nm, r2 = 55 nm and r3 = 75 nm.

Nanodisk section	e-beam position: radial distance / height	V _{InGaN} V _{total}	c _{In-meas} (%)	с_{In-REAL} (%)
Semipolar InGaN	0 / h _i	$\frac{r_2 - r_1}{r_3} = 0.4$	3.98	9.95
Polar InGaN	0 / h _{ii}	$\frac{r_1}{r_2}$ = 0.45	8.86	19.68

III. THEORETICAL MODELING

Continuum mechanical model (3D). Strain distribution in the nanostructure was modeled using the continuum mechanical model. Within this model, elastic energy of the structure is given as

$$W = \frac{1}{2} \sum_{i,j=1}^{6} \int d^3 \boldsymbol{r} \, C_{ij}(\boldsymbol{r}) [\epsilon_i(\boldsymbol{r}) + \epsilon_i^0(\boldsymbol{r})] [\epsilon_j(\boldsymbol{r}) + \epsilon_j^0(\boldsymbol{r})], \tag{S2}$$

where C_{ij} are the components of the elastic tensor, which, in the case of wurtzite nitride materials, has the form

$$C = \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\ C_{12} & C_{11} & C_{13} & 0 & 0 & 0 \\ C_{13} & C_{13} & C_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & C_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & C_{66} \end{bmatrix}.$$
 (S3)

 ϵ_i are the components of the strain tensor

$$\epsilon = [\varepsilon_{xx} \quad \varepsilon_{yy} \quad \varepsilon_{zz} \quad 2\varepsilon_{yz} \quad 2\varepsilon_{xz} \quad 2\varepsilon_{xy}], \tag{S4}$$

with ε_{ab} defined as

$$\varepsilon_{ab} = \frac{1}{2} \left(\frac{\partial u_a}{\partial x_b} + \frac{\partial u_b}{\partial x_a} \right).$$
(S5)

 u_a is the displacement in direction a ($a \in \{x, y, z\}$) of an atom from the position it would exhibit if the lattice constant of the substrate and the nanostructure material were the same. ϵ_i^0 are the components of the intrinsic strain tensor defined as

$$\epsilon^{0} = \begin{bmatrix} \frac{a_{s} - a(\mathbf{r})}{a(\mathbf{r})} & \frac{a_{s} - a(\mathbf{r})}{a(\mathbf{r})} & \frac{c_{s} - c(\mathbf{r})}{c(\mathbf{r})} & 0 & 0 \end{bmatrix},$$
(S6)

where a_s and c_s are the lattice constants of the substrate, while a(r) and c(r) are the lattice constants of the unstrained nanostructure material at point r.

Spatial distribution of atomic displacements $u_a(r)$ and strain tensor components $\epsilon_i(r)$ was found by minimizing the elastic energy of the structure. Discretization of the integral given in Eq. (S2) was performed using the finite element method. Finite elements in the shape of the prism whose base is equilateral triangle were used. First order Lagrange interpolation was used to express the functions at arbitrary point in space in terms of its value at element nodes. After the discretization of the integral given in Eq. (S2) one obtains that the elastic energy of the structure W is a quadratic function of displacements at element nodes inside the nanostructure region. Minimization of that function leads to a system of linear equations which is solved numerically using the PETSc package [41].

Poisson equation (3D). Electrostatic potential $\phi(r)$ in the structure caused by polarization induced charges was found by solving the Poisson equation

$$\nabla[\varepsilon_0 \varepsilon_r(\mathbf{r}) \nabla \phi(\mathbf{r})] = -\rho_p(\mathbf{r}) \tag{S7}$$

where ε_0 is the vacuum permittivity, $\varepsilon_r(\mathbf{r})$ is the relative dielectric constant of the material and $\rho_p(\mathbf{r})$ is the charge density of the charges induced by spontaneous polarization $\mathbf{P}_{sp}(\mathbf{r})$ and piezoelectric polarization $\mathbf{P}_{pz}(\mathbf{r})$. Spontaneous polarization is a constant of a given material, while in wurtzite nitride semiconductors piezoelectric polarization components are related to strain tensor components as.

$$(\boldsymbol{P}_{pz})_{\chi} = e_{15}\epsilon_{5}$$

$$(\boldsymbol{P}_{pz})_{y} = e_{15}\epsilon_{4}$$

$$(S8)$$

 $(\mathbf{P}_{pz})_z = e_{31}(\epsilon_1 + \epsilon_2) + e_{33}\epsilon_3$ The density of the charge induced due to spontaneous and piezoelectric polarization is then given as

$$p_p(\mathbf{r}) = -\nabla \cdot \left[\mathbf{P}_{sp}(\mathbf{r}) + \mathbf{P}_{pz}(\mathbf{r}) \right]$$
(S9)

Poisson equation was solved by minimizing the following functional

$$W_{el} = \frac{1}{2} \int d^3 \boldsymbol{r} \,\varepsilon_0 \varepsilon_r(\boldsymbol{r}) (\nabla \phi) \cdot (\nabla \phi) - \int d^3 \boldsymbol{r} \,\rho_p(\boldsymbol{r}) \phi(\boldsymbol{r}). \tag{S10}$$

The same type of discretization of the integrals in Eq. (S10) as in the case of strain calculation was performed which leads to a linear system of equations for the values of the electrostatic potential at element nodes. This system of equations was solved numerically using the PETSc package [41].

8-band $k \cdot p$ Hamiltonian for wurtzite semiconductors. When spin-orbit effects are neglected (due to very small value of spin-orbit splitting in GaN and InGaN of only a few meV) this Hamiltonian reduces to two identical 4x4 blocks which read [42]: $H_{aut} = H_{but}^{k} + H_{aut}^{s}$

$$H_{4x4}^{k} = \begin{bmatrix} E_{c0} + \frac{\hbar^{2}k^{2}}{2m_{0}} & V & V^{*} & U \\ V^{*} & E_{v0} + F & K^{*} & H^{*} \\ V & K & E_{v0} + F & H \\ U & H & H^{*} & E_{v0} + \Lambda \end{bmatrix}$$
$$H_{4x4}^{s} = \begin{bmatrix} w_{1} & 0 & 0 & 0 \\ 0 & f & k^{*} & h^{*} \\ 0 & k & f & h \\ 0 & h & h^{*} & \lambda \end{bmatrix}$$

where

$$H = i \frac{\hbar^2}{2m_0} A'_6 k_z (k_x + ik_y)$$

$$K = \frac{\hbar^2}{2m_0} A'_5 (k_x^2 - k_y^2 + 2ik_x k_y)$$

$$\Lambda = -\Delta_1 + \frac{\hbar^2}{2m_0} [A'_1 k_z^2 + A'_2 (k_x^2 + k_y^2)]$$

$$\Theta = \frac{\hbar^2}{2m_0} [A'_3 k_z^2 + A'_4 (k_x^2 + k_y^2)]$$

$$F = \Delta_1 + \Lambda + \Theta$$

$$V = \frac{1}{\sqrt{2}} P_n (k_x + ik_y)$$

$$U = P_p k_z$$

$$w_1 = a_{c,xy} (\varepsilon_{xx} + \varepsilon_{yy}) + a_{c,z} \varepsilon_{zz}$$

$$\lambda = D_1 \varepsilon_{zz} + D_2 (\varepsilon_{xx} + \varepsilon_{yy})$$

$$\theta = D_3 \varepsilon_{zz} + D_4 (\varepsilon_{xx} + \varepsilon_{yy})$$

$$f = \lambda + \theta$$

$$k = D_5 (\varepsilon_{xx} - \varepsilon_{yy} + 2i\varepsilon_{xy})$$

In previous equations, E_{c0} and E_{v0} are the unstrained material conduction and valence band edge, Δ_1 is the crystal field splitting, P_p and P_n are the momentum matrix elements that couple the conduction and the valence

band and A'_1 - A'_6 are the parameters of the 8-band model which are related to usually reported parameters of the six band model A_1 - A_6 via

$$A'_{1} = A_{1} + \frac{2m_{0}}{\hbar^{2}} \frac{P_{p}^{2}}{E_{g}}$$

$$A'_{2} = A_{2}$$

$$A'_{3} = A_{3} - \frac{2m_{0}}{\hbar^{2}} \frac{P_{p}^{2}}{E_{g}}$$

$$A'_{4} = A_{4} + \frac{m_{0}}{\hbar^{2}} \frac{P_{n}^{2}}{E_{g}}$$

$$A'_{5} = A_{5} + \frac{m_{0}}{\hbar^{2}} \frac{P_{n}^{2}}{E_{g}}$$

$$A'_{6} = A_{6} + \frac{\sqrt{2}m_{0}}{\hbar^{2}} \frac{P_{p}P_{n}}{E_{g}}$$

 D_1 - D_6 are the valence band deformation potentials, while $a_{c,xy}$ and $a_{c,z}$ are the conduction band deformation potentials, while E_g is the band gap of unstrained material. $a_{c,xy}$ and $a_{c,z}$ are related to usually reported interband deformation potentials a_1 and a_2 via $a_1 = a_{c,z} - D_1$ and $a_2 = a_{c,xy} - D_2$ (see Yan et al [43]).

Strain contribution to band edge energies. The contributions of strain to conduction band-edge energy $f_c(\varepsilon)$ and to the valence band edge energy $f_v(\varepsilon)$ were calculated by diagonalizing the 8-band strain-dependent $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian for wurtzite nitride semiconductors at the Gamma point (k = 0). At the Gamma point (k = 0), the Hamiltonian reduces to

$$H_{4x4} = \begin{bmatrix} E_{c0} + w_1 & 0 & 0 & 0 \\ 0 & E_{v0} + f & k^* & h^* \\ 0 & k & E_{v0} + f & h \\ 0 & h & h^* & E_{v0} + \lambda - \Delta_1 \end{bmatrix}$$

The conduction band edge at the point of space (x, y, z) is then obtained as $E_c(x, y, z) = E_{c0} + f_c(\varepsilon)$
where
 $f_c(\varepsilon) = a_{c,xy}(\varepsilon_{xx} + \varepsilon_{yy}) + a_{c,z}\varepsilon_{zz}$,
while the valence band edge at that point reads
 $E_{v(x,y,z)} = E_{v0} + f_v(\varepsilon)$
where $f_v(\varepsilon)$ is the largest eigenvalue of the 3x3 matrix
 $\begin{bmatrix} f & k^* & h^* \\ k & f & h \\ h & h^* & \lambda - \Delta_1 \end{bmatrix}$

Schrödinger equation (1D). To estimate the effects of quantum confinement on the energies of electrons in the top part of polar InGaN, we have solved an one dimensional Schrödinger equation

$$\left[-\frac{\hbar^2}{2m_{eff}}\frac{\mathrm{d}^2}{\mathrm{d}z^2} + V(z)\right]\psi(z) = E\,\psi(z),\tag{S11}$$

where m_{eff} is the InGaN effective mass, V(z) is the conduction band energy on the *z*-axis (presented in Fig. 5(d) right, of the main article), $\psi(z)$ is the wave function of the electron and *E* its energy. The confinement energy ΔE_e was then calculated as $\Delta E_e = E_0 - E_{2c-min}$, where E_0 is the lowest eigenenergy obtained by solving Eq. (S11) and E_{2c-min} is the conduction band edge energy at the top of the polar InGaN, where it is minimal. Confinement energies in other two directions were neglected since wave function localization length in these directions is much

larger than in the z-direction, leading to negligible confinement energies. In what follows, we provide the justifications for: (a) the use of single band effective mass model instead of more elaborate multiband $\mathbf{k} \cdot \mathbf{p}$ model; (b) the use of 1D model instead of fully 3D model; (c) the use of effective mass of $\ln_{0.2}$ Ga_{0.8}N instead of position dependent effective mass.

(a) Justification for the use of single band effective mass model. The use of single band effective mass model is justified by the fact that the effective mass model gives a rather accurate dispersion within first 200 meV from the bottom of conduction band. In Fig. S3 we compare the conduction band dispersion of GaN obtained within the effective mass model and within more elaborate 8-band $\mathbf{k} \cdot \mathbf{p}$ model. The energy differences between two models are less than 15 meV in the spectral region within first 200 meV from the bottom of conduction band.



FIG. S3. Comparison of the conduction band dispersion of unstrained GaN obtained within the effective mass model and within the 8-band $k \cdot p$ model

Because of small differences between bulk dispersion obtained with the two methods, it is expected that the energy of the quantum well ground state is as accurate in the effective mass method as in the 8-band $\mathbf{k} \cdot \mathbf{p}$ model. To check that this is the case, using the two methods we calculate the ground state energy of the quantum well, in the potential $V(x = 0, y = 0, z) = E_{c0} - |e|\phi(x = 0, y = 0, z)$, see Fig S4. Using effective mass model we obtain the ground state energy of -739.1 meV, while using the $\mathbf{k} \cdot \mathbf{p}$ model we obtain -738.4 meV, suggesting that a single band model is sufficient to get accurate energy of the ground conduction band state.



FIG. S4. The potential on z-axis of the structure near the top of middle InGaN region and the wave function moduli squared of two lowest quantum well states.

(b) Justification for the use of 1D (instead of 3D) Schrödinger equation. Since the wave function localization lengths, in the *x* and *y* directions, are much larger than the localization length in the *z* direction, it is expected that ground state energy in the conduction band can be accurately obtained by solving the 1D instead of 3D Schrödinger equation. To check if this is the case, we have compared the results obtained by solving the 1D Schrödinger equation in the potential $V(x = 0, y = 0, z) = E_{c0} - |e|\phi(x = 0, y = 0, z)$ and the results obtained by solving the 3D Schrödinger equation in the potential $V(x = 0, y = 0, z) = E_{c0} - |e|\phi(x, y, z) = E_{c0} - |e|\phi(x, y, z)$. We obtain, respectively, the ground state energy of -739 meV and -722 meV, suggesting that 1D approach introduces an error less than 20 meV for transition energies, which affects the transition wavelengths by less than 3 nm.

(c) Justification for the use of constant, rather than position dependent effective mass. Due to the facts that: (i) effective masses of $In_{0.2}Ga_{0.8}N$ and GaN are rather similar ($0.174 m_0$ versus $0.2 m_0$) and (ii) ground state conduction band wave function is predominantly localized in $In_{0.2}Ga_{0.8}N$ region, it is expected that the model with constant $In_{0.2}Ga_{0.8}N$ effective mass gives essentially the same result as the model with position dependent effective mass; we find that the model with position dependent effective mass yields the ground state energy of -740.8 meV in excellent agreement with -739.1 meV obtained from the model with $In_{0.2}Ga_{0.8}N$ effective mass.