Sub-meV linewidth of excitonic luminescence in single GaN nanowires: Direct evidence for surface excitons

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We investigate the low-temperature photoluminescence of GaN nanowires grown catalyst free on Si(111). For single nanowires dispersed on Si(111), we observe excitonic transitions with linewidths below 300 μeV and at energies clearly above the donor-bound exciton in the bulk. We show that these transitions are due to donor-bound excitons close to the surface. The broadening of about 3 meV observed for the nanowire ensemble is shown to be a natural consequence of the energy dispersion of bound-exciton states as a function of their distance from the surface.

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I. INTRODUCTION

Heteroepitaxy is mainly constrained by the degree of structural mismatch between substrate and adsorbate.[1] Even if epitaxy prevails, the crystal quality of the adsorbate may be handicapped by defects generated to accommodate the structural differences between the two materials involved.[1] These defects have, in general, detrimental consequences for the optical and electronic properties of the epitaxial film. A particularly prominent example of this fact is the continuing effort to achieve an integration of compound semiconductors with Si technology (see Refs. 1 and 2, and references therein).

The synthesis of nanowires (NWs) (Refs. 3 and 4) is believed to overcome many of these limitations. In fact, the NW geometry inhibits the propagation of threading dislocations along the NW axis. Dislocations are confined at the interface between the substrate and the NW or bend to the sidewalls at the bottom of the NWs.[5] In addition, the free surface at the sidewalls permits an efficient elastic relaxation of strain.[6–8] As a result, the dislocations and lattice strain, which are inevitable in the epitaxy of mismatched materials, are believed to be absent in these nanostructures.[7,9]

For these reasons, II–VI and III–V NWs are believed to be among the most promising candidates for the monolithic integration of direct band-gap semiconductors onto Si.[2,9,10] In particular, the realization of GaN NWs on Si is thought to be a first but decisive step toward the realization of efficient light emitters[11–13] integrated with Si microelectronics.[2,10,14] However, despite the common belief that these structures are free of strain, they exhibit optical transitions which are significantly broader than those observed for a bulk crystal.

The low-temperature photoluminescence (PL) linewidth of bound-exciton transitions is one of the most sensitive measures for the presence of inhomogeneous strain within the crystal. The typical linewidth observed for these transitions in an essentially strain-free crystal is <100 μeV (see, for example, Ref. 15 for the case of GaN), which translates into a maximum inhomogeneous strain of $\pm 10^{-5}$ considering the typical deformation potentials of compound semiconductors. However, the narrowest linewidth reported so far for NWs, regardless of the materials system and whether being isolated or as ensemble, still exceeds 1 meV.[16–21] This fact has been largely ignored in the literature but in a recent investigation the unexpectedly large broadening has been recognized and attributed to residual strain.[22]

Here, we present a study of the low-temperature PL of GaN NWs grown catalyst free on Si(111). For single nanocolumns dispersed on Si(111), we observe excitonic transitions with linewidths below 300 μeV and provide direct evidence for the existence of surface excitons in these structures. The broadening observed for the NW ensemble is shown to be a direct consequence of the energy dispersion of bound-exciton states as a function of their distance from the surface.

II. EXPERIMENT

GaN NWs are grown directly on Si(111) substrates by plasma-assisted molecular-beam epitaxy (PAMBE). A detailed discussion of the substrate preparation and the growth of the NWs can be found elsewhere.[23] Briefly, the NWs are grown under extremely N-rich conditions (N/Ga=5) and at a substrate temperature of 780 °C, i.e., similar to the conditions reported by other groups.[16,24] The as-grown NW ensemble is transferred ultrasonically to 2-propanol and dispersed onto a Si(111) carrier for studying single, isolated NWs.

The NWs, both as ensemble and isolated, are imaged by scanning electron microscopy (SEM) and studied by macrophotoluminescence (MPL) and microphotoluminescence (μPL) spectroscopies. In either case, we use the 325 nm line of a Kimmon He-Cd laser for excitation. In MPL, the spot is about 75 μm in diameter, and the MPL signal is dispersed by a 1 m Jobin-Yvon double monochromator equipped with a cooled charge-coupled-device (CCD) detector. μPL measurements are carried out using a Cryovac microscope cryostat, with the laser focused to an about 3-μm-sized spot by a 15× microscope objective with a numerical aperture of 0.32. The μPL signal is collected by the same objective and analyzed by a 0.8 m Jobin-Yvon monochromator equipped with a CCD detector. The spectral resolution of this setup is about 250 μeV. All measurements are done at 10 K. The excitation density is given in units of $I_0=20$ kW/cm^2. Prior to all measurements, the samples are subjected to the *in situ* ozone cleaning described in Ref. 25.

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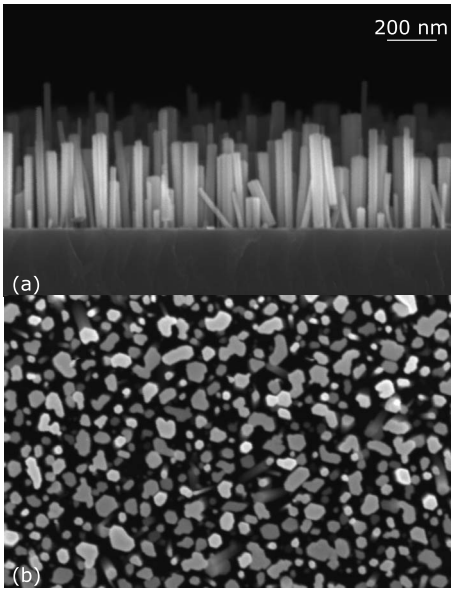

FIG. 1. (a) SEM side view and (b) SEM top view of the as-grown NW ensemble.

III. RESULTS AND DISCUSSION

Figure 1 shows a side (a) and a top view (b) of the as-grown NW ensemble, respectively. In (a), a high-density NW array with an average height of 350 nm is observed. In (b), the density of the NW array can be estimated to be higher than 10^{10} cm^{-2}. Furthermore, we see that a significant number of NWs has undergone coalescence with adjacent wires. As a result, individual NWs exhibit quite different morphologies, and their diameter varies between 20 and as much as 100 nm.

Figure 2 displays MPL spectra of the as-grown NW ensemble at 10 K and two different excitation densities. Three major near-gap transitions are observed. The dominant one at 3.472 eV has a linewidth of 2.7 meV and is attributed to the decay of the donor-bound exciton in strain-free GaN.[26–28] In

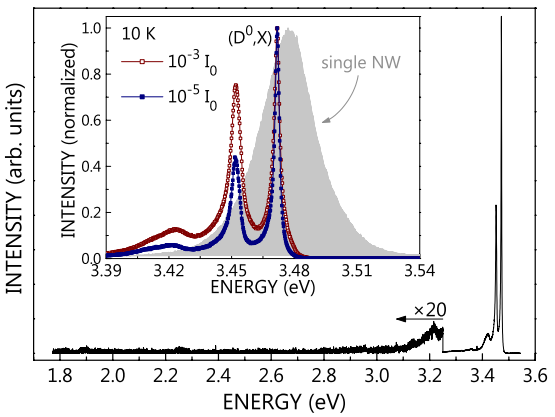

FIG. 2. (Color online) MPL spectrum of the NW ensemble displayed in Fig. 1 at 10 K and an excitation density of $10^{-5}I_0$. The inset shows an expanded view of the near-band-gap PL lines at two different excitation densities as indicated in the figure. The shaded area depicts a spectrum of a strained single NW for comparison.

μPL spectra, we observe a basically identical linewidth since the exciting spot still covers about 10^3 NWs. The two transitions at lower energy (3.422 and 3.450 eV) are characteristic for GaN NWs (Refs. 21 and 29) but more difficult to identify. These transitions are not observed for GaN layers grown in the same MBE system, suggesting that they are most likely not caused by impurities. Furthermore, these transitions scale linearly with excitation density (up to $10^{-1}I_0$) unlike the donor-bound exciton which starts to saturate in intensity at the highest excitation density shown here (see inset of Fig. 2). This finding is consistent with reports from the literature, where these transitions have been attributed to excitons bound to planar defects such as stacking faults and inversion domain boundaries.[29–33] which are generally not as easily saturable as excitons bound to point defects.[34] Note, however, that a linear dependence would also be observed for a point defect which is present in abundance, such as a surface defect.[21] Attempts to saturate these lower-energy lines were not successful, as they broaden and merge at excitation densities above $10^{-1}I_0$. This broadening is easily understood; the lack of carrier diffusion in these spatially restricted systems results in carrier densities that are beyond those attainable in planar structures.

Next, we have investigated single NWs dispersed on Si(111) as described in Sec. II. Each of these single NWs we have examined exhibits its own, individual PL spectrum. Most of them feature a PL band of a width (>20 meV) far exceeding that observed for the ensemble (cf. inset of Fig. 2). This line is usually blueshifted by about 10 meV compared to the donor-bound exciton transition of the ensemble (cf. inset of Fig. 2). These findings indicate that single NWs dispersed on Si(111) are frequently under inhomogeneous and overall compressive stress, caused by the interaction with the underlying substrate.[22]

However, among the dozens of single NWs we have investigated, we also found some which apparently did not suffer from adhesion-induced stress in that they display narrow, well-resolved transitions. In particular, the two-column agglomerate shown in Fig. 2 exhibits PL spectra which signify that this particular NW is free of strain.

In fact, while the spectrum of this selected single NW (Fig. 3) is dominated by the (presumably) stacking-fault-induced transition at 3.422 eV (i.e., at the same energy as in the ensemble), the bands at 3.450 and 3.472 eV observed for the ensemble now resolve into several sharply defined transitions. To avoid any confusion due to the sudden plethora of PL lines between 3.42 and 3.48 eV, let us stress once again that each NW we have investigated exhibits a different PL spectrum. Some NWs, for example, are completely free of PL lines below 3.46 eV, others (such as the one shown in Fig. 3) are not. The ensemble spectrum is formed by the superposition of about 10^3 entirely different spectra. Next, let us categorize the lines observed in the spectrum shown in Fig. 3.

(i) The band at 3.422 eV is observed for just a few NWs and is always broad. It occurs preferentially in coalesced NW agglomerates, and we believe that it is in fact related to a structural defect (particularly, a stacking fault) in GaN.

(ii) All lines between 3.44 and 3.46 eV are designated to belong to the defect band centered at 3.45 eV in the en-

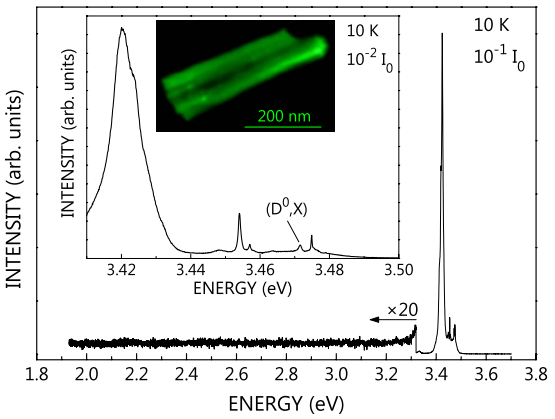

FIG. 3. (Color online) μPL spectrum of the dispersed single NW displayed in the inset at 10 K and an excitation density of $10^{-1}I_0$. The inset also shows an expanded view of the near-band-gap PL lines at an excitation density of $10^{-2}I_0$.

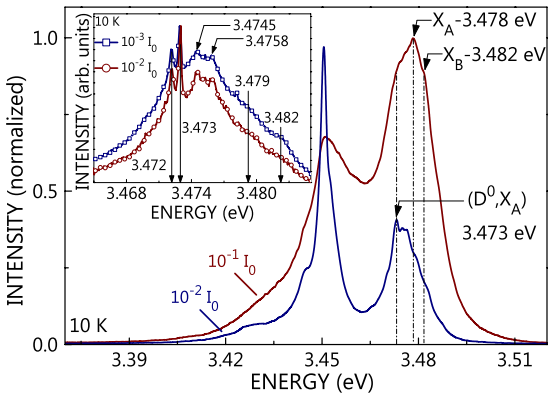

FIG. 4. (Color online) μPL spectra of another dispersed single NW at 10 K and excitation densities of $10^{-1}I_0$ and $10^{-2}I_0$. The position of the most important transitions is marked by dashed lines. Arrows point to the labels of these transitions. The inset shows high-resolution spectra of the near-band-gap PL lines at excitation densities of $10^{-2}I_0$ and $10^{-3}I_0$. All relevant transitions are marked by arrows and their energies are denoted in the figure.

semble. We will later comment on the origin of these transitions characteristic for most GaN NWs.

(iii) The lines between 3.46 and 3.47 eV are most likely belonging to acceptor-bound (A^0,X) excitons. It is not surprising that *individual* NWs have also incorporated acceptors.

(iv) Most interesting, however, are the lines between 3.474 and 3.48 eV, an energy range for which no strong transitions have ever been observed in bulk or unstrained layered GaN. Indeed, the spectral position of these transitions does not correspond to that of the corresponding bands in the ensemble. For example, the prominent high-energy transition in the single NW occurs at 3.475 eV, clearly above the donor-bound exciton line (D^0,X) at 3.472 eV (cf. Fig. 2).

Transitions in the energy range between the donor-bound and the free A exciton are usually resolved only in high-quality epitaxial layers, and originate there from excited states of the donor-bound A and the ground state of the donor-bound B exciton.[35] These transitions are orders of magnitude weaker than the donor-bound A exciton and cannot account for the line at 3.475 eV observed in Fig. 3.[35]

Given the fact that most of the dispersed NWs we have studied are heterogeneously strained (cf. Fig. 2), it appears to be at least possible that some NWs experience a purely homogeneous strain which would result in a rigid shift of the spectrum without causing a broadening. However, the energy separation between the two dominant high-energy transitions is only 3 meV while that between the (A^0,X) and the (D^0,X) amounts to 6–7 meV, just as the one between the (D^0,X) and the free A exciton (X_A). The energies observed are thus inconsistent with the assumption of a purely homogeneous strain.

For a definitive assignment of the transitions observed, we turn to another dispersed single NW whose PL spectra are also distinguished by narrow lines as shown in Fig. 4. For an excitation density of $10^{-2}I_0$, the spectrum of this NW is dominated by the transition at 3.45 eV. At higher energies, a peak at 3.473 eV is visible, which is assigned to the (D^0,X) transition, as well as higher-energy bumps at 3.478 and 3.482 eV. These bumps dominate the spectrum for an excitation density of $10^{-1}I_0$ (and higher). The linear dependence of the intensity of these features on excitation density, and their dominance at high excitation density, identifies them as the free A and B excitons (X_A and X_B). The energies of these features (without any attempt of a deconvolution of the lines) are within 1 meV of the values reported for bulk, strain-free GaN. In other words, there is no detectable shift of the spectra due to a homogeneous strain.

The inset of Fig. 4 displays the high-energy range of the PL spectrum of this NW recorded with a higher spectral resolution. The (D^0,X) line is now resolved into two transitions and the free-exciton shoulders at higher energies are clearly visible. Most important, however, is the presence of lines in the intermediate energy range between 3.4745 and 3.476 eV, i.e., in the same range as that discussed in the context of Fig. 3. Knowing from the above discussion that these energies are indeed accurate, strain can be excluded as a reason for their occurrence. The question about the origin of these transitions thus remains.

Other potential reasons for an energy shift of optical transitions include electric fields and varying carrier concentrations due to depletion at the NWs' side facets. The energy of *excitonic* transitions in GaN is, however, insensitive to electric fields which might be present at the surface, as well as screening by free carriers. Regarding the former, Camacho *et al.*[36] and Pedrós *et al.*[37] have shown that fields up to 10 kV/cm do not result in a discernible shift of the exciton transition in GaN but in a pronounced quenching (note that the redshift of <1 meV observed by Camacho *et al.*[36] is due to strain-induced band-gap modulation and not the electric field). The same results were obtained for bound excitons in CdS, a material with virtually the same excitonic parameters as GaN.[38] In fact, the reduction of the exciton binding energy by the electric field is almost exactly canceled by the Stark shift of the excitonic state. An analogous conclusion applies to screening: here, the reduction of the exciton binding energy by free-carrier screening is almost exactly cancelled by band-gap shrinkage, basically due to charge neutrality of the

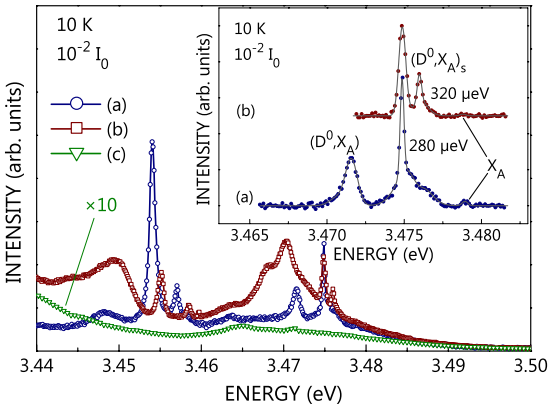

FIG. 5. (Color online) μPL spectra of the single NW displayed in Fig. 3 at 10 K and an excitation density of $10^{-2}I_0$ taken (a) right after, (b) one, and (c) five months after its dispersion on the Si(111) carrier. The inset shows the first two spectra after background subtraction. The symbols represent the data, the solid lines Gaussian fits to determine the spectral position and linewidth of the individual transitions.

exciton. This fact, together with the quenching of the excitonic resonance near the Mott density, has been shown both experimentally.[39,40] and theoretically.[41,42] Concluding, neither electric fields nor screening can be responsible for an energy blueshift in the meV range as required to explain the transition at 3.475 eV.

Prior to attempting a further discussion on the origin of these transitions, let us point out an as surprising as important observation. Figure 5 compares three different measurements taken on the single NW of Fig. 2: the first trace (a) right after dispersing the column onto Si, the second (b) one, and the third (c) five months after. Additionally, following the measurement shown in trace (b), the sample was subjected to a prolonged exposure to a 10 keV electron beam while recording the micrograph depicted in Fig. 3. Spectra (a) and (b) in Fig. 5 both exhibit several sharply defined transitions in the 3.45 and 3.47 eV spectral range. However, none of these transitions is equal in spectra (a) and (b) with regard to spectral position and intensity, and most of them are absent in spectrum (c), which, in addition, is a 100-fold lower in intensity as compared to the earlier spectra. We believe both of these latter observations to be a consequence of the inevitable C contamination induced by the high-energy electron beam in the SEM. In any case, it is important to note that all of these spectra were recorded at least three times after intentionally altering the adjustment in each respective measurement session and were found to be identical in each case.

The above results foremost demonstrate that the electronic properties of a single GaN NW evolve with time. This important finding may be understood based on the fact that both the ionization energy of defects and the energy of excitons bound to them are altered in the vicinity of a surface. For example, the ionization energy of a hydrogenic donor located directly at the surface is only one fourth of that of the same donor in the bulk, assuming that the vacuum represents an infinite barrier.[43] Recent tight-binding calculations refraining from this approximation qualitatively arrive at the same conclusion, namely, that the ionization energy of impurities close to the surface is always lower than the that of the corresponding bulk defect.[44] As shown by Satpathy,[45] the same applies to Wannier excitons close to semiconductor surfaces; the exciton binding energy decreases smoothly from its bulk value (reached for distances three times the Bohr radius a_B) to one fourth of this value directly at the surface. This result can be extended to the binding energy of excitonic complexes (which, in the bulk, are spectrally located at 3.471–3.473 eV for shallow donor-bound and 3.465–3.466 eV for shallow acceptor-bound excitons).[27,28] In other words, surface donor-bound excitons are expected to have a transition energy in between the bulk donor-bound and the free excitons. Note that exactly the same observation has led to the proposal of surface excitons in CdS, CdSe, and ZnO more than two decades ago (see Refs. 46 and 47, and references therein).

To accurately determine both the spectral position and linewidth of the highest-energy transitions in Figs. 5(a) and 5(b), we subtract the background they are riding on and fit the resulting spectra with a set of Gaussians as shown in the inset of Fig. 5. The spectrum obtained right after dispersion is displayed as trace (a) and exhibits two transitions we are familiar with from bulk GaN, namely, the donor-bound exciton $[(D^0,X_A)]$ at 3.4716 eV and the free A exciton at 3.4789 eV.[26,48] The dominant line at 3.4749 eV with a linewidth of 280 μeV, however, is attributed to a surface donor-bound exciton $[(D^0,X_A)_s]$, as are its high-energy shoulders at 3.4754 and 3.4758 eV. The spectrum obtained one month later in (b) also shows the free A exciton at 3.4788 eV, and two surface donor-bound excitons at 3.4749 (the same as before) and 3.4760 eV. The latter has a linewidth of 320 μeV.

We have found similar results for two further dispersed NWs. Having investigated several dozens of dispersed single NWs, this is a rather low yield which does not allow us to investigate the effects reported above in a systematic fashion. In what follows, we instead make use of the fact that the as-grown NW ensemble thins out considerably toward the edge of the wafer due to a temperature gradient. Figure 6(a) shows a contrast-enhanced SE micrograph of a region about 1 cm from the edge where the NW ensemble exhibits inter-NW distances of 1 μm and more. In the micrograph, three well-formed NWs are observed with a length of about 300 and a diameter of 30 nm. Due to the low density, coalescence of NWs is entirely absent. Furthermore, we see that there is absolutely no parasitic growth in between the NWs. This fact allows us to optically address individual free-standing NWs which are free of strain by their very nature. This necessary absence of strain follows directly from the principle of Saint-Venant,[49–51] which in the present case may be expressed as follows: *biaxial stress applied to one end of an otherwise free elastic rod of diameter d will decay to zero approximately within a distance d from the clamped end of the rod* (an explicit solution of this problem can be found in §144 of Ref. 50).

Figure 6(b) displays a trichromatic PL intensity map of this low-density edge region. The individual spectral components are represented by the bulklike donor-bound exciton (red), the surface donor-bound exciton (green), and the free

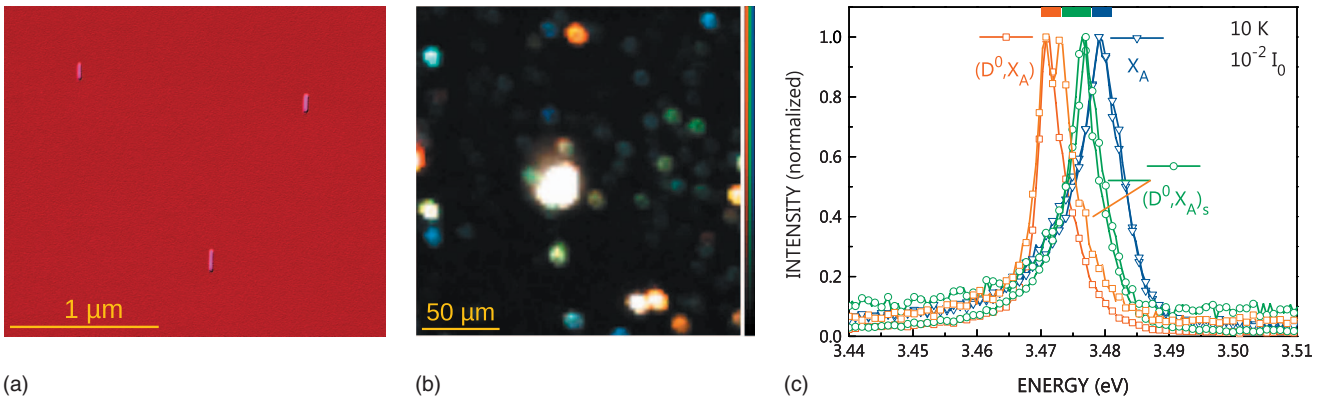

FIG. 6. (Color) (a) Bird's-eye view (30°) of the low-density edge region of the NW ensemble under investigation. The raw SE micrograph is color coded to enhance the material contrast between GaN and Si. Evidently, no parasitic growth takes place in between the free-standing NWs. (b) Trichromatic PL intensity map of the low-density edge region of the NW ensemble under investigation. The spectral range characteristic for bulklike and surface donor-bound excitons is displayed in red and green, respectively, while the free-exciton transition is decoded in blue. (c) Representative PL spectra from six individual NWs within the map displayed in (b). The spectral range for each of the transitions is denoted at the top.

exciton (blue). Note that we have confirmed these assignments by excitation-density-dependent experiments (not shown here) analogous to the ones in Fig. 4. Figure 6(c) shows the energy range used for each of these transitions, as well as representative spectra taken from two different NWs for each energy range. Owing to the excitation density required to record the map (which consists of 4900 spectra) in a manageable time span, the transitions are comparatively broad. Decreasing the excitation density to $10^{-3}I_0$ reduces the width of these peaks to about 1 meV. Apparently, the excitation of free-standing NWs is more efficient than that of dispersed ones (the PL lines of which do not show any discernible broadening at $10^{-2}I_0$), suggesting that the coupling of light into the NW (and thus the carrier generation within the NW) is enhanced in the free-standing excitation configuration.

The map in Fig. 6(b) clearly exhibits more blue and green than red spots. Mixed spectra, represented by yellow and orange, also occur but purely red spots are found only a few times (the large white spot near the center of the map is caused by the broad spectra of NWs inadvertently dispersed on this area during cleavage). In other words, in this low-density region, where NWs have been formed at even higher temperature than those at the center of the wafer, columns with dominant free or surface donor-bound excitons are more numerous than those displaying a bulklike donor-bound exciton transition. This finding is certainly at least partly due to the reduced diameter of these NWs but it assures us nonetheless that the occurrence of surface donor-bound excitons is not a statistical anomaly; it is the rule rather than the exception.

Not that this finding is actually surprising from a geometrical point of view. As a matter of fact, an unexpectedly large fraction of donors or acceptors *is* in the vicinity of the surface. If the radius of the (assumed to be cylindrical) column is a and the Bohr radius of the hydrogenic impurity $a_B \ll a$, a fraction η of the total number of impurities are effectively "at the surface" (i.e., within the cylindrical shell with $a-a_B<r<a$) if they are presumed to be distributed randomly,

$$\eta = 1 - (1 - a_B/a)^2. \quad (1)$$

For the present case, $a=20$ nm, $a_B=4.8$ nm,[37] and $\eta=0.42$. Also note that the volume of a (noncoalesced) single column is smaller than 10^{-15} cm^3. Assuming an impurity concentration of 10^{17} cm^{-3}, which is at the lower limit for common GaN layers, we would expect about 100 impurities in a single column and 40 of these would be at the surface. If the ionization energy of impurities changes close to the surface as much as predicted theoretically, the transitions observed would all be broadened to values much above 1 meV. The very fact that we observe significantly narrower lines suggests that these NWs have an impurity concentration much lower than what is commonly observed.

There is, indeed, additional experimental evidence suggesting that the NWs do contain fewer point defects than even state-of-the-art layers. First, we have not observed any trace of the "omnipresent" yellow luminescence (which is caused by a transition between a shallow donor and a native defect, probably a Ga vacancy[33]) for any of our GaN NW ensembles (cf. Fig. 2), in contrast to GaN layers grown in the same MBE system. Second, the PL intensity of these NWs is high regardless of the N plasma cell used for their growth. This finding is in complete contrast to GaN layers, whose PL intensity drops by orders of magnitude when using a plasma cell with high N-ion flux.[37] These two empirical facts inspire the following important hypothesis: *point defects such as, e.g., Ga vacancies, tend to diffuse or segregate to the NW surface and annihilate during growth. GaN NWs may thus indeed possess an inherently superior optoelectronic quality than GaN films.*[52,53]

This diffusion/segregation process may not only occur for native point defects but also for impurities such as Si and O as indeed suggested on theoretical grounds[54] and known in the nanocrystal community as "self purification."[55] Considering that a few atomic hops would suffice to obtain a detectable change in spectral position, this mechanism would provide a plausible explanation of the gradual conversion of bulklike into surface donor-bound excitons. If this process

continues with the impurity finally emerging at the surface being thus rendered inactive, it is even conceivable that the free-exciton decay may become the dominant recombination channel as observed in Fig. 6. The spectrum of the individual NW will thus depend on its initial impurity density and the presence of coalescence-induced defects but also on its size and the growth temperature.

We stress that this model accounts only for energy changes in point defects. It is difficult to imagine a mechanism which would change the energy of excitons bound to a planar defect such as a stacking fault in a similar way. Thus, we believe the emission at 3.45 eV to be related to an abundant surface defect (analogous to the present understanding for ZnO (Ref. 56) and in agreement with Ref. 21 given the following three empirical facts: (i) no emission at this energy is observed for GaN layers grown in the same PAMBE system, (ii) the band, in contrast to the donor-bound exciton line, scales linearly with excitation density, and (iii) the lines at 3.45 eV change their spectral position with time (cf. Fig. 4).

IV. CONCLUSION

Summarizing and concluding, we have shown that the PL spectra of GaN NWs grown on Si(111) can have a linewidth of at least below 300 μeV (limited by our spectral resolution). Our results furthermore suggest that point defects and impurities are driven to the NW sidewalls during growth, resulting in an overall crystal quality even higher than that of state-of-the-art homoepitaxial layers.

The spectral broadening commonly observed for single NWs is *not* intrinsic. The main broadening mechanism responsible for the minimum reported linewidth of 2–3 meV for NW ensembles, however, *is* intrinsic and *not* caused by residual strain but rather by the energy difference between excitons bound to bulk and surface defects of 3–4 meV. While this energy difference shows up in individual NWs as spectrally distinct lines, it will manifest itself in a corresponding broadening when summing over the hundreds or thousands of NWs excited in the ensemble.

Finally, we note that our experimental findings confirm two recent theoretical predictions: they directly show that impurities have the tendency to segregate to the NW surface[54,55] and they also directly reflect the lower binding energy of these surface defects.[44]

Note added in proof. We recently became aware of the work of Corfdir *et al.*[57] Based on time-resolved PL data on GaN NW ensembles, these authors interpreted the comparatively large PL linewidth they observed (5 meV) in *literally* the same way as we do in the present study: "Thus, the random distribution of donor sites could be an *intrinsic* origin of the broadening of all PL lines involving donors." Note that Corfdir *et al.* went one step farther and attributed the transition at 3.45 eV to the two-electron satellite of the surface donor-bound exciton.

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