Polarized photoluminescence from single GaN nanorods: Effects of optical confinement

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Abstract: By measuring linearly polarized photoluminescence (PL) from single, isolated gallium nitride (GaN) nanorods with the rod diameters in the subwavelength regime (30–90 nm), we present clear evidence for size dependence of polarization anisotropy. The maximum polarization ratio at room temperature (~0.9 with emission and excitation light polarized parallel to the long axis of nanorod) occurs at the rod diameter of ~40 nm. The experimental data are compared with the recent theoretical model proposed for thick semiconductor nanowires. It is concluded that the optical confinement effects in this size regime play an important role in the observed giant polarization anisotropy. Furthermore, we have performed a temperature-dependent study of polarized PL to show the importance of internal emission anisotropy at low temperatures.

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1. Introduction

Optical properties of one-dimensional (1-D) nanostructures (nanowires, nanorods, nanopillars, and nanotubes, etc.) have been intensively studied during the last decade. One of the unique properties in the 1-D nanostructures is the strong polarization anisotropy of emission/absorption spectra. Due to the material and/or geometry anisotropy, the emission/absorption intensity of I_{\parallel} , in which the emission/absorption light is polarized

parallel to the long axis of 1-D nanostructures, can be very different from that of I_{\perp} , in which the emission/absorption light is polarized perpendicular to it. The observed polarization anisotropy is typically defined in terms of the polarization ratio (also called as the degree of polarization)

$$\rho = \frac{I_{\parallel} - I_{\perp}}{I_{\parallel} + I_{\perp}}$$

In the early studies, 1-D nanostructures were fabricated primarily by lithography [1–3] and epitaxy [4,5] and the III-arsenide (AlAs, GaAs, InAs, and their alloys) quantum wire structures were often used. Ils *et al.* suggested that the polarization ratio is a function of the lateral width [3]. It was found that the polarization ratio is proportional to $1/L_x$ (L_x is the lateral width of nanowires) for thick wires and it saturates in the case of thin wires at a level of 0.2–0.6 depending on the structure of quantum wire. Later on, a giant anisotropy (~0.9) in the band gap emission with the dominant polarization parallel to the wire long axis was reported for single InP nanowires prepared by a bottom-up method [6]. After that, many different kinds

of nanowires were studied and similar phenomena were also reported [7–10]. For example, the polarization dependence on the emission wavelength has been found for the case of single ZnO nanowires [7]. Especially, the band gap emission of ZnO nanowires shows a negative polarization ratio (-0.65). This property has also been observed in CdS nanorods [9]. Furthermore, polarized photoluminescence (PL) from individual CdSe nanowires grown parallel and perpendicular to the *c* axis of the wurtzite structure was reported [10]. Shan *et al.* concluded that the polarization anisotropy of emission from CdSe nanowires does not depend on the growth direction, but is mainly determined by the elongated shape (aspect ratio) of nanowires.

Although the present experimental results are not completely consistent, it is generally acknowledged that there are two major mechanisms responsible for these phenomena. The first kind of mechanism attributes these phenomena to the quantum size effects [11-16]. In this mechanism, the quantum confinement of carriers in 1-D nanostructures causes the quantization of energy spectrum and the changes in optical transition matrix. It is important to note that, in order to be applicable to this mechanism, the size of nanostructures should be smaller than the exciton Bohr radius. In contrast, the second kind of mechanism is based on the confinement of optical electric field [17,18] due to the variation in the dielectric constants of nanowires/nanorod (larger) and its environment (smaller), which can result in the anisotropic electric field distribution. This effect becomes dominant in thick semiconductor 1-D nanostructures and a strong size dependent behavior was predicted by Ruda *et al.* in the subwavelength regime, in which the rod diameter is smaller than the emission wavelength but much larger than the exciton Bohr radius [18].

Comparing with other semiconductor nanowires, reports on optical properties of single GaN nanowires or nanorods are relatively scarce. Recently, a polarization-resolved PL study of single wurtzite GaN nanowires has been reported by Schlager *et al.* [19]. Their experimental data showed that the observed excitonic transitions at low and room temperatures are in agreement with the optical selection rules of wurtzite crystal symmetry. However, the degree of PL polarization and its dependence on the sizes of GaN nanowires or nanorods have not been investigated in detail. In this work, by measuring PL from single, isolated GaN nanorods in the subwavelength regime (30–90 nm), we present clear evidence for strong size (rod diameter) dependence. Based on these results, it can be concluded that the optical confinement effects play an important role in the observed giant polarization anisotropy. Furthermore, we have performed a temperature-dependent study of PL polarization anisotropy to show the importance of internal emission anisotropy at low temperatures.

2. Sample preparation and optical measurements

Vertically aligned, wurtzite GaN nanorod arrays were grown on 3-inch Si(111) wafers by plasma-assisted molecular-beam epitaxy (PA-MBE). The details of growth process can be found elsewhere [20]. The grown GaN nanorod arrays have been confirmed to be single crystalline and epitaxially oriented with the epitaxial relationship of $\langle 2\overline{1} \ \overline{1} 0 \rangle_{GaN} \| \langle \overline{1} \ 10 \rangle_{Si}$ and $<1\overline{1}00>_{GaN} || <11\overline{2}>_{Si}$. Moreover, the GaN nanorods are nitrogen-polar with the axial direction pointed to the -c-axis ([0001] direction). The samples of single, isolated GaN nanorods for the polarized PL measurements were prepared by using a four-probe nanomanipulator (Zyvex) installed inside a field-emission scanning electron microscope (FE-SEM, Ultra 55, Zeiss). A movie showing the nanorod manipulation process is shown online (Media 1 [3.6 MB]). Figure 1(a) shows the FE-SEM image taken at the region of a 2-µm-long GaN nanorod-array sample near the cleavage edge and with a tungsten probe positioned nearby. In such a region, numerous single nanorods and bundled nanorods can be found, and we can utilize sharp tungsten probes to manipulate single GaN nanorods to designated positions and orientations. During the manipulation process, when the distance between the tungsten probe and the GaN nanorod is small enough, single nanorods can be attracted to the commonly grounded tungsten probe by using the electron-beam-induced electrostatic force.

As show in Fig. 1(b), one single GaN nanorod is attached to the probe by using this mechanism and is then removed far from the Si substrate. Afterwards, we can separate single GaN nanorods from the probe by using the van der Waals force when the contact area between the nanorod and the substrate is formed by moving the nanorod to the designated areas on a surface registered with a gold alignment pattern. Single nanorods can be separated from the probe because that the van der Waals force (with the substrate) is larger than electrostatic force (with the probe). Figure 1(c) shows four single GaN nanorods manipulated in this way onto a gold-patterned Si substrate. The inset in Fig. 1(c) shows the GaN nanorod with uniform diameter of 35 nm and length of 2 μ m. Utilizing this technique, we can manipulate single nanorods according to specific requirements. For the polarized PL measurements, we selected GaN nanorods ranging in diameter of 30–90 nm and in length of 1.2–2 μ m. The shorter nanorods were produced during the cleavage process of nanorod-array-covered Si substrates.

For the PL analysis, a home-built micro-PL (μ -PL) system was used in this study. Samples were excited with a continuous-wave HeCd laser operating at 325 nm, which was focused with a UV compatible object lens (Mitutoyo, NA = 0.5) to a spot diameter of about 1 μ m. The PL signal was collected with the same object lens and dispersed by a spectrometer (HR460, Jobin-Yvon) equipped with a liquid-nitrogen-cooled CCD detector. In the optical images, single GaN nanorods appeared to be small shadows. With reference to the FE-SEM images, we can position the focused laser spot to excite individually designated GaN nanorods by the relative positions between gold alignment pattern and GaN nanorods. The polarized PL data were carefully calibrated in order to eliminate the polarization dependence of the measurement setup. Using this procedure, the detected PL of *c*-plane GaN epilayers at similar emission energy has been confirmed to be unpolarized, as theoretically expected.



Fig. 1. FE-SEM images of nanorod manipulation process. (a) Image near the cleavage edge of a Si(111) substrate covered by a vertically aligned array of 2-µm-long GaN nanorods. A tungsten probe is positioned nearby. (b) A tungsten probe attached with a single GaN nanorod by electrostatic interaction. (c) Using this technique, four single, isolated GaN nanorods were positioned on a gold-patterned Si substrate with designated locations and orientations; the inset shows a GaN nanorod with uniform diameter of 35 nm and length of 2 µm. A movie showing the nanorod manipulation process is shown online (Media 1).

3. Results and discussion

In our u-PL setup, the polarized PL measurements at room temperature were performed in the geometry that the incident direction (wave vector) of laser light is perpendicular to the long axis of GaN nanorod ($\mathbf{k}_{exc} \perp \mathbf{c}$). Unlike the common $\mathbf{k}_{exc} \parallel \mathbf{c}$ geometry for thin-film studies, we can distinguish two PL polarizations in the plane where the GaN nanorods reside: the σ polarization corresponds to the electric field perpendicular to the long axis (*c*-axis); and the π polarization corresponds to the electric field parallel to the *c*-axis. The emission properties of bulk GaN in the $\mathbf{k}_{exc} \perp \mathbf{c}$ geometry have been reported by Paskov *et al.* [21] using the cleaved sample edge of bulk GaN. Here, we investigate the emission properties of single, strain-free GaN nanorods in the $\mathbf{k}_{exc} \perp \mathbf{c}$ geometry. Figure 2(a) shows the PL dependence on the excitation polarization. The inset shows the FE-SEM image of the measured GaN nanorod (length: 1.2 µm, diameter: 40 nm). This rod is labeled as #R1 according to the nearest gold alignment pattern mark. As shown in Fig. 2(a), both of the PL spectra exhibit band gap emission (~3.4 eV) at room temperature and a higher PL intensity were obtained in the configuration that $\mathbf{E}_{exc} \parallel \mathbf{c}$ (the electric field of excitation is parallel to the *c*-axis of GaN nanorod). This difference is mainly due to stronger optical absorption in the configuration that $\mathbf{E}_{exc} \| \mathbf{c}$. Similar results have also been reported on other semiconductor nanowires, such as InP [6] and CdSe [10].



Fig. 2. Photoluminescence spectra of single GaN nanorods at room temperature measured with the $\mathbf{k}_{exc} \perp \mathbf{c}$ geometry. (a) Dependence on the optical absorption polarization. The inset shows the FE-SEM image of the measured GaN (#R1) nanorod with a diameter of 40 nm. (b) and (c) show the polarized PL spectra with the electric field of optical excitation parallel and perpendicular to the *c*-axis, respectively. The measured polarization ratios are 0.9 and 0.84, respectively. (d) Polarization ratio measured from another GaN nanorod (#R3) using the same experimental configuration. The diameter of #R3 GaN nanorod is 80 nm, as shown in the inset.

The linearly polarized PL signal was measured by using a polarizer positioned in the luminescence collection pathway. In Figs. 2(b) and 2(c), we show the polarized PL of #R1 GaN nanorod with the electric field of luminescence parallel ($\mathbf{E}_{\mathbf{P}_{l}} \parallel \mathbf{c}$) and perpendicular $(\mathbf{E}_{PL} \perp \mathbf{c})$ to the long axis (c-axis) of GaN nanorods. In Fig. 2(b), the excitation configuration is $\mathbf{E}_{exc} \| \mathbf{c}$, and the intensity of polarized PL with $\mathbf{E}_{PL} \| \mathbf{c}$ is much larger than that with $\mathbf{E}_{PL} \perp \mathbf{c}$. The polarization ratio can reach as high as 0.9. This value is close to highest record that observed in individual InP nanowires [6]. Figure 2(c) shows the polarized PL using the other excitation configuration such that $\mathbf{E}_{exc} \perp \mathbf{c}$ and the polarization ratio can also reach about 0.84. Although the polarization ratios with different excitation configurations are not in full agreement, we believe that the polarization anisotropy does not depend on the polarization of excitation. And, the slight difference is mainly due to the misalignment between the axes of the polarizer and the nanorod during different measurements. Figure 2(d) shows the polarized PL from a different single GaN nanorod (labeled as #R3) using the same measurement configuration as that shown in Fig. 2(b). The #R3 GaN nanorod has a length of 2 µm and a diameter of 80 nm. And, the measured degree of linear polarization is only 0.16. This value is quite small compared with that of #R1 GaN nanorod. Therefore, in order to understand the origin of large difference in polarization anisotropy from different GaN nanorods, it is necessary to study a large number of singles GaN nanorods with different diameters. The nanorod manipulation technique adopted here is therefore very handy for these measurements.

As mention in introduction, there are two main mechanisms proposed for interpreting the giant linear polarization, which include the quantum size effects and the optical confinement effects. Both of these two mechanisms depend on the sizes of nanostructures [11–18]. However, for the case of carrier confinement, the size dependence not only determines the degree of polarization but also affects the emission energy [11–16]. Thus, the experiments for size-dependence of polarized PL should be carried out also with an analysis on the emission energy. For the size-dependent measurements, we selected and measured a number of GaN nanorods, which have diameters in the range of 30–90 nm and lengths in the range of 1.2–2 μ m. Figure 3(a) shows the relation between the diameters of selected nanorods and their PL peak energies at room temperature. Clearly, there is no significant indication that the peak energy of PL depends on the diameter of nanorods. Thus, the quantum confinement effect of carrier should be small and it can be safely neglected for these samples. This result is in agreement with the fact that the size of GaN nanorods in this work (30–90 nm) is much larger than the exciton Bohr radius in GaN (<10 nm).



Fig. 3. Size dependence of polarized photoluminescence from single GaN nanorods ($\mathbf{k}_{exc} \perp \mathbf{c}$). (a) The peak energies of PL from single GaN nanorods with different rod diameters. The dashed lines show the averaged values of peak energy with two PL polarizations. (b) The polarization ratio of PL from single GaN nanorods with different diameters using the measurement configuration that the electric field of excitation is parallel to the *c*-axis. The dashed line is the calculated result considering dielectric confinement of optical electric field with $\varepsilon = 6$ and isotropic internal emissions (see text for details).

On the other hand, the selection rules of optically active exciton emissions can be confirmed from these data. As shown in Fig. 3(a), the peak energy of $\mathbf{E}_{PI} \parallel \mathbf{c}$ (solid circles) is averagely higher than that of $\mathbf{E}_{PL} \perp \mathbf{c}$ (empty circles) by a few meV. The dashed lines are the average positions of the peak energies for $\mathbf{E}_{PL} \| \mathbf{c}$ and $\mathbf{E}_{PL} \perp \mathbf{c}$ polarization, respectively. The difference in energy is about 5-6 meV. According to the optical selection rules for A and B exciton emissions in wurtzite GaN, only the B exciton emission is allowed for the $\mathbf{E}_{\text{PI}} \parallel \mathbf{c}$ polarization (π polarization) so that the peak related to A exciton is completely absent [19,21]. For the $\mathbf{E}_{PL} \perp \mathbf{c}$ polarization (σ polarization), the *B* exciton emission was experimentally confirmed to be much weaker than the dominant A exciton emission [19,21]. Therefore, the observed difference in PL peak positions in two polarizations is mainly due to the energy difference between A and B excitons in GaN. For a more rigorous analysis, a spectral deconvolution of contributions from A and B excitons for the σ polarization might be necessary. However, it is difficult to be performed for the room-temperature PL spectra. In contrast to the observation that the emission energy has no obvious size dependence, the polarization ratio is highly dependent on the diameter of nanorod. Figure 3(b) shows the relation between the rod diameter and the polarization ratio measured with the excitation configuration of $\mathbf{E}_{exc} \| \mathbf{c}$. A large size dependence in polarization ratio can been found for the nanorods with smaller diameters (30-40 nm) and the measure polarization ratios can be as large as 0.8-0.9. In contrast, for the nanorods with larger diameters (80-90 nm), almost no polarization can be found. For comparison, the dashed line is shown according to the model proposed by Ruda et al. [18], in which the effects of optical confinement are considered. From the comparison shown in Fig. 3(b), it is clear that, for the studied GaN nanorods in the present size regime, mainly the effects of optical confinement are responsible for the observed polarization anisotropy.

In the past, many researchers have attributed the giant linear polarization to discontinuity of dielectric constants [6,8,10] and the related background can be found in Chap. 2 of Ref. [22]. For a thin cylindrical nanowire with diameter much smaller than wavelength of light, the internal field $\mathbf{E}_{i,\perp}$ is attenuated by a factor $2\varepsilon_0/(\varepsilon+\varepsilon_0)$ from the external electric field $\mathbf{E}_{e,\perp}$ for electric fields perpendicular to the long axis, where ε is the dielectric constant of the cylindrical nanowire. In contrast, the internal field $\mathbf{E}_{i,\parallel}$ remains the same as the external electric field $\mathbf{E}_{e,\downarrow}$ for electric fields parallel to the long axis. Therefore, the field distribution is essentially non-uniform for a thin cylindrical nanowire and this was suggested to be the cause for the observed polarized properties. For example, in the case of absorption, the total power of absorption is directly proportion to $|\mathbf{E}|^2$. However, using only the discontinuity of dielectric constants, the observed strong size dependence of polarization ratio can not be fully accounted for.

In contrast, the strong size dependence can be explained well using the optical confinement effects [17,18]. In this approach, the internal field \mathbf{E}_i is described by the Helmholtz equation. Using cylindrical coordinates (r, ϕ, z) , inside the nanorod $(r \le a, where a$ is radius of nanorod) \mathbf{E}_i can be expressed as

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \mathbf{E}_{i}}{\partial r}\right) + \frac{1}{r^{2}}\frac{\partial^{2}\mathbf{E}_{i}}{\partial \phi^{2}} + \frac{\partial^{2}\mathbf{E}_{i}}{\partial z^{2}} + \varepsilon\frac{\omega^{2}}{c^{2}}\mathbf{E}_{i} = 0, \qquad (1)$$

where ω is the light frequency and c is the light velocity. Outside the nanowires (r > a), the equations should be modified by replacing ε with ε_0 . The solution of this equation depends on the relative orientation of external electric field $\mathbf{E}_e(r,\phi,z)$ and the nanowire long axis [18,23]. Also, the factor of $2\varepsilon_0/(\varepsilon + \varepsilon_0)$ can be derived using the limit of $\omega a/c \rightarrow 0$. In general, the luminescence properties can be obtained by using the general formulas for a field created by a dipole with arbitrary position r_0 followed by integration over r_0 , which can only be performed

numerically [24]. Instead of this method, Ruda and Shik modeled their case simply by an effective emitting dipole $d(r, \phi, z)$ at the axis (r = z = 0).

According to the model proposed by Ruda *et al.* [18], for original dipole moment oriented along the long axis (d_{0z}), the effective dipole moment can be written as $d_z = d_{0z}A_z$, where A_z is a function of dielectric constant (ε), light frequency (ω), and radius of nanorods (a). Thus, A_z can be considered as an effective parameter that comes from the material properties and size. Similarly, for the perpendicular dipole (d_{0x}), the effective dipole moment can be written as $d_x = d_{0x}A_x$. Therefore, for cylindrical nanowire $d_x = d_y$, and the intensity ratio for different light polarization is given by

$$\frac{I_{II}}{I_{\perp}} = \frac{2d_z^2 + d_x^2}{3d_x^2},$$
(2)

where I_{\parallel} and I_{\perp} is the luminescence intensity with electric field parallel and perpendicular to the long axis, respectively. And, the polarization ratio can be given by

$$\rho = \frac{I_{I/} - I_{\perp}}{I_{I/} + I_{\perp}} = \frac{d_z^2 - d_x^2}{d_z^2 + 2d_x^2} \quad . \tag{3}$$

For isotropic internal emission ($d_{0z} = d_{0x}$), the polarization ratio can be described completely by effective parameters

$$\rho = \frac{A_z^2 - A_x^2}{A_z^2 + 2A_x^2} \equiv \rho_0 \,. \tag{4}$$

Here, ρ_0 depends only on the properties of dielectric media, which are dielectric constant (ε) and the ratio of the nanorod diameter to the emission wavelength in vacuum (a/λ_0) . Figure 4(a) shows the numerical results of ρ_0 with varying dielectric constants under the conditions of isotropic internal emission. In short, the high degree of polarization occurs prominently in thin nanorods (diameters in the subwavelength regime) with a high dielectric constant. And, independent of the dielectric constant, the polarization anisotropy would not be obvious when the nanorod diameter is much larger than the emission wavelength $(a/\lambda_0 \propto \omega a/c >> 1)$.

For anisotropic internal emission, we should go back to Eq. (3). Here, we define a parameter $\beta \equiv d_{0x}^2/d_{0z}^2$ to quantify the anisotropy of internal emission. Thus, for the case of $\beta = 1$, linear polarization can be described completely by ρ_0 as discussed above. Figure 4(b) shows the simulated polarization ratio with different values of β . Apparently, higher degree of polarization can be obtained under the situation of small β . Moreover, the value of polarization ratio converges to -0.5 with increasing β , which is in accordance with Eq. (3) at the limit of $\beta >> 1$. Furthermore, in the case of $\omega a/c \rightarrow \infty$, which can be considered as the case of bulk crystal, optical confinement can be ignored and the degree of polarization is mainly attributed to the anisotropy of internal emission.



Fig. 4. Numerical results for simulating the effects of optical (dielectric) confinement. (a) Results obtained by varying dielectric constants; (b) Results obtained by varying ratios of internal dipole moment along two orthogonal directions.

At room temperature, the internal emission strengths of measured for bulk GaN were measured to be almost isotropic [21]. Therefore, for simplicity, we assume the case of isotropic internal emission (β =1) for analyzing the measured polarization ratio. Figure 5(a) shows the simulated result of polarization ratio with a few possible values of dielectric constant measured for a GaN bulk film and nanorod arrays [25]. For comparison, empty circles showed the experimental data of polarized PL from single GaN nanorods. Using this fitting procedure, we can confirm that the optical confinement effects dominate the properties of polarized PL from single GaN nanorods at room temperature. Moreover, the red line with ε = 6 seems to be the best fitting curve for the cases of thin nanorods if we ignore the data points from thicker nanorods. From the known anisotropy of the dielectric constant in wurtzite GaN (less than 10%), this anisotropy effect would not significantly affect the fitting result. For the thicker nanorods, the degree of polarization is much reduced since thinker GaN nanorods are not "pure" single nanorods but coalesced bundles with two or more GaN nanorods. In that case, defect-related emissions, rather than free exciton emissions can also play an important role. According to the theoretical study by Gil and Briot [26], the β value can be estimated from the calculated excitonic oscillator strengths to be about 1.5. We have also fitted the measured polarization ratio with this β value and the result is shown in Fig. 5(b). It is obvious that the dielectric confinement effect still holds for the case of slightly anisotropic internal emission.



Fig. 5. Numerical results of polarization ratio using the dielectric confinement model. (a) Results obtained by varying dielectric constants for GaN nanorods; (b) Result obtained by using $(\beta, \varepsilon) = (1.5, 6.5)$.

In order to understand the temperature-dependent behavior of PL anisotropy, lowtemperature experiments were carried out with a continuous-flow microscopy cryostat (Cryo Industries of America). Under the low-temperature conditions, instead of the free exciton emissions, PL peaks associated with bound excitons are more dominant. Figure 6(a) shows the FE-SEM images taken for four isolated GaN bundles, which have been selected from a 1.2µm-long GaN array for the detailed understanding of defect-related bound-exciton emissions at low temperature. Using the nanorod manipulation technique, the #V5 GaN bundle was positioned vertically and the other bundles were arranged in the in-plane geometry on the Si substrate. Moreover, the diameter of GaN bundle depends on the number of single GaN nanorods that coalesced into the bundle. Typically, thicker GaN bundles indicate the possibility of a higher level of structural defects induced by coalescence of slightly misoriented nanorods during PA-MBE growth. Figure 6(b) shows the PL spectra of GaN bundles at 10 K and the inset shows a semi-logarithm plot for peak clarity. The PL spectrum of the #V5 GaN bundle ($\mathbf{k}_{exc} \parallel \mathbf{c}$ for the PL measurement of #V5 GaN bundle) shows a boundexciton emission (D^0X_A) and two structural-defect-related emission peaks (Y_2 and Y_7), which are consistent with the previous ensemble measurement of vertically aligned GaN nanorod arrays [20]. By contrast, the in-plane GaN bundles exhibit different PL spectra because of the different bundling conditions. As shown in Fig. 6(b), among three in-plane bundles, the #B2 GaN bundle has the highest PL intensity at the near-band-gap energy of GaN and the #B0 GaN bundle shows the highest PL emission at around 3.36 eV, which may have its origin in excitons bound to the structural defects at the surface (Y_4) [27] or some other unknown defects. These results indicate that defect-related emissions are more important at low temperatures.



Fig. 6. (a) FE-SEM images of four different GaN bundles, among which the #V5 GaN bundle stands vertically on the Si substrate. (b) Photoluminescence spectra of GaN bundles at low temperature (~10 K). For clarity, the inset shows the PL spectra in semi-logarithm scale.

Figure 7 shows the polarized PL measurement for the #R1 GaN nanorod at ~20 K, the strong near-band-gap emission is accompanied with a shoulder peak occurring at around 360 nm in the $\mathbf{E}_{PL} \perp \mathbf{c}$ polarization. Interestingly, the intensity of this shoulder can be varied by moving the focused laser spot with respect to the initial growth end of #R1 GaN nanorod, which corresponds to the interfaced region with the Si substrate during PA-MBE growth. Thus, the shoulder peak should correspond to the defect emission (Y_2) at the GaN/Si interface. From the normalized PL plot (inset), we can identify that the main exciton emissions correspond to D^0X_A (3.472 eV) and D^0X_B (3.477 eV), which are in good agreement with the selection rules of optically active exciton emissions in strain-free wurtzite GaN [19,21].



Fig. 7. Measurement of polarized photoluminescence from the #R1 GaN nanorod at low temperature (~20 K). The inset shows the corresponding PL peak positions in the plot of normalized intensity. Comparing with the room-temperature measurement on the same nanorod, the polarization ratio at low temperature is completely quenched due to the much enhanced anisotropy of internal emissions due to the neutral-donor-bound excitons.

Comparing with the room-temperature case, the polarization ratio of #R1 GaN nanorod at ~20 K is quenched to near zero (-0.05). This value is much smaller than 0.9 which has been obtained with the same measurement configuration at room temperature. We suggest that this depolarization phenomenon is mainly due to the change of anisotropic internal emission since the dielectric properties is nearly independent on temperature. Thus, the ratio of internal emission (β) should be highly anisotropic for the emissions of the neutral-donor-bound excitons at low temperatures. This suggestion can be justified with the simulation results that shown in Fig. 4(b) for nanorod diameter of 40 nm ($\omega a/c \approx 0.35$) and polarization ratio of -0.05. Indeed, a large value of $\beta \ge 20$ is needed for the observed value. In comparison, this figure is also consistent with the previous experimental work studying the polarized PL of the neutral-donor-bound excitons from the cleaved sample edge of bulk GaN [21]. Similar results of temperature dependence in polarized luminescence have also been reported for nonpolar films of III-N alloys [28,29], such that the intensity of polarized PL with $\mathbf{E}_{PL} \perp \mathbf{c}$ increases with decreasing temperature. This result demonstrates that the polarization ratio could be significantly modified by varying the anisotropy of internal emission at low temperatures.

4. Conclusions

In summary, size- and temperature-dependent polarized PL from single, isolated GaN nanorods have been measured. The excitonic emissions of GaN nanorod have been confirmed to follow the optical selection rules in wurtzite GaN crystal. It is concluded that the optical confinement effects dominate the linearly polarized properties of GaN nanorods with diameters in the subwavelength regime (30–90 nm). At room temperature, the size-dependence of polarization ratio can be described well using the recently proposed theoretical model based on optical (dielectric) confinement. At low temperatures, the degree of polarization could be dramatically modified by the anisotropy of internal emissions originating from the neutral-donor-bound excitons. Due to the their general applicability, we propose that the polarized luminescence properties reported here for GaN can also be found for other semiconductor materials and for other optical measurements, such as electroluminescence and cathodoluminescence.

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