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Photoluminescence enhancement associated with the small size of GaN nanorods

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Small size GaN nanorods (GaN NRs) show new features of the photoluminescence (PL) associated with the enhanced surface effects and the strong electron-phonon coupling. A dominant emission is observed at 3.43 eV; a lower energy with respect to the commonly reported D^0X_A peak for thicker NRs. The phonon replicas of the silent *B1l* acoustic phonon mode are well represented in the PL spectra at temperatures up to 150 K inferring the enhanced Fröhlich electron-phonon coupling. The *B1l* phonon mode was previously detected for the grown GaN NRs by resonant Raman scattering under resonance excitation. The enhanced electron-phonon coupling through Fröhlich interaction is proven by the calculated Huang-Rhys factor. The low-energy dominant peak intensity is around six times the D^0X_A peak intensity indicating that the majority of excitons occupy the surface shell of GaN NRs. This study provides new insights on small-size GaN NRs that greatly influence their physical properties for applications in optoelectronics, UV and blue lasers, and high-temperature/high-power electronic devices.

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1. Introduction

GaN is a polar semiconductor with a direct bandgap suitable for high temperature/high-power electronic devices, short wavelength optoelectronics, photodetectors and sensing applications [1–5]. GaN nanorods (NRs) integrated with Si microelectronics have shown advantages for nanoelectronics and nanoscale device applications such as photovoltaic devices, light emitting diodes and field-effect transistors due to the merits including high carrier mobility, low electron affinity and high thermal and chemical stability [6–8].

The enhanced surface effects associated with small size nanostructures are expected to play a dominant role in controlling their physical properties [1,9]. In particular, NRs of small size have an advantage for getting good crystallinity driven by self-purification where point defects tend to diffuse or segregate to the NR surface and disappear during growth [10]. The free surface of the NR permits elastic relaxation of the strain, and therefore dislocations are expected to be confined at the interface between the substrate and the NR or at the bottom of the NRs [11].

Excitons in nanostructures are subjected to quantum confinement; the exciton size is governed by the nanostructures' physical dimensions in addition to the electron–hole Coulomb interaction. Spatially confined phonon modes and the appearance of zone boundary phonon modes are reported for small diameter GaN NRs [12]. In nanostructures, the motion of electrons is perturbed by phonons due to electron-phonon interactions. The Coulomb interaction between electrons and the longitudinal electric fields produced by the longitudinal optical phonons results in the Fröhlich interaction. As a result of this interaction, phonons can participate both in the near-band emission and in the absorption processes leading to an enhanced absorption and emission peak broadening. The observation of Fröhlich dipolar forbidden modes due to the coupling of surface optical modes with electrons has been reported in nanostructured GaN [13].

The photoluminescence of GaN NRs frequently show a surface-defect related spectral line around 3.45 eV at low temperatures (< 50 K) [14–16]. The PL peak reported at 3.45 eV is attributed to donor-bound excitons close to the nanowire surface.

Here, the photoluminescence spectra of small-size GaN NRs with average diameter and length values around 12 and 100 nm are presented. The physical properties of GaN NRs having such a small size are not well explored in literature. The electron-phonon

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coupling in addition to the enhanced surface effects of the small size NRs are clearly represented in the PL spectra, which haven't been reported so far to our knowledge.

2. Experimental

GaN NRs samples have been grown by plasma-assisted molecular beam epitaxy (PA-MBE) on Si (111) substrates [1,17]. The growth of the GaN NRs was performed under N-rich conditions with a Ga/N ratio of 1/3. The Ga flux was fixed at 5.2×10^{-8} Torr, the N_2 flow rate was fixed at 1.0 sccm and the RF plasma power at 300 W. A GaN buffer layer was grown for 40 s at 450 °C and annealed at 800 °C in the plasma-enhanced N_2 to produce GaN nanoclusters. GaN NRs were grown at 800 °C for 30 min with the maximum growth rate fixed around 200 nm/h. The high quality 3D growth of GaN NRs is checked by Reflection high-energy electron diffraction (RHEED) patterns during growth. High-resolution field emission scanning electron microscopy (HR-FE-SEM), high-resolution transmission electron microscopy (HR-TEM) and θ-2θ X-ray diffraction (XRD) were used for the structural and morphological characterization of the sample.

PL spectra were collected for the as grown GaN NRs on the Si substrates using a He–Cd continuous wave laser (KIMMON IK series) with a wavelength of 325 nm. The laser power was fixed at 18 mW. The photoluminescence from GaN NRs was dispersed in a 1 m grating monochromator (JASCO CT-100) blazed at 300 nm. The direct laser light scattered by the sample was cut off using a low energy pass filter incorporated in the monochromator with both the entrance and exit slits of the monochromator set to 1 mm. The spectra intensities were recorded using a lock-in technique and a photomultiplier (650 U type). The low temperature measurements were conducted using an ACTI Cryogenics closed-cycle helium cryostat. The PL spectra were fitted with a Lorentzian function and multi-peak analysis.

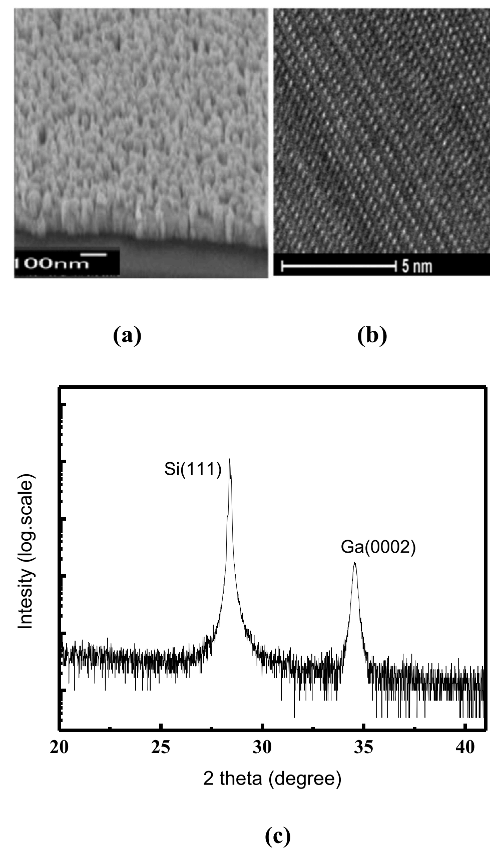


Fig. 1. (a) HR-FE-SEM bird's-eye image shows the as-grown GaN NRs on Si(111) substrate. (b) An HR-TEM image of the single crystalline structure of the grown GaN NRs. (c) X-ray diffraction patterns of GaN NRs sample in the axial (0002) direction.

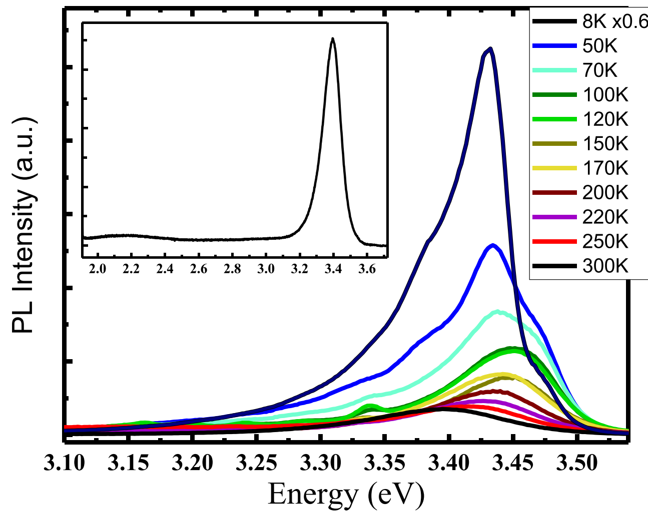
3. Results and discussion

Bird's-eye view of the as-grown GaN NRs on Si substrate by HR-FE-SEM is displayed in Fig. 1(a) and shows the vertically c-oriented GaN NRs. HR-TEM image shown in Fig. 1(b) infers the single crystalline structure of the grown GaN NRs. The estimated average diameter and length values of the grown GaN NRs are 12 and 100 nm, respectively. The c-axis growth of the GaN (0001) fibre texture was proven by XRD measurements shown in Fig. 1(c). The high structural quality and unstressed grown GaN NRs are confirmed by resonant Raman scattering studies [1].

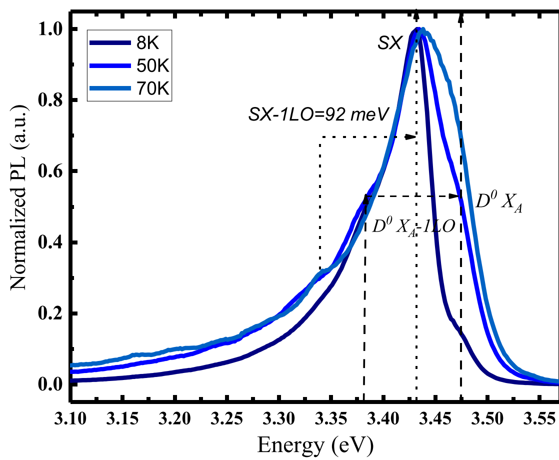
Fig. 2(a) presents the PL spectra of GaN NRs measured at temperatures from 8 K to 300 K. The PL shows a dominant peak at 3.43 eV, a shoulder of the D^0X_A emission at 3.47 eV and a lower energy shoulder at 3.38 eV corresponds to the D^0X_A-*1LO* phonon replica. The commonly reported PL spectra of bulk GaN are dominated by neutral donor bound exciton emission (D^0X_A) with a phonon peak corresponds to the recombination of D^0X_A exciton with emission of *1LO* phonons [18]. The dominant PL peak at 3.43 eV represented in Fig. 2(a) is attributed to emissions by excitons close to the surface and is referred as *SX*-surface peak in the following discussions. The PL of GaN NRs measured at 300 K is displayed in the inset of Fig. 2(a) and shows only the peak at 3.39 eV. The yellow luminescence caused by transitions between native defects and shallow donors is diminished compared with typical yellow GaN luminescence indicating the good quality of the grown samples [5,19,20].

The normalized PL spectra (with respect to the *SX*-surface peak) measured at 8, 50 and 70 K are displayed in Fig. 2(b) to clarify the relative temperature dependence of the *SX*-surface and D^0X_A emissions in this temperature range. The *1LO* phonon replica of the *SX*-surface peak is clearly represented at 3.34 eV. The dominant PL peak shows a small blue shift of 8 meV between 10 and 70 K. This small blue shift is frequently reported for D^0X_A "S-shape temperature dependence" and attributed to the thermal delocalization of carriers localized at potential fluctuations or surface traps [5,22,23]. The temperature dependence of the PL peak energy is a characteristic behaviour of localized carriers in semiconductors. At low temperatures, excitons are unable to escape from the local potential wells and hence the PL is dominated by the localized exciton recombination. With increasing temperature, the D^0X_A excitons are thermally released into free excitons resulting in an initial increase of the free excitons concentration up to a certain temperature where all the D^0X_A excitons will transfer into free excitons. The energy shift is small up to 100 K while the intensity damping is the effective parameter contributing to the spectra in this temperature range [21]. The thermal delocalization of carriers out of local potential minima into higher energy states results in the thermal quenching of the PL peak.

Fig. 3(a) shows the PL spectra measured at temperatures from 100 to 300 K to lighten the temperature dependence of the PL in the higher temperature range. The dominant peak is observed at 3.45 eV at 100 K in agreement with the free exciton peak of bulk GaN. The *1LO* phonon replica of the *SX*-surface peak is indicated by dotted arrows. The dominant peak shows redshift with increasing temperature due to the ordinary temperature-induced band gap shrinkage. Due to the red shift of exciton peaks with increasing temperature, overlapping of the free exciton peak and the lower energy peaks results in the wide width of the observed main peak. The phonon peak energy shows a small red shift with increasing temperature as commonly reported in literature [24]. The intensity of the phonon replica becomes remarkable at higher temperatures

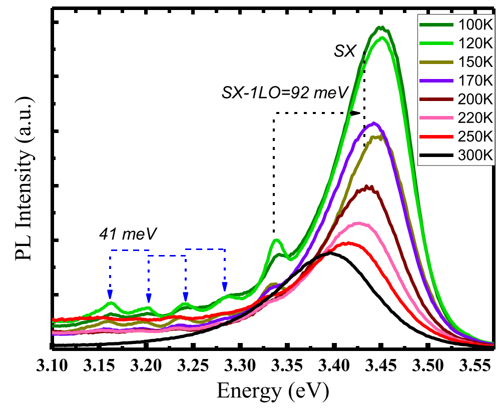


(a)

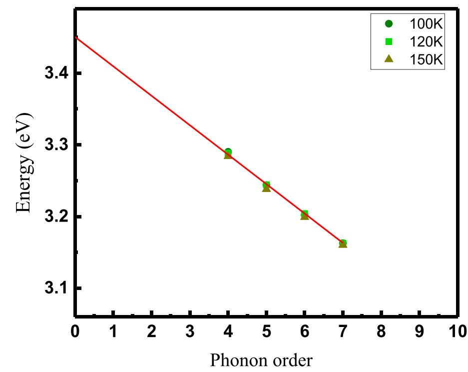


(b)

Fig. 2. **(a)** PL spectra measured at different temperatures from 8 to 300 K, from up to down, the represented PL intensity measured at 8 K is multiplied by 0.6; the inset show the PL of GaN NRs at RT. **(b)** Normalized PL spectra for the GaN NRs sample with respect to the *SX*-surface peak measured at 8, 50 and 70 K. The D^0X_A and D^0X_A *-1LO* are indicated by dashed lines, the surface *SX* peak and *SX-1LO* phonon replica are indicated by dotted lines,.



(a)



(b)

Fig. 3. **(a)** PL spectra for the GaN NRs sample measured at different temperatures from 100 to 300 K, from up to down; the dotted lines show the phonon replica *1LO* of the surface PL peak and the acoustic phonon replicas are indicated by dashed arrows. **(b)** A linear fit of the phonon replicas energy observed at the low energy side.

as a result of the thermal enhancement of the phonon-assisted excitonic transition probability.

The widely reported low-temperature PL of GaN is characterized by specific bands as follows: (i) a dominate near band edge (NBE) emission around 3.47 eV contributed to D^0X_A beside a higher energy shoulder of free-exciton emissions, (ii) a broad emission band around 3.26 −3.29 eV contributed to both donor–acceptor pair (DAP) and free electron–acceptor (e–A) recombinations; their 1LO phonon replica is around 3.18 eV (iii) a weak BL band around 2.9 eV in addition to a broad yellow-green band consists of GL band around 2.48 eV and YL band around 2.22 eV as well as RL around 1.8 eV. Most of these bands thermally quench at room temperature; only the red-shifted room temperature NBE around 3.41 eV and the broad yellow-green band (2.48 eV) can be observed [4,5,25]. The Y lines reported in GaN films or nanowires were ascribed to exciton-point defect complexes trapped at surface states [17,26–28]. The Y lines are mostly quenched when the temperature is increased to 60 K and almost disappear by 100 K.

GaN NRs grown on Si by MBE frequently show a strong emission around 3.45 eV at temperatures below 50 K and appears more strongly for the non-coalesced, low density and thin NRs (diameter < 50 nm) [4,14–16,19,29]. The peak intensity increases when the NRs average radius decreases. Some NRs exhibit luminescence at both D^0X_A transition and at the 3.45 eV transition while others show luminescence only at one of these lines. GaN NRs with large diameters (> 100 nm) do not show the 3.45 eV peak. Besides, the 3.45 eV line shows a spectral evolution with time [16]. Polarization-resolved luminescence and magneto-luminescence experiments attributed this emission to donor-bound excitons close to the surface [20]. Sample-dependent dominant peaks at energies from 3.405 to 3.421 eV, depending on the measured sample, were reported for GaN/Al_2O_3 NRs [26].

For our grown GaN NRs, the *SX*-surface peak shows peak intensity (I_s) more than six times the D^0X_A peak intensity (I_d) at 8 K indicating that excitons are mostly occupying the lower energy sites relative to the bulk donor bound sites D^0X_A. The integrated PL decreased to 0.4 of its value by increasing the temperature from 8 to50 K indicating a strong quenching associated with the reduction of ionization energy of defects and impurities in the vicinity of the surface. The ionization energy of impurities closer to the surface is always lower than that of the corresponding bulk defect due to the electrostatic potential change which the bound charge carrier experiences close to the surface. Recent tight-binding calculations showed that both the ionization of defects and the energy of excitons bound to these defects are altered in the vicinity of a surface [17,30]. The ionization energy of a donor located directly at the

surface is reduced to one fourth of that of the same donor in the bulk, assuming that the vacuum represents an infinite barrier.

The dominant peak shows a long tail in the lower energy range with a full width at half maximum around 60 meV. This slow damping of the PL intensity in a broad energy range infers a significant carrier population of the energy states occupying the lower energy band of the *SX*-surface peak. The wide tail of the main peak can be attributed to exciton scattered by acoustic phonons originating from the Fermi tail describing the thermal population of the exciton. The PL spectra of thin nanowires have frequently been reported to show a broadened tail toward the lower energy side attributed to the contribution of a large number of localized states due to the presence of potential fluctuations [31]. Here it is worth emphasizing that self-purification, known for NRs, enhances the tendency of both native point defects and impurities to accumulate close to the NR surface leading to a significant density of surface defect states. The surface effects will be significant when the exciton is close to the surface by less than the Bohr diameter describing the separation distance between the hole and the electron of the exciton (~ 6 nm). A surface shell around 6 or 7 nm is used to identify exciton surface states [14]. The emission transitions depend on the radial distribution of the exciton states along the NRs diameter, leading to a broad energy distribution rather than a well-defined energy. The smaller the NR radius, the larger the average influence of the surface on excitonic transitions.

Fig. 3(b) shows the energies of the phonon replicas displayed in Fig. 3(a). A linear fit of the phonon energies gives a slope of 41 meV in agreement with the observed Raman *B1l* silent mode [1]. The forbidden *B1l* mode was observed by resonant Raman scattering for the grown GaN NRs under resonance excitation using a laser line of 325 nm, the same excitation wavelength used for the PL spectra. The contribution of phonons to the temperature dependence of optical transitions and energy band gap of GaN stems from the electron-phonon interactions [32]. Two phonon energies of ~ 23 meV (186 cm^{-1}) and ~69 meV (557 cm^{-1}) have been reported by the two-phonon Pässler approach for the temperature dependence of GaN band gap [33]. The interactions between excitons and phonons ionize the exciton into a free electron and hole or scatter the exciton into discrete exciton states. The exciton lifetime is significantly reduced at high temperatures due to scattering by phonons.

The coupling strength between excitons and phonons is inferred from the Huang–Rhys factor (Sn) according to the equation $In = I_0 (Sn/n!)$ [34].

Here, n = 0, 1, 2, 3,... represent the number of LO-phonons involved in exciton recombination, *In* is the emission intensity of the nth phonon replica and I_0 is the intensity of the main emission line. Here the Huang–Rhys factor of *1LO* for the surface dominant PL peak of our grown GaN NRs shows a value of 0.32 at 100 K and increases to 0.38 at 120 K associated with a small red shift. The *Sn* values reported in literature for free exciton, D^0X_A and DAP recombinations are around 0.0077, 0.0013 and 0.7, respectively [19,24]. The larger *Sn* observed for the grown GaN NRs indicates the enhanced coupling between the LO phonons and *SX* peak.

The coupling strength increases with increasing the temperature due to an increase of the exciton kinetic energy which in turn increases the phonon assisted excitonic transition probability [35]. The *LO* phonon Fröhlich scattering dominates at high temperatures. The high density of defects or residual donors will reduce the phonon assisted free excitonic efficiency since scattering by impurities and defects will significantly reduce the emission intensity of phonon replicas [36]. The line width also increases linearly with temperature with a linear dependence of the integrated intensity ratio of the phonon replicas (*1LO/2LO*).

The observed peak energy of the free exciton tracks the temperature dependence of GaN bandgap and follows Varshni's equation [37].

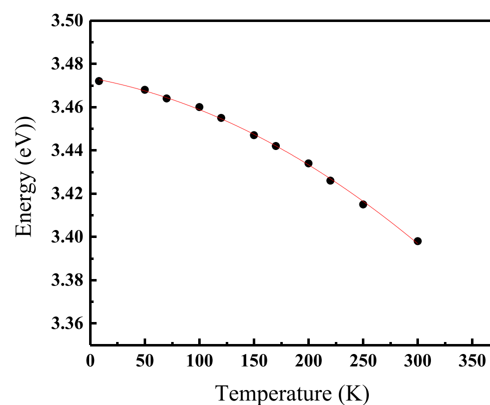


Fig. 4. Temperature dependence of D^0X_A peak energy, the solid line shows the Varshni equation fitting.

$$E(T) = E(0) - \alpha T^2/(\beta + T)$$

where *E(T)* and *E(0)* are the bandgap values at a temperature *T* and at 0 K, respectively; α and β are fitting parameters characteristic of a given material. The fitted values estimated from the PL temperature dependence shown in Fig. 4 are: *E(0)* = 3.473, $\alpha = 8.938 \times 10^{-4}$ eV K^{-1} and β = 680 K; in agreement with the previous work [4,5,17].

4. Conclusions

The enhanced emission of small size GaN NRs is discussed considering the enhanced surface effects and Fröhlich electron-phonon coupling. The silent B11 phonon replicas are well defined in the PL spectra up to 150 K; these phonon replicas have not been reported so far in the photoluminescence of GaN NRs to our knowledge. Stronger electron-phonon coupling is inferred from the calculated Huang–Rhys factor. The surface-related effects in addition to the enhanced electron-phonon coupling represented for small-size nanorods are essential factors to be considered for future application in nanotechnology like the enhanced photoemission of nanodiods. Moreover, these results clearly expose a key role played by size effects for the phonon assisted processes in nanostructures.

CRediT authorship contribution statement

M. Almokhtar: Conceptualization, Methodology, Writing – Original draft. **N. Abdell All:** Methodology, Visualization, Investigation, Writing – review & edit. **Jamal Q. M. Almarashi:** Visualization, Investigation, Writing – review & editing. **Hajime Asahi:** Methodology, Visualization, Writing – review & edit, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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