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RESEARCH ARTICLE

Size-dependent optical properties of InGaN quantum dots in GaN nanowires grown by MBE

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Abstract Quantum dots in nanowires (DINWs) are considered as important building blocks for novel nanoscale semiconductor optoelectronic devices. In this paper, pure axial heterojunction InGaN/GaN DINWs are grown by using plasma-assisted molecular beam epitaxy (PA-MBE) system. The InGaN quantum dots (QDs) are disk-like observed by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The diameter of QDs can be controlled by the growth conditions of nanowires (NWs), while the thickness of QDs can be controlled by the growth time of InGaN. Temperature-dependent photoluminescence (TDPL) measurements demonstrate that the PL peak of DINWs with small and uniform sizes shows a general red shift with increasing temperature. However, the PL peak of DINWs with non-uniform sizes shows an abnormal blue shift with increasing temperature, which is due to different internal quantum efficiencies of the DINWs with different sizes.

Keywords InGaN quantum dots (QDs), nanowires (NWs), photoluminescence (PL), molecular beam epitaxy (MBE)

1 Introduction

The spontaneously grown nanowires (NWs) with natural nanoscale diameters are extremely attractive for the fabrication of nanoscale devices. InGaN heterostructures and quantum dots (QDs) in GaN NWs have emerged as important building blocks for novel optoelectronic devices such as single photon emitters [1], light-emitting diodes [2] and nanolasers [3], because they have several advantages compared to the self-assembled InGaN QDs on GaN films. First the dot-in-a-nanowire (DINW) is more controllable in its dimensions, since the thickness and the diameter of the QD can be well controlled by growth time and NW diameter, respectively. Then the quantum confined Stark effect (QCSE) in DINW can significantly be reduced due to radial strain relaxation during epitaxy. The crystalline quality of DINW is relatively high also due to strain relaxation [4], leading to high internal quantum efficiency (IQE). Therefore, the InGaN/GaN DINWs are drawing more and more interests in the application of nanoscale optoelectronic devices.

In recent years, much efforts have been paid to understand the growth mechanisms of InGaN/GaN DINWs [5], indicating that the shapes of the InGaN dots depend closely on the growth conditions such as V/III ratio and growth temperature. Various devices have been demonstrated on DINWs [1,6–8], showing high IQE, low threshold currents or high temperature single photon emission. However, the comprehensive understanding on the relationship between the optical properties and the QD sizes is still lacking.

In this paper, InGaN/GaN DINWs with pure axial heterojunction have been grown on Si(111) substrates by plasma-assisted molecular beam epitaxy (PA-MBE). The influence of dot size on its optical properties has been investigated by temperature-dependent photoluminescence (TDPL) measurements, demonstrating that QDs with different sizes show different PL peak wavelengths and IQEs. The ensemble of DINWs with divergent diameter distribution presents an abnormal blue shift of PL peak with increasing temperature, due to the high IQE of small DINWs at high temperature.

2 Experiments

All InGaN/GaN DINWs in this work were grown on Si (111) substrates in an SVTA 35N PA-MBE system. The

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substrates were first thermally cleaned at 940°C, then the nitridation process was performed at 900°C for 120 s until the “8×8” reflection high-energy electron diffraction (RHEED) pattern disappeared, indicating the β-Si_3N_4 became amorphous SiN. The growth of GaN NWs was carried out at a substrate temperature in the range of 750°C – 770°C under N-rich condition, with the Ga source fixed at 1000°C and the N_2 flow rate maintained at 1.7 sccm. For the growth of InGaN dots, the growth temperature was decreased to around 580°C in order to enhance In incorporation. Finally GaN NW caps was grown on the InGaN dots.

The TDPL measurements were performed at temperature range of 8 – 300 K in a close-cycle cryostat. A HeGd laser with 325 nm emission wavelength was used to optically excite the DINWs, and the excitation power density was around 60 W/cm^2. The PL signal of DINWs was dispersed by a Horiba Triax550 monochrometer and detected by a photomultiplier. Scanning electron microscopy (SEM) was employed to analyze the morphologies of the NWs. High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and energy-dispersive spectroscopy (EDS) were used to analyze the shape, size and In content of the InGaN QDs.

3 Results and discussion

Figure 1(a) shows a typical cross-sectional SEM image of the as-grown InGaN/GaN DINWs in sample A. As seen from the image, GaN NWs around 350 nm in length were grown with c-axis aligned to the normal of the substrate. The average diameter of the NWs is about 35 nm, however the sizes of the NWs are not very uniform for the growth condition of sample A. The InGaN QD cannot be observed in the SEM image, but is clearly shown in the HAADF-STEM image in Fig. 1(b), where the disk-like QD with well-defined pure axial heterojunction is indicated by the bright contrast area near the NW top due to higher average atomic number of InGaN. The EDS measurement results in Fig. 1(c) from a line scan across the QD along the red arrow in Fig. 1(b) are also clear evidence of the realization of InGaN QD grown in the NW, as the count of Ga signal decreases while the count of In signal increases in the QD area. However, the spatial resolution of the EDS is not high enough to reveal the actual thickness of the InGaN QD due to the scattering of the electrons. The accurate size of the QD can be drawn from the STEM image, which shows that the diameter of the QD is about 35 nm, the same as the diameter of the NW, and the thickness of the QD is about 4 nm.

To study the influence of QD size on its optical properties, sample B with smaller InGaN QDs was prepared. The growth temperature of GaN NWs was increased by 20°C to reduce the average diameter of the

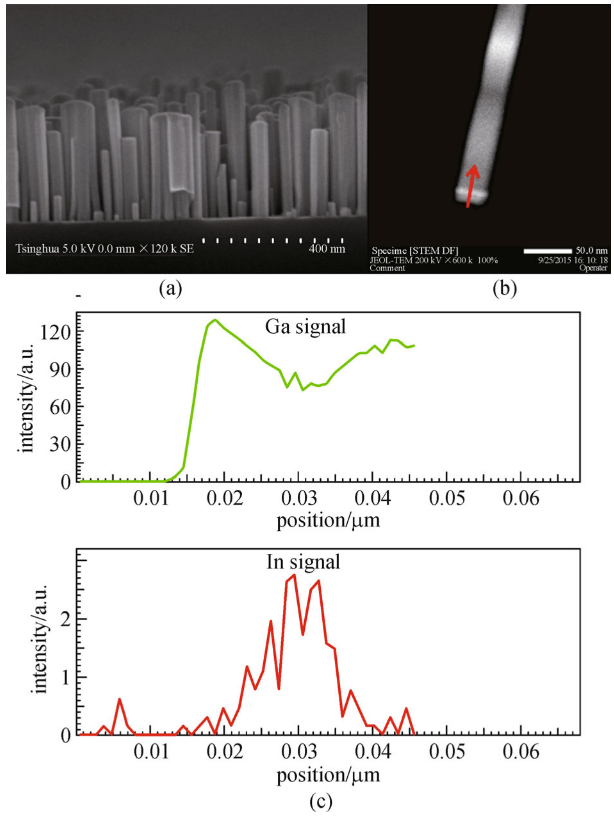

Fig. 1 (a) Cross-sectional SEM image of as-grown InGaN/GaN DINWs; (b) HAADF-STEM image of a single InGaN/GaN DINW, the area with bright contrast represents the InGaN QD; (c) EDS results of a line scan along the red arrow in (b), the green curve for Ga counts and the red one for In counts

NWs [9], resulting in an average NW diameter of 25 nm. The thickness of InGaN QDs was reduced to 2 nm by reducing the growth time, while the growth conditions were set the same as that for the growth of InGaN QDs in sample A. The distributions of the NW diameters of samples A and B are shown in Fig. 2. Due to the nature of spontaneous growth [9], larger average NW diameter usually means more heterogeneous diameter distribution, so the NW diameters in sample A are dispersed in a range from 15 to 85 nm. Compared to sample A, sample B exhibits more uniform NW diameter, and most of the NWs have diameters between 25 – 35 nm. In sample A, the NWs with diameters larger than 65 nm probably result from the coalescence of NWs, while sample B hardly shows NW coalescence since rising the growth temperature also reduces the NW density [10].

Figures 3(a) and 3(b) show the TDPL spectra of samples A and B with room temperature PL central peaks at 562 and 458 nm, respectively. Since the growth conditions for InGaN QDs of both samples are the same, the shorter PL peak wavelength of sample B can mainly be attributed to the enhanced quantum confinement and the weakened QCSE when the QDs get smaller in both diameter and

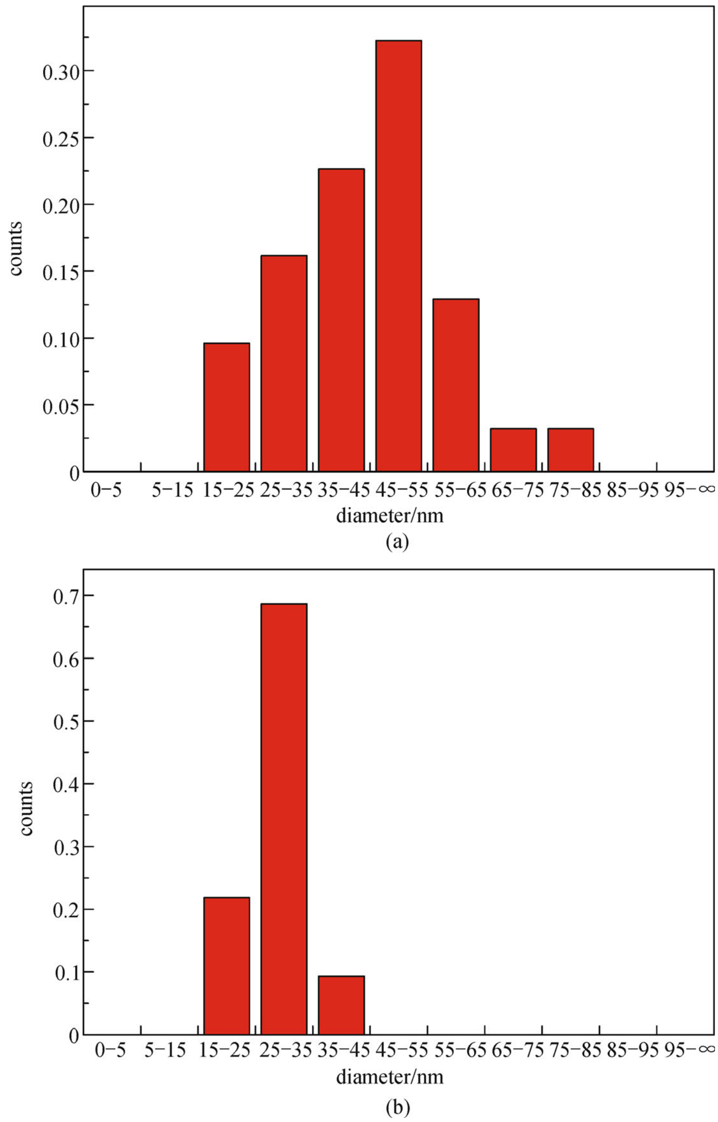

Fig. 2 Distribution of NW diameter in (a) sample A and (b) sample B

thickness. The full widths at half maximum (FWHM) of PL spectra of samples A and B are around 75 and 50 nm at 8 K, respectively. The broadening of PL peak of sample A is a result of the wider distribution of the DINW diameters.

The integral PL intensities and peak energies versus temperature are plotted in Figs. 3(c) and 3(d), respectively, for samples A and B. The PL intensities of both samples drop gradually with increasing temperature in low temperature range, but drop relatively faster when above 100 K. This can be explained as below. In the QDs, the excitons are strongly confined at low temperature, the PL intensity decreases a little with temperature. However at high temperature above 100 K, the excitons are no longer strongly confined due to thermal excitation, therefore the PL intensity drops fast. The PL peak energies of both samples exhibit little change in low temperature range. When the temperature is above 100 K, however, the behaviors of samples A and B are quite different. For sample B, the PL peak shows red shift with increasing temperature due to the bandgap shrinkage; for sample A, in contrast, the PL peak presents an abnormal blue shift. This can be explained by the different IQEs of QDs with different sizes, as discussed below.

It is commonly believed that InGaN QDs with diameter below 30 nm have quasi-three dimensional quantum confinement effect (3D QCE) [11]. As demonstrated in Fig. 2(a), QDs with diameter below 30 nm account for nearly 30% in sample A. Therefore the QDs can be divided into two types: QD-Is with smaller size as well as quasi-3D QCE, and QD-IIs with larger size as well as weak 3D QCE. Meanwhile, the low- temperature PL spectrum of sample A can be deconvolved to two Gaussian peaks, as shown in Fig. 4. Peak 1 presents a shorter wavelength and lower intensity, while Peak 2 presents a longer wavelength and

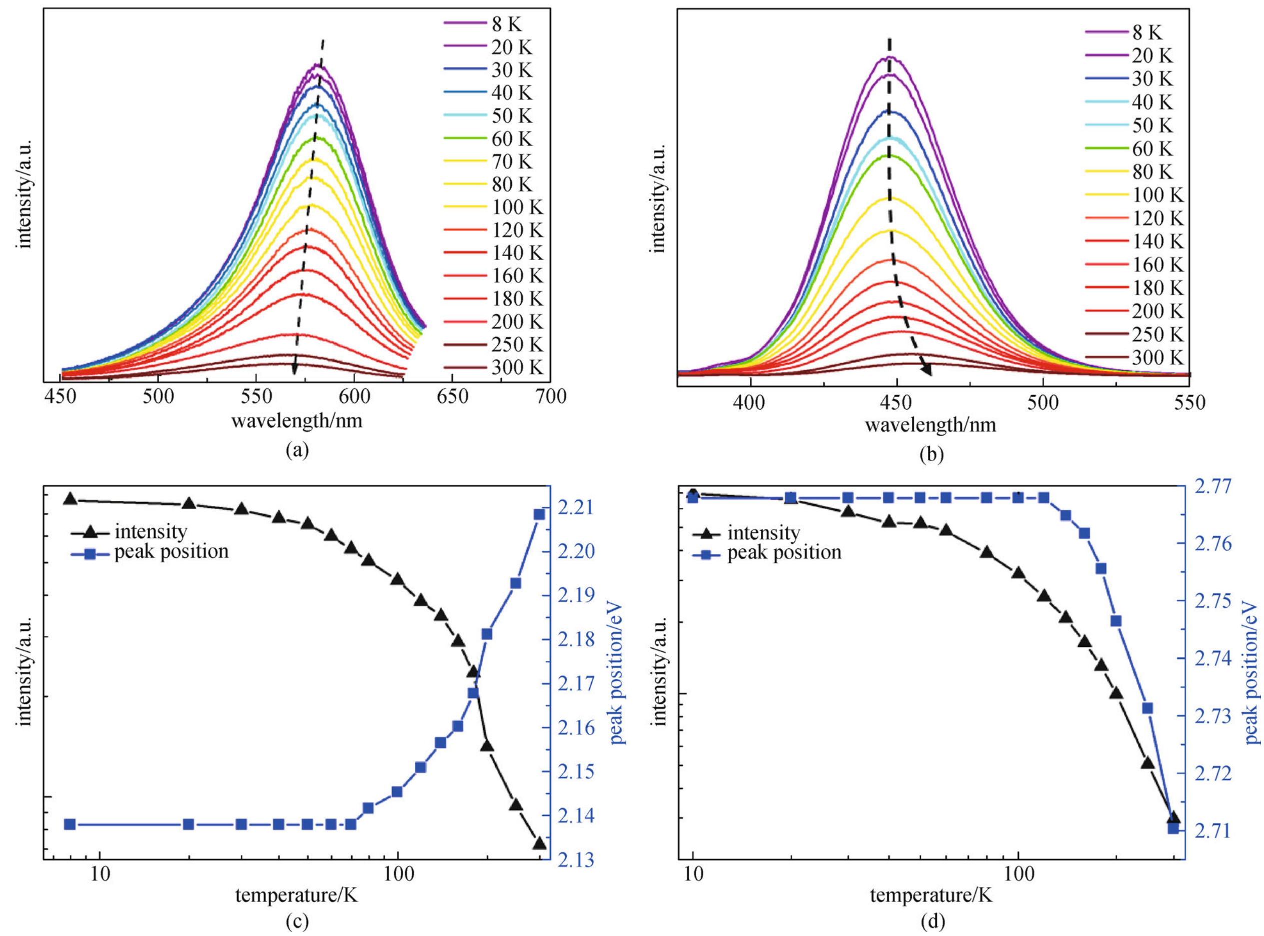

Fig. 3 (a) and (b) are TDPL spectra of samples A and B; (c) and (d) are the integral PL intensities and peak energies versus temperature for samples A and B

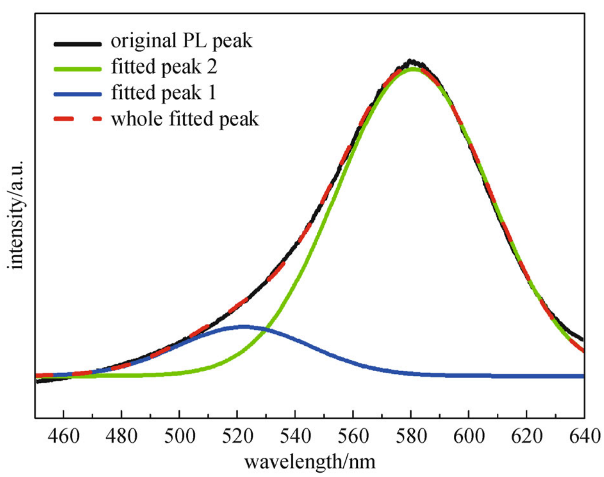

Fig. 4 Low temperature PL spectrum of sample A fitted by two Gaussian peaks

higher intensity. The integral intensity of Peak 1 is about 22% of the whole PL peak. Therefore, it can be considered that Peak 1 corresponds to the contribution from QD-Is and Peak 2 corresponds to QD-IIs, since small QDs with stronger QCE have shorter PL peak wavelength and account for a small percentage as discussed above. Given that the IQE is 100% at 8 K, QD-Is and QD-IIs present IQEs of 18.7% and 8.2%, respectively, at room temperature. That means in high temperature range, the PL intensity of QD-Is decreases slower than QD-IIs, thus the blue shift of the ensemble PL spectrum is observed. On the contrary, as shown in Fig. 2(b), sample B possesses narrow distribution of QD diameter and most of the QDs are small enough to have quasi-3D QCE, therefore the PL spectra present nearly single Gaussian peaks and a general red shift with temperature is observed.

4 Conclusion

In summary, InGaN/GaN DINWs have been grown by PA-MBE, SEM and TEM measurements show that well-defined InGaN QDs are embedded in GaN NWs with the same diameter as the NW. TDPL measurements show that, the PL spectra of the QDs with uniform sizes present red shift with increasing temperature, while the PL spectra of the QDs with non-uniform size present abnormal blue shift with increasing temperature due to different IQEs of QDs with different sizes.

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