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A polarity-driven nanometric luminescence asymmetry in AlN/GaN heterostructures

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Group III Nitrides nanowires are well suited materials for the design of light emitting devices. They have many applications for light emission. In particular, nanowires (NW) of these materials are good candidates for device design, considering their defect free crystal structure and possible light guiding capabilities. Axial confinement due to heterostructures (such as AlN/GaN/AlN quantum wells, QW) allows the fine tuning of light emission by controlling the exciton binding energy. These materials and their nanostructures are typically grown with the wurtzite structure along the [0001] direction, resulting in spontaneous and piezoelectric polarizations. The internal electric field due to heterostructures (such as AlN/GaN/AlN quantum wells, QW) allows the fine tuning of light emission by controlling the exciton binding energy. These materials and their heterostructures are typically grown with the wurtzite structure along the [0001] direction, resulting in spontaneous and piezoelectric polarizations. The internal electric field caused by polarization greatly affects the optical properties of QWs. Manipulation of this field by applying an external potential on GaN quantum discs allows control over the disc emission wavelength. The internal field can be assessed using electron holography. However, techniques allowing nanometer scale characterization are required to probe the effects of such field on optical properties at sub-wavelength scales. Recently, it has been demonstrated that cathodoluminescence (CL) in a scanning transmission electron microscope can be used to characterize the emission properties of individual QWs in NWs with a spatial resolution of the order of 5 nm. Also, the collection efficiency of this system is sufficiently high to detect single photon emission from Nitrogen-Vacancy (NV) centers in diamond nanoparticles.

Here, we have used the same experimental approach to characterize the excitation probability at the nanometer scale of a single GaN QW in an AlN NW, grown in the [0001] direction. We have observed that the excitation probability is highly asymmetric along the [0001] direction, which we attribute to the influence of the internal electric field on charge carriers. In fact, CL experiments have been routinely used to measure carriers’ diffusion length in different semiconductor heterostructures, including NW. The improved spatial resolution here allows the observation of the asymmetry described.

Cathodoluminescence experiments have been performed in a VG HB 501 scanning electron microscope operated at 60 kV (1 nm wide electron beam with typical current 50 pA). Light emitted by the sample has been collected using a system which is described elsewhere. Light spectra have been measured using an optical spectrometer (300 grooves diffraction grating blazed at 300 nm, 4.133 eV) equipped with an EMCCD (electron multiplying charge coupled device). Spectrum images have been acquired by scanning the electron beam in a 2 dimensional array and acquiring at each pixel the spectrum of emitted light. In parallel to spectrum images, annular dark field (ADF) images have been acquired. These contain, basically, information from the average projected (along the electron path) mass and can be used to distinguish GaN and AlN. Alternatively, energy filtered intensity images (2 dimensional images of the total light intensity in a fixed energy range) have been acquired using a photomultiplier tube and a bandpass optical filter (3.647 eV, 340 nm, to 4.133 eV, 300 nm, range). Atomically resolved ADF images and chemical maps using electron energy loss spectroscopy (EELS) have been acquired using the aberration corrected USTEM Nion 200 electron microscope at 200 kV. Convergent beam electron diffraction (CBED) experiments have been performed in an Akashi Topcon EM-002B at 100 kV.

Nanowires were grown by catalyst free plasma-assisted molecular beam epitaxy (PA-MBE) on Si [111]. Growth was performed at 830 °C. Short GaN NWs were used to start growth, followed by AlN NWs. A thin GaN QW was grown on top of the AlN NW, which was finally capped by AlN. The end result has been an AlN/GaN/AlN heterostructure, as shown in Fig. 1(a).

Energy filtered CL intensity images of individual GaN QWs in AlN NWs (Figs. 1(a) and 1(b)) show an asymmetry
of the total emission intensity between both sides of the quantum well. This behavior is observed on all NWs from this sample containing single QWs.

GaN NWs grown by PA-MBE on Si (111) are known to be of wurtzite symmetry, with the [0001] axis parallel to [111] Si axis.\textsuperscript{12,17} As this structure is not centrosymmetric, it gives rise to internal spontaneous and piezoelectric fields, which govern to a large extent the optical properties of NW heterostructures.\textsuperscript{14} Moreover, specific surface states are expected to appear depending upon the chemical nature (N or metal terminated) of the upper surface.\textsuperscript{15} In practice, it has been demonstrated that the experimental conditions used in the present work lead to the formation of N-polar NWs.\textsuperscript{12} This feature has been confirmed by CBED measurements shown in Fig. 1. Comparison to simulations (using JEMS software\textsuperscript{16}) demonstrates that the NWs heterostructures under study are N-polar (Fig. 1(d)). High resolution ADF images (Figs. 1(e)–1(i)) show that the QWs are epitaxial, interfaces have similar and abrupt chemical profiles (Ga to Al transition). Intensity maps have been measured using the 2-area-fitting method for background estimation\textsuperscript{17} for the Ga L and Al K edges (Fig. 1(i)).

An average spectrum from the region (80 nm × 80 nm area) of highest CL intensity shows three distinct peaks at 3.83 eV, 3.74 eV, and 3.67 eV (324.2 nm, 331.8 nm, and 338.3 nm, Fig. 2(a)). The first peak can be attributed to the emission originated from the confined exciton in the GaN QW. The other two peaks, with energy differences to the main peak of 90 meV and about 160 meV, can be attributed to 1 LO (longitudinal optical) and 2 LO phonon replica.\textsuperscript{18} No emission from AlN has been observed; either the near band emission at 6.25 eV (Ref. 19) or the violet band\textsuperscript{20} (inset Fig. 2(a)). The yellow band emission from GaN has not been observed either.\textsuperscript{20} This indicates that all light detected originates from the QW. Therefore, when the electron probe is away from the QW, luminescence occurs due to minority carriers’ diffusion and drift.\textsuperscript{21}

In addition to the excitation asymmetry, 2D spectrum images (Fig. 3) evidence two other effects: a small energy shift of 0.008 eV (between 3.831 eV and 3.823 eV) and a full width at half maximum (FWHM) change of 27 meV (between 71 meV and 44 meV) of the QW emission peak (the reason for these is the appearance of a supplementary peak, as described later). These effects, along with the excitation efficiency described above, are shown in total intensity, peak energy position, and FWHM maps retrieved from Gaussian curves fits to the data (Figs. 3(a)–3(c)). In these maps, the NW contour and the QW positions are marked by white lines.

In Fig. 3(d) the spectral shift and the apparent increase of the FWHM can be seen in two spectra with similar intensities taken from different sides of the QW. To understand the FWHM increase we have fitted the two spectra using 2 Gaussian curves. The result is a good fit on the right and a bad one (large residue) to the left side. On the left (right) side the QW the surface is Ga-polar (N-polar). The residue decreases if 3 Gaussians are used to fit the spectrum from the left side. This behavior is true for all spectra on this side of the QW. We believe that the increase in FWHM and the shift occur due to the appearance of a different emission peak which we cannot resolve at our current spectral resolution and temperature. The same excitation probability asymmetry

![Figure 1](image1.jpg)

**FIG. 1.** (a) ADF image of a NW (growth direction from left to right) containing a QW. (b) CL emission intensity acquired in parallel to (a). An asymmetry of the total emitted intensity can be seen. The scale in (b) is the same as in (a). (c) and (d) Experimental and simulated CBED patterns of the NW shown in (a). They show that the growth direction is the [0001] with N termination (scale bars are 2 nm\textsuperscript{-1}). (e) and (f) High resolution ADF image of a typical QW. (g) ADF image and EELS chemical map of the same QW. The Ga-L and Al-K edge intensities are represented in yellow and purple, respectively. (h) Line profiles from the bottom to the top of (g). (i) Typical EELS spectrum showing integration windows. In (g)–(i), the growth direction is upwards. The scale bars in (g) and (f) are 0.5 nm.

![Figure 2](image2.jpg)

**FIG. 2.** (a) CL Spectrum of the QW shown in Fig. 1. Three peaks can be seen at 3.83 eV, 3.74 eV, and 3.67 eV. They are attributed to the exciton confined in the GaN quantum well, the 1 LO and the 2 LO emission lines. No emission from the AlN has been observed in this sample (inset).
has been observed in other NWs. Energy shifts have also been observed. However, in some NWs, the energy shift occurs to higher energy.

To understand the presented effects we must take into consideration two characteristics of the analyzed material: (1) the GaN QW is unintentionally n-type doped and (2) the non-centrosymmetric crystal structure leads to an internal electric field. The first point suggests that CL is controlled by hole (minority carrier in the GaN QW) dynamics. The direction of the internal electric field dictates how holes drift in the material. The direction of the electric field was determined from CBED measurements and a detailed theoretical model of a similar heterostructure developed by Mojica and Niquet.15 These elements have allowed us to use a simple model to explain the observed differences in excitation efficiency. In Fig. 4(a), we show a sketch of the conduction and valence bands across the QW region. On the right side (N-polar) of the QW holes drift towards the QW, while on the left (Ga-polar) they drift away from the QW, due to the electric field. Hence, for a constant electron current, we expect more holes (and, thus, higher emission intensity) to reach the QW when the beam is located at a given distance from the center of the QW on its right than on its left. To confirm this hypothesis, we have solved the one dimensional drift-diffusion equation for holes using the linear finite difference method. The number of holes expected to reach the QW can then be estimated from the density of holes at a fixed time. The best solution has been found with the parameters for AlN: $D_h = 2 \times 10^{-4} \text{m}^2/\text{V} \cdot \text{s}$ (diffusion coefficient for holes), $\mu_h = 3 \times 10^{-5} \text{m}^2/\text{V} \cdot \text{s}$ (hole mobility), $\tau_h = 18 \text{ ps}$ (hole non-radiative lifetime), and $E = 5 \times 10^6 \text{ V}/\text{m}$, $D_h$ and $\mu_h$ have been chosen to be close to typical values for a thin film of AlN. $\tau_h$ has been chosen to match the length of the tail of the emission profiles (these can be fitted with exponential with decay constants 18 nm and 22 nm). Finally, $E$ has been chosen to match the asymmetry (it is close to expected values, considering the size of the AlN section). The qualitative agreement (Fig. 4(b)) shows that the simple model captures the physical origin of the observed asymmetry. The direction of the effect is insensitive to the parameters magnitude in a large parameter range. A more precise knowledge of $D_h$ and $\mu_h$ for NWs would allow a measurement of the magnitude of the electric field outside the QW. Alternatively, the electric field could be measured using electron holography,5 as in Ref. 6, allowing an estimate of $D_h$ and $\mu_h$.

This model cannot explain the appearance of an additional peak observed in the emission to the left of the quantum well. We should note that, according to our simple model, the energy at which the holes arrive at the QW from the left and from the right is different. We speculate that this might result in the excitation of different energy levels of the exciton inside the QW, resulting in different emission peaks and possibly energy shifts.

To conclude, we have investigated the reasons behind the highly asymmetric excitation probability of GaN QWs in AlN NWs. Our interpretation is based on the effect of the
internal electric field on hole drift. A qualitative model has been constructed based on nanometer resolved structural and luminescence measurements. We believe that the observed phenomenon could be used to determine the polarity of various group III Nitrides and III-V material with known doping. However, the presence of piezoelectric field may complicate such interpretation. In this case, the use of CBED for polarity measurements would allow the detection of large strain fields (which induce piezoelectricity) through the observation of reversed asymmetries in CL maps. Finally, a precise knowledge of AlN electrical properties would allow the measurement of the internal electric field.

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