Probing Local Emission Properties in InGaN/GaN Quantum Wells by Scanning Tunneling Luminescence Microscopy

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1. Introduction

III-nitride heterostructures have found wide use in optoelectronic devices, in particular as energy-efficient light sources. However, although the nitride light-emitting diodes (LEDs) made of InGaN/GaN quantum wells (QW) have outstanding performance at low injection current and low indium content, the extension of nitride light-emitting devices to high current densities and long wavelengths is still challenging. When compared to other conventional III–V semiconductors such as GaAs, the crystalline quality of nitride materials is poor, with large dislocation and defect densities. Furthermore, InGaN/GaN QWs present broad photoluminescence (PL) spectra compared to conventional semiconductors. The origin of this inhomogeneous broadening is still debated and may be due to both small- and large-scale composition fluctuations arising from intrinsic alloy disorder and extrinsic causes such as clustering or large-scale variations in indium incorporation related to defects and morphology. For example, near-field PL mapping indicates fluctuations in the emission properties related to composition fluctuations at the micrometer scale and underlines the link between the indium incorporation and the surface morphology in InGaN thick layers. Another near-field technique based on scanning tunneling electroluminescence (STL) microscopy was used to identify the origin of PL broadening in nitride materials. However, experiments were performed on inappropriate heterostructures with multiple QWs or a single QW too far from the surface which led to degraded spatial resolution, allowing only for emission fluctuations at large spatial scales to be observed. More recently, STL has nonetheless shown its capability to assess smaller-scale sources of PL broadening.

2. Experimental Section

In this work, we used STL microscopy to map the optoelectronic properties of an InGaN/GaN QW specifically designed to permit electroluminescence imaging down to the nanometer scale. The technique (schematized in Figure 1a) relies on the use of a scanning tunneling microscope (STM) tip both to image the surface topography and to locally inject electrons into the p-doped single In$_{0.23}$Ga$_{0.77}$N QW located only 30 nm below the surface. Electron recombination with majority holes in the p-type InGaN QW resulted in light emission, which was collected and analyzed by a spectrometer. Figure 1b shows a schematic view of the...
The spatial resolution of the STL measurement depends on several factors. First, the resolution of the electron injection at the surface, which can be evaluated by the quality of the topography image, is typically smaller than 1 nm. Then, the electrons form an injection cone whose angle is given by momentum conservation during the crossing of the near-surface band bending region of ≈4 nm width due to the aforementioned p-type doping. An estimation of an upper limit for this angle is $\pi/4$ radians, which corresponds to a radius of the injected area. Finally, 2D lateral transport inside the QW could occur before the recombination process, with a diffusion length that depends on the properties of the QW and on the temperature. More precise information on the actual resolution of the measurement was obtained through the analysis of the experimental results as described.

### 3. Results

The experiment was performed in an ultrahigh-vacuum (UHV) STM, with chemically etched tungsten tips. A typical STM topography image taken at 100 K is shown in Figure 2a for a tunneling voltage of 4.4 V and a tunneling current of 0.5 nA. Note that STM on large-bandgap semiconductors requires the use of severe injection conditions. A bias voltage of several volts had to be applied between the tip and the metallic contact at the sample surface, with particularly high values at low temperature due to contact and access resistance. Despite this, excellent topographic images could be obtained. The topography image in Figure 2a shows a structure of atomically flat terraces separated by mono- or biatomic steps arising from the substrate miscut. A 150 nm-wide hexagonal hole is also observed in the field of view. Two topography profiles along lines labeled 1 and 2 are shown below the STM image. Profile 1 shows the 80 nm-wide atomically flat terraces separated by atomic steps. Profile 2, taken across the hexagonal hole, shows that this defect is 2.5 nm deep and has a flat bottom. Note that the topography profile inside the hole exhibits atomic steps, which indicates that the STM tip is indeed in tunneling contact at the bottom of the hole.
Based on the morphology, we believe that the sidewall facets are either \( f_{110} \) or \( f_{111} \) as these are stable natural facets for GaN.\(^{[20]}\) Also, the intentional miscuts of the sapphire substrate yield \( a \)-direction \( \frac{1}{3} \langle 1 \overline{1} 20 \rangle \) steps which are approximately parallel to two of the edges of the hexagonal defect (step line direction from the upper left corner toward the lower right corner of the STM image in Figure 2). A possible origin for this defect could be a polarity inversion region with a reversed pyramidal shape that has grown in the GaN \( p \)-type layer present below the QW, inducing different growth rates on top of it and leading to a hexagonal flat-bottom hole in the topography.\(^{[21]}\) Such a hole would then lead to a discontinuity of the QW position and probably to variations of the QW thickness on its edges.

The electroluminescence measurement was then carried out on the same area, which was easily recognizable thanks to the wide defect. A set of 1024 electroluminescence spectra were recorded on a \( 32 \times 32 \) array of points regularly spaced in the \( 500 \text{ nm} \times 500 \text{ nm} \) scanned area. The light emission was measured with a spectrometer, with a spectral resolution of 15 meV and an integration time of 10 s per spectrum. The tunneling current was set to 4 nA, to insure a sufficient signal-to-noise ratio with a reasonable acquisition time. Reducing the temperature to 100 K was also essential to increase the electroluminescence signal, thanks to a strong reduction in nonradiative processes. The 32 spectra along the scan lines number 3 and 12 (indicated on the topography image) are shown in Figure 2b. Surprisingly, in addition to the expected green emission peaked at about 2.33 eV, which is observed almost over the entire sample area, several distinct, highly localized, contributions appear, including an intense blue emission peaked at about 2.85 eV.

As an illustration, two individual spectra, numbered 78 and 497, are plotted in Figure 3. The corresponding positions where these spectra were recorded are indicated on the topography image of Figure 2a. These selected spectra exhibit four emission peaks, respectively, at 2.33, 2.44, 2.63, and 2.85 eV, which are representative of the four main emission peaks present in the scanned area. Each of them is accompanied by a phonon replica, appearing 91 meV lower in energy. To extract quantitative information on the intensity, peak energy, and width of these four contributions, data fitting was performed using pairs of Gaussian functions. The two Gaussians of each pair represent a peak and its phonon replica, have identical widths, and are separated by 91 meV. Respective fits for the two spectra 78 and 497 are also shown in Figure 3.

4. Discussion

Figure 4 shows maps of intensity, peak energy, and FWHM of the four contributions present in the scanned area, obtained by fitting all spectra as explained earlier. The intensity maps are plotted in logarithmic scale because of the large intensity fluctuations.
Furthermore, the local emission spectrum exhibits nearly two times smaller than the width of the PL spectrum. Figure 1c, its lineshape is well fitted by a single Gaussian function plotted in red, green, blue, and gray, respectively. For each contribution, the Gaussian functions used to fit the main peak and its first phonon replica are respectively plotted as solid lines and dashed lines of the same color.

As already mentioned, the contribution at about 2.33 eV corresponds to green light emission, which is the expected emission from the 3 nm QW with 23% indium content. It is present on the whole scanned area, and its phonon replica has a relative intensity of about 0.22, in agreement with already reported values obtained by PL spectroscopy measurements performed on green-emitting InGaN QW. This emission line corresponds to the dominant contribution in the PL spectrum. However, in contrast with the PL spectrum shown in Figure 1c, its lineshape is well fitted by a single Gaussian function (and its phonon replica), with FWHM of about 70 meV, nearly two times smaller than the width of the PL spectrum. Furthermore, this local emission spectrum exhibits fluctuations in intensity, energy, and width both at large (100 nm) and small (a few nm) spatial scales. These fluctuations do not appear to be correlated to the surface morphology. Note that the energy and spatial scales of the large-scale fluctuations seem compatible with measurements reported in the literature using near-field PL mapping.

The emission characteristics of this 2.33 eV contribution close to the hexagonal defect are also interesting. The emission energy is similar inside and outside the defect, but the spatial fluctuations of the peak energy are much smaller inside than outside the defect. Moreover, the emission energy map presents sharp edges that have the same shape as the defect and are shifted by only 20 nm in position. This indicates that there is no transport of electrons from inside to outside the defect and that the defect exists nearly exactly at the same spatial position in the QW as at the sample surface. This is compatible with the aforementioned possible origin of this hexagonal defect as being related to a polarity inversion region which would isolate the QW in the defect area from the surrounding QW area.

The three other contributions are very surprising as they correspond to emission at much higher energies than the green emission. Moreover, they can be very intense and spectrally narrow. Finally, as shown in their intensity, peak energy, and FWHM, they are observed in regions of small spatial extension (about 100 nm) near the hexagonal defect edges. However, although these contributions can be very intense in the local electroluminescence spectra, they are not visible in the macroscopic PL measurement presented in Figure 1, probably because they represent only a small fraction of the total emitted light, arising from a small fraction of emitting area.

We can distinguish two different behaviors. Contributions at 2.63 and 2.85 eV both appear at a fixed energy and width. They most probably arise from a modification of the QW on the edges of the defect, due either to thinning of the QW (2.63 and 2.85 eV would correspond to QW thicknesses of 2 and 1.25 nm, respectively, as estimated using an open-access 1D Poisson–Schrödinger solver or to a lower indium incorporation. When injection occurs outside the defect, the electron states leading to this high-energy emission are accessible from a distance of about 80 nm. This allows an estimate of the electron diffusion length as being shorter than 50 nm, taking into account that the radius of the electron injection area in the QW is of about 30 nm. This value of the diffusion length agrees with reported data in the literature. However, when injection occurs inside the defect, there is no diffusion to these states, which might be because the QW in the defect area is isolated from the surroundings. A surprising feature is that both peaks have very different phonon replica intensity, which suggests that they correspond to electronic states with very different localization lengths.

In contrast to this, the 2.44 eV contribution which appears along the bottom edge of the defect shows a significant gradient in emission energy over a few tens of meV. Furthermore, the central point of this gradient is positioned at about 100 nm from the defect edge. This additional contribution most probably does not originate from a fluctuation in QW width at the defect edge. Rather, it might be due to a gallium-rich region, whose formation could have been induced by the presence of the...
defect. Note that in all the regions where high-energy emission occurs, the main green emission is most often still present but less intense (Figure 4, left), which means that whatever the injection position, some of the injected carriers diffuse to green regions.

Finally, the electroluminescence map can be used to extract a lot of information about the QW structure close to the defect. Indeed, it shows that the main light emission inside and outside the defect is in the green spectral region and corresponds to the nominal QW thickness of 3 nm and indium concentration of 23%. On the edges of the defect, states higher in energy exist, which leads to an emission in the blue. Injected electrons can diffuse to these states only when they are injected outside the defect. Figure 5 shows a schematic of the QW structure close to the defect that can be deduced from the combination of the STM topography measurement and the light emission properties. From this schematic, it is clear that, due to the polarization field in the QW that confines electrons on the upper side of the well, electrons wavefunctions outside and inside the defect are spatially separated. This can explain the absence of transport between outside and inside the defect and the discontinuity in the green emission properties observed in the maps of Figure 4.

5. Conclusion

In this work, we show that STL microscopy can be used to image and quantitatively analyze the local recombination properties of
an InGaN QW down to the nanometer scale. We can distinguish large- and small-scale fluctuations in the emitted light spectrum and correlate them with the surface topography. The recombination properties in the vicinity of structural defects can be investigated in detail, as is done here in the case of a shallow hexagonal defect with a flat bottom visible in the surface topography. The resolution of the measurement is limited by the transport of carriers inside the material, which can be strongly position dependent, as is shown with the injection of carriers inside or outside the defect. The imaging of regions with specific emission properties allows an estimate of the electron diffusion length of a few tens of nanometers in the QW. Finally, the possibility to meter scale, in various congestions recombination properties in semiconductors down to the nanometer scale, opens the way to the observation of the radiative properties allows an estimate of the electron diffusion length of a few tens of nanometers in the QW. Finally, the possibility to meter scale, in various congestions recombination properties in semiconductors down to the nanometer scale, opens the way to the observation of the radiative fluctuations in emission arising from extrinsic causes, such as defects, step edges, or large-scale variations in the indium incorporation, or from intrinsic properties such as alloy disorder.

Keywords
electroluminescence, nitrides, scanning tunneling microscopy

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.