Exciton dynamics at a single dislocation in GaN probed by picosecond time-resolved cathodoluminescence
W. Liu, J.-F. Carlin, N. Grandjean, B. Deveaud, and G. Jacopin

Citation: Applied Physics Letters 109, 042101 (2016); doi: 10.1063/1.4959832
View online: http://dx.doi.org/10.1063/1.4959832
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/109/4?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Exciton recombination dynamics in a -plane (Al,Ga)N/GaN quantum wells probed by picosecond photo and cathodoluminescence
J. Appl. Phys. 107, 043524 (2010); 10.1063/1.3305336

Low-temperature time-resolved cathodoluminescence study of exciton dynamics involving basal stacking faults in a -plane GaN

Time-resolved cathodoluminescence of Mg-doped GaN

Time-resolved cathodoluminescence and photocurrent study of the yellow band in Si-doped GaN
J. Appl. Phys. 94, 2341 (2003); 10.1063/1.1592296

Carrier relaxation and recombination in an InGaN/GaN quantum well probed with time-resolved cathodoluminescence
Appl. Phys. Lett. 73, 1430 (1998); 10.1063/1.121966
Exciton dynamics at a single dislocation in GaN probed by picosecond time-resolved cathodoluminescence

W. Liu,a J.-F. Carlin, N. Grandjean, B. Deveaud, and G. Jacopina)

Institute of Physics, École Polytechnique Fédérale de Lausanne (EPFL), Lausanne, Station 3 CH-1015, Switzerland

(Received 7 April 2016; accepted 14 July 2016; published online 26 July 2016)

We investigate the dynamics of donor bound excitons (D*XA) at T = 10 K around an isolated single edge dislocation in homoepitaxial GaN, using a picosecond time-resolved cathodoluminescence (TR-CL) setup with high temporal and spatial resolutions. An ~1.3 meV dipole-like energy shift of D*XA is observed around the dislocation, induced by the local strain fields. By simultaneously recording the variations of both the exciton lifetime and the CL intensity across the dislocation, we directly assess the dynamics of excitons around the defect. Our observations are well reproduced by a diffusion model. It allows us to deduce an exciton diffusion length of ~24 nm as well as an effective area of the dislocation with a radius of ~95 nm, where the recombination can be regarded as entirely non-radiative.

To go beyond such limitations, picosecond time-resolved CL (TR-CL) has been proposed and used to investigate the exciton diffusion in InGaN/GaN quantum well (QW), core-shell nanowires, and bulk GaN on free standing substrate. Here, thanks to a picosecond TR-CL setup combining high temporal (<20 ps), spatial (~50 nm for the secondary electron image), and spectral (~3 meV) resolutions, we investigate the exciton dynamics at T = 10 K on a single dislocation in bulk GaN grown by MOVPE. We obtain the distributions of emission peak energies, effective lifetimes, and CL intensities around such a dislocation. By fitting our experimental data with a simple diffusion model, we are able to define the type of dislocation and extract the R_eff and diffusion length of exciton. These results give precious information about the carrier dynamics around a dislocation and confirm the relevance of such a technique to elucidate the influence of dislocations on GaN-based LEDs. The structure of the sample is the following: 2 μm GaN layer on a free standing GaN (0001) substrate with non-uniform dislocation density (ranging from ~10^6 cm^-2 to ~10^8 cm^-2), followed by a capping layer of 10 nm Al_{0.05}Ga_{0.95}N used as a protecting layer to avoid surface recombination. In Fig. 1(a), temperature dependent photoluminescence (PL) demonstrates that donor bound exciton D*XA (3.473 eV at 15 K) dominates the spectrum in the range of 15–50 K, whereas free exciton FXA gradually thermally activates and only becomes predominant above 70 K. The strong luminescence from D*XA most probably comes from the high n-type doping (≥2.5 x 10¹⁸ cm⁻³) inherited from the GaN substrate. Fig. 1(b) shows the CL intensity mapping of the (0001) surface of the sample in continuous excitation mode at T = 10 K, and the linear grayscale indicates the CL intensity. The mean electron current is around 5 nA and the acceleration voltage is 6 kV. Such an acceleration voltage has been chosen as a trade-off between the CL interaction volume

...of R_eff, carrier diffusion, and the excitation volume on the CL intensity on a dislocation are still elusive.

Blue InGaN/GaN light emitting diodes (LEDs) on sapphire (0001) substrates are currently the most efficient lighting sources for energy saving, achieving above 85% internal quantum efficiency (IQE) at low current density. Indium ingots for energy saving, achieving above 85% internal solutions, remains difficult to access. Furthermore, the influences simultaneous picosecond temporal and nanoscale spatial resolutions, remains difficult to access. Furthermore, the influences...
and the yield of CL signal when working in pulsed excitation mode. The dark spots with lower CL intensity suggest the locations of dislocations acting as non-radiative centers. The average density of dislocation within this area (~240 µm²) is ~8 × 10⁶ cm⁻², which is within the order of magnitude of the dislocation density in the GaN substrate. Indeed, it has been reported that the distribution of dislocations in the GaN substrate is non-uniform. In our sample, the density of dislocations revealed by the CL can be as large as ~4 × 10⁷ cm⁻² in some small regions (~25 µm²), whereas we do not observe any dislocations in some other parts of the sample.

To further understand the recombination dynamics of carriers on a single dislocation, we perform our measurements in pulsed excitation mode at 10 K. Low temperature allows us to reduce the spectral linewidth due to the suppression of phonon scattering. It is a prerequisite to precisely observe whether or not the dislocation induces a local energy gradient and to probe whether or not this gradient could affect the carrier dynamics. Fig. 2(a) presents the integrated CL intensity mapping around a dislocation at T = 10 K. The blue spot in the center with ~200 nm diameter shows the signature of a non-radiative dislocation. The CL intensity on the dislocation is ~50% lower than that of its surroundings. Fig. 2(b) presents the corresponding mapping of the emission peak energy in the same area. The average energy all over the area is ~3.473 eV, which is consistent with D³X₄ emission. More importantly, there is a dipole-like energy shift in the order of ~1.3 meV difference around the dislocation, as illustrated by the blue and red circles. Such a dipole behavior has been observed at several dislocations under different excitation conditions in our sample. Fig. 2(c) presents the average spectra within each circle indicated in Fig. 2(b). The blue spectrum, which corresponds to the emission from the blue circle, shows a clear blue shift compared to the red spectrum from the red circle. This dipole-like energy shift clearly indicates the presence of an edge or mixed type of dislocation, which induces local strain fields.

In Fig. 2(d), a continuum elastic model reproduces the dipole-like energy shift with the ~1.3 meV difference and ~400 nm separation between the two poles, which are consistent with the experimental results. In detail, supposing a pure edge dislocation with the Burgers vector $b = \frac{1}{3} (11 \bar{2} 0)$, the dipole-like energy shift $\Delta E_g$ is given by

$$\Delta E_g = \langle a_1 + b_1 \rangle \varepsilon_{zz} + \langle a_2 + b_2 \rangle \langle \varepsilon_{xx} + \varepsilon_{yy} \rangle,$$

$$\varepsilon_{xx} = \frac{b}{4\pi} \left[ \frac{(3c_{11} + c_{12})x^2 + (c_{11} - c_{12})y^2}{c_{11}(x^2 + y^2)^2} \right],$$

$$\varepsilon_{yy} = \frac{b}{4\pi} \left[ \frac{-(c_{11} + 3c_{12})x^2 + (c_{11} - c_{12})y^2}{c_{11}(x^2 + y^2)^2} \right],$$

where $a_{i=1,2}$ and $b_{i=1,2}$ are hydrostatic deformation potentials, $c_{ij=11,12}$ are the elastic constants of GaN, and $\varepsilon_{xx,yy,zz}$ are strain tensors, in which $\varepsilon_{zz} = 0$, considering the Burgers vector is parallel to the $xy$ plane. All the above values are taken from Ref. 19. Considering the interaction volume and the diffusion of injected carriers, $\Delta E_g$ is convoluted with a two-dimensional (2D) $(xy)$ plane Gaussian distribution with a full width at half maximum $FWHM = 220 \pm 30$ nm, which is the only fitting parameter and determines both the energy shift and the separation distance of the dipole. Therefore, our simulation, which includes the $FWHM$ for the interaction volume, shows that the optical properties around this particular dislocation are dominated by the edge component, with $(11 \bar{2} 0)$ orientation, as the red mark of dislocation in Fig. 2(b) shows. In Ref. 19, the distance between the two poles was found to be ~5 µm, which is one order of magnitude more than what we observe here (~400 nm). The reason behind this discrepancy is the following: the strain fields around a dislocation present a divergence exactly at the center of dislocation. As a result, the distance between the observed maximum and minimum in energy only comes from the limited spatial resolution.
Thanks to a much better spatial resolution of CL compared to micro-photoluminescence, the two peaks appear closer in our experiments, which confirms the relevance of our approach.

The above analysis is useful to determine the type of dislocation and its influence on the local bandgap; however, we do not observe any significant impact of this dipole on the carrier diffusion. Indeed, the CL intensity around the dislocation in Fig. 2(a) does not exhibit any favored direction. This could be explained by the small energy shifts induced by the dislocation (\(\Delta E \approx 0.7 \text{ meV}\)) which is less than the thermal energy at \(T = 10 \text{ K}\) and as such does not play a significant role in the exciton dynamics.

TR-CL is then employed to probe the exciton recombination dynamics around the dislocation. The effective lifetime \(\tau_{\text{eff}}\) is measured along the red dashed arrow plotted in Fig. 2(a). Fig. 3(a) shows a streak image recorded all along the red dashed arrow in Fig. 2(a) and demonstrates that the decay time is not energy dependent in our sample. Fig. 3(b) presents the decay curves probed at different distances from the dislocation, as denoted by the solid arrows (on, near, and off the dislocation) in Fig. 2(a), and reveals two main features when closer to the dislocation: (1) The CL intensity \(I_{\text{CL}}(t = 0)\) at early delay time is lower and (2) the effective lifetime \(\tau_{\text{eff}}\) is shorter. In Figs. 3(c) and 3(d), more details are given by the one-dimensional (1D) mappings of \(I_{\text{CL}}(t = 0)\) and \(\tau_{\text{eff}}\), extracted from a mono-exponential fit. \(I_{\text{CL}}(t = 0)\) drops by 40\% at the dislocation compared to the surroundings, and \(\tau_{\text{eff}}\) reduces from 125 ps to 90 ps.

To explain both the spatial dependent effective lifetime \(\tau_{\text{eff}}\) and the CL intensity at early delay time \(I_{\text{CL}}(t = 0)\), we propose a model based on a 2D (xy plane) diffusion equation which considers a region around the dislocation, defined by an effective radius \(R_{\text{eff}}\), where the recombination is treated as entirely non-radiative and instantaneous. This assumption is based on the fact that, when the electron beam is located at the dislocation, a drop of intensity appears at early delays, which indicates the non-radiative processes at the dislocation are faster than our temporal resolution (\(< 20 \text{ ps}\)). For simplicity, we set the dislocation at the origin of the simulation area.

When an exciton is outside the \(R_{\text{eff}}\) of dislocation, it can diffuse and recombine either radiatively or non-radiatively,
The initial exciton density distribution is supposed to follow the Gaussian distribution \( N(r_0, \sigma^2) \), namely, \( n(r - r_0, t = 0) = n_0 N(r_0, \sigma^2) \), where \( n_0 \) is initial exciton density and \( \sigma \) is its standard deviation. Since the FWHM has been determined to be \( \sim 220 \) nm as mentioned before, the remaining fitting parameters to derive \( n(r - r_0, t) \) are: \( R_{\text{eff}}, D \), and \( \tau_{\text{eff}, \infty} \), where \( \tau_{\text{eff}, \infty} \) corresponds to the effective lifetime far from dislocation; we set \( \tau_{\text{eff}, \infty} = 125 \) ps as observed experimentally. As a result, the CL intensity of exciton can be described as \( I_{\text{CL}}(r - r_0, t) \propto n(r - r_0, t)/\tau_r \), and \( \tau_{\text{eff}, \infty} \) can be modelled by fitting \( n(r - r_0, t) \), supposing a constant radiative lifetime \( \tau_r \). Here, it is important to notice that the two remaining parameters \( D \) and \( R_{\text{eff}} \) are independent and can be tuned almost independently during the fitting procedure. Indeed, the ratio between the effective radius \( R_{\text{eff}} \) and the FWHM of the interaction volume mostly defines the drop observed in the CL intensity at early delays \( I_{\text{CL}}(t = 0) \), since we consider that the dislocation instantaneously acts on the carrier inside the effective dislocation area. On the other hand, the diffusion coefficient \( D \) mainly governs the dynamical behavior and hence acts on the effective lifetime \( \tau_r \). Such an independence between the two fitting parameters allows us to extract them confidently and provides robustness to our fitting procedure.

As shown in Figs. 3(c) and 3(d), the good agreement between our experimental results and the fits (blue lines) allows us to clearly identify the effective radius of dislocation.
$R_{\text{eff}} = 95 \pm 3 \text{ nm}$, the diffusion coefficient $D = 4.5 \pm 0.2 \text{ nm}^2/\text{ps}$, and the effective lifetime $\tau_{\text{eff}} \propto 125 \pm 3 \text{ ps}$. Thus, the diffusion length of $D'X_A (L_{\text{diff}} = \sqrt{D \tau_{\text{eff}}})$ can be estimated to be $\sim 24 \text{ nm}$, which indicates that $D'X_A$ are able to diffuse on a short distance by hopping between donor levels.\cite{14,21} Note that the CL intensity at initial delay is defined as $I_{\text{CL}}(r_0, t = 0) \propto n_0 N/\tau_r$. Considering that no diffusion happens at this moment, the decrease of $I_{\text{CL}}(r_0, t = 0)$ near the dislocation can be due to either the increase of $\tau_r$ or the reduction of $n_0$. However, the convoluted $\sim 1.3 \text{ meV}$ local potential fluctuation of dislocation unlikely changes the wave functions overlap of exciton, which justify why we consider $\tau_r$ to be constant over the sample. Therefore, the decrease of $I_{\text{CL}}(r_0, t = 0)$ near the dislocation should be ascribed to the non-radiative and instantaneous depletion of exciton within $R_{\text{eff}}$. In particular, $R_{\text{eff}} = 95 \pm 3 \text{ nm}$ is close to the previously reported value ($\sim 50 \text{ nm}$).\cite{22} Even though it is admitted that the core of a pure edge dislocation is at the atomic scale,\cite{22} our results clearly demonstrate that its impact on non-radiative recombination occurs up to the nanoscale, potentially because of the decoration of such dislocation by point defects.\cite{23,24} In addition, in Fig. 3(b), the decay curves at different positions are well reproduced by the fitting equation $I_{\text{CL}}(r_0, t) \propto n(r_0, t)/\tau_r$, which further attest our model.

In conclusion, using our picosecond TR-CL, we study the exciton dynamics at 10 K around a single dislocation in bulk epitaxial GaN grown on a free standing GaN substrate at $T = 10 \text{ K}$. Edge dislocations are identified by observing and simulating the $\sim 1.3 \text{ meV}$ dipole-like energy shift due to the local strain fields induced by such type of dislocation. We determine the effective radius of dislocation $R_{\text{eff}} \sim 95 \text{ nm}$ and the diffusion length $L_{\text{diff}} \sim 24 \text{ nm}$ of $D'X_A$ by fitting the 1D mappings of CL intensity at early delay time $I_{\text{CL}}(r_0, t = 0)$ and effective lifetime $\tau_{\text{eff}} (r_0)$ around this dislocation, through a 2D exciton diffusion model. Therefore, our results pioneer an innovative way to quantitatively study the influence of dislocation on the efficiency of optoelectronic devices.

The present work was financially supported by the Swiss National Science Foundation under Project Nos. 153620 and 154853 and by the EPFL fellowship program co-funded by Marie Curie (FP7 Grant Agreement No. 291771). We also thank the Attolight Company for the constant improvement of the TR-CL setup.