Picosecond Time-Resolved Cathodoluminescence to Probe Exciton Dynamics in $a$-Plane (Al,Ga)N/GaN Quantum Wells


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Introduction
Picosecond and femtosecond spectroscopy allow for a detailed study of carrier dynamics in nanostructured materials [1]. In such experiments, a laser pulse usually excites several nanostructures at once. However, spectroscopic information may also be acquired using pulses from an electron beam in a modern scanning electron microscope (SEM), exploiting a phenomenon called cathodoluminescence (CL) where electrons are promoted from the conduction band to the valence band upon impingement of the high energy electron beam onto a semiconductor. This approach offers several advantages over the usual optical spectroscopy. The multimode imaging capabilities of the SEM enable the correlation of optical properties (via CL) with surface morphology (secondary electron mode) at the nanometer scale [2] and the large energy of the electrons allows the excitation of wide-bandgap materials. Here, we present results obtained with an original time-resolved cathodoluminescence (TRCL) setup [3]. This setup uses ultrafast UV laser pulses to create short photoelectron pulses. The light pulses from an ultrafast UV laser illuminate a metal photocathode from which the electrons are extracted and accelerated inside the high voltage column of the microscope and focused on the sample surface. The collected CL signal is dispersed in a spectrometer and analyzed with an ultrafast STREAK camera to obtain high time resolution.

Our current setup reaches combined space and time resolutions of 50 nm and 10 ps, respectively. Measurements can be carried out at temperatures between 25 K and 300 K. We will describe the TRCL setup in detail and will also present results obtained on $a$-plane (Al,Ga)N/GaN quantum wells (QW) [4] that explain the large range of exciton radiative lifetimes that have been reported in literature so far.

Since Waltereit et al. demonstrated ten years ago the realization of polarization free (Al,Ga)N/GaN quantum wells (QWs) [5], a lot of work has been carried out on non-polar nitride-based heterostructures. Built-in electric field is absent in $a$-plane GaN (non-polar GaN), which allows for the growth of wide QWs without decreasing the radiative recombination probability of electrons and holes. However, even when processing techniques such as epitaxial lateral overgrowth (ELO) are used, non-polar GaN grown on sapphire presents high densities of extended defects. While dislocations are considered as non-radiative recombination centers, basal plane stacking faults (BSFs) are optically active and give rise to an emission centered 50 meV below the excitonic bandgap of GaN [6].
Results
We present a low-temperature TRCL study of exciton dynamics as a function of the local BSF density in $\alpha$-plane (Al,Ga)N/GaN single QWs grown by molecular beam epitaxy on an ELO-GaN template. First, CL experiments demonstrate the existence of nearly BSF-free regions as well as the existence of regions with BSF bundles. This indicates that the BSF distribution of the underlying $\alpha$-plane GaN template [7] is reproduced in the QW. We confirm the results obtained by Badcock et al., who demonstrated that the intersection of BSFs with the QW leads to the formation of quantum wires (QWR) [8]. We then study the local relaxation-recombination dynamics of excitons in both QW and QWRs. In particular, we show that the dynamics of QW excitons is dominated by their capture by the BSFs. The QW CL decay time therefore exhibits a strong spatial dependency (FIG. 1), explaining the large range of values reported so far for exciton radiative lifetimes in non-polar (Al,Ga)N/GaN QWs [4]. We finally demonstrate that below 60 K, QWR excitons exhibit a zero-dimensional behavior, which we relate to their binding on localization centers such as QWR-width fluctuations.

FIG. 1 Low temperature (30 K) TRCL decays of QW and QWR excitons in regions with low (a) and high (b) local BSF densities. The decay time variations of the QW exciton show that the QW exciton dynamics is dominated by capture on the BSFs.

References