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Spatio-Time-Resolved Cathodoluminescence Spectroscopy Imaging: Microscopic Recombination Kinetics in Semi-Polar InGaN Quantum Wells

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We present results from ps-time- and nm-spatially resolved cathodoluminescence (CL) microscopy ($\delta t < 35$ ps, $\delta x < 40$ nm). The electron beam in our SEM based CL setup is extremely fast switched on, stays "on" for a selected time, and is then extremely fast switched off by an electrostatic beam-blanking. This allows the analysis of excitation from thermal equilibrium into true steady state condition and the relaxation back into thermal equilibrium. Consequently, we have chosen adequately long rectangular pulses (e.g. $\Delta t = 37$ ns) for excitation and moderate repetition frequency (e.g. 1 MHz). The light is detected by a cooled micro-channel plate photomultiplier in single photon-counting system mode in delayed coincidence. The focused electron beam is digitally scanned over the surface (256×200 pixels) while CL transients are recorded for a certain dwell time at each pixel. Subsequently, the resulting data set consisting of 51,200 local CL transients i.e. CL(x, y, t) is stored and evaluated ex situ to produce: local transients, sets of time delayed CL intensity mappings (TDCLI), transient line scans. Using a digital box-car method, finally a microscopic lifetime mapping can be generated, directly revealing the nano-scale kinetics in correlation with the morphology. One approach for beating of the negative impact of spontaneous polarization fields of group-III nitrides is the growth on semi- and/or non-polar crystal planes. The sample under investigation was grown using MOVPE. On top of a 2 µm thick GaN buffer a 200 nm thick SiO2 mask was patterned into hexagons (photolithography + reactive ion etching). Subsequent MOVPE overgrowth results in the formation of 3D inverse pyramids due to selective area growth. The resulting surface planes of the inverted pyramids are formed by {10-11} and {11-22} facets. An InGaN single quantum well (SQW) with a thin GaN cap was grown on top of the patterned surfaces. The differences in crystal structure, strain, polarization fields, In-incorporation and/or SQW-thickness result in an extremely complex interaction of relaxation, recombination (radiative + non-radiative), and transport in energy, space and time of the excited carriers. The spatially averaged transient shows strong non-exponential decay (Fig. 1a) as a result of the complex dynamics involved. Microscopic local transients taken at the center of the inverted pyramids exhibit a very short lifetime down to $\tau_{initial} = 200$ ps. Upon going up to the ridge of the pyramid, the life time dramatically increases reaching $\tau_{initial} > 10$ ns at the ridges. This giant change of lifetime with local position is visualized in a CL transient linescan across an inverted pyramid (Fig. 1c). Time delayed CL maps TDCLI directly reveal the strong difference of the local lifetime. In particular at the center of the pyramids the CL intensity vanishes very fast in less than a few ns after switching off the excitation, whereas at the ridges the CL persists even after 55 ns (Fig 2). The microscopic distribution of the initial lifetime is depicted in Fig. 3a in a log-scale. It directly correlates with the micro-morphology as well as local emission wavelength of the InGaN QW. At the center of the pyramid we find very short emission wavelength ($\lambda = 370$ nm) and here, the lifetime is very short (< 0.4 ns). Upon going up along the pyramids the emission wavelength increases up to 500 nm where the initial lifetime becomes > 13 ns. Here, the persistent decay time exceeds 55 ns. Normalizing the total lifetime map to the local quantum efficiency yields a microscopic map of the radiative lifetime (Fig. 3c), which directly correlates the local oscillator strength to the local SQW properties.

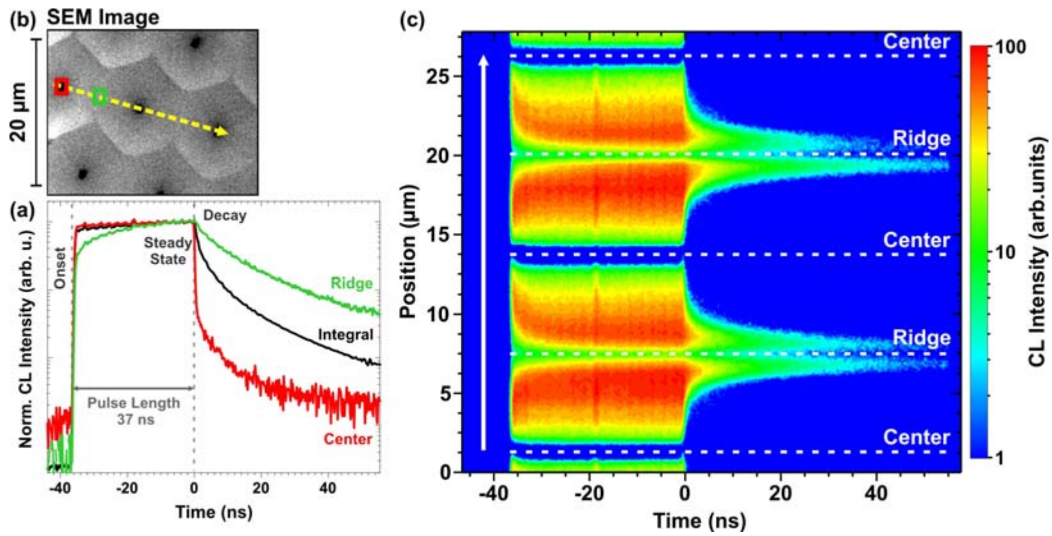

Fig. 1: (a) CL transients: black - spatially averaged, red - at the center of the inverted pyramid, green - at the ridge (b) SEM image, the positions of the local transients in (a) and the linescan (c) are indicated; (c) CL transient linescan.

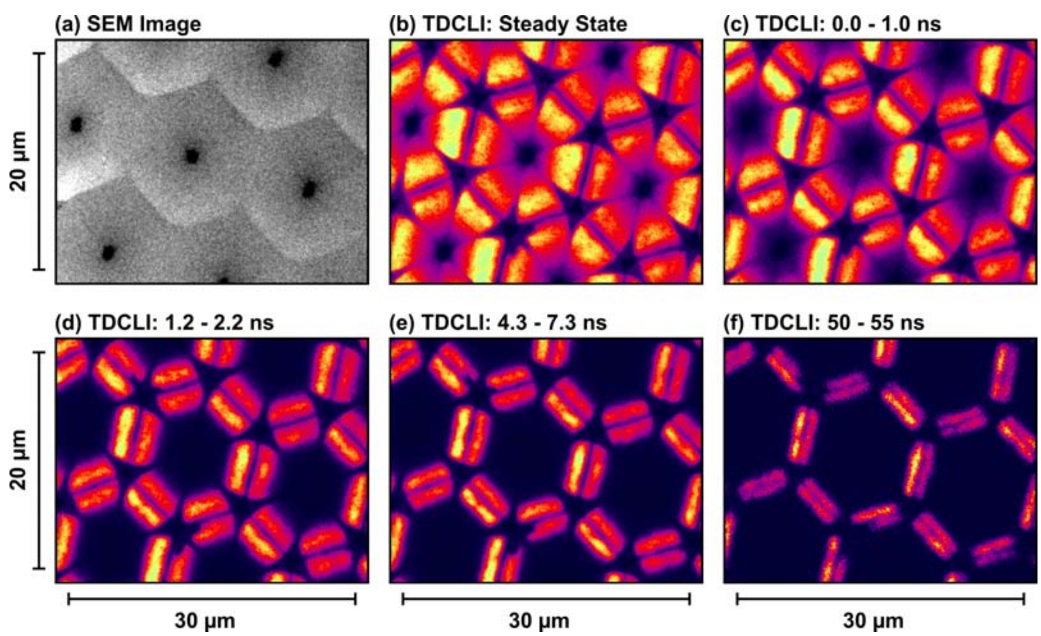

Fig. 2: (a) SEM image, (b-f) time delayed CL images (TDCLIs) in steady state (b), and at certain delay times during decay (c-f): Fast decay in center hexagon + strong CL persistence at the ridges.

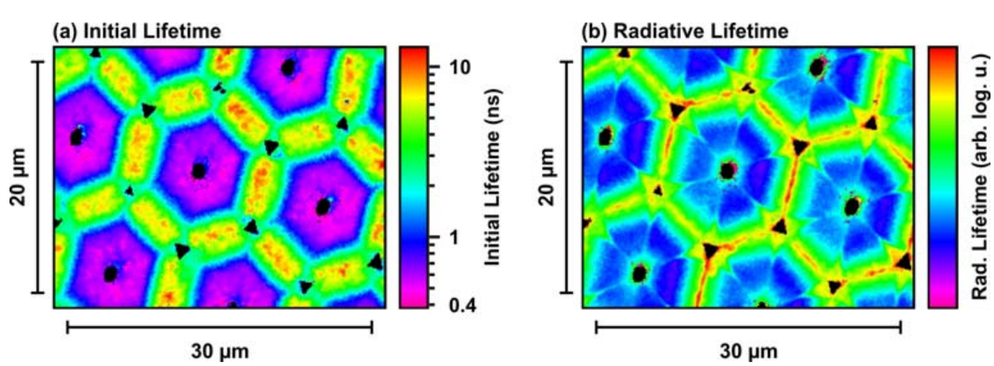

Fig. 3: spatial mappings of the (a) initial decay time and (b) the radiative lifetime (for SEM Image see Fig. 2a).

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