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Estimates of photoluminescence efficiencies in GaN nanowires at high injection levels from steady-state photoluminescence measurements

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Photoluminescence (PL) efficiencies were estimated for individual silicon-doped GaN nanowires grown by plasma assisted molecular beam epitaxy (PAMBE). Steady-state PL measurements reveal efficiencies that depend on excitation intensity, temperature, and nanowire morphology. While many nanowires had their best efficiencies at base temperature (T ~ 4 K), some nanowires, given sufficient excitation intensities, had best efficiencies at elevated temperatures (T ~ 100 K). Room-temperature internal quantum efficiency (IQE) of PL was as high as 33 ± 5%. These steady-state results corroborate with time-resolved PL measurements reported earlier, and both methods can aid in optimizing the growth and processing of nanowires for future applications in nanoscale optoelectronics.

1 Introduction The internal quantum efficiency (IQE) of photoluminescence (PL) (henceforth PL-IQE) is an important parameter in assessing the bulk and surface qualities of semiconductor materials for optoelectronic applications [1, 2]. For GaN nanowires, with large surface-to-volume ratios, this parameter is particularly sensitive to nonradiative recombination of carriers at the surface and thus offers a means to evaluate and eventually optimize surface performance. Here, we measure steady-state photoluminescence from isolated GaN nanowires grown on Si(111) substrates by plasma assisted molecular beam epitaxy (PAMBE) as a function of temperature and excitation intensity. These steady-state results complement earlier time-resolved photoluminescence (TRPL) measurements [3] and reveal nanowire PL-IQE's that depend on excitation intensity, temperature, and nanowire morphology.

2 Experiment GaN nanowires were grown by PAMBE on Si(111) substrates as described elsewhere [4]. The wires for this study were n-type, silicon-doped nanowires from a single growth run with lengths of 10 – 20 μm and diameters of 200-1500 nm. These wires were removed from their growth substrates by sonication in isopropanol. The resulting nanowire suspension was then dispersed onto silicon substrates with aid of a pipette. The dispersed wires on their substrates were placed in a closed-cycle optical cryostat for temperature-dependent measurements of steady-state PL. The cryostat was placed under an optical microscope that enabled imaging of individual nanowires and the delivery of excitation light into a spot of a few micrometers in diameter. The excitation light (at 266 nm) came from a frequency-quadrupled, Q-switched $Nd{:}YVO_4$ laser operating at a repetition rate of 15 kHz with pulse durations of about 8 ns. These pulse durations were well above the typical ~ 1 ns PL decay times of these nanowires and thus quasi-steady-state conditions were assumed. Excitation intensities (I_{exc}) ranged from 0 to 50 kW/cm^2 and were kept well below the lasing thresholds, which, for these nanowires, were at least 250 kW/cm^2 [3]. Front-surface PL from the samples was collected, dispersed through a monochomator, and detected with a photomultiplier tube (PMT). A time-gated photon counter enabled low-noise detection of the PL signal. The PL output (PL_{out}) used to calculate PL efficiencies was determined by integrating under the PL spectral curves over the near-band-edge emission (350 nm to 390 nm, or 3.54 eV to 3.18 eV).

3 Results The PL efficiencies (PL_{out}/I_{exc}) as a function of I_{exc} were measured for several individually dispersed nanowires at different temperatures. Each nanowire exhibited an efficiency behaviour like that shown in Fig. 1, which represents the PL efficiency at three temperatures for a single, silicon-doped nanowire dispersed onto a silicon substrate (NW #1). This relatively large nanowire had a length of 19.1 μm and a diameter of about 970 nm. Optimal PL efficiencies occurred at lower intensities for lower temperatures as nonradiative recombination channels were evidently saturated similar to the saturation behaviour seen in ZnO nanowires [5]. The intensity-dependent data show that nonradiative processes exist at base temperature (T ~ 4 K) and optimal efficiencies do not occur at the same intensity for different temperatures. Calculation of IQE will necessarily involve the comparison of PL efficiencies at different excitation intensities across different temperatures. Similar behaviour was observed by Mickevičius et al. in AlGaN epilayers under strong photoexcitation [6].

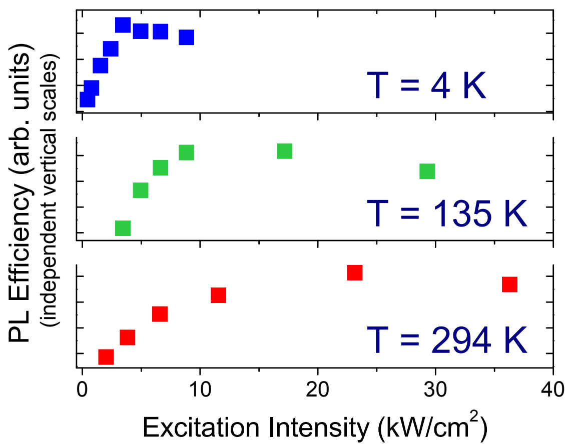

Figure 1 PL efficiencies (PL_{out}/I_{exc}) for a single, dispersed, silicon-doped nanowire (NW #1) as a function of excitation intensity at three different temperatures.

Figure 2 shows the temperature-dependent PL efficiencies of a second similarly dispersed silicon-doped nanowire (NW #2). This nanowire was 13.6 μm long with a diameter of about 348 nm; it had a clean hexagonal cross section and little taper as seen in the scanning electron micrograph (SEM) image of Fig. 3. With sufficient excitation intensity, optimal PL efficiency for this wire occurred at an elevated temperature around 100 K, where free excitons predominate over bound excitons. TRPL measurements reported previously indicated that wires from the same growth run exhibited unity PL-IQE at this elevated temperature. This was shown by matching temperature-dependent PL lifetimes with the temperature-dependent lifetimes expected for purely radiative free excitons in GaN that increase in proportion to $T^{3/2}$ [3]. With unity PL-IQE determined at this temperature, PL-IQE values at other temperatures will scale with the PL efficiencies as indicated by the right vertical axis of Fig. 2. The PL-IQE was as high as 33 ± 5% at room temperature for this nanowire.

As with the TRPL results, the steady-state PL results here show a temperature-dependent drop in PL-IQE for temperatures between 4 K and 50 K. Such behaviour is associated with the nonradiative recombination associated with the donor-bound excitons and may be more prevalent in more highly doped samples where efficient inter-donor exciton transfer is more likely to occur [7, 8].

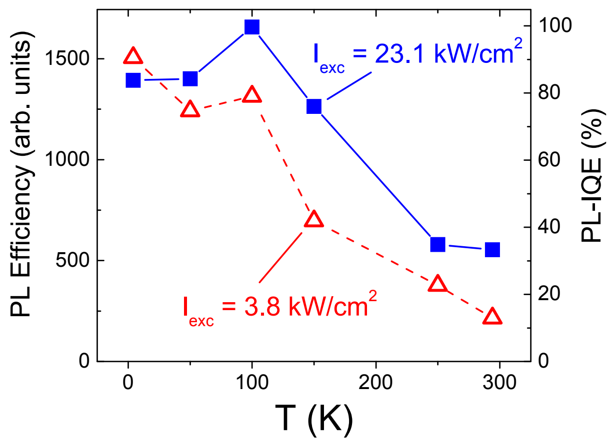

Figure 2 Temperature-dependent PL efficiency and PL-IQE for silicon-doped nanowire (NW #2) dispersed onto Si substrate at two excitation intensities.

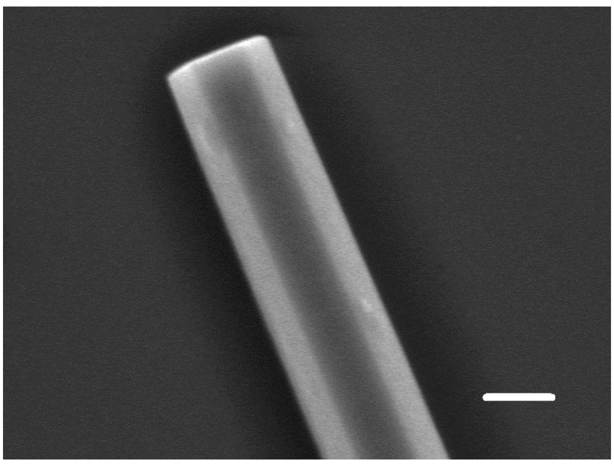

Figure 3 SEM image of the nanowire studied in Fig. 2 (NW #2). The nanowire is 13.6 mm long with a diameter of about 348 nm. The scale bar is 200 nm long.

The temperature-dependent behaviour shown in Fig. 2 was *not* common to all nanowires measured. Many wires exhibited optimal PL efficiencies at base temperatures with more rapid decline in efficiencies with increasing temperature. Room-temperature PL-IQE's for these nanowires were estimated to be a few percent or less. These wires also did not share the clean morphology of NW #2 shown in Fig. 3. They typically had a stronger taper and irregular cross sections. These wires may incorporate defects associated with abnormal growth. One mode of abnormal growth occurs when densely packed wires grow together and fuse, hiding defects at their growth interface and creating non-hexagonal nanowire cross sections.

4 Conclusion Steady-state PL measurements on silicon-doped nanowires show PL efficiencies that depend on excitation intensity, temperature, and nanowire morphology. Calculation of PL-IQE requires special attention to excitation conditions. Nanowires with clean hexagonal cross sections and little taper can have PL-IQE's as high as 33 ± 5% at room temperature with unity PL-IQE's at temperatures substantially above base temperatures. As earlier TRPL measurements demonstrated, steady-state PL measurements can show different recombination behaviours for donor-bound and free excitons in GaN provided nanowires are without growth defects. Such measurements should prove helpful in optimizing future nanowire surface treatments, nanowire growth, and nanowire device development.

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