Ultrafast optical characterization of carrier capture times in In$_x$Ga$_{1-x}$N multiple quantum wells

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Subpicosecond wavelength-degenerate differential transmission optical spectroscopy was used to characterize the electron capture time in a 10-period In$_x$Ga$_{1-x}$N multiple-quantum-well (MQW) structure. Photoluminescence and photoluminescence excitation spectroscopies demonstrated enhanced MQW emission for injection within ±50 meV of the barrier energy. Time-resolved differential transmission measurements for excitation in this region reveal efficient electron capture in the quantum wells with a time constant between 310 and 540 fs. A slower exponential relaxation, with strongly wavelength-dependent subnanosecond decay constants, is also observed. © 2000 American Institute of Physics.

Group-III nitride-based optoelectronics have emerged as an important technology, with the commercialization of a nitride-based laser being one of the most recent and important examples. However, there have been only limited investigations of carrier injection into the multiple-quantum-well (MQW) active regions of the lasers. The efficiency of radiative recombination in MQWs is known to depend on a number of time scales, including the transport and recombination times in the barriers, and the capture times from the three-dimensional (3D) unconfined states at the barrier energy to the confined two-dimensional (2D) quantum well states. In this letter, a measurement of the electron capture time is reported for a MQW sample that is similar to the active region of a laser described in the literature.

The sample was grown by metalorganic chemical vapor deposition (MOCVD) at the University of California at Santa Barbara in a modified two-flow horizontal reactor on double polished c-plane sapphire. It consists of a 10-period MQW with 25 ÅIn$_{0.15}$Ga$_{0.85}$N wells and 55 ÅIn$_{0.05}$Ga$_{0.95}$N:Si barriers. There is a 0.1 μm GaN cap layer on top of the MQW, and the structure is grown on top of a ~2 μm GaN:Si layer. The doping in the barriers is ~10$^{18}$ cm$^{-3}$.

Photoluminescence (PL) spectroscopy was performed on the MQW sample using a continuous-wave HeCd laser operating at 325 nm, and peak PL emission occurred at 2.99 eV (Fig. 1). To estimate the energy level structure, a simple, one-dimensional (1D) band-edge calculation was performed without including spontaneous polarization, piezoelectric (PZE) polarization, free charge, and space charge effects. Because of the indium phase separation problem in In$_{0.15}$Ga$_{0.85}$N, various band gaps (2.859–2.831 eV), conduction–valence band offsets (30:70–75:25), and potential fluctuations (up to 0.2 eV) are quoted in the literature. By contrast, the band gap for In$_{0.05}$Ga$_{0.95}$N is better constrained between 3.220 and 3.224 eV. Agreement between the PL and the calculation is within 60 meV, a satisfactory result given the potential fluctuations and quantum confined Stark redshift from the ~1 MV/cm strain-induced piezoelectric field.

The wavelength of the time-integrated PL emission peak was insensitive to variation of the excitation wavelength from 325 (3.83 eV) to 410 nm (3.03 eV), but the strength of the emission was wavelength sensitive. To quantify this behavior, photoluminescence excitation (PLE) spectroscopy was used to measure the intensity of the peak PL emission (2.99 eV) as a function of excitation wavelength. Excitation was provided by a standard 300 W Xe arc lamp-grating spectrometer arrangement with 1.5 nm resolution. The peak absorption of the MQW occurred at 3.22 eV (Fig. 1), in agreement with previously measured barrier band gap energies.

The observed 3.23 eV PL emission peak for a separate In$_{0.05}$Ga$_{0.95}$N sample, consisting of a 60 nm strained epilayer on ~2 μm of GaN grown on sapphire, corroborates this result. Finally, the calculation reveals only one conduction subband in the MQW, and the MQW PL and PLE data confirm the lack of any obfuscating intersubband transitions. Together, these data suggest that the appropriate wavelength to

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FIG. 1. PL-MQW: PL data for the InGaN MQW sample excited by a HeCd laser. PLE-MQW: PLE data for emission from the 2.99 eV PL peak of the MQW sample. PL-InGaN: PL data for a 60 nm epilayer of In$_{0.05}$Ga$_{0.95}$N.

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observe carrier concentrations of 0.5 and 2.3 after the pump pulse. The decay constant of the fast, sub-ps overlaid from the data, and the analysis was started 200–300 fs time increments between the pump and the probe. The beams overlapped on the sample with an incident angle approximately 5° from normal, and the pump beam was defocused to a spot diameter (100 μm) roughly 2.5 times larger than the probe beam to ensure uniform illumination of the probed region. The cross-polarized probe beam was passed through a polarizer to eliminate stray pump light on the high-speed Si photodetector, while the pump beam was varied in intensity with a variable reflector wheel and chopped at 2 kHz to permit lock-in detection.

Pump/probe energies between 3.145 and 3.272 eV were used with 3.5 and 14 mW (0.55 and 2.2 μJ/cm²) average incident pump powers and 0.2 mW probe power. Assuming an absorption coefficient of 10⁴ cm⁻¹, the pump creates carrier concentrations of 0.5 and 2 × 10¹⁷ cm⁻³, respectively. Relative to the 3.22 eV barrier energy, typical data for excitation above (3.262 eV), near (3.229 and 3.175 eV), and below (3.147 eV) the barrier are shown in Fig. 2. These pump/probe energies are far enough from the MQW subband that no resonant or coherent bound excitonic effects are expected. The pulse width was approximately 85 fs (15 meV) at these pump wavelengths. The feature at ~1 ps corresponds to a reflection artifact and should be ignored. The data were normalized to the bare probe transmission T₀, and the corresponding DT = ΔT/T₀ data reflected a transmission change of 0.1%–0.6%. Pump intensities were kept low to minimize the effects of carrier–carrier scattering and band gap renormalization.

Over a range of pump/probe energies near the barrier energy (3.169–3.262 eV), the data have similar shape, revealing a fast (f), sub-ps decay and a much slower (s), sub-ns decay. The decay portion of the DT data was fit with a simple biexponential (Fig. 2): Aₓ exp(−t/τₓ) + Aₛ exp(−t/τₛ). Because the features were significantly slower than the 85 fs pulse width, the width of the probe pulse was not deconvoluted from the data, and the analysis was started 200–300 fs after the pump pulse. The decay constant of the fast, sub-ps feature decreased slightly with increasing photon energy and excitation intensity (Fig. 3). The decay constant for the slow feature varied dramatically with wavelength, ranging from 20 ps near the barriers to 690 ps at 3.02 eV (not shown) near the QW state. The error in fitted fast τₓ (±5%–10%) and slow τₛ (±10%–20%) decay time constants arises primarily from noise in the data. The DT data near the barrier energy showed a pronounced oscillatory behavior, recently attributed to the effects of zone-folded acoustic phonon modes in similar strained InGaN MQWs. This oscillatory behavior is currently under investigation and will be discussed elsewhere.

The relative strength of the faster feature, Aₓ, and the slower feature, Aₛ, also varies with excitation wavelength but not with excitation intensity (Fig. 4). The fractional strength of the slowly decaying feature, f = Aₛ/(Aₓ + Aₛ), rises from 0 for excitation well above the barriers, plateaus between 0.20 and 0.35 for excitations within ±50 meV of the PLE-defined barrier energy (3.22 eV), then rapidly rises to 1.0 for injection below the barriers. A small value for this ratio suggests efficient removal of carriers from their injected energy states. For injection energies more than 50 meV above the barrier, the ratio is near zero and carriers relax rapidly. In this case, the PLE signal is reduced and a weak, sub-ps DT feature is observed, suggesting that multiple scattering events are required for carrier capture. By contrast, a large value for the ratio suggests inefficient carrier removal, especially for injection energies significantly below the barriers where again the PLE signal drops.

The PLE data suggest the absorption which leads to ef-
efficient QW emission occurs in the same region as the 100 meV-wide plateau in $f$. A separate time-integrated PLE measurement made using the frequency doubled mode-locked Ti:S laser, with tunable excitation between 3.0 and 3.4 eV, reveals that PL emission is almost a factor of 2 stronger for excitation in this plateau region than for anywhere else (Fig. 4). It has been suggested that the hole capture time is much too fast to be observed in this sort of experiment.\textsuperscript{15} Thus, the 310–540 fs $\tau_f$ values parameterize electron capture from the 3D barrier states to the 2D QWs, and the plateau signifies the region of efficient electron capture via emission of a longitudinal optical (LO) phonon.\textsuperscript{15–17} Electron capture times were preliminarily estimated to be a much slower $\sim 15$ ps for a GaN QW of the same width,\textsuperscript{15} but sub-ps capture times have been predicted by an improved model.\textsuperscript{16} Furthermore, the predictions of faster capture times in the GaN systems than in the GaAs system\textsuperscript{15} appear to be borne out in comparison with the measured 650 fs–1.7 ps capture times in wider GaAs wells.\textsuperscript{17}

Inhomogeneities in the QW structure and composition cause potential fluctuations and inhomogeneous broadening of the QW state. The MQW PL linewidth and the region of efficient carrier capture are both approximately 100 meV wide. Consistent with previous findings,\textsuperscript{11} this suggests that the potential fluctuations are approximately 100 meV in the MQWs. Moreover, the shape of the plateau implies the distribution of potential fluctuations is not uniform, and 1-$f$ crudely maps it out. For example, the most efficient carrier capture occurs for pump excitation energies near 3.19 eV, observed as a dip in Fig. 4.

The decay of the slower feature was strongly dependent on the excitation wavelength. For wavelengths near the QW state, the decay occurs with a 0.69 ns time constant consistent with previous measurements of recombination times using time-resolved PL.\textsuperscript{18} For wavelengths nearer the barrier, the decay varies from 300 ps to as fast as 20 ps in the plateau region before disappearing entirely above the plateau. Differ- ent relaxation mechanisms must be at work in these regions, including hot carrier (carrier–carrier and carrier–phonon) relaxation above the barrier region and increasingly important radiative recombination as the QW state is approached. Since no radiative recombination was observed from the barriers in the MQW PL measurements, we speculate that the sub-300 ps relaxation in the plateau region results from nonradiative processes. In particular, carrier distribution cooling processes in the QWs are increasingly effective in removing carriers from the probed region as the pump/probe energy is tuned above and away from the QW state.

In summary, PL, PLE, and time-resolved, wavelength-degenerate DT spectroscopies on InGaN MQW structures reveal optimal emission efficiency when carrier injection is within $\pm 50$ meV of the 3.22 eV barrier energy. In this 100 meV-wide region inhomogeneously broadened by QW potential fluctuations, the measured QW electron capture times were between 310 and 540 fs. A slower relaxation process was observed whose sub-ns rate depended much more sensitively on excitation wavelength.

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