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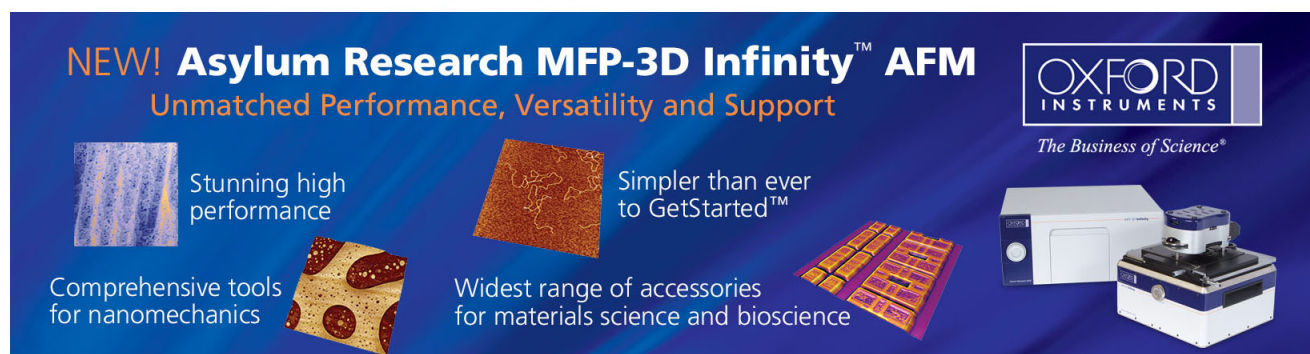
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Time-resolved photoluminescence studies of $\text{In}_x\text{Ga}_{1-x}\text{As}_{1-y}\text{N}_y$

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Time-resolved photoluminescence spectroscopy has been used to investigate carrier decay dynamics in a $\text{In}_x\text{Ga}_{1-x}\text{As}_{1-y}\text{N}_y$ ($x \sim 0.03$, $y \sim 0.01$) epilayer grown on GaAs by metal organic chemical vapor deposition. Time-resolved photoluminescence (PL) measurements, performed for various excitation intensities and sample temperatures, indicate that the broad PL emission at low temperature is dominated by localized exciton recombination. Lifetimes in the range of 0.07–0.34 ns are measured; these photoluminescence lifetimes are significantly shorter than corresponding values obtained for GaAs. In particular, we observe an emission energy dependence of the decay lifetime at 10 K, whereby the lifetime decreases with increasing emission energy across the PL spectrum. This behavior is characteristic of a distribution of localized states, which arises from alloy fluctuations. © 2000 American Institute of Physics. [S0003-6951(00)03702-5]

Recently, the quaternary InGaAsN alloy system has attracted a great deal of attention due to its potential application in devices such as next generation multijunction solar cells and optoelectronic devices for optical communications.^{1–7} The alloy is of fundamental and technological interest because it exhibits an extremely large band gap bowing coefficient ($b \sim -14$ eV) between the III–N and III–As binaries.⁸ The extremely large bowing coefficient permits the $\text{In}_x\text{Ga}_{1-x}\text{As}_{1-y}\text{N}_y$ quaternary alloy to maintain lattice match to GaAs, with a wide range of tunable band gap energies smaller than the GaAs band gap for $x \sim 3y$. Studies of InGaAsN solar cell structures with 1 eV band gap have shown that the quaternary suffers from a short minority carrier diffusion length.^{2,3} More recent work has found that significantly improved minority hole diffusion lengths may be obtained by thermally annealing the InGaAsN after growth, although minority electron diffusion lengths remain short.¹ In this letter, we report the results of time-resolved PL spectroscopy studies of an InGaAsN epilayer. This letter is one of the first investigations of the carrier dynamics within InGaAsN.

A 3- μm -thick, InGaAsN epilayer was grown at a growth temperature of 590 °C by metal organic chemical vapor deposition on a semi-insulating GaAs substrate and terminated with a 5 nm GaAs cap. Trimethylindium, trimethylgallium, arsine, and dimethylhydrazine were used as source gases. The nominal In and N molar fractions were 0.03 and 0.01, respectively. The In/N incorporation ratio of three has been shown to provide lattice match to GaAs.^{8,9} As grown, the unintentionally doped InGaAsN film was p type. After growth, the sample was annealed at 600 °C for 30 min in a nitrogen ambient in order to improve the electrical and optical properties of the material.¹ Photoluminescence (PL) measurements for various sample temperatures and excitation intensities were performed with the sample mounted on a cold

finger and cooled by a closed-cycle helium refrigerator. The sample was optically pumped with 580 nm laser pulses of 10 ps width and 9.5 MHz repetition rate, and a surface pump—surface emission geometry was used. PL emission was collected and analyzed with a 1.3 m grating monochromator equipped with a microchannel plate photomultiplier tube used in a single photon counting mode. The overall time resolution of the detection system is approximately 25 ps with deconvolution techniques.

PL spectra acquired at 10 K for several different relative excitation intensities, varied over two orders of magnitude, are shown in Fig. 1. The PL spectra have been corrected for the spectral response of the monochromator and detector. The peak position (solid line) and half-maximum location on the low energy side (cross) are indicated for each of the emission spectra. It is clear from Fig. 1 that the PL spectrum exhibits blue-shift and broadening with increased excitation intensity. The low energy side of each of the spectra shown in Fig. 1 is functionally very similar with a logarithmic slope $[d \ln(I)/dE]$ of approximately $(9 \text{ meV})^{-1}$. The form of the low energy side of the spectrum suggests a distribution of localized states with an exponential-like density of states (DOS).¹⁰ We attribute these states to local fluctuations in nitrogen concentration since the magnitude, nitrogen concentration dependence, and magnetic field dependence of InGaAsN PL linewidths [full width at half maximum (FWHM) ~ 18 meV for $\text{In}_{0.06}\text{Ga}_{0.94}\text{As}_{0.99}\text{N}_{0.01}$ at 10 K] are well described by an alloy fluctuation model.^{11,12}

The temperature dependence of the time-integrated PL spectra also provides evidence of exciton localization. Figure 2 shows PL spectra acquired for sample temperatures from 10 to 150 K along with indicated peak positions. A pronounced blue-shift of the PL is observed as the sample temperature is raised from 10 to 50 K. This blue-shift diminishes and reverses to the expected red-shift for temperatures above 50 K. A similar temperature dependent behavior has previously been observed for InGaN luminescence and is due to localized exciton effects.^{13,14} For the Ga-rich, InGaN system,

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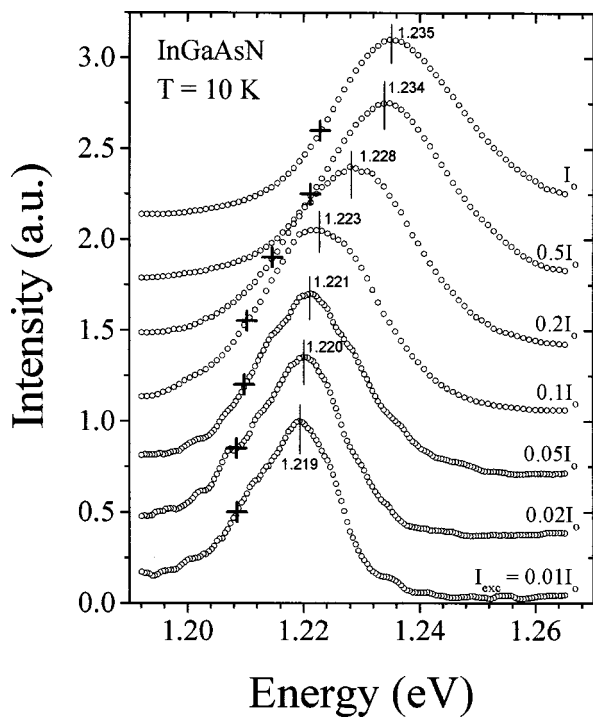


FIG. 1. PL spectra at 10 K from the InGaAsN sample measured for various excitation intensities, I_{exc} . The indicated peak and half-maximum positions show that blue shifting and broadening occur with increased I_{exc} .

exciton localization is attributed to fluctuation of indium content within the alloy.^{15,16} Qualitatively, a blue shift can be understood as a temperature related change of the average kinetic energy and distribution of excitons within the exponential tail of the DOS. However, a localized exciton distribution may be energetically and spatially frozen at 10 K, and discussion of thermal equilibrium may be inappropriate for our results. As described later, it is likely that the blue-shift

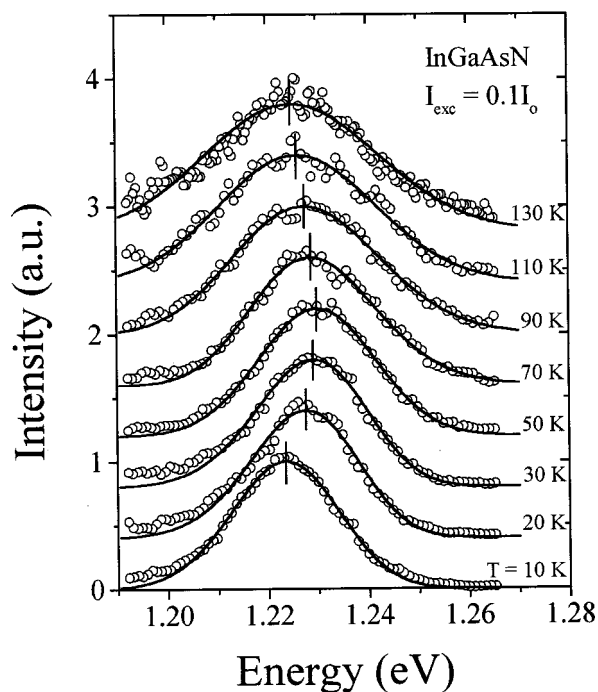


FIG. 2. Temperature dependence of the InGaAsN PL spectrum. Peak positions of individual spectra are indicated with vertical lines.

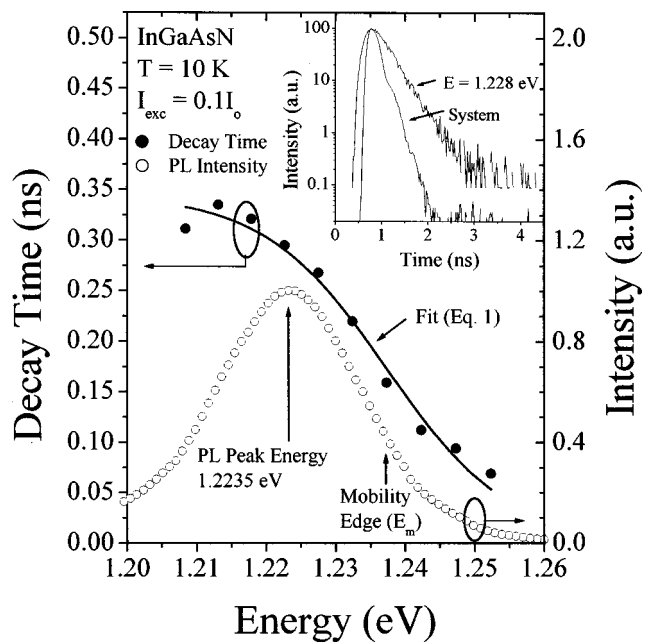


FIG. 3. Emission energy dependence of the temporal PL decay at 10 K. A time-integrated emission spectrum is also shown. The inset shows the PL decay at 1.228 eV and the system response to a 10 ps laser pulse.

is more related to carrier dynamics than to a thermal equilibrium distribution.

Figure 3 shows the measured PL decay time as a function of emission energy for the InGaAsN sample at a temperature of 10 K. Also shown, is a representative emission spectrum. The sample was pumped with an excitation intensity of $I = 0.1I_0$ in comparison with the data of Figs. 1 and 2. The inset of Fig. 3 shows an example of a PL decay near the PL spectral peak position (1.228 eV) and the system response to the laser pulse. All of the measured decays were predominantly single exponentials, $I(t) = I_0 \exp(-t/\tau)$. The exciton decay time, τ , was strongly energy dependent and varied over the range 0.07–0.34 ns. This lifetime is considerably shorter than low temperature exciton lifetimes observed for high purity GaAs¹⁷ and room temperature minority carrier lifetimes in surface-free GaAs.¹⁸ The shortened PL lifetime observed for InGaAsN could result in lower solar cell open circuit voltages and reduced power efficiencies. Also, short lifetimes in InGaAsN lasers could lower radiative efficiencies and increase threshold currents over similar GaAs devices.

The emission energy dependence of the PL decay is characteristic of a distribution of localized excitons.^{19,20} Decay data in Fig. 3 are fit with the function

$$\tau(E) = \tau_R / \{1 + \exp[\alpha(E - E_m)]\}, \quad (1)$$

which includes pathways for either exciton recombination or transfer out of the localized state with energy, E .¹⁸ In Eq. (1), E_m is defined by Oueslati¹⁹ as the energy where the recombination rate equals the transfer rate. The maximum decay lifetime is τ_R and α is a model dependent energy scale. The decay channel is predominantly radiative for the lowest energy ($E < E_m$), strongly localized excitons, while higher energy ($E > E_m$) excitons exhibit a shortened decay time due to transfer out of their sites. From the least squares fit of Eq. (1) to the data in Fig. 3, we find a value at 1.237 eV (indicated in

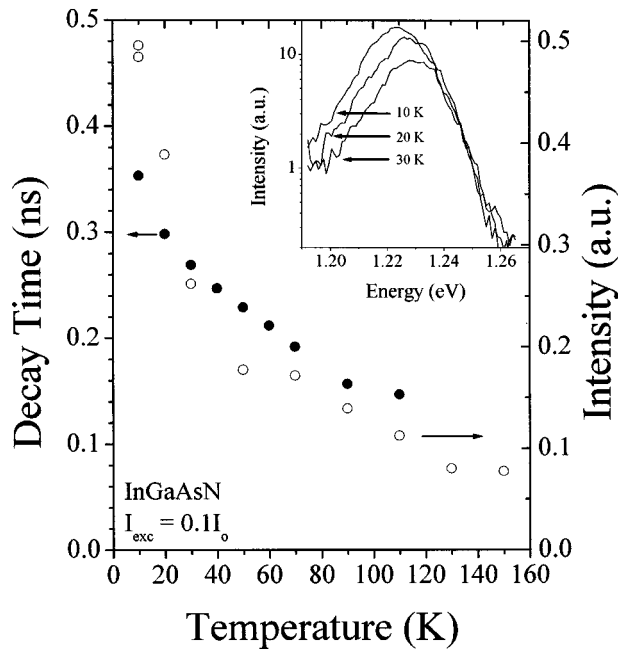


FIG. 4. Temperature dependence of the integrated PL intensity and PL decay time. The inset shows PL spectra at temperatures of 10, 20, and 30 K. Below 50 K, the temperature dependence of the integrated PL intensity is associated with the low energy side of the spectra.

Fig. 3 with an arrow) for E_m , approximately 14 meV above the 10 K PL emission peak. Values for α and τ_R are $(8.8 \text{ meV})^{-1}$ and 0.345 ns, respectively. Similar values for α and the low energy PL line shape parameter, $(9 \text{ meV})^{-1}$, are noted. Specific models for localized exciton transfer processes suggest a relation between α and the exponential tail state parameter.²⁰ However, we do not draw any conclusions regarding the specific transfer processes in this sample from our data.

The temperature dependence of the decay lifetime measured at 1.228 eV and integrated PL intensity are shown in Fig. 4. For temperatures greater than 50 K, the lifetime and PL intensity decrease with temperature at similar rates as might be expected for thermal activation to nonradiative recombination processes. For temperatures from 10 to 50 K, on the other hand, the PL intensity exhibits a much larger relative decrease with temperature than the decay lifetime does. We note that the 10–50 K temperature range exhibiting the strongest thermal quenching of PL intensity is coincident with the temperature region exhibiting the strongest blue-shift in Fig. 2, and we believe the two effects are correlated. Within the inset in Fig. 4, we show that the rapid reduction of PL intensity with increased temperature is realized on the low energy side of the PL spectrum while the high energy side of the spectrum is unaffected. As the temperature is increased from 10 K, we believe that other nonradiative recombination channels become available. The nonradiative channels could affect the PL spectrum in two different ways.

From a macroscopic viewpoint, these channels would compete more efficiently with the longer-lived excitons on the low energy side of the PL spectrum and produce a blue-shift. Alternatively, introduction of nonradiative channels with increased temperature could result in a blue-shift if such channels compete with the transfer of higher energy excitons into lower energy localization sites. On the other hand, we expect that the other nonradiative recombination channels have a much weaker emission energy dependence than the exciton transfer into lower energy localization sites.

In conclusion, time-resolved photoluminescence spectroscopy has been employed to study carrier dynamics in an InGaAsN (1% N) epilayer. Results show that low temperature PL emission is dominated by localized exciton recombination, and measured recombination lifetimes are shorter than the corresponding GaAs exciton lifetimes. The observed localization is believed to arise from alloy fluctuations, and InGaAsN incorporating larger levels of N than this sample will exhibit even stronger localization effects and shorter recombination lifetimes, possibly degrading the performance of minority carrier devices and lasers.

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