Recombination mechanisms in GaInNAs/GaAs multiple quantum wells

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Recombination processes in $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs multiple quantum wells (MQWs) were investigated as function of the nitrogen molar fraction. We found a pronounced S-shaped behavior for the temperature-dependent shift of the photoluminescence emission similar to the ternary nitrides InGaN and AlGaN. This is explained by exciton localization at potential fluctuations. Time-resolved measurements at 4 K reveal an increase of the decay time with decreasing emission energy. A model based on lateral transfer processes to lower-energy states is proposed to explain this energy dependence. The formation of tail states in the $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs MQWs is attributed to nitrogen fluctuations. © *2001 American Institute of Physics.*
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The strong interaction between the conduction band and a band formed by nitrogen states accounts for the reduction of the band gap energy in the quaternary material $Ga_{1-x}In_xN_yAs_{1-y}$.[1] The incorporation of small portions of nitrogen is sufficient to tune the band gap energy of the alloy to 1.3 and 1.55 μm and, therefore, provides an opportunity to realize long-wavelength semiconductor lasers for optical-fiber communications.[2] The proper x/y ratio allows us to grow the quaternary alloy lattice matched to GaAs[3] and the large band offsets in $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs quantum-well structures are expected to improve the temperature characteristics of long-wavelength laser diodes.[4] Recent publications report about the carrier dynamics in a $Ga_{1-x}In_xN_yAs_{1-y}$ epilayer[5] and localization effects in a $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs single quantum well.[6] In this letter, we report results of temperature-dependent and time-resolved photoluminescence (PL) of $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs multiple quantum wells (MQWs). The aim of this study is to investigate the influence of the nitrogen concentration on the recombination dynamics of type I $Ga_{1-x}In_xN_yAs_{1-y}$ MQWs.

The $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs MQWs under investigation were grown on GaAs (001) substrates followed by an AlAs/GaAs superlattice cladding layer applying plasma-assisted molecular-beam epitaxy. All samples contain eight quantum wells of 4 nm thickness with a nitrogen concentration of 1.5%, 1.9%, 2.3%, and 2.6%, respectively, determined by x-ray diffraction and are capped with 10 nm GaAs. The Indium molar fraction of 37% chosen to achieve type I band alignment results in highly strained MQWs. Details of growth are given elsewhere.[7] The samples were excited with a titan-sapphire laser system tuned to 790 nm. The laser was operated in continuous wave mode for time-integrated measurements and in pulsed mode for time-resolved measurements yielding a pulse width of 3 ps. The rise and decay time constants were obtained by fitting the convolution of two exponential decay functions and the system response to the measured transients providing a time resolution of approximately 10 ps.

Figure 1 shows the PL spectra of four $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs MQWs with various nitrogen concentrations at room temperature (RT). Increasing the nitrogen molar fraction shifts the emission to lower energies. In comparison with Ref. 4, the emission energies we observe are somewhat higher for corresponding nitrogen molar fractions as a result of the smaller well width in our samples. Furthermore, we find a correlation between nitrogen concentration and PL linewidth: the higher the nitrogen concentra-

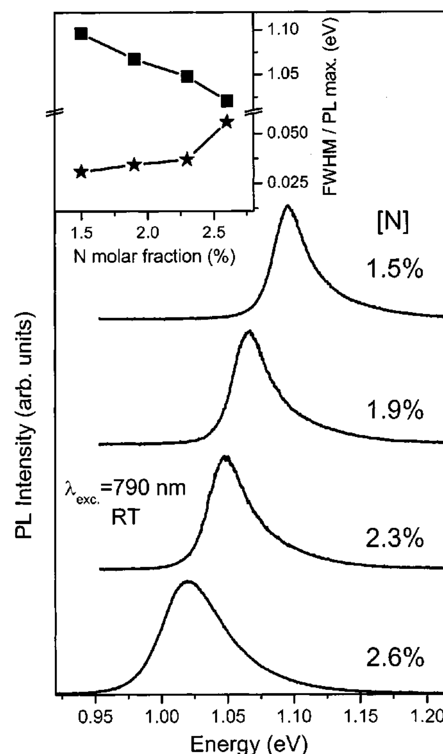

FIG. 1. RT PL spectra of four $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs MQWs with different nitrogen molar fractions. The inset shows the PL peak position (squares) and the full width at half maximum (stars) vs nitrogen concentration.

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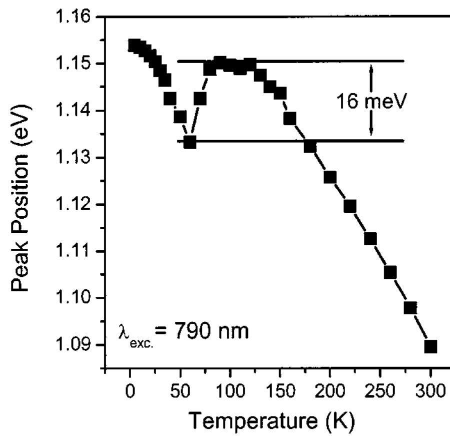

FIG. 2. Temperature dependence of the PL emission of the $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs MQW with 1.5% nitrogen exhibiting a pronounced S-shaped behavior.

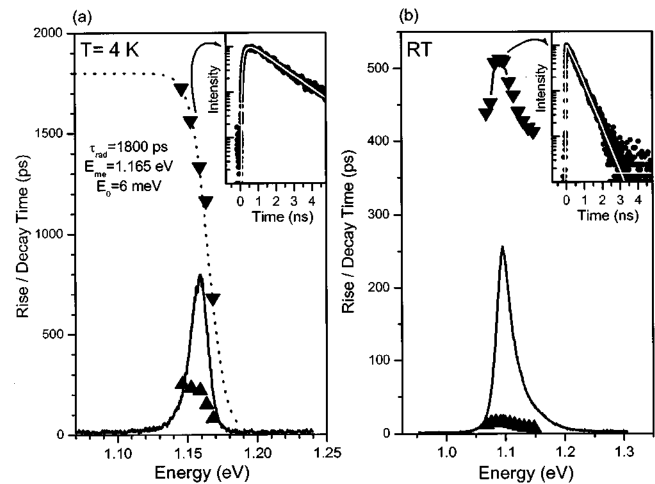

FIG. 3. Luminescence spectra of the sample with 1.5% nitrogen at 4 K (a) and RT (b) together with the corresponding rise (up triangle) and decay (down triangle) times of the luminescence measured at different energetic positions. The dotted line in (a) is a fit to the decay times using Eq. (1) with the given values. All other lines are guide to the eye only. The insets show transients of the luminescence (black dots) together with a fit (white line) taken at the PL maximum.

tion, the broader the PL linewidth (see inset). Such behavior suggests an increasing degree of compositional and structural disorder with increasing nitrogen concentration in the quantum well.

The temperature dependence of the PL emission energy of the sample with 1.5% nitrogen is depicted in Fig. 2. The other samples exhibit an analogous temperature behavior. The behavior differs remarkably from the change of the band gap energy with temperature usually described by the Varshni– or Bose–Einstein formula. The so-called S-shaped temperature dependence, i.e., a red-blue-redshift of the emission energy with increasing temperature, is well known for the ternary nitrides InGaN (Ref. 8) and AlGaN.[9] It was proposed that the luminescence in these materials originates from the recombination of excitons localized at potential fluctuations. The anomalous temperature-induced emission shift is attributed to band-tail states in the density of states (DOS).[10,11] Between 4 K and the first minimum a redshift appears, because the excitons gain sufficient thermal energy to overcome small potential barriers and become trapped in adjacent lower levels of the DOS from where the recombination takes place. The following blueshift of 16 meV is a result of the thermal population of higher energy states of the DOS. Here, a transition from the localized states in the quantum well takes place. At even higher temperatures the temperature shift of the emission is mainly determined by the behavior of the band gap. A similar effect as described in the ternary nitrides is assumed to be the origin of the S-shaped temperature behavior in $Ga_{1-x}In_xN_yAs_{1-y}$. Compositional and structural inhomogeneities such as fluctuations in the alloy composition and well thickness may account for the described band-tail states in the DOS. In Figs. 3(a) and 3(b) the variation of the PL decay and rise time with the energetic position is depicted for 4 K and RT, respectively. At 4 K the PL emission peak is asymmetric with a low-energy tail. Both the rise (250–80 ps) and the decay (1720–680 ps) times decrease with increasing energy (1.146–1.168 eV). The rise time is determined by how fast the excitons can relax into the states where they recombine. In our experiment we do not excite the quantum wells resonantly, but the GaAs barriers which explains the relative long rise times only in part. We think that transfer processes between different localized states mainly determine the rise times. Three different processes account for the decay of localized excitons: radiative, nonradiative recombination, and transfer processes. Assuming the density of tail states is proportional to $\exp(-E/E_0)$ the PL decay time for excitons in localized states as function of the spectral position can be described by

$$\tau(E)=\frac{\tau_{\mathrm{rad}}}{1+\exp\{(E-E_{\mathrm{me}})/E_0\}}, \quad (1)$$

where E_0 is a characteristic energy for the density of states, τ_{rad} the radiative lifetime, and E_{me} the energy for which the radiative lifetime equals the transfer one.[12] The underlying model presumes that excitons transfer to lower-energy localized states by the tunnel effect. In Ref. 5 a similar recombination dynamic was found for a $Ga_{1-x}In_xN_yAs_{1-y}$ epilayer, but with lower absolute values for the PL decay time. Figure 3(b) shows the decay and rise times measured at RT. In contrast to the results at 4 K the decay time first increases with decreasing energy, but after passing the PL emission maximum decreases again. However, the relative difference between maximum (510 ps) and minimum (420 ps) decay time is much smaller compared with the low temperature measurements. The absolute values for the decay constants are by more than a factor of 3 smaller than at 4 K. The rise time remains constant with energy within the experimental accuracy and is now given by the quickness of the carrier capture from the barriers in the wells only. The recombination dynamics at RT is determined by quantum well properties. Exciton scattering with rough interfaces and nonradiative processes, such as phonon-assisted and defect-related processes, dominate the recombination. In Fig. 4 we compare the decay times of the four samples at RT and 4 K. At He temperature the decay time increases from 1560 to 4000 ps varying the nitrogen molar fraction from 1.5% to 2.6%. This is due to an increasing degree of localization leading to a smaller overlap of the electron and hole wave function. At RT the decay time increases up to 2.3% nitrogen (510–760 ps) but drops significantly for the 2.6% sample (185 ps). This

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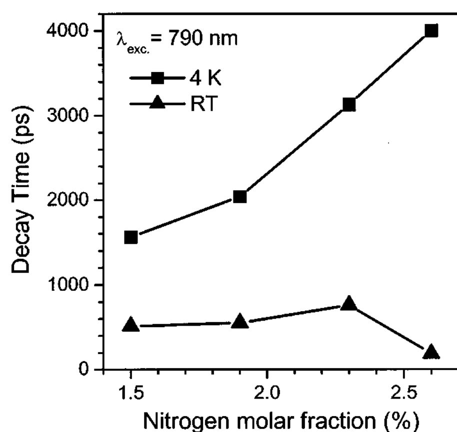

FIG. 4. Decay constants measured at the PL maximum of four different samples at RT (up triangles) and 4 K (squares), respectively.

can be explained by a higher defect concentration in this sample due to the incorporation of more nitrogen thus leading to a stronger influence of the defect-related nonradiative processes.

In conclusion, temperature-dependent and time-resolved photoluminescence experiments have been employed to investigate the recombination mechanisms in $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs MQWs with various nitrogen concentration at different temperatures. We have strong indication that band-tail states play a key role for the recombination process in $Ga_{1-x}In_xN_yAs_{1-y}$/GaAs MQWs at 4 K. The band-tail states in our samples may originate from composition nonuniformity of nitrogen and indium, as well as structural inhomogeneities. Since similar results were found for a $Ga_{1-x}In_xN_yAs_{1-y}$ epilayer with much lower indium concentration,[5] we suggest that local fluctuations in the nitrogen concentration have the highest impact on the recombination dynamics in the quaternary alloy. For the dynamic at RT nonradiative recombination processes become important. The behavior can be understood assuming nonideal quantum wells including rough interfaces and defects. Due to a higher concentration of defects and therefore increased nonradiative processes the decay time of the sample with the highest nitrogen concentration is much shorter than for the other three samples.

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