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Recombination dynamics of localized excitons in InGaN quantum dots

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Indium-rich fluctuations in ultrathin InGaN layers act at low temperatures as a dense ensemble of quantum dots (QD). This leads to a complex potential landscape with localization sites of widely varying depth for excitons. We report on investigations of the recombination mechanisms of excitons localized in InGaN/GaN QD structures by time-resolved and spatially resolved photoluminescence (PL) measurements. The structures were grown by metal-organic chemical-vapor deposition on Si (111) substrates. Sharp lines originating from single QDs could be observed. Their PL decays show monoexponential behavior. Similar transition energies have different time constants. Thus, the well-known nonexponential PL decay of the QD ensemble is assigned to the summation of monoexponential decays originating from individual QDs with different exciton lifetimes. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1790599]

Group III nitride-based optoelectronic devices are used in a wide range of applications due to their large band gap range. InGaN/GaN heterostructures form the active layer in blue-green light-emitting diodes (LED's) and laser diodes commercially available today.[1] Quantum dot (QD) structures could still improve their performance enormously.[2] In particular, InGaN QD lasers are expected to have low-threshold current density and better temperature stability compared with conventional blue lasers with InGaN quantum wells (QW).[3] Consequently, InGaN QD structures have been intensively investigated in recent times. Growth of nanometer-scaled islands, similar to the well-known Stranski-Krastanov growth mode, as well as QD properties of In-rich fluctuations were reported.[3–7] Spatially resolved luminescence experiments demonstrated sharp emission lines originating from single QD states with δ-like density of states.[7,8] Time-resolved luminescence investigations on such emission lines were reported.[8] Nevertheless, the processes that govern the recombination dynamic of an exciton localized in individual InGaN QDs are still poorly understood. For a deeper comprehension of these processes a single QD state and its interaction with the whole QD ensemble is to be studied.

In this letter we present time-resolved photoluminescence (PL) investigations on InGaN QD structures. Monoexponential decay of the PL is determined for emission lines from single QDs. Meanwhile a nonexponential decay was observed for the luminescence of the entire QD ensemble. This behavior is attributed to disorder in the size and in the composition distribution of the QDs.

The sample was grown by low-pressure metal-organic chemical-vapor deposition (MOCVD) on Si (111) substrate. The growth process is described elsewhere.[7] Previous cross-section TEM measurements showed composition fluctuations in the InGaN layer with indium-rich domains. Their lateral size of about 5 nm is comparable to the exciton Bohr radius, thus small enough to provide not only strong localization of excitons, but also confinement of their wave functions. The sample was masked with a 70 nm thick metal cap leaving apertures of about 200 nm diam for spatially resolved investigations. Previous cathodoluminescence investigations demonstrate emission lines originating from single QD states.[7]

The InGaN QD sample was investigated by time-resolved and time-integrated PL spectroscopy. The QD luminescence was excited at 353 nm by the second harmonic wave of a mode-locked Ti:sapphire laser. The temporal width of the laser pulses was 2 ps at a repetition rate of 80 MHz. The PL measurements were performed in a helium-flow microscope cryostat at a temperature of 5 K. The luminescence light was collected through a microscope objective. While the luminescence of the entire QD ensemble was detected from unmasked areas, single QD luminescence was detected through 200 nm apertures in the metal mask. The detection system consisted of two 0.35 m McPherson monochromators in subtractive mode and an ultrafast photo detector (multichannel plate) providing a spectral resolution of about 1 meV and a time resolution of better than 30 ps.

The PL spectrum of the entire QD ensemble [Fig. 1(a)] has its maximum at 3.05 eV and a full width at half of maximum (FWHM) of 75 meV. Sharp emission lines of discrete QD states were observed from 2.8–3.2 eV indicating QD origin of the entire emission.[8] In Fig. 1(b) the decay of the integral PL intensity is shown. No rise time was found. A nonexponential decay was observed. Therefore, we fitted our data with stretched exponential model

$$I(t) = I_0 \exp[-(t/\tau^*)^{\beta}], \tag{1}$$

developed for heavily disordered materials[9] [dashed line in Fig. 1(b)]. Here, τ^* is the time constant and β is the stretching parameter. The variation of β from unity is a measure for the degree of disorder in the material. For the investigated QD ensemble $\beta \sim 0.80$ was obtained. This well-known nonexponential behavior[5,6,8] was previously interpreted as a temporal dependence of the decay time. It was attributed to the screening of piezoelectric fields, which influences electron-hole wave-function overlap through the quantum-confined Stark effect (QCSE). It was suggested that at high excitation densities, reached by pulsed lasers, piezoelectric fields could be screened by nonequilibrium charge carriers. This screening would initially lead to a stronger wave-function overlap,

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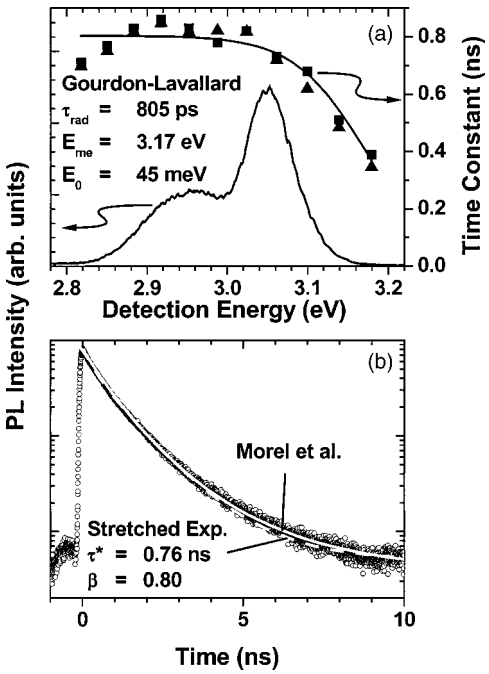

FIG. 1. (a) Time-integrated ensemble PL, time constant τ_e of the $1/e$ decay (squares) and the fitting parameter τ^* (triangles), fit with Eq. (2) (solid line). (b) Time-resolved ensemble PL, Fits with Eq. (1) (dashed line) and Morel *et al.* (Ref. 5) (solid line).

while progressive depopulation would be followed by electron-hole separation and result in increasing lifetimes.[10] This effect should be dependent on excitation density. We observed that a decrease of the excitation density of two orders of magnitude did not result in any change of the shape or the time constant of the transient PL.

An alternative explanation can be related to a broad distribution of exciton lifetimes in QDs with similar transition energies. Morel *et al.*[5] attribute this variation of lifetimes to an in-plane separation of electrons and holes in different localization centers. Though this model yields a good approximation of our results [solid line in Fig. 1(b)], it will be shown that individual QDs have different exciton lifetimes and that this leads to the nonexponential decay of the QD ensemble.

In Fig. 1(a) the spectral dependence of the PL time constants is shown. The fitting parameter τ^* from the stretched exponential as well as the $1/e$ decay time τ_e are presented. Both time constants show a similar spectral dependence. Their decrease with increasing energies is generated by the rise of transfer processes from QDs providing smaller localization energy. Hence, the decrease of the decay time with increasing energies indicates the rising contribution of nonradiative recombination of localized excitons. To determine the radiative lifetime of the localized excitons as well as their average binding energy and mobility properties, we evaluated the results with a model introduced by Gourdon and Lavallard[11] accounting for lateral energy transfer in a high-density QD ensemble. Assuming the density of tail states is proportional to $\exp(-E/E_0)$ the PL decay time for excitons in localized states as a function of the spectral position can be described by

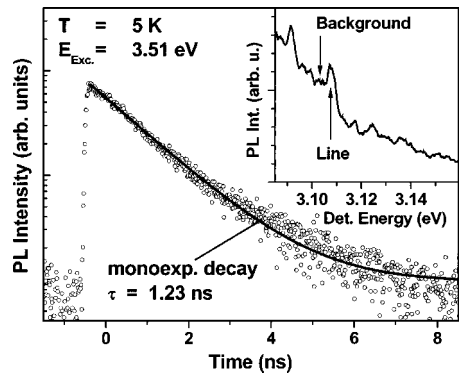

FIG. 2. Time-resolved PL of a single QD emission line (inset), fitted by a monoexponential decay (solid line).

$$\tau(E) = \frac{\tau_{rad}}{1 + \exp[(E - E_{me})/E_0]}, \tag{2}$$

where τ_{rad} is the radiative lifetime, E_{me} the energy for which the radiative lifetime equals the lateral transfer time and E_0 is a characteristic energy for the density of states. The latter parameter is a measure for the average localization energy of the QDs. E_{me} denotes the change from three-dimensional-localized QD states to QW states with higher exciton mobility. Above this energy the transfer of excitons is more probable than their radiative decay.[12] Applying Eq. (2) [solid line in Fig. 1(a)] yields a radiative lifetime of $\tau_{\mathrm{rad}} \sim 805$ ps, an average binding energy of $E_0 \sim 45$ meV and an $E_{\mathrm{me}} \sim 3.17$ eV. The most striking result is that nearly the whole PL is emitted at energies below E_{me}. Thus, the spectral dependence of τ^* and τ_e yields another unambiguous proof for the QD origin of the luminescence. While Eq. (2) gives a good approximation for our results at higher energies, both, τ^* and τ_e differ from the fitting curve on the low energy tail of the QD ensemble emission band. We attribute the decrease of the decay times with lower energy to an increasing confinement of excitons in QDs with higher localization energy and thus with a higher electron-hole overlap.

The dynamics of excitons in single QDs were investigated using μ-PL. Figure 2 shows the transient behavior of a single QD line emphasized in the inset. Superimposed to the studied line there is a background signal that made up to 70% of the PL intensity. This background signal is attributed to luminescence light from the QD ensemble transmitted through the mask and scattered light from other mask holes. To eliminate the transient behavior of the background and isolate the single QD emission line, two transients for each QD line were recorded: one detected at the maximum of the line and one 5–10 meV aside. Since the changes in the PL dynamic of the entire QD ensemble are negligible for energy differences of a few meV (see also Fig. 1), the latter transient was subtracted from the first to obtain the true PL decay of the single QD state as suggested by Robinson *et al.*[8] As opposed to the ensemble luminescence, single QD lines show a single-exponential behavior. Their decay times range between 0.4 and 1.6 ns.

In Figure 3 the observed decay times as a function of the transition energy are depicted for the ensemble luminescence (squares) and for the investigated single QDs (triangles). A large spread of the latter was observed even at similar detection energies. Consequently, QD states with the same transition energy do not necessarily have the same exciton life-

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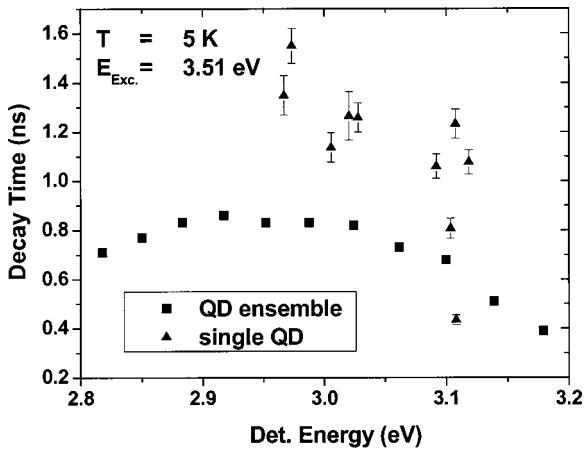

FIG. 3. Spectral dependence of time constants for single QD lines (triangles) and the QD ensemble (squares).

time. In fact, the dynamics of luminescence are governed by two competing processes: the transition probability which primarily depends on electron-hole wave-function overlap and the probability of nonradiative escape. The wave-function overlap is affected by QD size, shape, and the depth of its localizing potential. The latter is not given by its absolute indium content, but depends on the difference of the indium concentration in the QD and its adjacent area. Due to the disordered distribution of the indium content in the investigated sample,[7] there is no correlation of the localization energy to the transition energy. Luminescence at a certain detection energy originates from a QD subensemble with the same transition energy. However, this subensemble still consists of QDs with a broad distribution of electron-hole wave-function overlap. The transfer probability shows a similar behavior. Generally, the trend towards shorter time constants for higher transition energies (Fig. 3) can be explained by an increasing importance of escape processes. Nevertheless, the transfer probability also differs inside one QD subensemble. For QDs of a subensemble this leads to different time constants. Thus, the nonexponential PL decay is assigned to the summation of monoexponential decays originating from individual QDs.

In conclusion, the recombination dynamics of excitons localized in a single InGaN QD was compared to the recombination dynamics of the entire QD ensemble. The dynamic of the exciton decay was found to be monoexponential. The nonexponential PL decay of the entire QD ensemble gives an unambiguous proof that the disorder of the QD system governs the recombination dynamics.

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