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Relaxation and recombination dynamics in GaN/Al_2O_3 epilayers

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Abstract. The dynamics of free- and bound-exciton recombinations in n-type GaN are studied as a function of temperature. The very short lifetime of 6 ps determined for the free exciton points to a strong influence of non-radiative processes introduced by impurities and defects. The high concentration of shallow donors gives rise to thermal equilbrium between the donor-bound-exciton and the free-exciton populations at higher temperatures governing the temperature dependence of the recombination dynamics.

1. Introduction

The high potential of the InGaAlN material-system for optoelectronic devices in the blue and ultraviolet spectral region [1] has triggered efforts of physicists world-wide to grow high-quality p- and n-type GaN epitaxial layers, characterize, and understand their properties. Excitons play a key role for an understanding of the electronic and optical properties of these wide-gap semiconductors. Beside the spectral position and lineshape of excitonic luminescences especially their dynamical behaviour reveals useful information on physical processes involving shallow impurities necessary to control the electronic properties of the material. Picosecond time-resolved luminescence experiments of free- and bound-exciton luminescences of epitaxial GaN/SiC showed that non-radiative processes introduced by relatively high defect and carrier concentrations impact upon

the lifetimes of excitons [2]. To further understand the physical mechanisms involved we studied the temperature dependence of excitonic emissions from a state-of-the-art GaN/Al_2O_3 sample.

2. Results

The sample investigated is a n-type, 400 μm epitaxial layer of GaN grown by hydride-vapour-phase epitaxy on (0001)-Al_2O_3 with a room temperature carrier density of 10^{17} cm^2. Details of the growth procedure have been published elsewhere [3].

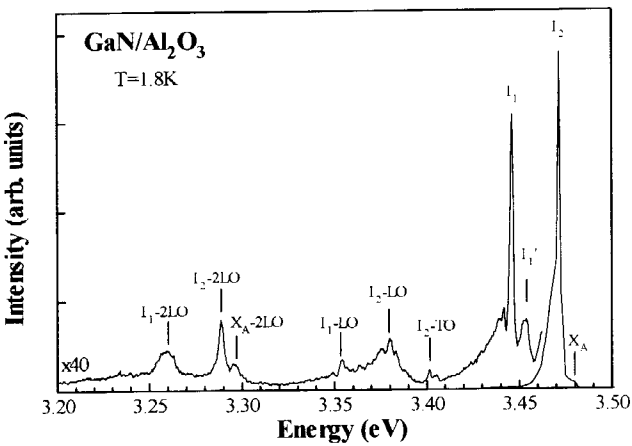

Figure 1. Low-temperature cw photoluminescence spectrum of the excitonic luminescences of GaN/Al_2O_3.

In Fig. 1 the low-temperature cw photoluminescence spectrum excited by a HeCd laser at 325 nm is shown. The phonon sideband is expanded by a factor of 40. The very intensive donor-bound-exciton line I_2 at 3.4728 eV dominates the luminescence. Its full width at half maximum (FWHM) amounts to 920 μeV. We find TO, LO, and 2LO phonon replica of I_2, the phonon energies precisely in accordance with those determined from Raman measurements performed on the same sample [4]. On the high-energy side of I_2 emission from the free exciton (labelled X_A) is resolved. An acceptor-bound-exciton line I_1 appears at 3.4464 eV with a FWHM of 2.1 meV, followed by LO and 2LO phonon-replica. Monemar and Lagerstedt reported this line before (I_{L5} in [5]). 6.7 meV above I_1 we find a weak line labelled I_1' at 3.4531 eV. The chemical nature of the acceptor giving rise to these two lines is unknown. We attribute both lines to one acceptor for reasons explained below.

Luminescence transients of X_A, I_2, and I_1 after band-to-band excitation at 3.6 eV with 6 ps laser pulses were recorded at temperatures between 4.2 K and 65 K. Decay time constants were determined employing standard convolution techniques. The first important result is that the luminescence rise processes of all three emissions at all temperatures chosen are faster than the resolution of our setup (15 ps). This points to a very fast capture of free excitons at the shallow impurities. In this context it is interesting to follow the temperature dependence of the decay of the free-exciton emission in Fig. 2.

An offset of time as well as intensity was added to each transient to allow a comparison for all temperatures. A very fast decay is observed between 4.2 K and 12 K. Then, up to 28 K it slows down significantly. Further raising the temperature results in a faster decay again, until at 65 K it is close to the limit of our time resolution. This

the intensity decay for more than the first decade. It is shown in Fig. 3 by the open circles. The time constant rises from ≤15 ps to 63 ps between 12 K and 28 K, and drops back down to 17 ps at 65 K.

To understand this behaviour it is necessary to compare it with that of the lifetime of the donor-bound exciton (D^0, X), given by the diamonds in Fig. 3. The decay of this luminescence is monoexponential for more than two orders of magnitude. A lifetime of 70 ps is observed at 4.2 K, decreasing continuously as a function of temperature. It is striking and a further essential result of our experiment that above 30 K the time constant of (D^0, X) agrees with that of the free exciton within the margin of error. We ascribe this to a thermal equilibrium established between the donor-bound-exciton and free-exciton populations mediated by the phonon-assisted dissociation of the bound exciton from the donor.

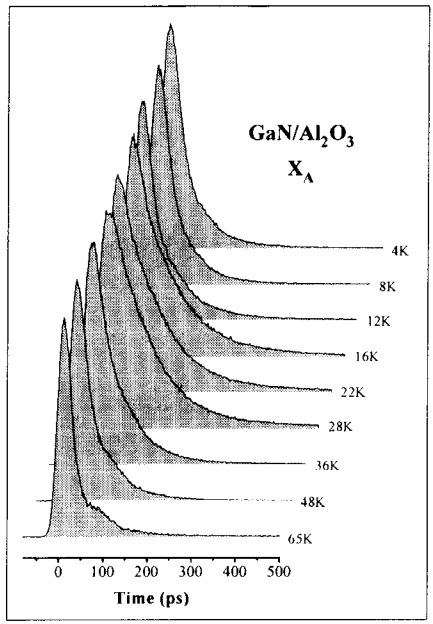

Figure 2. Transients of the free-exciton emission recorded at various temperatures.

A confirmation of this interpretation is provided by the calculation of the lifetime of the donor-bound exciton in the framework of a three-level system with thermal equilibrium in the upper two states, cf. the inset of Fig. 3 . Given the low-temperature lifetime τ_1 of (D^0, X) and its localization energy ΔE of 7.2 meV the decay of the total population $n = n_{(D^0,X)} + n_{X_A}$ has to be calculated using a Boltzmann factor for the population ratio of the excited states. An exponential decay is derived [6], governed by the time constant

$$\tau = \tau_1 \left[1 + \exp\left(-\frac{\Delta E}{kT}\right)\right] \cdot \left[1 + \frac{\tau_1}{\tau_2} \exp\left(-\frac{\Delta E}{kT}\right)\right]^{-1} \qquad (1)$$

The solid curve shown for (D^0, X) in Fig. 3 is a fit to the experimental data obtained by using a free-exciton lifetime $\tau_2 = 6 \pm 1$ ps. This value agrees with our observation that the free-exciton lifetime is below the limit of our time resolution. It shows that despite the good quality of the sample expressed by the low half-width of I_2 there is still a severe influence of impurity scattering on the lifetime of the free exciton.

At this point it is clear that the decay constant of the free-exciton-emission intensity plotted in Fig. 3 cannot be attributed to the decay of the free-exciton population.

It is well known that from a luminescence transient one cannot distinguish between population-rise and -decay processes. The faster process determines the luminescence rise and the slower the luminescence decay. We therefore attribute the observed decay-constants of the free-exciton *emission* X_A to the rise of the free-exciton *population*. At temperatures above 30 K the repopulation of the free-exciton state by the donor-bound excitons is dominating due to the high concentration of shallow donors. At low tempe-

framework of a simple three-level system as shown by the first results of a more detailed analysis to be published soon. A spectral broadening of the relaxation bottleneck as observed for free excitons in high-purity CdS [7],[8] cannot be seen in our sample due to high impurity scattering rates.

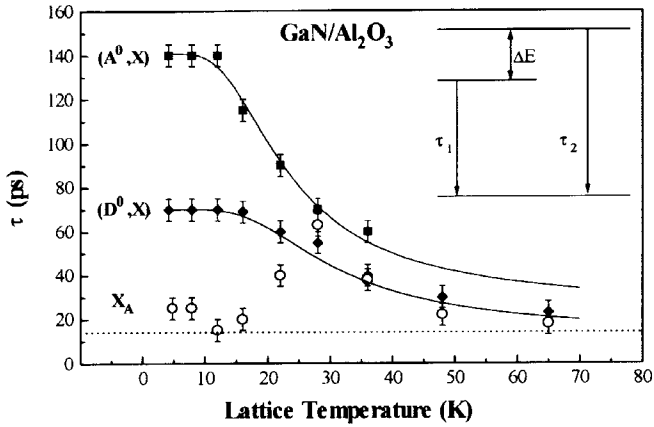

Figure 3. Decay time-constants of the free exciton, the donor-bound exciton and the acceptor-bound exciton as a function of temperature. The solid curves were calculated using eq. (1) derived for the three-level system shown in the inset.

For the acceptor-bound exciton a similar mechanism as for the donor-bound exciton is seen to govern the dependence of the lifetime on temperature. Here, a thermal equilibrium with an excited state 6 ± 1 meV above (A^0, X) whose lifetime is 11ps is found to be responsible for the decay of the lifetime. This energy difference is in good agreement with the splitting between I_1 and I_1' as determined from Fig. 1. We therefore assign line I_1' to an excited state of the (A^0, X)-complex giving rise to I_1. The small energy difference between these lines excludes the $A_{n=2}$ state of the hydrogen-like exciton series as origin of this excited state. The most likely explanation at this point seems to be the splitting of the bound-exciton state caused by exchange interaction between the impurity- and the exciton-hole.

References

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