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LUMINESCENCE DYNAMICS OF EXCITON REPLICAS IN HOMOEPITAXIAL GaN LAYERS

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Photoluminescence of excitons and their phonon replicas in homoepitaxial MOCVD-grown gallium nitride (GaN) layers have been studied by picosecond (ps) time-resolved photoluminescence spectroscopy. The time-resolved photoluminescence spectroscopy has shown that the free excitons and their replicas have the fastest dynamics (decay time of about 100 ps). Then, the excitons-bound-to-donors emission rises (with the rise time similar to the free excitons decay time) and decays with $t = 300$ ps. The excitons-bound-to-acceptors has the slowest decay (about 500 ps). It has been found that the ratio of excitons-bound-to-acceptors and excitons-bound-to-donors amplitudes and their decay times are different for 1-LO replicas and then for zero-phonon lines, whereas the ratio of amplitudes and the decay time of the 2-LO replicas are similar to the ones of the zero-phonon lines.

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Gallium nitride is a very promising material for optoelectrical devices. The performance of such devices depends upon the dynamics of recombination processes. We have measured the luminescence dynamics of excitons and their replicas in GaN.

The GaN samples used in our experiments were homoepitaxial layers grown by metalorganic chemical vapour deposition (MOCVD) on GaN single crystals. Their thickness was about 1 μm. Measurements have been done at the temperature of 1.8 K. A mode locked solid state Ti:sapphire laser has been used as an excitation source. This produces 2 ps pulses, which are frequency doubled with a BBO crystal to an energy of about 3.53 eV. For spectral measurements the emitted PL has been detected with a cooled CCD camera, while the time resolved measurements have been performed with a synchroscan streak camera.

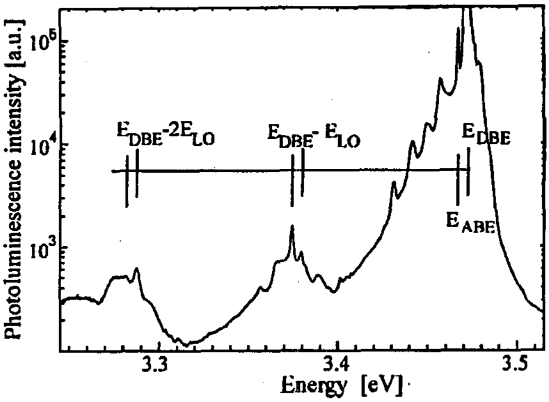

Fig. 1. Photoluminescence spectra showing excitons lines and their phonon replicas. Vertical lines separated by the energy of LO phonon ($E_{\mathrm{LO}} = 92$ meV) indicate expected positions of the replicas.

The excitonic luminescence spectrum of homoepitaxial GaN consists mainly of two strong lines connected with recombination of excitons bound to donors (DBE) and acceptors (ABE) [1]. In addition there are observed relatively weak lines originating from the free excitons recombination [1] (see Fig. 1). Interaction with longitudinal optical phonons leads to creation of phonon replicas. Energy of the LO phonon ($E_{\mathrm{LO}} = 92$ meV [2]) has been plotted in Fig. 1 to indicate expected positions of the replicas. The two-phonon replica is usually hardly visible because it lies close to the position of the donor–acceptor emission. In the presented spectrum the donor–acceptor emission has been numerically subtracted. It could be observed that the first replica of DBE is very weak. In fact it is 7000 times weaker than the zero-phonon line. The ABE replica is only 90 times lower than its zero-phonon line. As a result the ABE replica is stronger than the DBE replica even in the samples where the DBE zero-phonon line is much stronger than the ABE one. The ration of amplitudes changes back for the second replicas which have proportions similar to the zero-phonon lines. The DBE+2LO line is stronger than the ABE+2LO line (they are attenuated 9000 times and 900 times, respectively). The small relative intensity of 1-LO DBE replica is probably caused by a higher radius of the DBE wave function [3]. A bigger DBE radius indicates that the volume of DBE in k-vector space will be much smaller than the ABE volume. Thus the number of interacting LO states will be small. For the second phonon replica many different combinations of the two-phonon wave vectors are possible for a given k-vector of the exciton, so the exciton–phonon interaction can be efficient even in a small volume of wave-vector space.

The time evolution of the excitonic lines has been observed for a few samples with different donor to acceptor ratios. It has been observed that in all samples the fastest kinetics are connected with the free excitons luminescence. The bound excitons have slower kinetics, but one can see that the DBE kinetics are faster than the ABE ones.

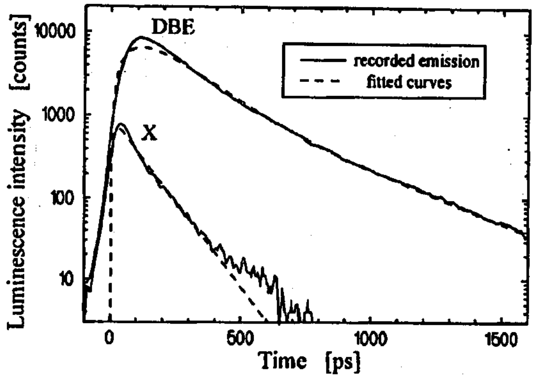

Fig. 2. Evolution of luminescence of free excitons (X) and donor bound excitons (DBE). The curve fitted to free exciton emission has a single decay time of 104 ps. The DBE emission is fitted with the double-decay curve.

Let us assume that creation of excitons is fed from the source which is excited at $t = 0$ and which decays exponentially *ca.* $\exp(-t/t_{\mathrm{H}})$. Growth and decay of the population of excitons which are fed from this source and which have the effective decay time t_{X} ($t_{\mathrm{X}} > t_{\mathrm{H}}$) can be described by the following expression:

$$I(t) = I(0)[-\exp(-t/t_{\mathrm{H}}) + \exp(-t/t_{\mathrm{X}})]. \tag{1}$$

This relation fits well to the temporal evolution of the free excitons (Fig. 2). The obtained growth rate $t_{\mathrm{H}} = 10$ ps corresponds rather to the time resolution of the streak camera system than to the hot carriers thermalization, which is about 1.5 ps [4]. The free exciton decay time $t_{\mathrm{X}} = (104 \pm 10)$ ps has been obtained. Fitting Eq. (1) to the DBE decay gave $t_{\mathrm{DBE}} = (283 \pm 30)$ ps, but the poor quality of the fit suggested that the process is not the single-exponential decay. Therefore we have used the double-decay curve

$$I(t) = I(0)[-\exp(-t/t_{\mathrm{H}}) + a\exp(-t/t_{\mathrm{DBE1}}) + (1-a)\exp(-t/t_{\mathrm{DBE2}})] \tag{2}$$

with the decay times $t_{\mathrm{DBE1}} = (124 \pm 10)$ ps and $t_{\mathrm{DBE2}} = (344 \pm 20)$ ps. The growth rate of DBE $t_{\mathrm{H}} = (92 \pm 10)$ ps is similar to the free excitons decay time and is much longer than the hot carriers lifetime. This fact suggests that the DBE are formed rather by binding of free excitons than by independent capture of holes and electrons. The fit of Eq. (1) to the ABE emission gives $t_{\mathrm{ABE}} = (460 \pm 60)$ ps. The obtained decay times of excitons are nearly as long as the radiative ones, which have been observed in bulk GaN grown by hydride vapour phase epitaxy [5]. This suggests that nonradiative recombination in our sample is weak.

The contour plot of the streak camera image of the first replica region is shown in Fig. 3. It is observed that the temporal sequence of the replicas is the same as for the zero-phonon lines. The fastest one is the free exciton replica ($t_{\mathrm{XR}} = (163 \pm 20)$ ps), then the DBE replica $t_{\mathrm{DBER}} = (350 \pm 50)$ ps. The ABE has the slowest decay $t_{\mathrm{ABER}} = (570 \pm 50)$ ps. It seems that the decay times of replicas are longer than average decay times of zero-phonon lines. The DBE zero-phonon line has two decay times and the longer one is similar to the replica decay time, which indicates that phonons couple mainly with long-living population of excitons.

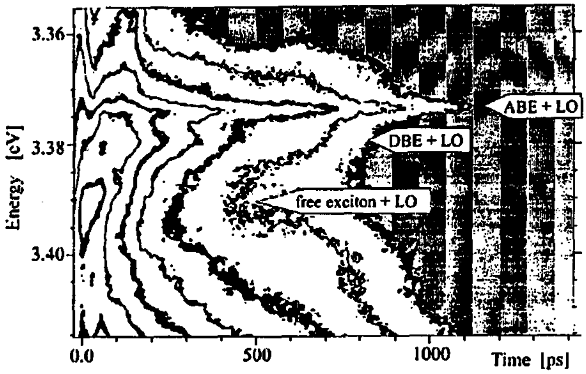

Fig. 3. Contour plot of the streak camera image of the first replica region. Values of contours are equidistant in logarithmic scale. The signal value changes twice from one contour to another.

It was also possible to fit Eq. (1) to the DBE second replica emission. Its decay time is $t_{\mathrm{DBE2R}} = (315 \pm 40)$ ps. This time seems to be faster than the first replica decay time and is similar to the average decay time of zero-phonon line t_{DBE}. This shows that the second replicas are similar to zero-phonon line not only in amplitude proportions but also in time constants.

In summary, the spectral and time resolved luminescence of excitons and their replicas in homoepitaxial GaN was analysed. The free excitons have the fastest kinetics ($t_{\mathrm{X}} = (104 \pm 10)$ ps), the bound excitons are slower. The comparison of t_{X} and growth time of DBE suggests that the DBE are formed rather by binding of free excitons than by independent capture of holes and electrons. It has been found that the ratio of ABE and DBE amplitudes and their decay times are different for 1-LO replicas than for zero-phonon lines, whereas the ratio of amplitudes and decay time of the 2-LO replicas are similar to the ones of the zero-phonon lines.

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