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EFFECT OF THE EXTRINSIC AND TEMPORAL CARRIERS ON RADIATIVE RECOMBINATION OF III-NITRIDE NANOSTRUCTURES*

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Abstract – Due to many important applications, the group III-Nitride semiconductors have recently attracted remarkable attention among semiconductor researchers and engineers. In this paper, we report on the impact of extrinsic and temporal carriers on the screening of polarization internal fields. The optical efficiency of GaN/AlGaN multiple quantum well (MQW) nanostructures were studied by means of photoluminescence (PL) and time-resolved PL measurements. Extrinsic carriers come from Si doping in the barriers, while temporal carriers originate when the samples are excited by laser beam. The emission peaks of MQWs in PL spectra of the undoped and low-doped samples show a shift towards higher energy levels as excitation intensity increases, while the other samples do not exhibit such a phenomenon due to the dominance of the extrinsic carriers. The transient data confirm the results of the PL measurements.

Keywords – Nanostructures, photoluminescence (PL), GaN/AlGaN multi quantum well, time Resolved PL (TRPL), polarization fields, exciton, III-Nitride semiconductors, quantum well

1. INTRODUCTION

III-nitride material family, especially GaN, have attracted much interest during the last few years. Such materials are particularly well suited for optoelectronic applications, e.g. as UV light emitters in the lighting solutions that are expected to replace today's incandescent and fluorescent lamps. Some other applications include laser diodes (LDs) as well as high temperature and high power electronic devices [1, 2]. In order to optimize the function of such devices, It is necessary improve our knowledge of the material properties and, in particular, understand the recombination mechanisms in III-nitride QWs. A peculiarity of nitrides is the significant macroscopic polarization with both spontaneous and piezoelectric components [3, 4]. These polarization charges create internal fields in the QWs that have a fundamental influence on optical properties, strongly affecting the oscillator strengths of excitons and the spectral position of the corresponding photoluminescence (PL) peaks. The radiative recombination processes are intensely modified by these built-in electric fields in the [0001] direction, resulting in a substantial quantum confined stark effect (QCSE). Screening by doping (extrinsic carriers) and/or photo-generated carriers (temporal carriers), strongly affects the polarization-induced properties [5]. The modulation doping technique (doping in the barriers) can be used to screen the macroscopic polarization fields in nitride quantum wells. The screening of the fields increases the overlap between the confined electron and hole wave functions, enhancing the radiative transition probability across the gap.

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In this paper, we discuss some recent results of MOCVD grown GaN/AlGaN MQWs nanostructures, all grown without an additional capping layer. In addition to the polarization fields inside the MQWs, the influence of the outer surface in the absence of the capping layer is, indeed, present.

2. SAMPLES AND EXPERIMENTAL PROCEDURE

A set of GaN/AlGaN MQW structures with Si doping concentration in the range of nominally undoped (with a residual donor doping of about 1×10^{16} cm^{-3}) up to 9×10^{19} cm^{-3} doping level in the AlGaN barriers has been studied by means of PL and time-resolved PL (TRPL). The MQW structures were grown on (0001) sapphire substrates by means of metal organic chemical vapor deposition (MOCVD) [6].On top of the substrate, an AlN layer with a thickness of 20 nm and a thick nominally undoped GaN buffer layer of about 2 μm were grown. This was followed by a multiple quantum well structure with five-period of 3 nm wide GaN quantum well layers separated by 7 nm wide $Al_{0.07}Ga_{0.93}N$ barriers. No additional capping layer was used so that the outermost QW barrier faces the surface.

For optical excitation in the PL measurements, we have used the fourth harmonic of a Nd:Vanadate laser with a wavelength of 266 nm. The PL signal was dispersed by a single-grating monochromator and detected by a UV enhanced liquid nitrogen cooled CCD camera. For the transient PL data, we used the third harmonic (λ_{exc} = 266 nm) of a Ti: sapphire femto-second pulsed laser. The PL transients were also detected by a UV sensitive Hamamatsu streak camera system with a temporal resolution better than 10 ps. The samples were then placed in a variable temperature cryostat for measurements at low temperature (2 K).

3. RESULTS AND DISCUSSION

As mentioned in our previous reports, [7, 8] the existence of polarization fields in III-nitrides QWs is a well-known concept. The abrupt variation of the polarization at surfaces and interfaces gives rise to large polarization sheet charges that in turn create a huge internal electric field of the order of MV/cm. The piezoelectric (P_{pz}) and spontaneous (P_{sp}) polarizations in GaN well layers for the undoped sample have been estimated at about 0.002 (C/m^2) and -0.034 (C/m^2), respectively [7], and the total internal polarization field (E_w) has been calculated -0.48(MV/cm). The internal field in the growth direction causes band bending and forces the electrons and holes to the opposite side of the well decreasing the overlap of the electron-hole wave functions, resulting in the reduction of the transition probability. In addition, the internal polarization field reduces the energy emission of the recombination, leading to a red shift on the PL peak position. This well known effect is referred to as the Quantum Confined Stark Effect (QCSF).

The cw (continuous wave) PL spectra of GaN/AlGaN MQW with different doping levels measured at T=2 K reveals a blue shifted broad QW emission with respect to the 3.48 eV PL peak from the GaN buffer layer due to the quantum confinement. The PL spectra for undoped and heavy-doped samples can be seen in Fig.1. The QW emission is attributed to excitons localized at the energy minima induced by well width fluctuations [9]. The internal field is screened more efficiently by increasing the doping level, consequently the QCSF is reduced and the QW's peak shifts to higher energies.

The time integrated PL spectra of the samples at low temperature clearly indicates that the enhancement of the optical efficiency in the heavy-doped sample corresponds to the undoped one (Fig. 1), which can be explained if we take into account the more effective screening of the internal polarization fields of the doped samples by the electrons originating from the doping in the barriers. Screening the fields increases the overlap between the confined electron and hole wave functions, and the optical efficiency is enhanced by increasing the modulation doping [9]. The variation of optical efficiency versus temperature has been shown in Fig. 2. As can be seen, in all temperatures optical efficiency which have

been deduced from $\eta = \tau_{nr} /(\tau_{nr} + \tau_{rad})$ is larger in the high doped sample in comparison to the undoped one. τ_{nr} and τ_{rad} are radiative recombination time and non-radiative recombination time respectively.

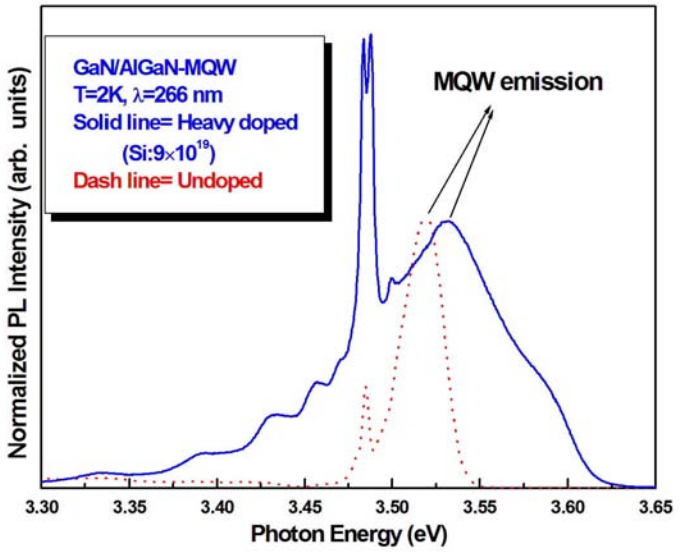

Fig. 1. PL spectra of undoped and heavy doped samples at 2 K (clearly the optical efficiency in the heavy doped sample is higher than that of the undoped one)

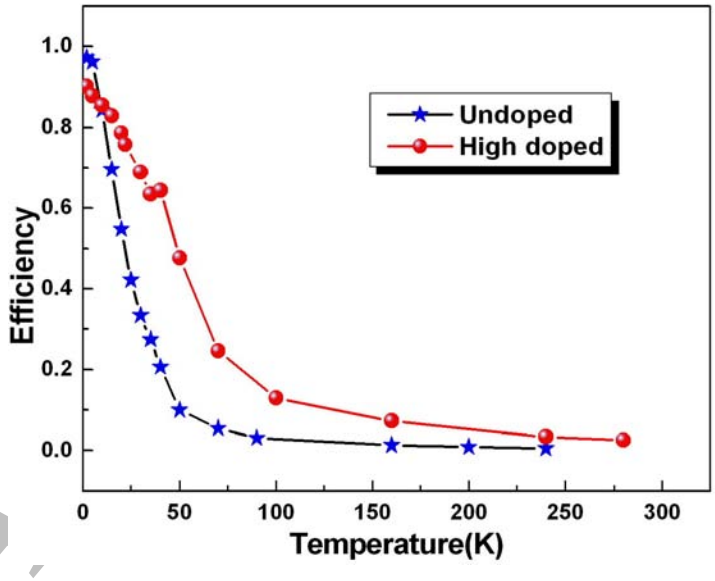

Fig. 2. Optical efficiency variation versus temperature for high doped and undoped samples

The outermost barrier (outermost AlGaN layer) faces the surface. Therefore the QWs will experience a strong depletion field [10], which is also expected to vary strongly between the different QWs in the MQW region. Figure 3 shows the variation of the full width at half maximum (FWHM) of the QW peak versus the doping level. FWHM increases by increasing the doping level, which can be explained by varying the internal polarization fields due to the variation of the depletion fields in different samples, as well as the large contribution of structural defects induced by high Si doping and nonuniform absorption of the excitation energy at different QWs. FWHM in the undoped sample is 23 meV, comparable with similar GaN/AlGaN QWs structures grown by molecular beam epitaxy (MBE), exhibiting a typical FWHM of about 20–25 meV [11], which shows that the quality of our MOCVD grown samples is comparable with the best MBE grown ones.

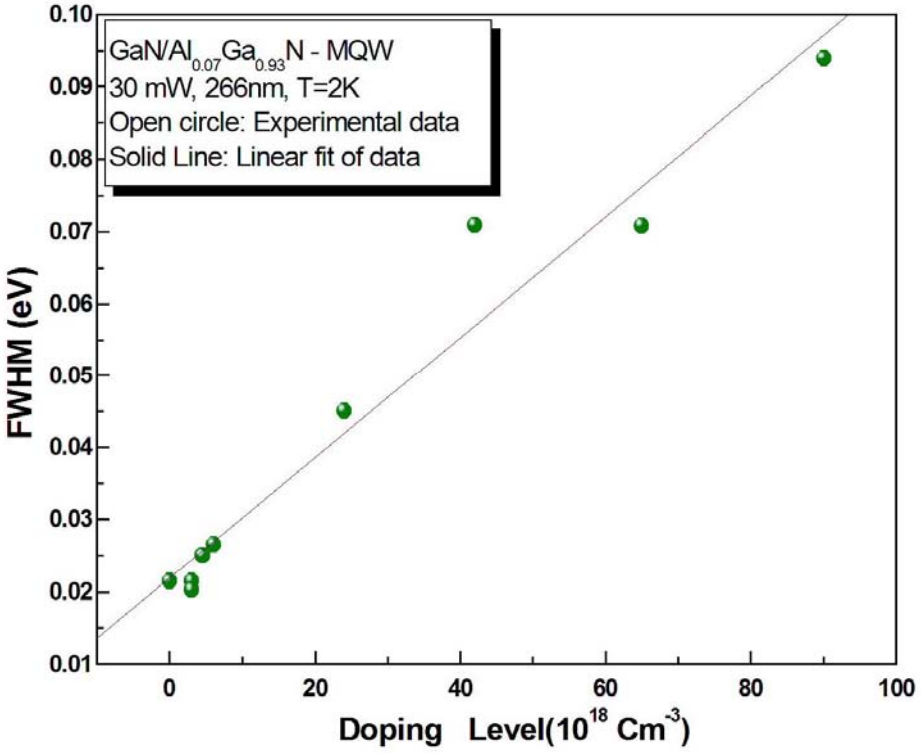

Fig. 3. Si doping dependence of the MQWs photoluminescence peak width (FWHM), measured at T=2 K

The PL temporal response has been evaluated at the PL peak position for undoped and heavy-doped ($9\times10^{19}cm^{-3}$) samples at different excitation powers (Fig. 4). The PL decay time corresponding to the stronger excitation regimes in the undoped sample is faster than the corresponding one for the weaker excitations and the PL transient curves show a non-exponential behavior at different excitation powers (Fig. 4a). The faster part of the PL transient in the undoped sample decreases from 360 ps at the lowest excitation intensity to 260 ps at the highest one. This part of the PL decay curve is attributed to the relaxation time for the excitons to relax to the localization centers. By increasing the excitation intensity the photo-generated carriers/exciton density will increase and partly screen the localization potentials. In the case of the undoped sample, the built-in electric fields are more effectively screened in the stronger excitation regime, photo-generated carriers recombine more quickly, and the slower part of the PL decay time curves becomes faster. However, in the case of the heavily doped sample, free carriers originating from the modulation doping effectively screen the built-in electric fields and the photo-generated carriers recombine at nearly flat band condition. In the case of the heavily doped sample, the extrinsic carrier density is much higher than the photo-generated density [12] and a large number of free electrons originating from the barrier doping are available to recombine with the localized holes, even at a low excitation power. The PL transient curves then show a mono-exponential behavior with a nearly similar decay time at different excitation intensity.

The effect of screening the polarization fields by the extrinsic carriers originating from the Si doping in the barriers can, in principle, be verified by PL measurements. Fig. 5 shows the PL spectra of undoped and heavy samples at different excitation powers at T=2 K. Power dependence PL spectra show that by decreasing excitation intensity from 100% to 0.002%, the PL peak position of the undoped and low-doped samples red shifted about 3.8 meV and 1 meV respectively, but there was no shift for the heavy-doped sample.

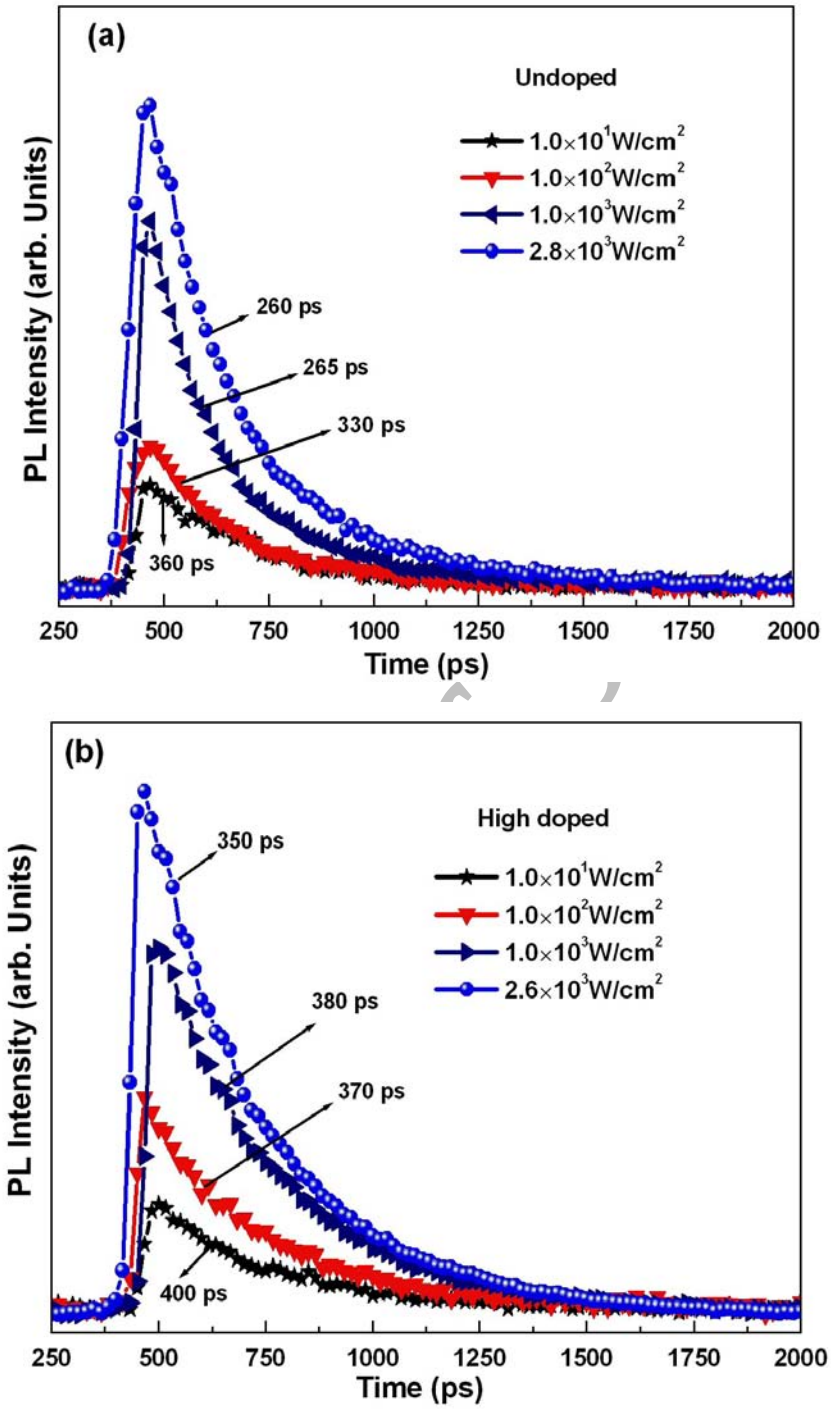

Fig. 4. (a) The time decay corresponding to the stronger excitation regimes in the undoped sample is faster than the corresponding one for weaker excitations. (b)The PL transient curves of the highest doped sample show almost the same decay time at different excitation intensity powers.

This behavior can be explained in terms of the screening of the internal fields by the photo-generated carriers and the relaxation rate to lower energy states in the undoped and low-doped samples. As we reported before, the maximum photo-generated carrier concentration (2×10^{18} cm^{-3}) is less than the extrinsic carrier concentration originating from doping in the barriers in the medium and heavy doped samples [12], which means that the extrinsic carriers dominate in the medium and heavy doped samples, and the photo-generated carriers have, accordingly, a small influence on the electron concentration, consistent with the transient PL data (Fig. 4).

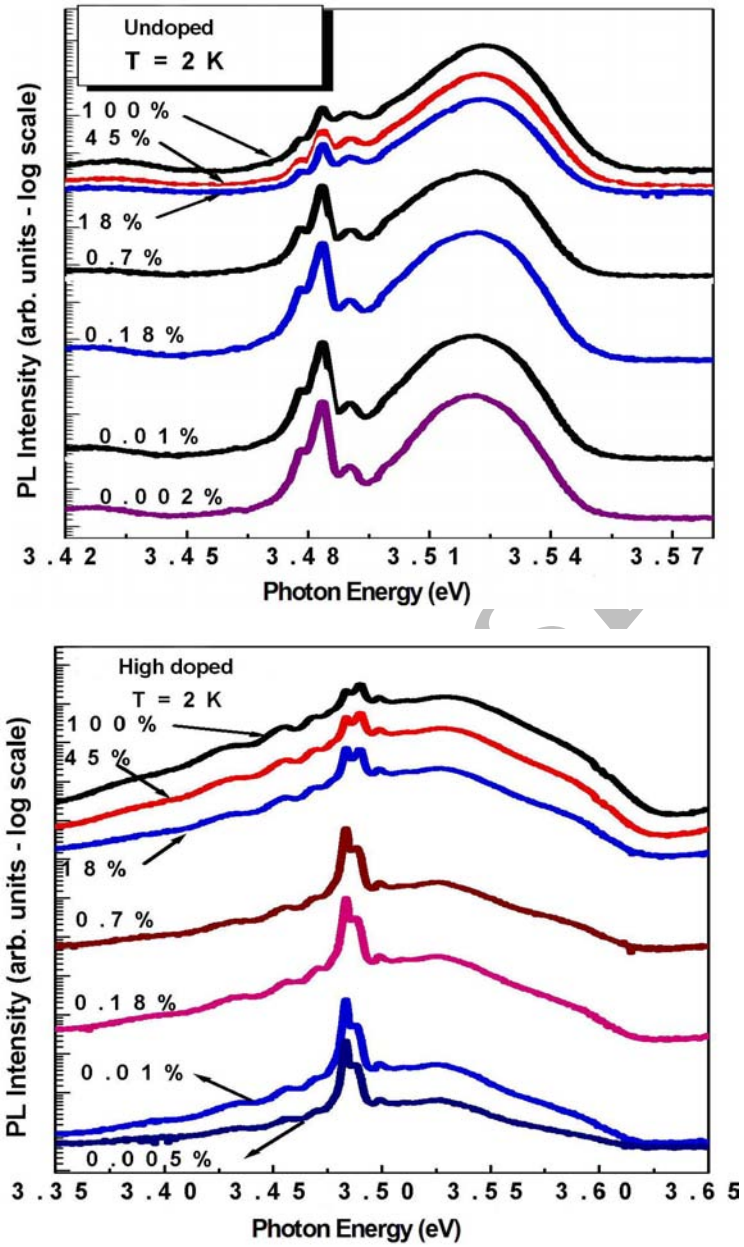

Fig. 5. PL spectra of the undoped sample show a clear red shift by decreasing the excitation power, but there is no shift for the heavy-doped sample

4. SUMMARY AND CONCLUSIONS

The screening effect of the internal polarization fields due to the extrinsic and/or temporal (photo-generated) carriers on the PL peak position, radiative recombination dynamic and optical efficiency of GaN/AlGaN MQW nanostructures have been investigated by means of photoluminescence and time resolved PL spectroscopy. It was found that the screening of the internal polarization fields increases the overlap between the confined electron and hole wave functions, therefore enhancing optical efficiency. Decreasing the excitation power resulted in a red shifted QW emission for the undoped and low-doped samples due to less screening of the internal polarization fields by the temporal carriers, but there is almost no shift for the medium and heavy doped samples due to the highly effective screening of the internal fields by the extrinsic carriers originating from the modulation Si doping.

By varying the excitation power, the faster part of the PL transient in the undoped sample decreases from 360 ps at the lowest excitation intensity to 260 ps at the highest excitation intensity due to partly screening the localization potentials. However in the heavy-doped sample, the decay time is nearly constant at different excitation intensities due to screening of the internal fields by the extrinsic carriers.

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