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Relationship between Sample Morphology and Carrier Diffusion Length in GaN Thin Films

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Scanning and spot-mode cathodoluminescence investigations of homo- and hetero-epitaxial GaN films indicate a surprisingly small influence of their microstructure on overall intensity of a light emission. This we explain by a correlation between structural quality of these films and diffusion length of free carriers and excitons. Diffusion length increases with improving structural quality of the samples, which, in turn, enhances the rate of nonradiative recombination on structural defects, such as dislocations.

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1. Introduction

GaN-based opto-electronic devices, emitting amber-violet light, were commercialised in recent years [1]. These devices contain a very large concentration of edge and threading dislocations, presently in the range of 10^8 cm^{-2} [2]. Light emission should be totally quenched at such high dislocation densities. Despite of this fact, a relatively high efficiency of light emission is observed for diodes with

InGaN quantum wells (QW). This property of GaN-based light emitting devices is still not well understood.

A low efficiency of nonradiative recombination at dislocations was related to strong localization effects in InGaN QWs, i.e., in the active regions of all commercialised GaN-based devices. Strong localization reduces diffusion length of free carriers and excitons, which have a limited chance to approach centres of nonradiative recombination, such as dislocations [3]. Simple calculations indicate that diffusion lengths in the range of 50–100 nm are sufficient to explain a relatively efficient light emission from InGaN-based light emitting devices. Much weaker localization effects, and thus larger diffusion lengths, are expected in GaN epilayers. This, if confirmed, would explain why GaN/AlGaN QW structures are not suitable for opto-electronic devices.

In this work we study strength of localization effects in GaN epilayers grown by metal-organic chemical vapour deposition (MOCVD) on either sapphire or on bulk GaN substrate. We argue that the localization of carriers/excitons in such epilayers is related to a microstructure of GaN samples and that a diffusion length of carriers and excitons correlates with a structural quality of the films.

2. Experimental

A range of GaN films grown by MOCVD on sapphire or on bulk GaN was studied using cathodoluminescence (CL) and scanning electron microscopy (SEM). CL and SEM investigations were performed at either 4 K or at room temperature, using SEM systems equipped with monoCL2 set-up of Oxford Instruments.

3. Experimental results and discussion

We performed scanning CL and SEM investigations to evaluate in-plane fluctuations of the excitonic edge emissions in GaN and to determine their origin. Figure 1 shows the scanning CL images taken for two types of MOCVD-grown GaN samples. These samples, due to modifications in a growth process, were grown with grains of 1 μm size (first type samples) or about 100 nm size (second type) [4]. In-plane fluctuations of the CL intensity were observed in the scanning CL study. Their intensity variations and also their in-plane size varied significantly, depending on a sample studied, as can be seen in Figs. 1a and b.

A comparison of the SEM and CL data, taken from the same regions of the samples, indicates that there is a direct link between a microstructure of GaN epilayers and in-plane fluctuations of the edge CL intensity. The observed fluctuations of the CL intensity not only correlate with the size of grains, but also with their in-plane positions. A brighter CL emission comes from the grain centres and a weaker CL comes from the grain boundaries, where dislocations are present. Apparently, at least a part of carriers excited at grain centres recombines there and

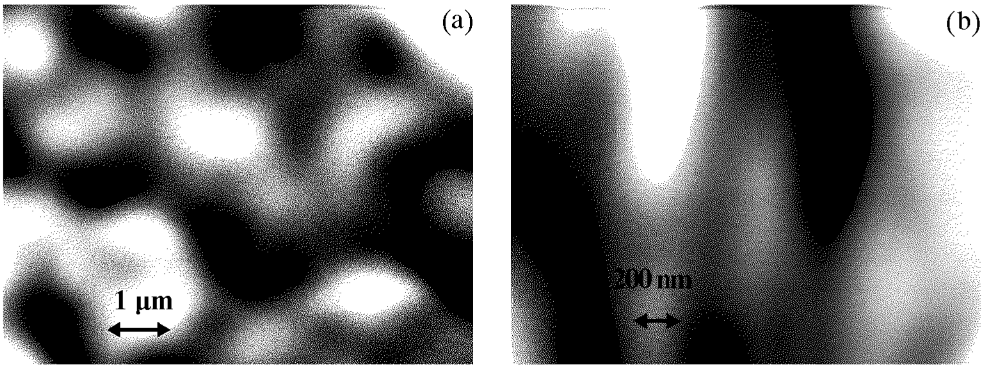

Fig. 1. (a) Scanning CL spectrum of the GaN sample grown by MOCVD on sapphire. (b) Scanning CL spectrum of the MOCVD-grown GaN sample grown on sapphire with grains of a smaller size than those for the sample shown in (a). The spectra were measured with the detection set at the excitonic edge emission.

does not migrate to grain boundaries. The diffusion length of free carriers must thus be smaller than a grain size, as verified separately by spot-mode CL investigations taken for both types of the samples [4]. Otherwise, for larger diffusion lengths in-plane homogeneous and weak CL emission should be observed, with emission intensity limited by efficiency of nonradiative recombination at dislocations present at grain boundaries.

Present studies indicate thus that the localization effects are also present in GaN epilayers. Moreover, in-plane fluctuations of the CL intensity we observed not only at 4 K but also at room temperature. This indicates that the localization effects are relatively strong, but apparently must be weaker than in InGaN films, where they are additionally enhanced by in-plane fluctuations in In fraction [3].

Our conclusion on strong localization effects in GaN epilayers, and on small diffusion lengths of carriers/excitons, is supported by the results of CL investigations taken for the two types of the MOCVD films discussed above. We first compared CL intensities of their edge emissions, which were excited at the same conditions either from relatively large areas of the samples, or from given spots (in the spot-mode CL). Samples with grains of about 100 nm size have a large density of dislocations, in the range of 10^{10} cm^{-2}. Samples with grains of about 1 μm size, have a lower density of dislocations, in the range of 10^{8} cm^{-2} [4]. Consequently, the CL spectra averaged from large areas should vary in intensity, if dislocations act as efficient centres of nonradiative recombination. However, we observed a surprisingly small difference in intensities of the large area integrated CL emissions. Moreover, spot-mode CL spectra indicate that large in-plane inhomogeneity of the CL is not accompanied by a large magnitude of the CL intensity changes. The CL spectra excited at different spots on surfaces of the two types GaN samples are of a similar intensity. In particular, the difference in intensity between CL emissions excited at grain centres, out of grain centres and at grain boundaries is surprisingly small. The latter we already observed in our previous spot-mode CL investigations, taken for GaN epilayers grown by different techniques [5, 6].

The above outlined results indicate that with an improving microstructure of GaN samples only relatively small changes of the CL intensity are observed, despite of a very reduced dislocation density. Our spot-mode CL investigations indicate that this is the direct consequence of, first, strong localization effects, and, second, an increased diffusion length of carriers/excitons in structures with larger diameter grains. Once a diffusion length is increased carriers/excitons can more readily approach dislocations and decay there. The role of dislocations in recombination processes is increased. This effect counterbalances reduced rate of nonradiative recombination at dislocations due to their decreased density.

To verify such possibility, we performed CL investigations of MOCVD-grown GaN homo-epitaxial films. These are high structural quality samples with a fairly low density of dislocations, which is typically below 10^4 cm^{-2}. In the SEM images of GaN/GaN films we observed large areas of atomically flat surfaces. These areas are interrupted by growth steps, which often are of a single monolayer size. Large growth steps (edges), of at least several monolayers size, and micro-defects, such as dislocations or hexagonal pits, are also observed. Their concentration is however relatively low.

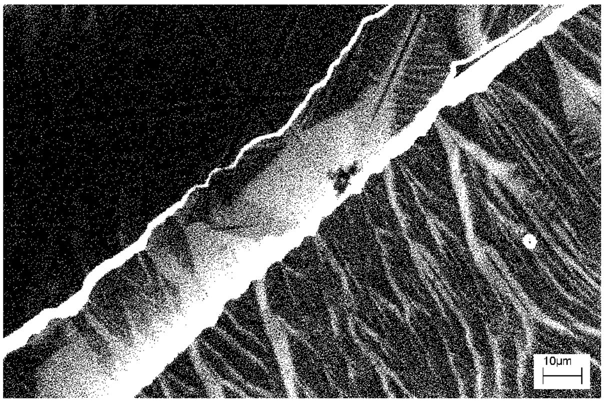

Fig. 2. Scanning CL spectrum of 80×100 μm region of the MOCVD-grown homoepitaxial GaN sample, measured with the detection set at the edge DBE CL emission.

In the scanning CL experiments we set the detection at the dominant donor bound exciton (DBE) CL of GaN/GaN and we followed in-plane variations of the intensity of this emission. The DBE CL is in-plane homogeneous in flat regions, but is significantly enhanced at the growth steps (see Fig. 2). Intensity variations of this CL are very large, as is shown in Fig. 3. CL from flat regions of the sample

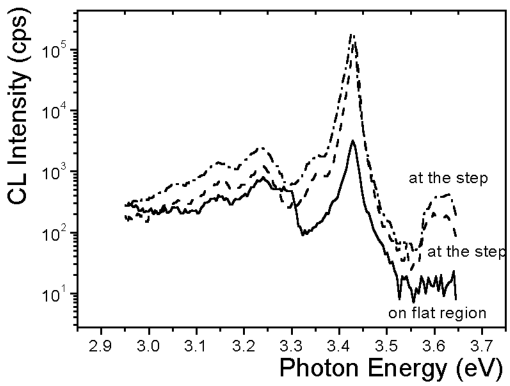

Fig. 3. Spot-mode CL spectra of the MOCVD-grown homoepitaxial GaN sample with the excitation set at three different regions on the sample, i.e., at the growth steps and on the flat region between two growth steps.

is much weaker than the one coming from the growth steps. These data suggest decoration of the growth steps with donor impurities and indicate relatively large diffusion lengths of carriers and excitons, which must be larger than distances between steps, i.e., larger than 2 μm, which is a typical distance between steps. The diffusion length of carriers/excitons is thus considerably larger than that deduced for the best hetero-epitaxial samples, grown by MOCVD on a lattice mismatch sapphire.

There is no accepted explanation of the correlation between a microstructure of the films and a diffusion length of carriers and excitons. The most likely one, as suggested by our results (see also [6]), is decoration of grain boundaries and dislocations with impurities, resulting in their charging and in introduction of in-plane fluctuations of electrostatic potential. Free carriers can then be either repulsed from the regions of grain boundaries and dislocations or be trapped there, forming DBE excitons, which will limit the efficiency of their nonradiative recombination.

4. Conclusions

The present studies demonstrate very complicated in-plane variations of the CL emissions in GaN epilayers. These variations are directly related to details of the microstructure of GaN epilayers. When a structural quality of the sample is improved, e.g., when much flatter surfaces of the films are observed, or when grains of a larger size are found in the SEM study, diffusion length of free carriers and of free excitons increases. In the consequence, despite of a reduced density of dislocations, nonradiative recombination at dislocations may be more efficient. This explains why improvements in a structural quality of GaN samples often result in only a very small increase in the quantum efficiency of light emission.

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