PIEZOELECTRIC PROPERTIES OF GaN SELF-ORGANIZED QUANTUM DOTS

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ABSTRACT

It is demonstrated that GaN quantum dots with the wurtzite structure grown by molecular beam epitaxy on AlN exhibit optical properties which, depending on the size of the dots, may be dominated by piezoelectric effects. In "large" quantum dots with an average height and diameter of 4.1 and 17 nm, respectively, the photoluminescence peak is centered at 2.95 eV, nearly 0.5 eV *below* the bulk GaN bandgap, which is assigned to a piezoelectric field of 5.5 MV/cm present in the dots. The decay time of the photoluminescence was also measured. A comparison is carried out with theoretical calculation of the radiative lifetime.

INTRODUCTION

Despite the successful realization of blue LDs by Nakamura et al., serious problems are still to be overcome, related to the lack of adapted substrate. In particular, a reduction in the number of cristallographic defects is highly desirable in order to improve the lifetime and to reduce the threshold current of the LDs. This is partly achieved through the recent development of lateral overgrowth for GaN which leads to the reduction of the dislocation density by several orders of magnitude [1]. Alternately, the realization of devices with quantum dots (QDs) in the active layer appears promising, based on the theoretical prediction of a low threshold current and of a weak temperature dependence of the threshold current [2]. Furthermore, due to the reduced size of the QDs, they are expected to be virtually perfect, with most cristallographic defects out of the dots, which should result in a decrease in non radiative recombination. It is the aim of this article to adress this point in the case of self-assembled GaN QDs, with a particular attention paid to the peculiar properties resulting from the presence of a huge piezoelectric field which is one of the most fascinating aspect of nitrides [3,4].

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EXPERIMENT

The samples were grown by molecular beam epitaxy (MBE) in a commercial MECA 2000 machine. The substrate was (0001) sapphire. After the nitridation step of the sapphire, a low temperature AlN layer, about 15 nm thick, was deposited followed by the growth of a 1.5 µm thick AlN buffer layer. The details of the growth procedure have been published elsewhere [5]. The GaN QDs were grown by depositing the equivalent of 3 monolayers of GaN on AlN at 700°C (Ref. 6, 7). Next, they were covered by AlN in order to smooth the surface again and the operation was repeated several times to obtain a superlattice of GaN QDs layers. The size of the dots was varied, depending on whether they were « ripened » under vacuum or not before further covering with AlN. The « large » dots were ripened under vacuum during about one minute before further capping with AlN. They were typically 4.1 ± 0. 4 nm high (17 nm diameter). The « small » unripened dots were capped with AlN with no growth interruption. They were 2.3 ± 0.2 nm high (8 nm diameter). These figures were extracted from the analysis of high resolution electron microscopy (HREM) pictures. As shown in Fig.1, analysis of the HREM pictures taken along two directions rotated by 30° allowed to conclude that the GaN quantum dots are coherently matched to the (0001) plane of AlN, then experiencing a strong biaxial stress. As a consequence of the biaxial stress and of the cristallographic symmetry, a strong piezoelectric field is expected to be present in the dots [3,4].

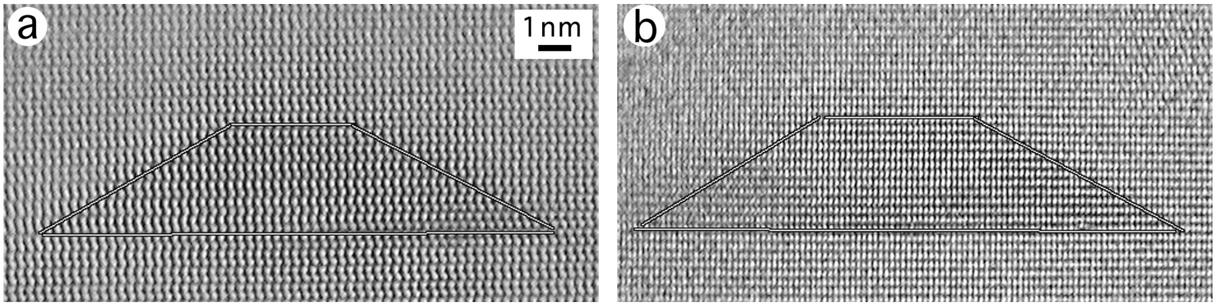

Fig.1 HREM images of the same GaN island taken along (a) the 1/3[2,-1,-1,0] and (b) the [10$\underline{1}$0] directions, differing from 30°

This hypothesis was confirmed by measuring the photoluminescence of the large dots. The result, plotted in figure 2, reveals that the PL peak of the large dots is observed at 2.95 eV, i.e. 0.5 eV *below* the bulk GaN energy gap.

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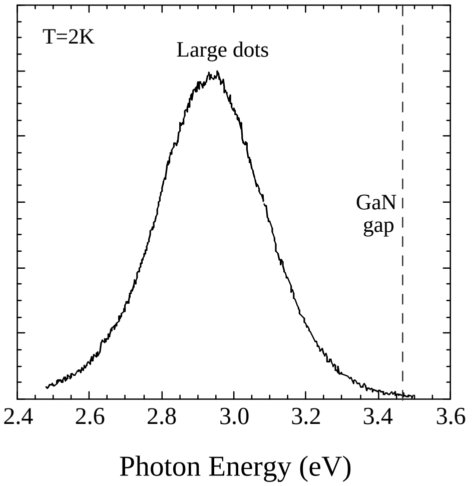

Figure 2: Photoluminescence of « large » GaN QDs

Actually, it has been shown elsewhere that the photoluminescence emission energy of GaN QDs was dramatically dependent on the QD size [8], with the « small » QDs exhibiting a PL peak centered at 3.75 eV, nearly 0.3 eV higher than the GaN bandgap. Consistent with the HREM results, this striking QD size effect was attributed to the presence of a giant piezoelectric field in the QDs along the c-axis [8]. This hypothesis was supported by PL experiments as a function of excitation power which revealed an increasing blue shift for increasing power, as an evidence of the screening of the piezoelectric field for an increased density of photocreated carriers. Finally, theoretical calculations were performed, allowing to conclude that the GaN QDs matched to the AlN matrix experience a piezoelectric field as high as 5.5 MV/cm, consistent with the experimental data available on the piezoelectric field values in GaN and related alloys [9-11].

The strong localization effects resulting from the reduced size of the GaN QDs was further confirmed by measuring the photoluminescence intensity as a function of temperature. As shown in figure 3 , it was found that, by contrast to quantum wells or Mg-doped bulk GaN, the PL intensity of large dots was practically unaffected by temperature and was even increasing slightly, as an evidence of strong localization effects.

The hypothesis of a strong piezoelectric field to account for the energy position of the PL peak of large dots was supported by optical power dependent PL spectra, the center of gravity of the PL peak being significantly blueshifted for increased power density [8]. This behaviour is typical of piezoelectric nanostructures and is due to partial screening of the piezoelectric field by the photoexcited electron-hole (e-h) pairs. In the case of large dots, a 70 meV blueshift was observed as the power density varies from 60 to 450 W/cm^2.

Further evidence of the strong piezoelectric field present in the GaN QDs is provided by PL decay measurements. The time-resolved PL experiments presented here were performed with a standard streak camera setup at T=10K on

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a QD sample with 72 layers of small QDs. In this sample, a relatively broad QD size distribution is available as manifested by the 500meV-broad PL spectrum centered at 3.6eV. The excitation was provided by 265nm pulses of 1.5ps duration at 76MHz repetition rate, resulting from frequency-tripling a 795nm line of a picosecond Ti-Sapphire laser. Typical average power densities used in these measurements were 50W/cm^2 and the corresponding sheet exciton density created per pulse is estimated to be about 10^{11}cm^{-2}, which is several times smaller than the QD density. In Fig. 4, we plot by solid squares the measured PL decay times, τ_D, as a function of photon energy. On the high energy side, we observe a continuous increase of the PL decay time with decreasing photon energy. This is consistent with the picture of piezoelectric QDs, since the piezoelectric field in the larger dots is more efficient in separating the electron and hole wavefunctions and hence increases their radiative recombination time. However, as the photon energy is further decreased we observe that the PL decay times reaches a plateau not exceeding the 3.6ns. We attribute this plateau to the presence of nonradiative channels and set τ_{NR}=3.6ns. If we assume that τ_{NR} is independent of energy, then, based on the equation $1/\tau_D = 1/\tau_{NR}+1/\tau_R$, we can tentatively estimate the radiative recombination time τ_R as a function of energy. The result is plotted as open-cross squares in Fig. 4. The most striking feature is the dramatic increase in the decay time which is observed for decreasing energy. In particular, when extrapolating down to 3 eV, corresponding to « large » dots emitting in the blue range a very large decay time is expected.

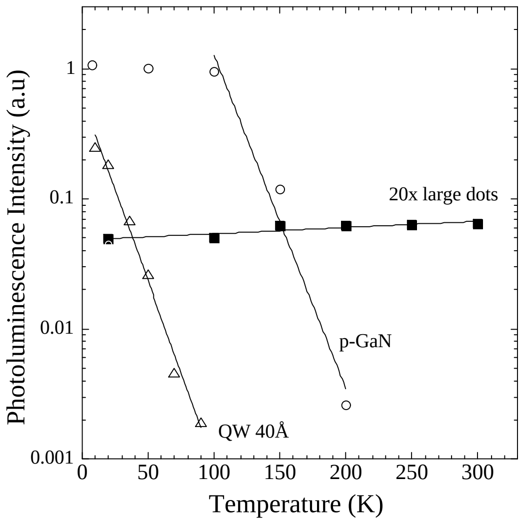

Figure 3: Temperature dependence of the photoluminescence for a) bulk GaN, b) quantum wells and c) large quantum dots

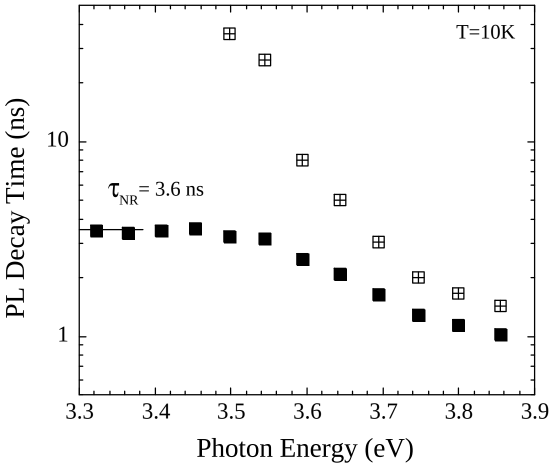

Figure 4: Photoluminescence decay time as a function of energy. Crossed squares are an estimation of τ_R deduced from the experimental data (full squares) by substracting the influence of τ_{NR}

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DISCUSSION AND CONCLUSION

In an attempt to understand the above results, the influence of the piezoelectric field on the oscillator strength was investigated. We first calculated the exciton radiative lifetime τ in the bulk semiconductor using the usual approach [12,13]. It was found that in GaN τ was of the order of 25 ns. Then, in order to determine the radiative lifetime in a QD, we use the model of Ref.14, which allows to calculate the oscillator strength, and therefore the radiative lifetime, in a cubic box provided that the bulk radiative lifetime is known. For that purpose, we have defined an equivalent cubic box which has the same volume as the pyramidal QD of heigth h. The radiative lifetime τ_h without piezoelectric field was then obtained. In a further step, the piezoelectric field F was taken into account via a two-dimensional quantum well, as reported in Ref. 8. Note that this approach is meaningful as the exciton Bohr radius parallel to the quantum well is smaller than the side of the equivalent cubic box (this would not apply to very small pyramids). We then obtain the relations linking $\tau_{h,50}$, τ_h and τ, where $\tau_{h,50}$ is the radiative lifetime in a QD taking into account a piezoelectic field F = 5 MV/cm. The last step of the calculation is to link the size of the pyramid to the transition energy [8]. The final result is shown in Fig.5. The key information is that the influence of the piezoelectric field is not very important (an increase in the lifetime by a factor of about 2 at 4 eV, h = 20 Å) for a small box (large energy). By contrast, this influence is very large in a large box (an increase in the lifetime by a factor of about 200 at 3eV, h = 40Å).

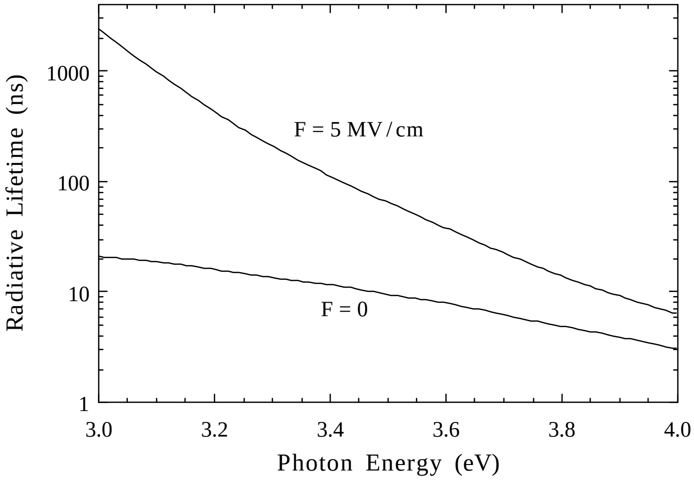

Figure 5: Theoretical calculation of the radiative lifetime in GaN QDs as a function of the photon energy, with and without piezoelectric field

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In conclusion it has been experimentally and theoretically demonstrated that the optical properties of GaN quantum dots with wurtzite structure grown on AlN by MBE of GaN result from the competition between confinement and piezoelectric effects. The piezoelectric effects were found to be dominant for QD height larger than 3nm. This is a very unique situation resulting from the combined effect of the QD crystallographic symmetry (wurtzite with a [0001] growth direction) and of the huge piezoelectric coefficients specific to the nitride family. The photoluminescence decay time was theoretically calculated and found to be strongly dependent on the piezoelectric field, consistent with experimental data. Finally, these results open the way to the achievement of high efficiency blue light emitting devices with no In.

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