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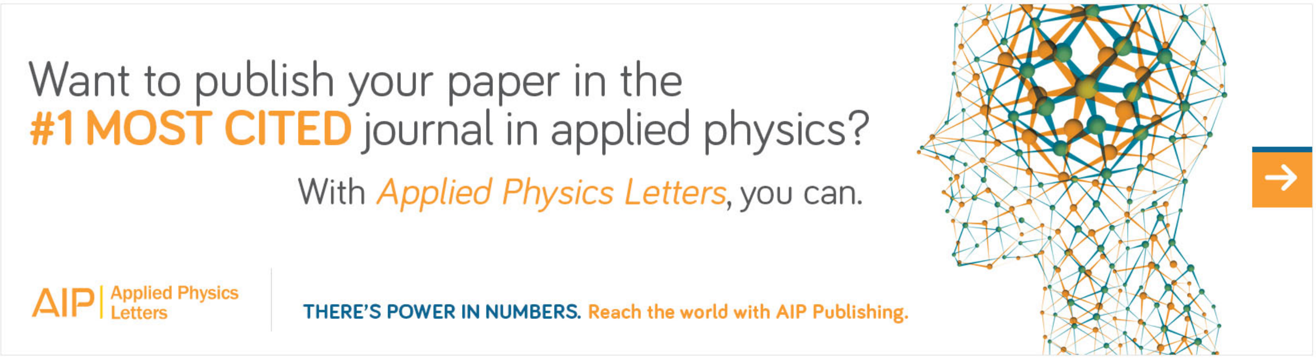

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Strong electric field and nonuniformity effects in GaN/AlN quantum dots revealed by high pressure studies

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The photoluminescence (PL) from GaN quantum dots (QDs) embedded in AlN has been investigated under hydrostatic pressure. The measured pressure coefficient of emitted light energy $[dE_E/dP]$ shows a negative value, in contrast with the positive pressure coefficient of the GaN band gap. We also observed that increasing pressure leads to a significant decrease of the light emission intensity and an asymmetric broadening of the PL band. All these effects are related to the pressure-induced increase of the built-in electric field. A comparison is made between experimental results and the proposed theoretical model which describes the pressure behavior of nitride QDs. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2219381]

A general peculiarity of the nitride heterostructures (with wurtzite symmetry) is the existence of a large built-in electric field caused by piezoelectric and spontaneous polarizations. Self-organized QDs in wurtzite nitrides usually have an approximate shape of truncated pyramids with a hexagonal base located in the wetting layer.[1] The main open questions concerning the GaN/AlN quantum dot system are related to (i) the built-in strain and (ii) the internal electric field. Nonuniformity in the size and/or shape of QDs is an issue as well. There are large differences in parameter values characterizing magnitudes of (i) and (ii). Experimentally deduced values of the electric fields for GaN/AlN heterostructures [quantum wells (QWs) and QDs] vary ranging from 4 MV/cm (Ref. 2) up to 9 MV/cm.[3] Theoretical calculations presented by Andreev and O'Reilly[4] predicted that the electric field along the axis of the QD exceeds 6 MV/cm at the bottom of the dot and decreases to a value of around 4 MV/cm when moving from the pyramid base to its top.

It has been demonstrated that in hexagonal InGaN/GaN and GaN/AlGaN QW structures changes in the emitted light energy with pressure are determined mainly by a contribution originating from an increase in piezoelectric polarization.[5] The latter effect leads to a decrease of dE_E/dP and competes with the opposite tendency resulting from the pressure-induced increase in emission energy originating from conduction-to-valence band transitions (which is related to the semiconductor band gap increase with pressure).

The purpose of this work was to use pressure studies of photoluminescence (PL) in GaN/AlN QDs to obtain information on not well understood effects related to both built-in strain and electric field. Theoretical model enabling calculations of dE_E/dP in nitride QDs was developed. Obtaining a satisfactory agreement between experimental and calculated magnitudes of dE_E/dP supports the predicted properties of strain and electric field as well as their distributions in GaN/AlN QDs.

Two samples of GaN/AlN QDs have been studied in the present work. The growth of sample A was performed by using ammonia-based molecular beam epitaxy (MBE) on a sapphire substrate with a GaN buffer (3 μm) and an AlN layer of 0.3 μm thickness to relax the strain. Then, ten periods of GaN QDs separated by 9 nm AlN barrier material were deposited. The dots have a truncated hexagonal pyramidal shape of about 4.3 nm height. The pyramids were formed on GaN wetting layers, approximately 2 ML (monolayers) thick. The dots are nominally undoped. Their low-temperature PL consists of a broad line centered at about 2.6 eV with a half-width of approximately 0.3 eV. Sample B has been deposited by using plasma assisted MBE on a 6*H*-SiC substrate with a 10 nm AlN buffer layer. The period of GaN quantum dots was repeated 100 times. From previous studies by transmission electron microscopy (TEM) and atomic force microscopy (AFM), GaN dots are found to be vertically correlated, with a typical height of about 4 nm.[6] Each layer of GaN QDs was separated by 8 nm of AlN. A broad PL line centered at about 2.9 eV (half-width of 0.35 eV) characterizes sample B. The broad photoluminescence lines in both samples likely correspond to ensembles of QDs with varying heights.

The high pressure experiments were performed in a low-temperature diamond anvil cell filled with solid argon as pressure transmitting medium. Samples were polished down to a thickness of about 30 μm. The measurements were performed at 80 K, and the photoluminescence from a small ruby crystal placed next to the samples was used to monitor the pressure. PL of GaN/AlN QDs was excited by the 325 nm line of a He–Cd laser with a power density of about 5 W/cm^2. The emission from the samples, collected in backscattering geometry and dispersed by a SPEX500M spec-

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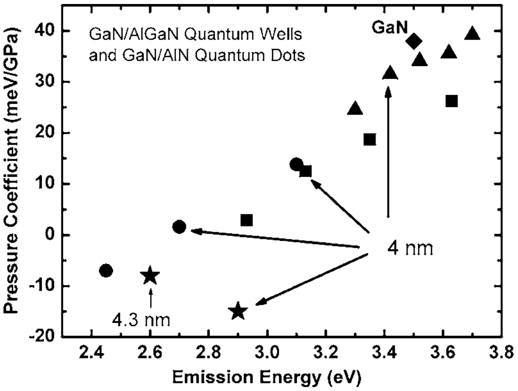

FIG. 1. Dependence of pressure coefficient of emitted light energy dE_E/dP on emission energy E_E measured in GaN/Al$_x$Ga$_{1-x}$N quantum well samples and GaN/AlN quantum dot samples (stars) this work. Solid diamond corresponds to GaN bulk crystal; solid triangles to GaN/Al$_x$Ga$_{1-x}$N: $x=0.17$, Ref. 7; solid squares: $x=0.5$, Ref. 8; and solid circles, $x=0.8$, Ref. 8.

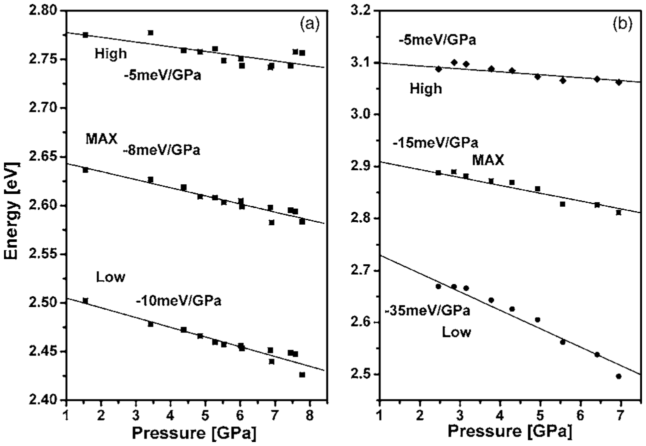

FIG. 2. Experimental results for GaN/AlN quantum dots A (a) and B (b). The photoluminescence peak position shift with pressure for maximum and half maximum of the intensity.

trometer, was detected by a GaAs photomultiplier.

Our measurements at ambient pressure show that the energies of the PL emission from the ensembles of GaN QDs of samples A and B are smaller than the energy gap of GaN. It was found that the energy of emitted light from wurtzite GaN QDs was dramatically reduced when the QD height was larger than about 3 nm.[1] Indeed, the blueshift of the PL band due to quantum confinement is clearly overcompensated. This is evidence of the existence of strong built-in electric fields that affect the emission energy via the quantum-confined stark effect (QCSE).

Increasing pressure causes a shift of PL lines toward lower energies, implying a negative pressure coefficients dE_E/dP in the studied QDs. This is unusual, especially when compared with the positive value of the pressure-induced increase of the GaN band gap (40 meV/GPa). The latter value characterizes interband transitions responsible for light emission in bulk GaN. We also observed that increasing pressure leads to a significant decrease of light emission intensity and to an asymmetric broadening of the PL band.

Figure 1 shows the relation between dE_E/dp and PL peak position (i.e., the energy at the maximum of intensity) of both studied QD samples (stars). A large, negative magnitude of dE_E/dp is clearly seen. This behavior is compared with results reported earlier for GaN/Al$_x$Ga$_{1-x}$N QW samples characterized by different Al contents.[7,8] Each sample was grown as a set of GaN QWs with various widths d. Lower values of E_E and dE_E/dp correspond to higher Al contents in the matrix material, mainly due to QCSE. Moreover, in each set of samples, lower E_E characterizes QWs with larger d. We marked QW samples with $d \approx 4$ nm to compare them with the QD samples studied here. One can also see a strong decrease of dE_E/dp with decreasing emission energy E_E. We can associate the correlation between E_E and E_E/dp with both stronger built-in electric field in samples with higher Al content and more pronounced influence of the QCSE in samples with higher QW width or QD height. One can also see that for the studied QD samples, sample A follows the considered trend well, whereas sample B shows less good agreement.

Dependences of PL band shift on pressure in our QD samples are presented in Figs. 2(a) (for sample A) and 2(b) (for sample B). The experimental points labeled "max" represent the energy corresponding to the maxima of the PL intensity. We also established pressure changes of the two energy values characterizing half maximum of intensity on the high- and low-energy shoulders of the PL band. They are labeled "high" and "low." We note significant differences in the pressure evolution of these energies, more pronounced for sample B. In this case, the extremely low negative pressure coefficient characterizes the lower energy shoulder of the spectrum. It is worth to point out that within the experimental error, no asymmetry of PL bands at high pressures has been observed in GaN/AlGaN QW samples studied by Vaschenko *et al.*[8] and InGaN/GaN QWs studied by us.[9] For a first tentative interpretation of the asymmetric broadening of PL band with pressure, one can use the following arguments. The high energy shoulder of the QD spectra moves with a less negative pressure coefficient and likely consists of contributions from the dots with smaller height (less pronounced influence of QCSE). Simultaneously, the lower energy wing related to the higher dots exhibits more pronounced negative pressure coefficient (stronger influence of QCSE).

Another possible cause of asymmetric broadening of PL peaks with pressure in the studied QD samples can be related to a difference in the structure of our samples A and B. The first one consists of ten stacks of QDs layers whereas the second one is formed by 100 stacks of QDs. In the paper by Gogneau *et al.*[10] it is shown that with increasing number of stacks in the GaN/AlN QDs lateral dimensions of GaN dots increase, keeping roughly a similar height the QDs. This effect can be additionally responsible for a larger PL band width in our sample B as well as for more dispersed values of pressure coefficients among individual QDs. We will demonstrate below by means of theoretical calculations how the latter effect influences pressure coefficient.

We performed calculations of the pressure coefficients of the light emission from GaN/AlN QDs using the model based on second-order elastic theory and the $k \cdot p$ method (with 8x8 Hamiltonian).[4,11] Strain-dependent piezoelectric coefficients of GaN and AlN are used.[12] Spontaneous polarization is also taken into account in the solution of the Poisson equation.[13] External hydrostatic pressure is introduced by applying appropriate nodal forces to finite element mesh. For each QD, the strain field, the electrostatic potential and the energy levels have been calculated for several values of external hydrostatic pressure in the range from 0 to 5 GPa.

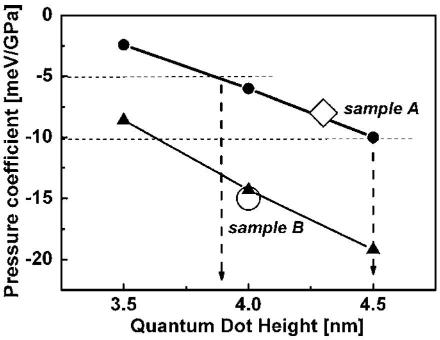

FIG. 3. Comparison of pressure coefficients of the emission energies for two measured samples (open symbols) with results of theoretical calculation (solid symbols) obtained for individual dots of different geometries. Solid circles and triangles correspond to the values of the pressure coefficients obtained for the quantum dots with the base diameter equal to 20 and 26.5 nm, respectively. Dot- and dashed-lines illustrate a way of estimating an average size of QDs corresponding to energy values "high" and "low" at Fig. 2(a) (sample A).

The obtained fundamental interband transition energies have shown almost linear dependences on pressure, which enables us to determine the values of the pressure coefficients.

We performed calculations for two sets of GaN/AlN QDs. The first set contains dots with the base diameter equal to 20 nm, whereas in the second set, the base diameter of the dots is 26.5 nm (this mimics an increase in the lateral dimensions of QDs with increasing number of QD stacks in the sample B, Ref. 10). In each set, the height of the dots changes from 3.5 to 4.5 nm. The angle between the base of the dot and the side walls is always equal to 30°. Calculated pressure coefficients are shown in Fig. 3. Solid circles and triangles correspond to results obtained for the first and the second set of the dots, respectively. One can see that the magnitude of the dE_E/dp decreases with (i) increasing the height of the dots and (ii) increasing the base diameter of the dot. We have found that both effects are related to the increase with pressure of the average vertical component of the built-in electric field, $d|E_{z,\mathrm{av}}|/dP$, in the QD region. The values of $d|E_{z,\mathrm{av}}|/dP$ are about 0.12 and 0.14 MV/(cm GPa) for dots from the first and the second sets, respectively. The magnitude of dE_E/dp depends, via the QCSE, on $d|E_{z,\mathrm{av}}|/dp$. This dependence leads to a linear decrease of dE_E/dp with the height of the dots, as shown in Fig. 3. The decrease of dE_E/dp is stronger for larger values of $d|E_{z,\mathrm{av}}|/dP$, i.e., for the dots from the second set.

Experimentally determined pressure coefficients are equal to −8 and −15 meV/GPa for samples A and B, respectively (at the maximum of the PL intensity). Very good agreement with the calculated value of dE_E/dp for the first set of QDs with a diameter of 20 nm and experimental data for sample A is seen (average QD height is 4.3 nm). The same observation concerns sample B and calculations related to the second set of QDs (with diameter 26.5 nm).

Now we attempt to use the calculated dependence of PL energy pressure coefficients versus QD height for the first set of dots, (i.e., the line determined by solid circles) to associate experimentally observed values corresponding to high ($dE_E/\mathrm{dP} \approx -5$ meV/GPa) and low (≈ -10 meV/GPa) energies of the spectra with QDs of approximately 3.9 and 4.5 nm heights. The described findings are obtained by a simple procedure illustrated by dot/dashed lines seen at Fig. 3. The applied procedure is less justified in case of sample B (with average QD size of 4 nm) where the value of dE_E/dP (Max) is −15 meV/GPa (−10 and −35 meV/GPa for low and high points of the spectrum) and differs significantly from the calculated results. This suggests that in case of sample B much higher built-in electric fields and/or less uniform distribution of QD heights and lateral dimensions is present. Our calculations confirmed the earlier findings of PL pressure studies in wurtzite nitride quantum wells, i.e., that the main reason for strongly reduced values of the pressure coefficients is the strong increase of the built-in electric field with pressure. Moreover, the related QCSE causes a spatial separation of electron and hole wave functions and then a reduction of the emitted light intensity. A clear decrease in the emission line intensity with increasing pressure was observed in our experiments for both QD samples.

In summary, we presented results of pressure studies of photoluminescence from GaN/AlN QDs. We observed several effects related to the pressure-induced increase of the large built-in electric field in the studied structures. The measured pressure coefficient of emitted light energy shows a negative value, which is in contrast with the positive pressure coefficient of the GaN band gap energy. We also observed that rising pressure leads to a significant decrease of light emission intensity and to an asymmetric broadening of PL band. We showed that the pressure induced asymmetry of the PL peak can be associated with the distribution in height and lateral dimensions of GaN dots embedded in AlN matrix.

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