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2002 J. Phys.: Condens. Matter 14 13329

(http://iopscience.iop.org/0953-8984/14/48/385)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **14** (2002) 13329–13336 PII: S0953-8984(02)54176-X

Optical and structural characterization of GaN/AlN quantum dots grown on Si(111)

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Received 27 September 2002
Published 22 November 2002
Online at stacks.iop.org/JPhysCM/14/13329

Abstract

GaN/AlN-based heterostructures made from stacked GaN quantum dots (QDs) have been studied by means of the cathodoluminescence (CL), photoluminescence (PL), near-field scanning optical microscopy (NSOM) and micro-Raman techniques. The influence of the number of stacked layers (2–85) and of the different electron beam injection conditions on the main optical emissions was studied by means of CL, revealing transitions from 2.5 and 4.4 eV. Power-dependent cross-sectional CL studies revealed a large (87–180 meV) blue-shift only for the optical bands located in the 2.5 and 3.1 eV spectral range. This observation enabled us to assign a zero-dimensional character to those bands. The results were confirmed by PL and NSOM studies. Different values of the blue-shift were found for specimens with different numbers of stacked layers. This suggested the presence of different residual strains inside the structures, as confirmed by micro-Raman studies. An inhomogeneous distribution of the QD emissions was also observed both in the plane and along the growth direction.

1. Introduction

GaN-based self-assembled quantum dots (QDs) are of great interest for a wide field of industrial applications from advanced emitting devices [1, 2] (high emission efficiency, low threshold currents etc) to quantum computing (small phonon coupling among the dots).

0953-8984/02/4813329+08$30.00

Table 1. Structure of the specimens investigated.

Sample	First AlN buffer layer (nm)	GaN buffer layer (nm)	Second AlN buffer layer (nm)	GaN QD layer (nm)	AlN barrier layers (nm)	Number of stacked layers
S2	30	300	300	2.6/0.8	45	2
S4	34	1100	330	2.8/2.3/1.6/1.0	9/9/11	4
S40	30	430	700	1.6	6.7	40
S85	30	430	700	1.6	6.7	85

One of the problems affecting the growth of III nitrides is the choice of suitable low-cost (e.g. Si) substrates also able to minimize the lattice mismatch. This is important for exploiting new structures in industrial applications. In particular, in the case of Si(111), despite the high density of defects, self-assembled Stranski–Krastanov MBE growth of wurtzite-type (WZ) GaN/AlN QDs ($\sim$2.5% lattice mismatch) has been demonstrated to give an intense visible luminescence at RT [3].

Defect formation, QD stacking misalignments, in-plane inhomogeneities (QDs size and distribution) etc negatively affect the optical properties of the heterostructures. Therefore, the use of complementary optical and structural techniques is necessary to evaluate the correlation between luminescence, plastic relaxation and internal fields.

Three major contributions affect the optical transitions of WZ GaN QDs based heterostructures:

(i) confinement effects,
(ii) the presence of a large strain (compressive for GaN on AlN) both leading to a blue-shift of the QD emission with respect to the GaN bulk emission and
(iii) the presence of huge internal electric fields (spontaneous and piezoelectric [4]) inducing a red-shift of the emission bands.

The relative weights of the various contributions are mainly influenced by the size of the islands, as revealed by combined photoluminescence (PL) and TEM investigations (see for instance [5, 6]) and by theoretical studies (see for instance [7]).

This work presents a preliminary study of different GaN/AlN stacked systems by micro-Raman, cathodoluminescence (CL), PL and near-field scanning optical microscopy (NSOM) techniques, carried out to examine the effect of the lateral and vertical homogeneity of the QDs, the relaxation processes inducing crystal defects and the effect of the internal fields. The results were thus analysed in terms of the different structural parameters, taking into account the nature of the various optical probes. Preliminary TEM studies confirmed the reported data.

2. Experimental details

The samples were grown by MBE on Si(111) after AlN and GaN buffer layer growth to produce a complete relaxation of the last layer, prior to the growth of the QDs, as described elsewhere [3]. Using the SK method, samples with 2, 4, 40 and 85 stacked layers were produced. The nominal thicknesses of the layers for the four samples are shown in table 1. A final AlN capping layer of 45 nm was deposited on top (omitted in table 1).

CL analyses, both in plane view and cross-section, were performed using a Gatan MonoCL2 system installed on a Cambridge 360 Stereoscan SEM with an alkali halide PMT detector. The electron beam energies (E_b) varied between 0, 5 and 20 keV.

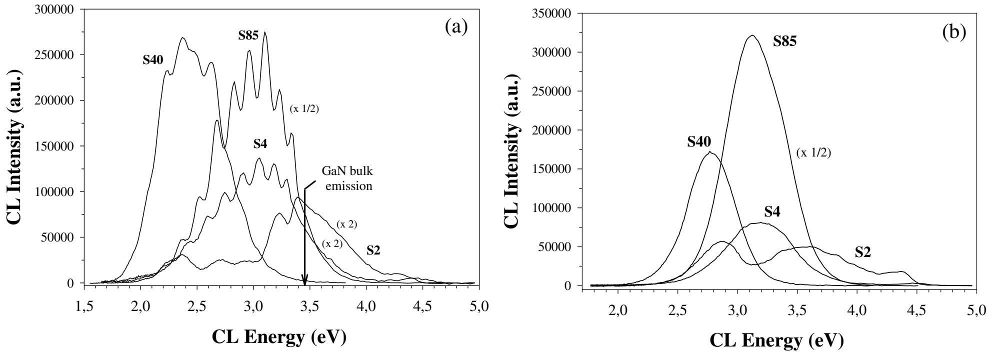

Figure 1. RT CL spectra of the samples studied at $E_b = 20$ keV, $I_b = 5$ nA: (a) plan-view and (b) cross-sectional geometry.

Table 2. Peak energy positions of the different bands fitting the CL spectra of figure 1(b).

Sample	Peak energy positions (eV)					
S2		$\sim$2.8		$\sim$3.4	$\sim$3.7	$\sim$4.3
S4		$\sim$2.9	$\sim$3.1	$\sim$3.4	$\sim$3.7	$\sim$4.3
S40	$\sim$2.5	$\sim$2.7	$\sim$2.9			
S85		$\sim$2.7	$\sim$3.1			

μ-Raman spectra were excited with a 632.8 nm He–Ne laser at RT and collected with a Raman confocal microspectrometer (LabRam ISA) through a microscope optical apparatus.

The NSOM spectra were taken at room temperature with near-field and far-field detection. The samples were excited through the NSOM fibre by a frequency-doubled, mode-locked Ti:sapphire laser in the range 2.9–3.5 eV. Standard PL measurements were also made using a frequency-doubled ps dye-laser, synchronously pumped by the second harmonic of a mode-locked Nd:YAG laser.

TEM studies were performed using a JEOL 2000-FX microscope working at 200 keV.

3. Results and discussion

The CL, PL and NSOM results were cross-checked to study the optical emissions of the samples, taking advantage of the superior spectral and lateral resolution of the PL and NSOM techniques and of the depth-resolved and power-dependent capability of the CL technique.

Figure 1 shows the CL spectra recorded at RT, both in the plan-view and the cross-sectional geometry, for the four samples under the same experimental conditions. The CL transitions in the plan-view geometry (figure 1(a)) are strongly modulated by Fabry–Perot interference effects due to the light reflecting from the Si substrate. This negatively affects the interpretation of the results even after complex deconvolution procedures. For this reason, cross-sectional studies have been carried out (figure 1(b)), allowing a more reliable band assignation.

The electron beam energy used to obtain the CL spectra of figure 1 was chosen in order to allow analysis of the complete heterostructure for each sample. Thus, contributions from the different layers are expected to be present. The results of deconvolution of the CL spectra are reported in table 2, which shows the number of bands used for the fitting procedures and their energy positions.

From a careful look of figure 1(b) it emerges that the bands at $\sim$3.4, 3.7 and 4.3 eV are visible only in some of the spectra and that their intensities decrease as the number of

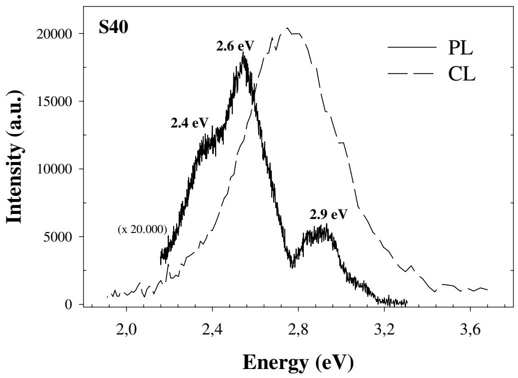

Figure 2. Comparison between RT PL and CL cross-sectional spectra for sample S40. The CL spectrum was recorded at $E_b = 20$ keV, $I_b = 5$ nA.

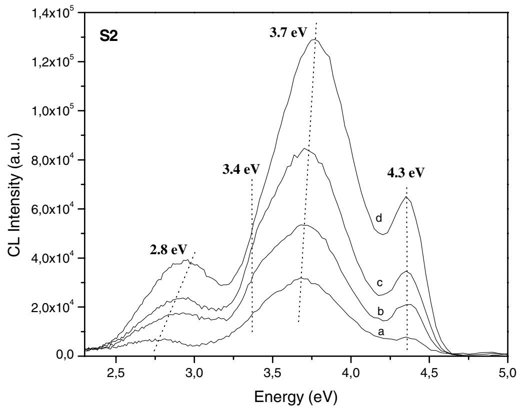

Figure 3. RT cross-sectional CL spectra recorded for sample S2 under different power injection conditions at 20 keV and (a) 15 nA, (b) 50 nA, (c) 150 nA, (d) 300 nA.

the stacked layers increases. This suggests that those emissions come from GaN and AlN buffer and capping layers. In fact, according to the structures of the samples (see table 1), higher integrated intensities of the aforesaid emissions are expected for the samples with lower numbers of stacked layers. In contrast, figure 1(b) also reveals that the bands in the energy range from 2.5 to 3.1 eV are present for all the samples. According to literature data [8], these transitions should be due to QD emissions. The presence of different subbands (table 2) for the same sample suggests an inhomogeneous QD distribution, as far as the average dimensions and vertical coupling are concerned.

Figure 2 shows, as an example, the comparison between the cross-sectional PL and CL for sample S40. The PL emissions are in substantial agreement with the band assignation previously established by CL investigations, as seen in table 2 where three subbands for sample S40 are reported, at nearly the same PL energy positions. The higher integrated intensities and the slight blue-shift of the CL bands are due to higher injection conditions and to the consequent larger screening of the internal fields (see the discussion below).

Power-dependent CL spectra recorded in cross-sectional geometry at constant electron beam energy showed a large blue-shift of the bands previously assigned to the QD transitions. As an example, figure 3 reports the results obtained for sample S2. The band at 2.8 eV shifted

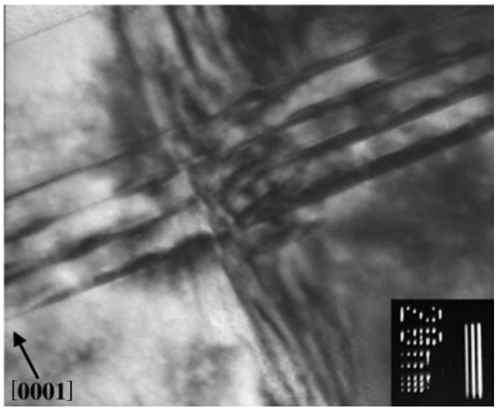

Figure 4. A conventional cross-sectional TEM image of sample S4 obtained in BF with $g = 0002$.

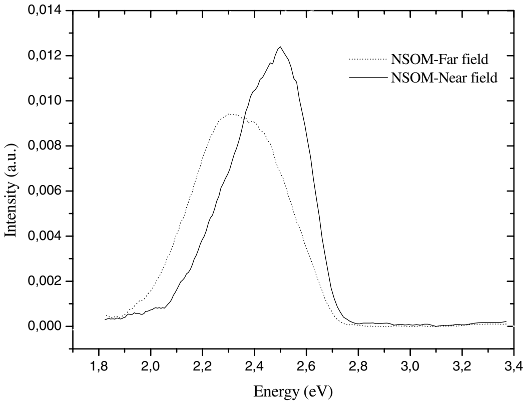

Figure 5. Results of NSOM in-plane studies of sample S40.

by nearly 200 meV in the range studied, while a smaller blue-shift for the band at 3.7 eV and no shift at all for the other two bands at 3.4 and 4.3 eV were observed. This result confirmed the nano-dimensional character of the transitions below 3.2 eV and the bulk character of the bands at 3.4 and 4.3 eV, the emission at 3.4 eV being ascribed to the GaN buffer layer. As for the nature of the transition at about 4.3 eV, it is not yet clear. Literature data [9] ascribed a similar band at 4.6 eV to a bidimensional GaN wetting layer. However, this interpretation cannot be applied to our results, since our power-dependent data do not reveal any shift due to quantum confinement effects. Similarly, an AlGaN wetting layer due to Al/Ga intermixing in the stacked layer sequence cannot be considered responsible for that emission. On the other hand, deep levels in the AlN layers or Al intermixing at the GaN/AlN buffer interfaces cannot be ruled out. Finally, the partial shift of the band at 3.7 eV suggests a mixed character of that emission, related to strained bidimensional GaN wetting layers [3] or to families of QDs with smaller dimensions. The presence of this band only for samples S2 and S4 can be explained by incomplete QD formation, as shown by the TEM micrograph reported in figure 4. This is consistent also with the variable thickness in the GaN/AlN stacking sequence in these samples.

The CL results were confirmed by the NSOM studies; typical examples are reported in figure 5. The spectrum acquired in far-field conditions (dotted curve; probe–sample

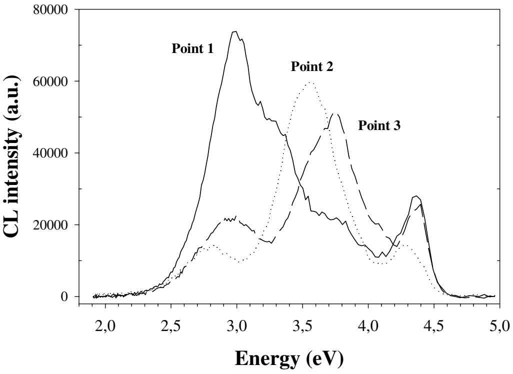

Figure 6. RT cross-sectional CL spectra of sample S2 recorded at different points at $E_b = 20$ keV, $I_b = 5$ nA.

distance: ~10 μm) showed a large band (FWHM ≈ 400 meV) peaked around 2.32 eV due to inhomogeneous broadening related to the spread of the QD dimensions. No differences in spectral shape were observed when spanning the excitation wavelength from 300 to 420 nm, except a decrease of the integrated PL efficiency. This seems to exclude the possibility of radiative recombination inside the GaN buffer layer, confirming that in our samples the QDs are excited directly by absorption in the excited states forming a quasi-continuum. The spectrum acquired in near-field illumination conditions (solid curve; probe–sample distance: ~10 nm) displayed a shift of the peak toward higher energies of about 180 meV and an overall reshaping of the spectral emission. No sharp peaks, the fingerprint of the single-dot recombination, were evident, as expected, given the large number of dots stacked on several planes simultaneously excited by the NSOM aperture, even in near-field illumination conditions. In agreement with [8, 10] and with our CL results, the blue-shift can be attributed to screening effects probably due to a preferential coupling of the near-field components with the uppermost QD layers. We remark that the screening of the internal fields induced by the photogenerated carriers yields both a blue-shift of the emission energy and a strong increase of the oscillator strength due to an increased spatial overlap of the electron–hole wavefunctions inside the dot and this can probably explain the reshaping of the emission spectrum. This is not so evident in the CL spectra (see figure 3), probably because it is rather difficult to work at injection conditions as low as the NSOM allows.

CL spectra obtained in cross-sectional geometry at different points revealed optical inhomogeneities only for the samples S2 and S4. Figure 6 shows the results for sample S2. The analysis of the behaviour of the relative intensities reveals an anticorrelation only between the QD emission at 2.8 eV and the band centred around 3.6–3.7 eV. This has been interpreted as a further evidence of the presence of families of QDs with different dimensions. Indeed, possible variations of the GaN wetting layer thickness competing with the QD formation cannot be ruled out, as shown for instance in figure 4 for S4.

Finally, the effect of the increasing number of stacked layers on power-dependent CL studies is shown in figure 7 for samples S40 and S85, where noticeable energy blue-shifts are shown. Different slopes of the lines corresponding to the deconvolution subbands of table 2 are apparent, suggesting that the two specimens present different internal fields, average dot dimensions and residual strains, but that it is not possible to separate their contributions. This made it necessary to use a complementary technique sensitive only to one of the previous parameters. In fact, micro-Raman studies [11, 12] allowed us to obtain the biaxial strain in

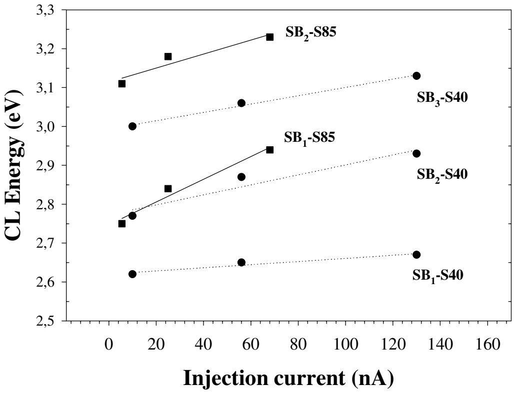

Figure 7. CL energy positions of the subbands fitting the cross-sectional CL spectra of samples S40 and S85 under increasing power injection conditions.

samples S40 and S85. The average values of the buffer layers were almost the same for both samples and of the order of the thermal strain (0.1% for GaN and 0.18% AlN). As for the QDs, the biaxial strain was −2.37 and −2.25%. This indicates that the residual strain is close to the nominal one for lattice-matched GaN/AlN (2.5%) and that a higher strain relaxation occurred in sample S85. This is consistent with the higher number of stacked layers and with the power-dependent CL data.

4. Conclusions

GaN/AlN self-assembled QDs grown on Si(111) have been studied by CL, micro-Raman, PL and NSOM spectroscopies. A good agreement among the optical techniques has been found as far as the energy band positions and internal field screenings are concerned. The CL cross-section studies allowed us to distinguish among the contributions from the different layers of the samples. The power-dependent CL enabled us to assign the origin of the transitions and to reveal the presence of different families of QDs, as confirmed by NSOM studies. A possible competition between the GaN wetting layer and island formation was also evidenced by cross-sectional CL results and confirmed by XTEM studies. Finally, micro-Raman and power-dependent CL findings clearly evidenced a different residual strain in samples with different numbers of stacked layers.

Acknowledgments

One of the authors (O Martinez) gratefully acknowledges financial support from the ‘Ministerio de Educacion y Cultura’ of the Spanish Government through a post-doctoral position.

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