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Study of isolated cubic GaN quantum dots by low-temperature cathodoluminescence

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Abstract

We report single dot spectroscopy of cubic GaN/AlN self-assembled quantum dots. Typical linewidths of the zero-phonon line between 2 and 8 meV are observed and interpreted in terms of charge fluctuations around a given quantum dot. The phonon sideband contribution in this emission, even at low temperature, reveals the importance of the acoustic phonon broadening mechanism which controls the exciton dephasing and may impose the real limits to the optical properties of GaN single QDs emission.

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1. Introduction

Although self-assembled GaN/AlN QDs present much interest for optoelectronic applications in the UV spectral range, their electronic and optical properties are still far less well known than for more mature QD systems such as InAs/GaAs. In particular, the optical study of a single GaN cubic QD has not been reported to date. The development of single QD spectroscopy in this system is particularly appealing since numerous specific properties are expected for GaN QDs [1] (stronger coupling to phonons [2,3], large excitonic effects for the cubic phase [4], very large single-particle

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charging effects for QDs in the hexagonal phase [5]). We report here a cathodoluminescence study of single GaN QDs, grown in the cubic phase.

2. The samples

The GaN QDs were grown by plasma-assisted MBE [6]. The metal fluxes were provided by conventional effusion cells, while active nitrogen resulted from radio frequency dissociation of N_2 using a plasma cell. The cubic pseudo-substrates consist of a 3 μm thick 3C–SiC layer grown by chemical vapor deposition on Si(0 0 1) substrates [7]. Both the surface morphology, and the strain relaxation of the GaN layer were monitored in situ by using reflection high-energy electron diffraction (RHEED). A smooth cubic AlN buffer layer with a thickness of about (~40 nm) was first obtained for a growth temperature $T_S = 720\,^{\circ}C$ under stoechiometric conditions. The roughness profile of AlN deduced from (1 μm × 1 μm) topographic atomic force microscopy (AFM) images shows superficial steps with a maximum height of 8 Å. The structural quality of the AlN buffer layer was checked by high-resolution X-ray diffraction and the purity of the cubic crystallographic phase was assessed by Raman spectroscopy measurements. After the deposition of 3MLs GaN under stoichiometric conditions, GaN islands appear as revealed by the abrupt change of the RHEED pattern [6]. These islands were subsequently overgrown with AlN, recovering a smooth surface after typically 15 nm. These capped GaN QDs were examined by transmission electron microscopy (TEM), and compared with data obtained by atomic force microscopy: a mean island height of 1.6 nm and a mean diameter of 13 nm are extracted with a density of a few $10^{10}\,cm^{-2}$.

A (1 1 0) cross-sectional TEM dark-field image in Fig. 1 clearly reveals that the GaN islands are formed on top of a two-dimensional wetting layer whose thickness is estimated to about 2 monolayers. Furthermore, the size of the GaN QDs agrees fairly well with that measured by AFM. One can also notice the presence of numerous stacking faults along the <1 1 1> directions. The TEM image shows that the islands grow preferentially within each domain bounded by these stacking faults. The large density of stacking faults may be at the origin of the broad size distribution of the islands since they can lead to local relaxation at the growth front and, thus, to an inhomogeneous strain distribution on the surface.

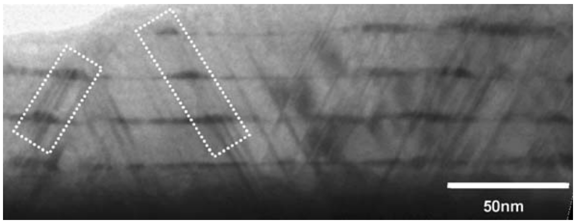

Fig. 1. TEM image in cross section of cubic GaN/AlN showing the GaN QD growing along the staking faults.

In order to isolate single GaN QDs within such a dense ensemble, square mesa structures were defined by e-beam lithography and reactive ion etching using a $SiCl_4$ plasma and a Ni mask.

3. Cathodoluminescence

A cathodoluminescence setup with variable temperature (4–300 K) sample holder was used to study these mesas. For the smaller mesa structures (100 nm), single QD emission spectra could be resolved in the high-energy tail of the QD ensemble (see Fig. 2). At low temperature (5 K), these spectra consist of a narrow line with a full-width at half-maximum (FWHM) between 2 and 8 meV (setup resolution 200 μeV) depending on the dots (see Fig. 2), and a strong low-energy acoustic phonon wing. With increasing temperature (Fig. 3), the emission peak shows a redshift, a reduction of its amplitude, and an energy broadening due to the increase of the phonon contribution. We mention that at about 80 K the FWHM is 7 meV for Fig. 3(a) respectively 9 meV for Fig 3(b), which is still much lower than the results previously reported in cathodoluminescence spectroscopy for the emission of a single hexagonal GaN dot [8].

The narrow line which is assigned to the excitonic zero phonon line is much larger than the intrinsic homogeneous linewidth due to the population relaxation as given by the decay time observed in time-resolved spectroscopy [9]. As a consequence, the origin of the observed linewidth should be attributed to other mechanisms such as

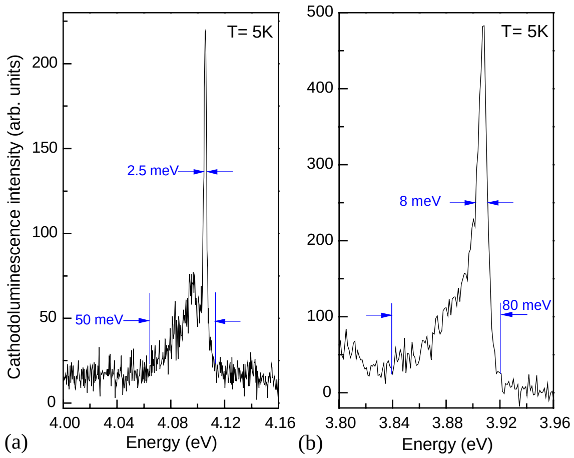

Fig. 2. (a)–(b) CL spectra obtained at 5 K for two single GaN QDs in 100 nm mesa structures.

dynamical broadening processes. Charge fluctuations at the mesa sidewalls, at traps in the vicinity of the QD (GaN wetting layer and AlGaN cap layer), could change the electrical field locally and cause a significant homogeneous line broadening of the order of several meV [10]. That would explain the FWHM values obtained here by cathodoluminescence for which large electron fluctuations are expected. This contrasts with micro-photoluminescence data with which FWHMs of several hundred micro-eV have been reported [11], even for nitrides dots such as InGaN ones [12].

As concerns the acoustic phonon sideband, it is stronger and broader than in the usual QD semiconductor systems such as CdTe/ZnTe [13] or InAs/GaAs [14]. To describe this band, an extension of the Huang–Rhys theory of localized electron–phonon interaction to the exciton system in a QD has been proposed by Besombes et al. [13]. In this model, we no longer consider the exciton–phonon interaction as a perturbation, but take

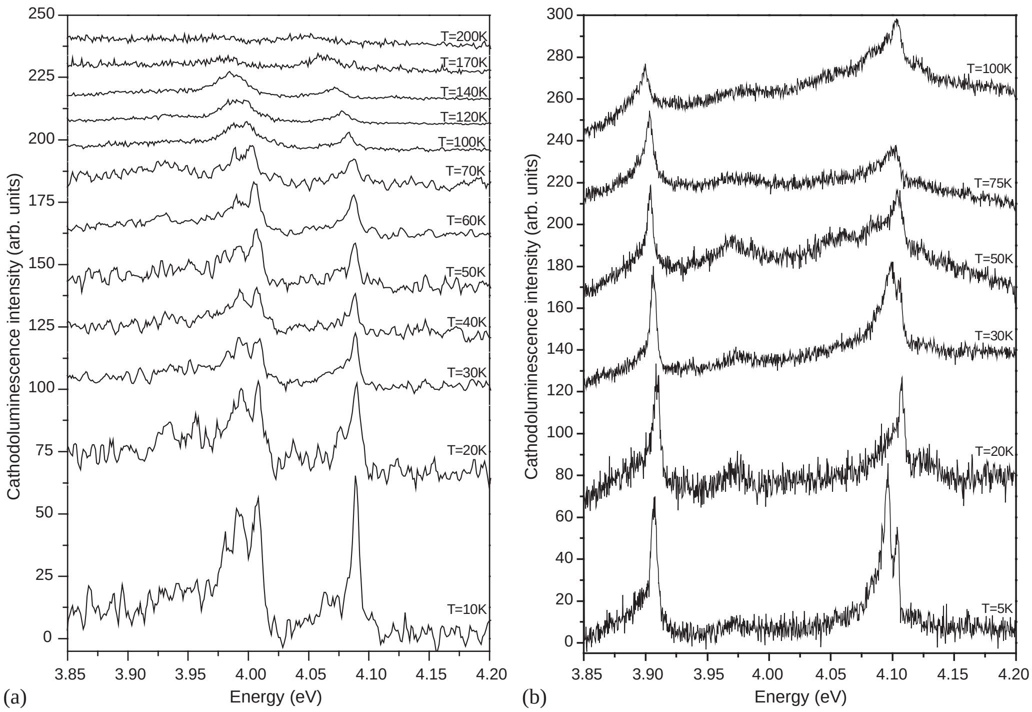

Fig. 3. (a)–(b) CL spectra for two single GaN QDs at different temperatures. The QD in Fig. 3(b) is the same as the one presented in Fig. 2(b).

into account the new eigenstates resulting from the coupling of a discrete excitonic state with the continuum of acoustic phonons. This gives rise to the exciton–phonon band surrounding the zero–phonon line which can be viewed as due to a pure dephasing mechanism, that is a loss of phase coherence within the mixed exciton–phonon state. Besides, the knowledge of the potential deformations in cubic nitride system, the importance of the strain-induced electric fields has to be taken into account in the present case in order to quantitatively fit the experimental data (see Fig. 3) as a function of temperature [15].

4. Summary

We have reported single-dot spectroscopy of cubic GaN/AlN self-assembled quantum dots grown by molecular beam epitaxy. By reducing the number of quantum dots using sub-micron mesa structures, we have obtained several cathodoluminescence peaks emitted by individual quantum dots. Typical linewidth of the zero-phonon line between 2 and 8 meV are observed and interpreted in terms of charge fluctuations around a given QD. The importance of the phonon sideband in this emission evidences the efficiency of the phonon broadening mechanisms which controls the exciton dephasing and which may impose the real limits to the optical properties of GaN single QD emission.

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