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Study of InGaN/GaN quantum dot systems by TEM techniques and photoluminescence spectroscopy

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Abstract: InGaN/GaN multilayer quantum dot structures produced by MOCVD techniques on c-plane sapphire were studied by transmission electron microscopy (TEM) and photoluminescence (PL) techniques. Indium fluctuations ranging from 1-4 nm were observed with both energy filtered TEM (EFTEM) and high angle annular dark field (HAADF) scanning TEM. The existence of V-shaped defects with nucleation centres at the termination of threading dislocation were observed in HAADF images. There was also evidence of the formation of large quantum dots at low densities from lattice HRTEM images. This was further confirmed by PL measurements through the observation of a single sharp line at low power with the typical saturation behaviour at higher power excitation.

1. Introduction

Self-organized quantum dot structures (SOQDs) are currently being studied for device applications. Ternary III-V alloys employing indium are the most promising materials for applications spanning the frequency range from Terahertz to visible/near UV. InGaN SOQDs have recently been used in the active region of laser diodes to improve their performance [1]. Despite the wealth of potential applications the growth of desirable In-rich quantum structures is still a challenge because of the high lattice mismatches, and therefore it is important to obtain a detailed understanding of the formation process and the atomic arrangements of the group III elements. In this work high angle annular dark field (HAADF) (where contrast arises from the sensitivity of the scattering intensity to the atomic number), energy filtered transmission electron microscopy (EFTEM) (which shows up compositional changes in the sample structure and provides concentration maps) as well as high resolution transmission electron microscopy (HRTEM) and photoluminescence (PL), are combined to study quantum dots in the InGaN/GaN system.

2. Experimental

Samples were grown on c-plane sapphire by metal-organic chemical vapour deposition (MOCVD): a thick GaN buffer-layer was grown at 1120 °C followed by 5 repeats of 15nm GaN at 765 °C and 2 nm InGaN at 745 °C with nominally 15% In (top layer was uncapped). HRTEM, EFTEM and HAADF images were acquired with a Tecnai F30 FEG-TEM microscope operating at 300 kV. EFTEM maps were produced by passing electrons which have lost at least 443 eV, corresponding to the In M4,5 absorption edge, through a Gatan imaging filter. The images were taken under short periods of...
irradiation to prevent electron beam induced artefacts [2], and the electron current density was kept as low as possible to further reduce any electron beam damage. Care was also taken to capture images from a fresh area within 2 minutes to reduce damage due to prolonged exposure [3]. The specimens for cross-sectional TEM analysis were prepared by mechanical polishing followed by ion beam milling.

A 100 nm thick Al film was evaporated on to the sample surface which was covered by specific diameter polystyrene beads (~650 nm). The sample was then placed in liquid nitrogen to remove the beads. This produced sub-micron apertures on the sample surface, through which microscopic photoluminescence (µ-PL) spectra were taken using the 405 nm line of a cube diode laser as the excitation power source. The laser beam was focused on the sample surface by an objective lens. The PL signal was collected by the same objective lens, and detected by a liquid-nitrogen-cooled charge coupled device (CCD) detector through a monochromator. The laser spot and its position on the sample surface were monitored by a CCD camera. The sample was mounted in a cold finger cryostat cooled to about 3.8K.

3. Results and Discussion

Figure 1(a) shows a HRTEM image of the bottom 3 quantum dot layers. There is evidence for a large quantum dot on the bottom layer (dark pyramidal region). The dot diameter and height are estimated to be 15 nm and 2.5 nm respectively. Similar quantum dots were observed by other groups [4] The dark region is due to strain caused by lattice mismatch between the InGaN region and the GaN barrier. Figure 1(b) shows a HRTEM image of all five GaN/InGaN layers and part of the thick GaN buffer (bottom left). From AFM images the density of the surface dots was evaluated to be about $6 \times 10^8$ cm$^{-2}$ [5].

Figure 1. HRTEM image of (a) the bottom 3 quantum dot layers, (b) all five repeats of GaN/InGaN layers and part of the thick GaN buffer layer (bottom left)

Figure 2(a) shows an HAADF scan in which four quantum dot layers can be identified. The image was acquired from a relatively thick part of the sample; therefore the dot layers appear very uniform. The dark area at the bottom left of the image is the vacuum. The InGaN layer width is measured to be approximately 2 nm, which agrees well with the nominal growth thickness. The thickness of the GaN barrier is measured slightly less than the nominal thickness of 15 nm. Figure 2(b) is a HAADF scan from an area of the sample which is thinner than in (a). Indium fluctuations within the quantum dot layers can be seen here, which range from 1 to 4 nm in length. Bright stripes parallel to the basal plane are InGaN layers (Z of In = 49) and dark ones are GaN layers (Z of Ga = 31). Figure 2(c) is an HAADF scan from a thicker area of the sample. Two V- shaped defects [6] can be seen at the surface, one of which nucleates at the termination of a threading dislocation.
Figure 2. HAADF images of (a) four quantum dot layers, (b) an area thinner than in (a) and (c) an area with V-shaped defects and threading dislocation. (d) EFTEM map of In distribution in the quantum dot layers.

In order to understand the compositional information of the sample EFTEM was carried out. Figure 2(d) represents an EFTEM map produced by passing electrons through the filter that have lost 443 eV corresponding to the In M4,5 absorption, to show the indium distribution in the structure. It can be seen that the distribution in the quantum dot layers is not uniform, with fluctuations leading to small-scale clustering of indium.

In order to see manifestation of quantum dot formation in the optical behaviour of the sample PL experiments were carried out. Figure 3 represents PL spectra for two different samples at 4K as function of excitation power taken through a sub-micron size aperture (~550 nm); the upper spectrum in (a) is from an unmasked sample (macro-PL). The usual excitonic emission (the large band) in (a) consists of several sharp lines while the macro-PL spectrum exhibits a continuum (broad peak). The discrete structure of the emission indicates that the epilayer is strongly disordered due to the immiscibility of indium in GaN within the wetting layer and to well width fluctuations, suggesting that there are localized exciton centres. Figure 3 (b) shows a series of spectra from a sample with a longer deposition time. At low excitation powers there is a single sharp line. This line reaches saturation point as the excitation power increases, as is typical of a quantum dot. We attribute this line to a single quantum dot. The AFM studies by Davies et al. [5] suggest a low dot density of about $6 \times 10^8$ cm$^{-2}$ and statistical studies of the number of apertures (~550 nm apertures) containing at least one dot (using PL) agree with this AFM density.

Figure 3. PL spectra at 4K as function of excitation power, taken through a submicron size aperture (~550 nm) for two samples with InN deposition times of 12s and 17.5s. Spectra are displaced vertically upwards with increasing excitation power.
4. Conclusion
EFTEM images using the In M4,5 absorption edge showed that In distribution is not uniform in the quantum wells and that there are fluctuations leading to clustering of In. There is agreement between EFTEM and HAADF showing the indium clusters within the quantum dot layers range from 1 - 4 nm in length. V-shaped defects were observed, some of which nucleated at the termination of a threading dislocation. There is evidence of large quantum dots in HRTEM images with estimated diameter and height of 15 nm and 2.5 nm respectively. This is confirmed by micro PL results which show a single sharp PL line at lower excitation power similar to a typical quantum dot emission which saturates with increasing power. AFM suggests a low density of $6 \times 10^8$ cm$^{-2}$ surface dots.

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References