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SHORT COMMUNICATION

Confocal microscopy and spectroscopy of InGaN epilayers on sapphire

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Summary

We report a preliminary investigation of spatial inhomogeneities in an InGaN epilayer using scanning confocal microscopy as the investigative tool. The Daresbury confocal microscope SYCLOPS provides simultaneous high quality reflection and fluorescence images of InGaN sample areas up to 500 μm square, even at room temperature. Sample cooling increases the brightness and quality of the fluorescence image, as expected. Spectral selection using interference filters permits identification of features close to sample edges resulting from the nitridation of indium droplets. The unexpected non-coincidence of fluorescence and reflection features below 10 μm in size is tentatively attributed to the differing absorption strengths of different crystallites.

Group III nitride epilayers, grown by metallorganic vapour phase epitaxy (MOVPE) on sapphire substrates, are currently of interest for use in light-emitting devices (LEDs) and lasers, primarily in the blue–green spectral region (see for example Gil, 1998). While it has recently been shown that epitaxial lateral overgrowth (ELO) techniques produce material that is almost single crystalline in habit, conventional, non-ELO growth, on a low-temperature buffer of GaN or AlN, produces material that is intrinsically polycrystalline (Yu *et al.*, 1998). The individual crystallites, resolvable by electron and scanning probe microscopies, are grains several micrometres across with a consistent orientational relationship to the sapphire substrate and therefore to each other (Trager-Cowan *et al.*, 1996). Occasionally, a crystallite, or several together, will grow to an exceptional size, perhaps greater than 100 μm in diameter. However, most of the light emission from nitride epilayers is found to be related to smaller, possibly nanometric, features of the growth (Chichibu *et al.*, 1997).

Recently, much attention has been paid to the emission of InGaN layers containing different fractions of indium, since luminescent devices are 'tuned' to different wavelengths by changing the amount of indium incorporated in the active layer. The present work was undertaken to test the possibility of examining 'statistical fine structure' (i.e. a statistical variation of the spectral character with excitation spot position) of conventional InGaN layers, by using confocal laser scanning microscopy to recover the spectra of individual crystallites. Complementary work using cathodoluminescence imaging and spectroscopy, mainly on larger crystallites, will be reported elsewhere (O'Donnell *et al.*, 1998a).

InGaN films 100–200 nm thick were grown by MOVPE on top of thicker (*c.* 1000 nm) GaN layers on thick (0001)-orientated sapphire substrates in a Thomas Swan rotating disc reactor. The indium content of these films was controlled by adjusting the growth temperature and growth rate (Van der Stricht *et al.*, 1997).

Confocal microscopy was carried out using the Daresbury Laboratory microscope SYCLOPS (Van der Oord *et al.*, 1992). This microscope has confocal illumination and detection optics arranged on an optical bench for flexibility of application, and features an attached scanning mirror head, objective lens mount, and sample stage. Laser sources are commonly used for high resolution imaging, while synchrotron radiation is used for fluorescence lifetime spectroscopy.

The setup of the microscope for the experiments to be described here is illustrated in Fig. 1. A 488-nm (2·54 eV) Ar^+ laser (145 mW, Omnichrome) is focused through the

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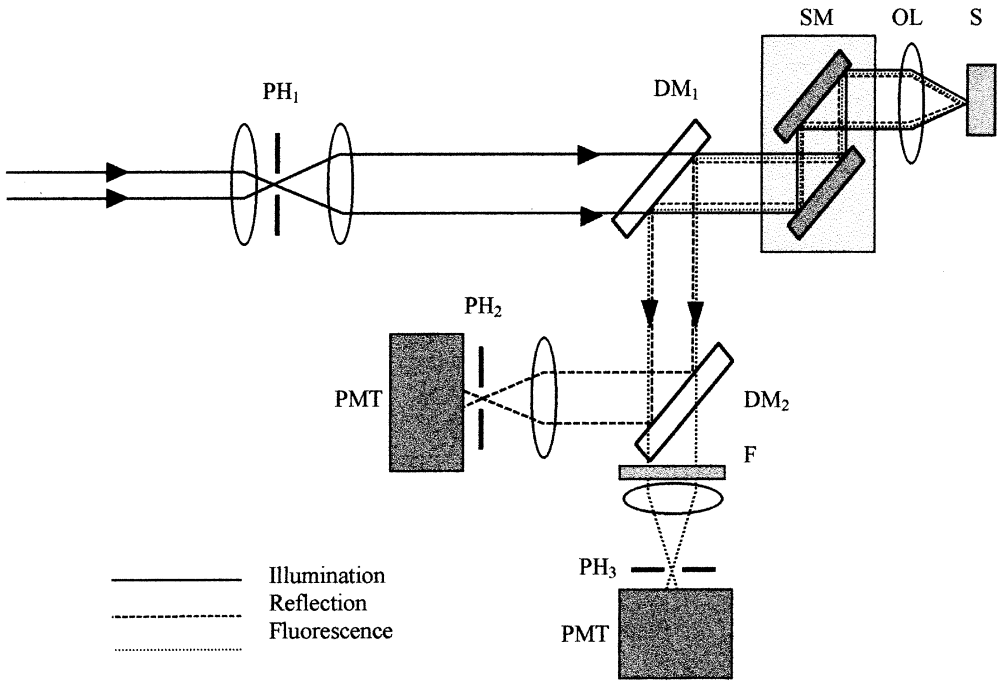

Fig. 1. Layout of SYCLOPS, as described in text.

source pinhole (PH_1) and collimated to a parallel beam. This passes through a short pass dichroic mirror (DM_1, Tech Optics), with a reflecting cut-on at 495 nm, to the scanning mirror head (SM). A Zeiss 10× infinity-corrected objective lens (OL) gives a large field of view, $(500\,\mu m)^2$. Reflected and fluorescence light from the sample (S) is collected by the objective and passes back through the scan head. The fluorescence, plus a small portion of the reflected light, is deflected by DM_1 into the detection beam-path. A second dichroic mirror (DM_2, Nikon), reflecting below 510 nm, separates the reflected and fluorescence light into two detection channels. These each contain a focusing lens, pinhole ($PH_{2,3}$), and cooled photomultiplier tube (PMT, Hamamatsu R3896). This arrangement allows simultaneous collection of fluorescence and reflection images from the same focal plane within a sample. Further spectral selection of the fluorescence beam can be achieved by inserting bandpass filters in the fluorescence light path at position F. Sample temperature control from 80 K to room temperature is achieved by using a Linkam HFS 91 liquid nitrogen cooled stage. We are primarily interested here in the greater lateral resolution and image clarity offered by confocal techniques. The well-established capability of the confocal microscope to prepare vertically separated image slices is not expected to reveal very much additional detail in thin layers.

As described above, a single laser scan in SYCLOPS produces two images: one corresponding to reflected 488 nm laser light from the image plane and the other to fluorescence, at a longer wavelength which is determined in practice by inserting an optical filter in the path of the beam. To enhance viewability of the images, we adopt a convention of colouring the reflectance and fluorescence images green and red, respectively, using the image processing software package PaintShopPro (Jasc Software, Inc., Eden Prairie, MN). The image processing software allows images to be overlaid; the composite (reflectance + fluorescence) of a region of sample in the vicinity of a large crystallite image is reproduced as Fig. 2. While features can be seen in both red and green images which appear to represent crystallites with a narrow range of diameters around 5 μm (the width of a pixel is about 1 μm at this magnification) there are very few true coincidences between the red and green images, even at the single pixel level. Note that a perfect registration of a red and a green pixel with equal brightnesses would produce a yellow composite. On the other hand, within the area covered by the large crystallite in Fig. 2, there is a close registration of the pattern of the fluorescence image with the crystal shape. We believe that such patterns, which appear in the form of 'stars', 'flowers' or 'butterflies', are caused by refraction of the exciting light and total internal reflection of the fluorescent light at crystal faces.

It is well known that the conversion efficiency of luminescent materials increases as the sample temperature is lowered, through a reduction of the quenching effect of non-radiative processes. Lowering the sample temperature increases the brightness and quality of a fluorescence image of InGaN layers, as illustrated in Fig. 3. Wavelengths above

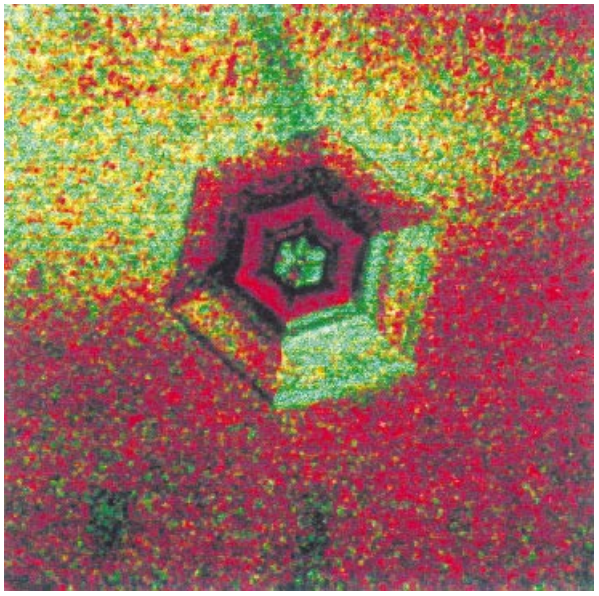

Fig. 2. Composite reflection (green) and fluorescence (red) image of region of an InGaN epilayer, approximately 500 μm square.

500 nm are detected in this image; again, false colour has been used. A close examination of the figure shows that different crystallites appear to have rather different temperature dependences.

In order to carry out spectroscopic investigations in the confocal fluorescence microscope, we introduce different interference filters, successively, into the fluorescence beam path, whilst keeping the excitation conditions and the sample temperature constant. This procedure is beset with calibration problems. To date, we have achieved only semiquantitative results on available samples. For example, Fig. 4 compares fluorescence images, centred at 550 nm and 600 nm, respectively, from the edge of a bilayer of InGaN/GaN grown on sapphire. There are clear and obvious differences between Figs 4(a) and 4(b), chiefly in the relative intensities of two groups of features towards the top right and bottom left of the images.

Conventional PL spectroscopy effectively averages the fluorescence emission from a region of a sample of macroscopic size. Spot-by-spot spectroscopy of the sample used in the present work shows a macroscopic gradient of

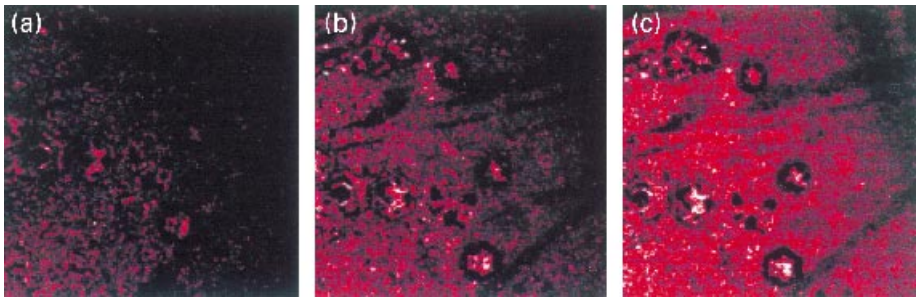

Fig. 3. Temperature dependence of fluorescence from InGaN layer at (a) 273 K, (b) 173 K and (c) 123 K.

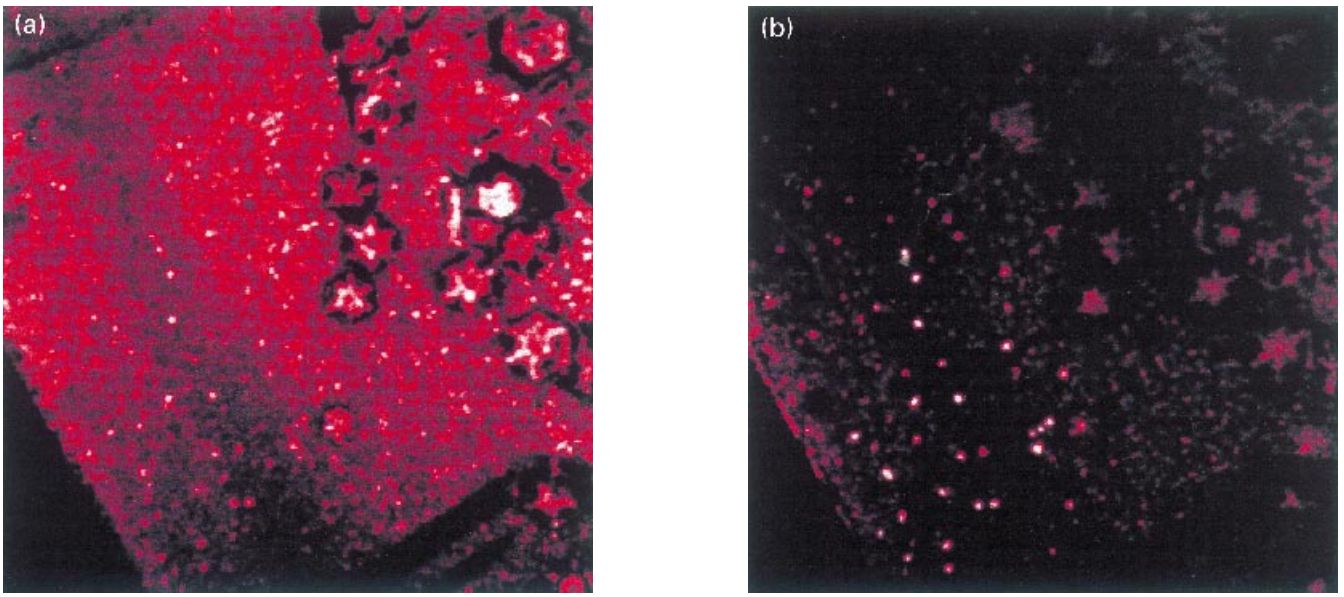

Fig. 4. Fluorescence micrographs centred at (a) 550 nm and (b) 600 nm.

the composition that is associated with differential indium incorporation in different regions of the wafer (Bayliss *et al.*, 1998). A composition gradient will tend in the first instance to broaden luminescence lines and also to distort them. However, the disposition of the compositional inhomogeneities cannot be deciphered from spot measurements along a line on the sample surface. On the other hand, confocal microscopy provides this information directly. It is noticeable from Fig. 4 that luminescence at 600 nm is due almost entirely to a number of small crystallites situated close to the edge of the sample. Studies of growth of samples with high indium content show that these features form when indium droplets react with ammonia on the sample surface (Van der Stricht *et al.*, 1997), suggesting that the small crystallites in Fig. 4(b) are probably composed of pure InN. In order to confirm this conjecture it will be necessary to increase the spectral resolution of the present instrument. This enhancement is in hand. Alternatively, it may be possible to combine confocal fluorescence microscopy with high resolution (microfocus) X-ray diffraction by utilising the brightness of the synchrotron radiation source.

The different temperature dependences of features in the confocal micrographs obviously need to be evaluated individually. In principle, the confocal microscopy technique, with the resolution used in the present work, is capable of acquiring $512 \times 512 = 262144$ spectra simultaneously. Not all of these spectra will be independent of course. Sophisticated automated data evaluation techniques will need to be developed in order to process this glut of information.

Finally we address the problem of observing statistical fine structure, in other words a statistical variance in the spectra of a number of crystallites, caused by variation in indium content from one to the other. Figure 2 reveals an unexpected lack of coincidence, in the case of the smaller crystallites, between features observed in fluorescence and reflection. Similar anti-correlations have been noted in previous studies using near-field scanning optical microscopy (Vertikov *et al.*, 1998). The excitation laser has insufficient photon energy to excite the underlying GaN directly across its band gap. Moreover, these samples show negligible GaN defect luminescence in conventional PL, hence all red channel features are due to InGaN (or InN) fluorescence and all green channel features are due to InGaN reflection. The absorption spectrum of (macroscopic) InGaN has a broadened step-like profile (O'Donnell *et al.*, 1998b). We speculate that the spectrum of an individual crystallite may be somewhat sharper. Hence a sharp division may exist between crystallites that are capable of absorbing laser light, and subsequently fluoresce, and those that do not, which reflect. Interference in thin films of varying thickness may also play a role. Again, confirmation of these ideas requires further development of the technique.

In summary, we have applied confocal microscopy to the study of spectral inhomogeneity in InGaN epilayers. The preliminary results suggest that the technique may be developed for the study of statistical fine structure in the spectra of small crystallites.

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