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Ionoluminescence and ion beam induced secondary electron imaging of cubic boron nitride

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Abstract

In this work, we introduce the use of ionoluminescence (IL) with ion beam induced secondary electron (IBISE) imaging to correlate the surface topography and crystal faces with luminescence properties. Since both IL and IBISE require low beam currents of <1 pA, simultaneous analysis can be performed. The newly installed system for light and electron detection in the NUS micro-beam facility is described. The performance of the present setup is demonstrated with high-resolution IL and IBISE images collected on Al-doped (1 1 1) c-BN and undoped (1 0 0) c-BN under 2 MeV proton irradiation. Results show that blue luminescence of Al-doped c-BN appears as triangular patterns and they tend to be associated with triangular voids on the surface. This work demonstrates the importance of combined IL and IBISE for characterizing wide band gap semiconductors.

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1. Introduction

Ionoluminescence (IL) performed with a nuclear microprobe is an emerging technique for the characterization of optical properties of semiconductors [1,2] and geological samples [3,4]. IL measured simultaneously with other ion beam analytical techniques can often enhance the information obtained from focused ion beam analysis and aid in the interpretation of the results obtained. IL has previously been used in combination with particle induced X-ray emission (PIXE) to study the spatial distribution of optically active impurities [5], and ion beam induced charge collection (IBIC) to obtain complementary information of the electrical and optical properties of semiconductors [6,7]. In this work, we show the potential application of IL combined with ion beam induced secondary electrons (IBISEs) to provide a clearer understanding of the luminescence properties and its correlation with surface topography of the material.

A new IL system has recently been installed at the nuclear micro-beam facility, National University of Singapore. The system is capable of performing high-resolution IL imaging and spectroscopy. A channel electron multiplier (CEM) has also been installed in the target chamber to

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provide the ability to perform simultaneous IL and IBISE imaging. This is possible because the number of photons and electrons detected are of the same order and only low beam currents of <1 pA are required. This paper describes the instrumentation for combined light and secondary electron detection and its application to the study of Al-doped (1 1 1) c-BN and undoped (1 0 0) c-BN.

2. Instrumentation

IL imaging is carried out using a H7421-40 Hamamatsu Single Photon Counting Head. This photomultiplier tube (PMT) system was chosen due to its high sensitivity, compactness and low dark current. It utilizes a GaAsP as photocathode, which offers high quantum efficiency in a spectral range of 300–720 nm. Typically, a very low beam current of $\ll$1 pA is needed to collect an IL image and this limits the beam damage to the luminescence properties of the materials. The PMT is Peltier cooled in order to maintain the counts due to dark current to less than 20 Hz. The TTL output of the PMT is then plugged directly into the OMDAQ data acquisition system for panchromatic imaging. In the case of monochromatic imaging, a notch filter with a chosen wavelength bandpass is attached in front of the PMT.

Spectroscopic measurements are performed using a multichannel Ocean Optics USB2000 Miniature Fiber Optic CCD Spectrometer, fitted with a 300 lines/mm grating blazed at 550 nm, and a fixed entrance slit of 200 μm. The multichannel spectroscopic system allows for high-speed continuous spectral acquisition with integration times as low as 300 ms. This feature is useful for monitoring the ion beam induced damage on the sample being studied. The spectrometer is controlled via a USB port using the OOIbase32 software that is provided.

The Amptektron UHV Model MD502 is used for electron detection in ultra-high vacuum. The CEM is placed in the target chamber and electrically connected to the remote MD502 electronics module via three in-vacuum SHV feedthroughs. A positive bias of 500 V is supplied to the cathode by the MD502 module to attract the electrons, thus increasing the electron detection efficiency. The Amptektron CEM system has a TTL pulse output that can be directly plugged into the OMDAQ data acquisition system.

3. Experimental setup

All measurements were performed at the Research Centre for Nuclear Microscopy, National University of Singapore. A beam of 2 MeV protons focused down to less than 1 μm was used to image the ion-induced luminescence and secondary electron emission from the sample.

Fig. 1 shows the typical setup for simultaneous IL and IBISE experiments. The channeltron is placed at 20° to the sample surface and a distance of 2.5 cm away from the target. For convenience, the CEM holder is mounted onto the same flange as the SHV feedthroughs so that the CEM detector can be permanently attached to the electrical connectors. Light is collected into the spectrometer and PMT via the 45° optical microscope mounted to the target chamber. This method was first adopted by Bettiol et al. in MARC in 1994 [8]. Here, the PMT is screwed directly to the optical microscope with a C-mount adapter. Light is coupled to the spectrometer via an optical fiber that is attached to one of the eyepiece holders of the microscope. In this way, the IL detection system can be easily integrated into the current microprobe system. However, one disadvantage of the present setup is the UV absorption of the optical microscope glass optics and the port windows. For better collection efficiency, the IL images were collected in panchromatic mode.

In order to correct for the efficiency of the diffraction grating and absorption of the optical microscope, the spectral response of the spectrometer was calibrated using a LS-1-Cal NIST tungsten halogen light source purchased from Ocean Optics. Fig. 2 shows the sensitivity curve for the Ocean Optics Spectrometer. It can be seen that the highest spectral response occurs at around 560 nm, which corresponds closely to the blazed wavelength of the spectrometer. Due to the absorption of the optical microscope, the IL spectrum has a low-end cutoff wavelength of 400 nm. The cali-

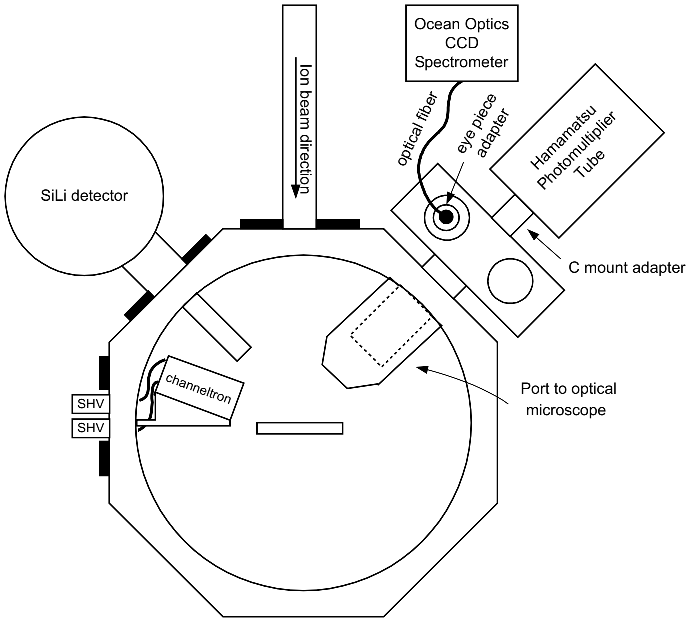

Fig. 1. Schematic diagram of simultaneous IL and IBISE setup.

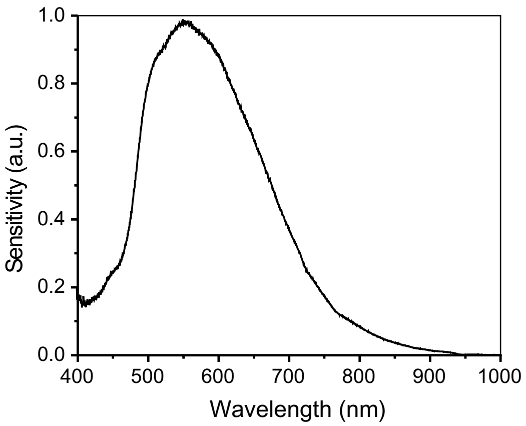

Fig. 2. Sensitivity curve of the Ocean Optics USB2000 spectrometer and light collection system.

brated spectrum allows accurate measurements of the peak wavelength and relative peak intensities.

4. Results and discussion

The new IL and IBISE system was used to analyze two c-BN samples. The c-BN crystals used in this study were recrystallized from hexagonal BN powder using the temperature gradient method in a belt-type reactor using ultra-high temperature, high-pressure conditions of 6–7 GPa and 1800–2000 °C.

4.1. Al-doped (1 1 1) c-BN

Fig. 3(a) shows the topography of the (1 1 1) c-BN surface measured with a Nomarski Optical Microscope. This image reveals that the surface consists of many $\langle 1\,1\,2 \rangle$ stacking faults and triangular voids composed of (1 1 0) planes twinned at 180°. An IL spectrum (Fig. 3(b)) from this sample was measured with a low beam current and a large scan area in order to minimize the modification of the sample luminescence by the ion beam. A single blue luminescence band peaked at 410 nm is observed in the IL spectrum. Cathodoluminescence measurements performed on the same sample (not shown) do not reveal any additional UV peaks in the range of 250–400 nm therefore IL imaging of the blue luminescence can be performed in panchromatic mode. Panchromatic IL imaging over a large scan area of 1×1 mm, revealed that the blue

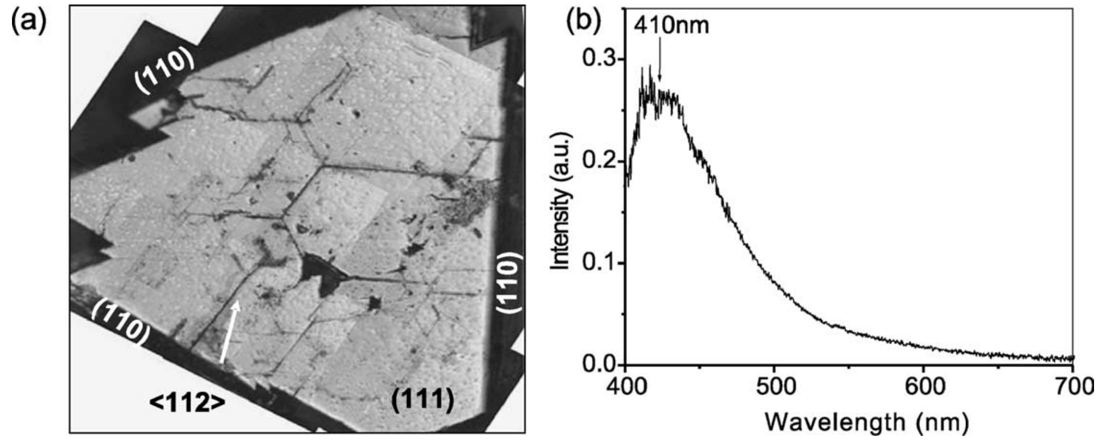

Fig. 3. (a) Nomarski optical micrograph and (b) IL spectrum of Al-doped c-BN sample.

emission appears as regular triangular structures (see Fig. 4(a)), aligned along the same direction. The corresponding IBISE image of the same region shows that the blue triangles tend to be associated with voids. It is possible to trace the region of interest back to a corresponding SEM micrograph in Fig. 4(a), which gives a better signal to noise ratio of the surface topography. Void 1 (Fig. 4(a)) is seen easily from both electron maps, however, void 2 can only be distinguished in the SEM map. In order to investigate the finer details of the triangles, the images were measured over smaller area of 400×400 μm (see Fig. 4(b)). The high-resolution IL map reveals that the blue emission surrounds the region immediately adjacent to the void. This suggests that it is related to

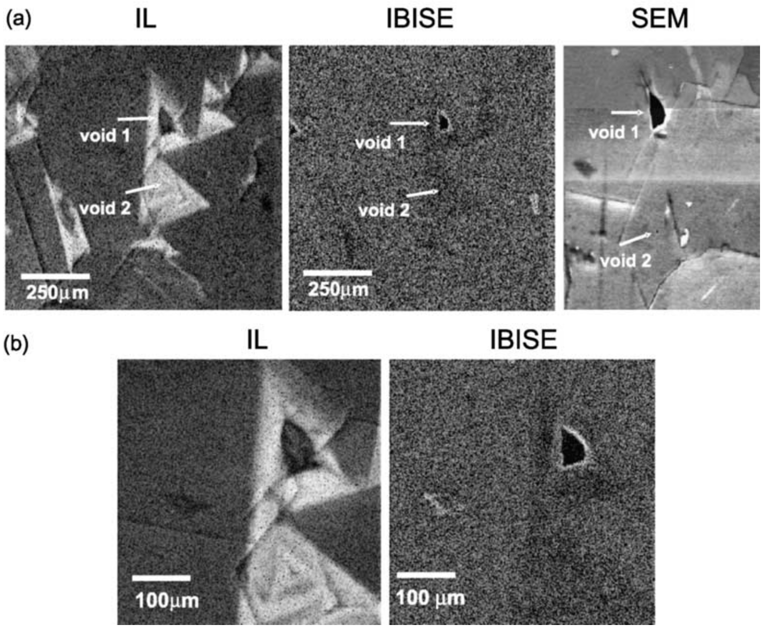

Fig. 4. (a) Panchromatic IL and IBISE images collected over an area of 1×1 mm, and SEM micrograph showing void 1 and 2. (b) IL and IBISE images over an area of 400×400 μm about the central area in (a).

the formation of the twinned (1 1 0) face. Concentric triangular patterns can be seen forming around the much smaller void 2. A closer look at the IBISE map reveals that the blue regions have a slightly lower electron yield. This contrast is not due to topography as the surface immediately adjacent to the void is flat under the SEM (see Fig. 4(a)).

Previous cathodoluminescence measurements on BN by Pipkin et al. [9] revealed a growth dependence of luminescence on different faces of $\langle 1\,1\,1 \rangle$ c-BN. It was also observed that the twin boundaries emit brighter luminescence. However, no explanation was offered at this time for the nature of luminescence. Other studies by Kanda et al. [10] showed that blue luminescence of c-BN could be due to nitrogen terminated $\langle 1\,1\,1 \rangle$ growth surface. The close correlation of the blue emission with twinned (1 1 0) triangular voids suggests that the blue luminescence is related to the (1 1 0) faces. Twinning of the (1 1 0) planes could happen at an impurity site, a common phenomenon on (1 1 1) surface.

4.2. Undoped (1 0 0) c-BN

Fig. 5(a) and (b) show simultaneous IL and IBISE images of two small grains of undoped (1 0 0) c-BN. The luminescence spectrum from both crystals shows a similar blue emission peak at around 405–415 nm. Panchromatic IL imaging of the first grain reveals a distinct V-shaped luminescence distribution (Fig. 5(a)). The second grain shows that the blue luminescence is concentrated

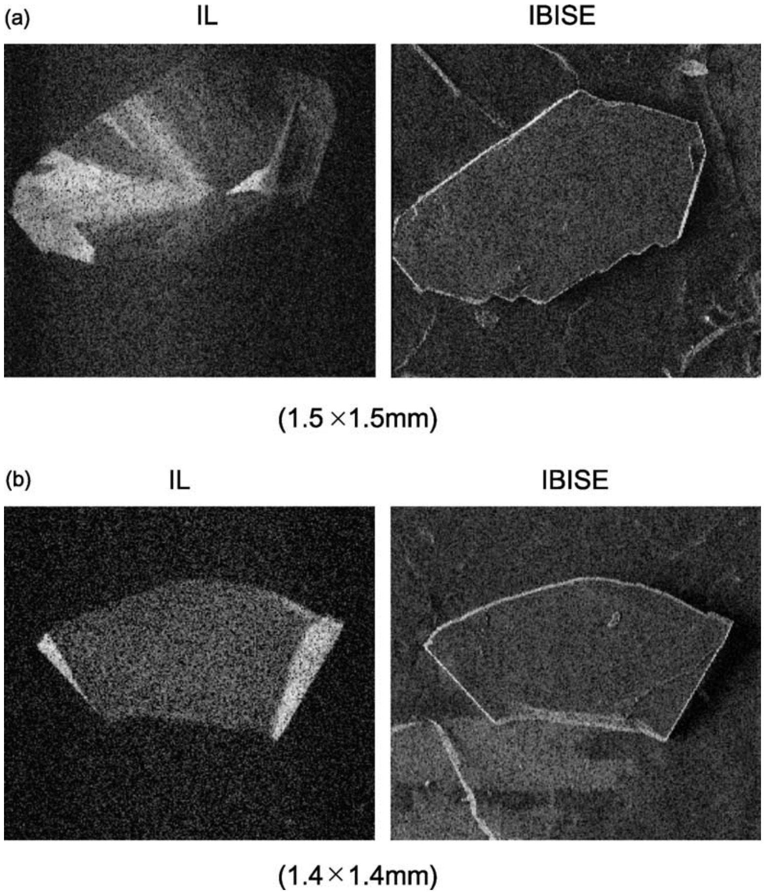

Fig. 5. IL and IBISE images of two grains of undoped (1 0 0) c-BN.

at the sides of the crystal (Fig. 5(b)). It is found that the topography of the crystals is rather flat and do not seem to indicate any correlation of the luminescence observed with different faces of the crystal or surface topography. From the regularity of the luminescence patterns, it is possible that the luminescence could be due to the imperfection of crystal lattice. No triangular patterns are observed from these (1 0 0) surfaces, in this case.

5. Conclusion

High resolution IL and IBISE imaging has been successfully employed to provide spatially resolved information of the luminescence properties and topography of c-BN under MeV proton irradiation. It is found that the blue luminescence of Al-doped BN appears as triangular contrast patterns, that tend to be associated with the formation of twinned (1 1 0) faces. The undoped c-BN that was studied shows no topographical dependence of the luminescence, nor does it show any dependence on crystal face.

Further improvements to the IL and IBISE system are currently being planned. A light pipe based IL light collection system is being developed that has improved collection efficiency, especially in the UV part of the spectrum. The signal to noise ratio of the IBISE imaging system is being improved by using a voltage modulated output signal, similar to that described in [11].

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