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The study of phosphors efficiency and homogeneity using a nuclear microprobe

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Abstract. Ion Beam Induced Luminescence (IBIL) and Ion Beam Induced Charge Collection (IBICC) have been applied in the study of the luminescence emission efficiency and investigation of the homogeneity of the luminescence emission in phosphors. The IBIL imaging was performed by using sharply focused ion beams or broad/partially-focused ion beams. The luminescence emission homogeneity in samples was examed to reveal possible distributed crystal-defects that may lead to the inhomogeneity of the luminescence emission in samples. The purpose of the study is to search for suitable luminescent thin films that have high homogeneity of luminescence emission, large IBIL efficiency under heavy ion excitation, and can be placed as a thin layer on the top of microelectronic devices to be analyzed with Ion Photon Emission Microscopy (IPEM). The emission yield was found to be low for organic materials, due to saturation of the light output dependence on the energy deposition of heavy ions. The emission yield of a typical Bicron plastic scintillator is about 70 photons/ion/micron. Inorganic materials may have higher IBIL yield under high-energy and heavy-ion excitation, but the challenging problem is the inhomogeneity of the IBIL emission. The IBIL image techniques are applied in the investigation of the homogeneity of a GaN epitaxial thin film, a zircon single crystal and a thin layer coated by Thiogallate (EuII) ceramic.

Introduction

The luminescence property of various materials has been a subject of research interests for a long time, however, the knowledge of IBIL excited by high-energy, heavy-ions in organic and inorganic materials is still very limited. The information on the IBIL yield in thin film phosphors is crucial to the success of a new nuclear emission microscopy: Ion Photon Emission Microscopy (IPEM), currently under development at Sandia National laboratory (SNL) [1]. The IPEM can be a very promising new tool for performing single ion effects microscopies without beam focusing, and potentially with a radioactive source instead of an accelerator. These luminescent thin films must be deposited or placed on the surface of microelectronic devices to enhance the production of IBIL. These IBIL photons are then projected with a high magnification lens system onto a position sensitive detector (PSD) that is sensitive to single photons. The efficient generation, transmission and detection of these photons is required using the IPEM technique to determine the arrival position of each ion which strikes the sample. The utility of IPEM therefore hinges on finding an optimum luminescent layer.

Plastic phosphors have several advantages over inorganic ceramic materials. They usually have a fast decay time (a few ns to tens of ns); they are clear and smooth (reducing or even eliminating light scattering); they are easily made thin and self-supporting; and such plastic scintillators are already commercially available. The drawback is that the emission yield is relatively low, due to saturation of the light output dependence on the energy deposition of heavy ions [2]. IBIL and IBICC are applied together to quantify the phosphors efficiency [3].

Inorganic materials may have higher IBIL yield under high-energy, heavy-ion excitation, but they have common problems of inhomogeneity of luminescence emission. Therefore, it is worthwhile to check the IBIL homogeneity prior to a further quantification analysis of the luminescence efficiency. In this paper IBIL image techniques were applied for the investigation of the homogeneity of the luminescence emission in the GaN

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epitaxial thin film, zircon single crystal and Thiogallate (EuII) ceramic.

Instrumentation

Measurements were performed on the UNT heavy-ion microprobe beam line in the Ion Beam Modification and Analysis Laboratory (IBMAL). A high quality optical microscope is one of the most important devices for the operation of a nuclear microprobe. A microscope (SM-OM40, JEOL USA, Inc.), which was originally designed for scanning electron microscopy (SEM), is installed in the nuclear microscope chamber for the observation of micrographic images with a very high magnification (x300) and a viewing field 650 microns in diameter. The optical microscope has an objective lens in a retractable long arm. The objective lens is a type of refracting lens with a hole drilled through its center through which the ion beam passes during nuclear microprobe experiments. The optical microscope has an easy working distance of 5 mm and numeral aperture (N.A.) of about 0.3.

With a large magnification, the microscope is ideal for viewing very small features (approximately one-micron resolution). The monocular eyepiece of the microscope can be easily replaced by a CCD camera or a luminescence detector such as PMT (photo-multiplier tubes) for Ionoluminescence applications. In this work, a PMT-based integrated photon counting head (Hamamatsu Model H5920-01) is used for a single photon counting detector. This luminescence detector has an output pulse height of 3.3 volts and 25 ns pulse width. The highest sensitivity of the detector is at about 400 nm wavelength with about 16% quantum efficiency. The dark current of the photon counting head is about 1-2 cps, after being stored in a dark room more than 24 hrs.

In this study, a PIN-diode (Hamamatsu, model S1223) is used to detect and count the incoming particles, which excite the IBIL photons. Thick and thin plastic phosphors can be placed on the top surfaces of the S1223 PIN-diodes. For the thick phosphors the ion beam is stopped completely, and only IBIL signals are detected. In uncovered area of the pin-diode, only IBICC signal produced and detected. The combination of the IBICC and IBIL information gives the quantitative information of the thick phosphor efficiency in the term of *photons/ions*. For the thin phosphors, the ion beam passes through the phosphors into the pin-diode and both IBICC and IBIL signals are detected. The experiments to determine the detected photon/ion efficiency proceeds just like an IBICC experiment on the PIN diode, where in addition, the IBIL signal is measured at the same time.

Two types of IBIL imaging methods are applied in this study. One is employing a broad (non-focused) ion beam. The IBIL image is captured through a video system attached to an optical microscope. In the second type of operation, a focused ion beam is applied and the IBIL image can be simultaneously obtained with other techniques in nuclear microscopy, such as IBICC, PIXE and RBS. This approach utilizes the full power of nuclear microscopy.

The measurement of the IBIL efficiency of organic scintillation materials

Nature of the luminescence in organic materials is characteristic of the molecular structure, because the interaction of inter-molecules is much weaker than that of intra-molecules. The organic phosphors are commonly aromatic hydrocarbon compounds containing linked or condensed benzene-ring structure. The luminescence can be explained with free valence electron transitions in the energy scheme of π-molecular orbitals [4].

The scan analysis with an alpha particle beam was carried out in the thick samples mounted on PIN-diodes. The scans were made to include 1) an area for IBICC analysis of the PIN-diode surface that remained uncovered and 2) part of sample area covered by the thick plastic which produced an IBIL signal. A typical IBIL and IBICC image for such a thick plastic phosphor is displayed in Figure 1, where a 6.0 MeV

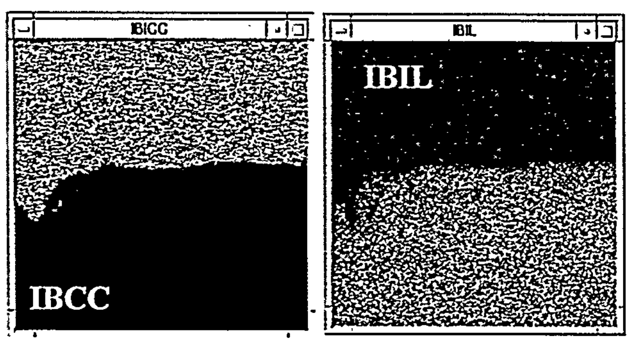

Figure 1. IBICC and IBIL images of a think phosphor over a pin-diode. 6.0 MeV alpha particle beam is applied in the study. The quantitative information of *ions/pixel* is extracted from the IBICC image area where the PIN-diode is not covered by the thick sample. Similarly, the quantitative information on *photons/pixel* is extracted from the IBIL image area where the PIN-diode is covered by the thick sample. The combination of the IBICC and IBIL information gives the quantitative information of the phosphor efficiency in terms of *photons/ions*.

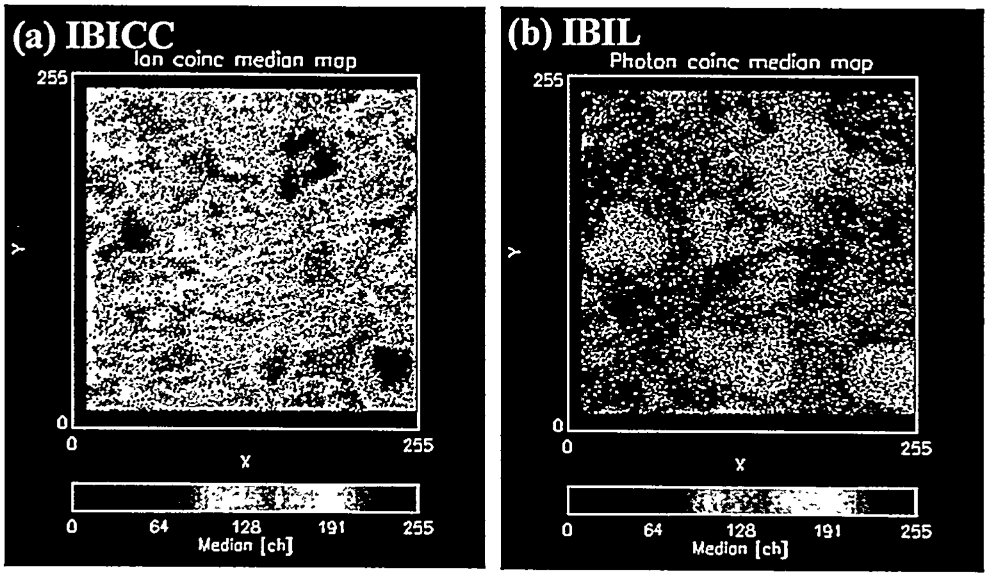

Figure 2. (a) IBICC (on the left side) and (b) IBIL (on the right side) images over the layer made by coating the inorganic ceramic Thiogallate (EuII) phosphor on a pin-diode. A 20 MeV carbon beam is used in the study. It is evident that the phosphor grains dominate the IBIL photon distribution in the sample. (data from Sandia National Laboratory).

alpha particle beam was scanned. The quantitative information of *ions/pixel* is extracted from the IBICC image area where the PIN-diode is not covered by the thick sample. Similarly, the quantitative information on *photons/pixel* is extracted from the IBIL image area where the PIN-diode is covered by the thick sample. Therefore, by combining this information, the IBIL efficiency in term of *photons/ion* can be easily derived. The investigation on the plastic scintillation materials, such as the Bicron samples: BC 400, BC 404, BC-408 and BC 430, indicates the relative low IBIL efficiency ~ 70 photons/ion/micron [3]. The image of IBIL from the organic material usually shows a homogenous distribution of the IBIL yield. The high homogeneity of the IBIL emission is essential for the successful IPEM application.

The investigation of the homogeneity of the IBIL emission in inorganic phosphors

The mechanism of luminescence in inorganic materials can be explained in semiconductor band structure theory [5] or crystal field theory [6]. It may be relatively easy to make thin films by coating a device with a fine powder of efficient luminescent materials; but it can not be used for the IPEM applications if there is a variation in mass density. The variation of the mass density in the thin film can lead to a large variation of the IBIL homogeneity. Figure 2 displays the IBICC (Figure 2a) and IBIL (Figure 2b) images over the layer made by coating the inorganic ceramic Thiogallate (EuII) phosphor on a pin-diode. A 20 MeV carbon beam is scanned over the sample. It is evident that the phosphor grains dominate the IBIL photon distribution in the sample. A further effort will be made to fabricate this type of phosphors thin films with a large improvement of the homogeneity.

The other causes of the IBIL variation in samples may not be associated with the variation of the mass density, but instead, for instance, related to internal crystal defects [7], even though the crystal may appear to be very smooth. Figure 3a reveals the line dislocation defects in the IBIL image produced with a broad 2 MeV alpha particle beam excitation in zircon, which is a large-band-gap crystal. The IBIL image was captured with a video system attached to the optical microscope. The size of the "broad beam" applied in this analysis is about 200 by 160 microns. The structure of the crystal defects that may occur in various forms is usually invisible when investigated by an optical microscope with reflected light, the broad beam IBIL image method can be easily used to screen the inorganic crystals for homogeneous thin films for the IPEM applications.

In Figure 3b, GaN (Mg doped) made by an epitaxial growth on an aluminum oxide substrate yields a bright green and homogeneous IBIL emission distribution when 2.0 MeV alpha particle beam with a partially focused beam size of 100 microns is applied. The high homogeneity of the luminescence emission

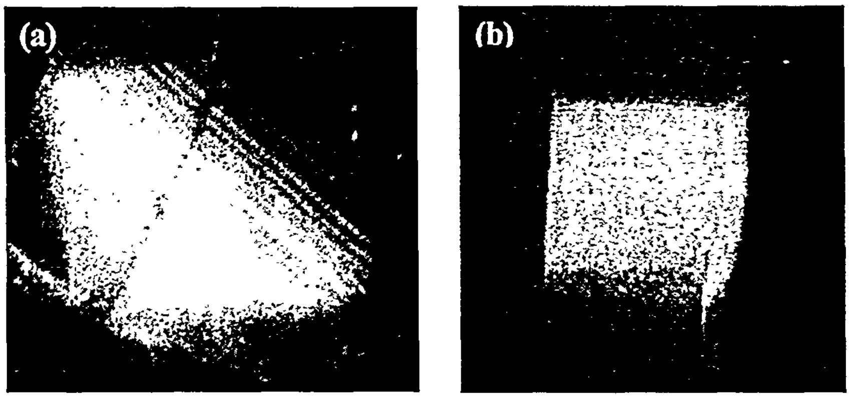

Figure 3. (a) The crystal dislocation defects in a zircon mineral are revealed by the IBIL image using partially focused beam (200 x 160 microns); (b) Smooth IBIL distribution in an epitaxial thin film made of GaN (Mg-doped). The thin film yields a bright green IBIL emission. Both IBIL images are excited with a 2 MeV alpha particle beam.

distribution, as revealed in the IBIL image, makes it a good candidate thin film for the IPEM application.

Conclusion

Plastic phosphors have the advantage of being relatively easy to produce homogeneous luminescent layers over some inorganic materials. The disadvantage is that the emission yield is relatively low. The challenging problem of inorganic phosphors for the IPEM application is the inhomogeneous distribution of the IBIL emission due to crystal defects or variation of mass density. The IBIL and IBICC with the nuclear microprobe can be applied to quantify the IBIL efficiency and study the homogeneity of the IBIL emission in the search for the candidate films suitable for the IPEM application.

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