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Observation of changes in ion beam induced luminescence spectra from organics during focused microbeam irradiation

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ABSTRACT

Continuous measurement of ion beam induced luminescence (IBIL) spectra was demonstrated with organic targets of nicotinamide adenine dinucleotide (NADH), tryptophan, riboflavin, and a polycyclic aromatic hydrocarbon (PAH), which are typically used as markers of biological contaminants in airborne particles. A 3 MeV external proton microbeam from a single-ended accelerator at QST/Takasaki was used to probe for changes in the IBIL spectrum using micro-optics sharing a focal point with the microprobe. We find that the decay of IBIL spectra from NADH and riboflavin varied by target organic species. Moreover, new peaks in the IBIL spectrum were recorded by continuous IBIL spectroscopy from the PAH target after destruction of a peak originally obtained in the initial measurement. These results suggest that IBIL monitoring can detect changes in the chemical composition of organics under focused beam irradiation.

1. Introduction

Among the various ion beam analysis techniques, ion beam induced luminescence (IBIL) analysis is sensitive to the chemical composition of various targets [1,2]. The range of analytical targets for IBIL is expanding [3–6], because IBIL can be used in conjunction with other ion beam analysis techniques such as particle-induced X-ray emission (PIXE), particle-induced γ-ray emission (PIGE), and Rutherford backscattering spectroscopy (RBS) [7,8]. IBIL is sensitive to chemical composition [9–11] and also the radiation damage occurred in inorganic target therefore it gives additional information to the other elemental composition analysis techniques [12–14]. In addition, IBIL using a microbeam probe can also be combined with micro-PIXE analysis as a complementary technique for the characterization of chemical composition of targets [15–17]. Previous studies developed a microscopic spectroscopy system using micro-optics for high-sensitivity IBIL detection [18]. Although there is limited information related to IBIL spectroscopy, several peaks have appeared in the IBIL spectra from standard targets in agreement with luminescence database information obtained by other excitation probes [19,20]. Through such micro-optics, IBIL spectra from several organic standards used for the recognition of biological aerosols have been successfully recorded and identified. IBIL analysis has also been utilized to characterize individual environmental aerosols [17,21]. For some particulate targets, strong IBIL emissions at wavelengths from 400 to 700 nm that do not correspond to the elemental composition of the inorganic component of the aerosols have been observed [22]. These IBIL spectra have broad peaks with peak wavelengths similar to a subset of atmospheric aerosols known as primary biological aerosol particles (PBAP) that bear viruses, bacteria, and living or dead microorganisms, including fragments of larger plants and animals [23–26]. Comparing IBIL with organic standards suggests that IBIL from these particulate targets may arise from surface organic contaminants [22]. There are also supporting results of IBIL analysis in which obtained different luminescent properties were obtained from road dust samples [27]. These preliminary experimental results expand the potential application range of IBIL to organic target characterization in complementary conditions for general micro-PIXE analysis; PIXE is assumed to be a non-destructive analysis technique, despite the potential for ion beam impacts to alter the chemical state of targets, particularly in organic compounds. These changes could be monitored by observing the structure of the IBIL spectra, and photons in the energy range of several electronvolts may be sensitive to changes in the

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chemical composition of targets. However, there remains insufficient knowledge about expected changes in the IBIL spectra from organics to determine their origin. The irradiation effects occurring in organics under intense beam irradiation are also unknown. From these motivations, we evaluated several standard organic materials by IBIL under external microbeam irradiation [28]. Continuous observation of IBIL spectra is desirable for evaluating these features of IBIL analysis, particularly for organic materials.

In this study, we demonstrate continuous IBIL spectroscopy with a 3 MeV external microbeam probe. A high-sensitivity spectrometer was used to continuously record IBIL spectra from several inorganic microscopic targets and from standard organics, namely, nicotinamide adenine dinucleotide (NADH), tryptophan, riboflavin, and the polycyclic aromatic hydrocarbon (PAH) benzo[*a*]pyrene. Decay and changes in IBIL structures were observed for these organic and microscopic targets. Their decay in particular may provide additional information related to the chemical structures of the target materials, which is not obtained by conventional ion beam analysis techniques.

2. Materials and methods

2.1. Experimental setup for continuous IBIL spectroscopy using microbeam

Fig. 1 shows a schematic illustration of the experimental setup for continuous IBIL spectroscopy. The external proton microbeam setup for in-air micro-PIXE analysis [29,30] on a microbeam line of the single-ended accelerator facility at TARRI/JAEA [31] was used for the IBIL analysis. A focused microbeam with a typical diameter of approximately 1 μm [30] was evaluated using a secondary electron image of copper mesh. Voltages applied to beam scanning electrodes were controlled by external digital to analog (D/A) converter installed in a computer. For IBIL imaging, beam scan was performed with maximum beam scanning area of 800 μm × 800 μm. In addition to the uniform scanning scheme, scanning command following binary data of desired structure image could be acceptable for beam scanning with any desired patterns. Beam extraction window with sample holder was placed at the end terminal of an external proton microbeam setup [29–31]. A Faraday cup was placed behind the sample to observe the beam current. A beam current of less than 100 pA was used for IBIL spectroscopy in conjunction with micro-PIXE analysis.

IBIL spectroscopy was performed under the same irradiation conditions using an electrically cooled back-thinned charge-coupled device (CCD) spectrometer (Solid Lambda CCD, Spectra Co., Ltd.) with an effective wavelength of 200–1000 nm and affected by optics. The reliable wavelength range was therefore limited to approximately 300–900 nm [32]. The system also contained specially designed confocal micro-optics for IBIL to accomplish high detection efficiency at the focal plane of the scanning proton microbeam [18]. Each optical lens had an anti-reflection coating to increase the overall optical throughput of the micro-optics. The lens had an effective focal point at the focal point of the microbeam at the target position. The effective region of

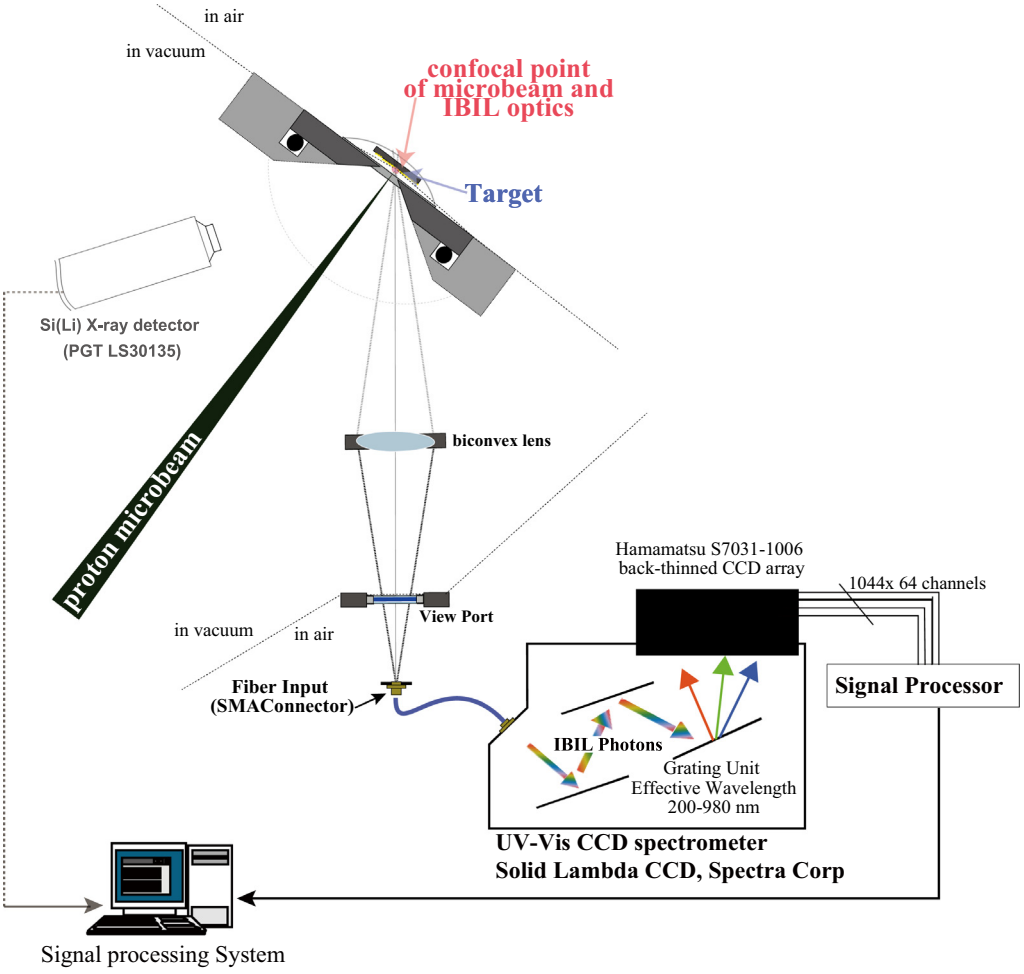

Fig. 1. Schematic illustration of the confocal setup for continuous IBIL spectroscopy. An electrically cooled CCD spectrometer was connected to confocal optics for IBIL spectroscopy using a 3 MeV external proton microbeam. Regions of interest for micro-optics were shared with a maximum beam-scanning area of Φ800 μm.

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