Low Energy Ga Ion Beams Bombardment onside Insulating Materials and X-ray Emission

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Abstract. X-ray spectra from the surface of insulating samples were measured using a 30 kV Ga+ focused ion beam instrument equipped with an energy dispersive X-ray spectrometer, showing an intense emission of low energy X-rays from insulating materials such as Al-K and O-K of Al2O3. This low energy ion induced X-ray emissions (LIIXE) from insulating materials is expected as a new analytical technique for light elements in insulating samples. It was found that X-ray yield induced by Ga+ ion beam irradiation depends on the incident beam energy. It was also found that high voltage of a secondary electron detector located near the sample strongly influences the intensity of X-ray emission, implying that bombardment of electrons, which may come from the chamber wall, onside the sample surface is a key phenomenon for the X-ray emissions.

1. Introduction

Recently, we have reported a phenomenon that characteristic soft X-rays are induced from insulator targets by irradiation of positive light ions with the energies ranging from several keV to 100 keV [1]. We call it as low-energy ion induced X-ray emission (LIIXE). This would be different from PIXE (particle induced X-ray emission). The most remarkable feature of the LIIXE from the insulating target is that intense emission of low energy X-rays can be obtained, while a small intensity emission of X-ray from metallic materials under the low energy Ga+ ion irradiation has been reported [5]. Conventional PIXE techniques need ions with primary energy of several millions of voltages to bombard the surface of the target samples especially in case of use of light gas ions such as proton and helium [2]. It has been reported that characteristic X-ray signals in the PIXE could be enhanced if the samples are of insulating [3]. Disadvantage of the PIXE is that use of MeV ion source requires large and expensive facilities.

Terasawa has found that an abnormal X-ray emission from Al2O3 and SiO2 insulators can happen under irradiation of protons even when their energy ranges from 10 to 120 keV [4]. Intense emission of characteristic X-ray with low energy may make it possible to detect a small amount of light elements in insulating and non-conducting organic materials sensitively. Thus, we have been now attempting to develop LIIXE to a new analytical tool for light elements in insulating and organic materials. For that purposes, comprehensive studies of LIIXE phenomena, features and capability is necessary. In this paper, we investigate the dependency of LIIXE on the primary ion energy and the target materials. We assume that the key mechanism of the enhancement of LIIXE signals was...
attributed to the bombardment of electrons on the target surface. To confirm it, influence of a high voltage of a secondary electron detector located near the sample on LIIXE spectra is observed.

2. Experimental procedure

The LIIXE instrument was constructed based on a focused-ion-beam (FIB) machine, JEOL JEM-9130-FIB. An energy dispersive X-ray spectrometer (EDS), JEOL EX-2300BU, equipped with a Si-Li detector is attached to the specimen chamber of the instrument with a take-off angle of 40°. Ga+ ions were irradiated on insulating samples in a direction normal to the sample surfaces. Non-doped Al2O3 and SiO2 were used as insulating samples. Characteristic X-ray emission from them was measured using focused Ga+ ion beams with primary energy from 5 to 30 keV to investigate the features of X-ray signals of light elements (Al, Si, O) with K-shell electron excitation energy, and the influence of the irradiation beam energy on the X-ray yield was systematically examined on the same condition of the total dose and the beam current (1 nA and 0.4 nA were used). The pressure in specimen chamber of the FIB machine is less than 1×10⁻³ Pa.

3. Results and discussion

Prior to the experiment, it was confirmed that the X-ray yield was independent of the vacuum pressure in range of 1×10⁻³ to 1×10⁻⁴ Pa.

Figure 1 shows X-ray spectra from Al with a disk shape of 3 mm in diameter and 0.3 mm in thickness (a) and Al2O3 with a square plate of 6×6 mm² and thickness is 0.3 mm (b) taken by 30 keV Ga+ beam irradiation. The beam current and acquisition time for the former were 6 nA and 1200 sec and those for the latter were 1 nA and 50 sec. It is noted that the low energy Ga ion irradiation onto the metal Al target resulted in a small intensity emission of X-ray of Al-K. This result is consistent with the spectra reported in ref [5] although the mechanism of emission of such characteristic X-ray from the metal by irradiation of low energy ions has been in argument. In contrast, a remarkably intense emission of low energy X-rays (O-K and Al-K) from the insulating Al2O3 is observed under irradiation of the 30 keV Ga+ beam. We confirmed the reproducibility of this LIXXE phenomenon for various insulating materials such as SiO2, MgO and more other specimens. Figure 2 shows X-ray spectra from SiO2 taken by irradiation of a 30 keV, 1 nA Ga beam. The total dose was 5×10⁻⁸ C.

Figure 1 X-ray spectra was taken by 30 keV Ga+ beam irradiation from (a) Al with a disk shape of 3 mm in diameter and 0.3 mm in thickness; and (b) Al2O3 with a square plate, size is 6×6 mm² and thickness is 0.3 mm. The total doses for (a) and (b) were 7.2×10⁻⁶ C and 5×10⁻⁸ C, respectively.

Figure 2 X-ray spectra from SiO2 taken by irradiation of focused Ga+ ion beams of 30 keV in energy and 1 nA in current. The total dose was 5×10⁻⁸ C.
focused Ga$^+$ ion beam of 30 keV in energy and 1 nA in current (e.g., the same condition as spectra shown in Fig. 1(b)). Si-K peak at 1.74 keV and O-K peak at 0.52 keV were successfully detected.

Figure 3 shows the dependency of the X-ray yield on the incident Ga ion energy for Al$_2$O$_3$ (a) and SiO$_2$ (b) samples. Samples were square plates with the size of 6×6 mm$^2$ and 0.3 mm in thickness. The total doses for 1 nA ion beam and 0.25 nA ion beam were 5×10$^{-8}$ C and 1.25×10$^{-8}$ C, respectively. X-ray signals are plotted for different ion energy of 10, 20 and 30 keV (in case of SiO$_2$, 5 keV ions were also employed). From Fig. 3, it was found that X-ray yield induced by Ga ion beam irradiation was independent of the beam current (the total dose was controlled to be constant, so that our data is comparable). But the yield enhancement was associated with the primary beam energy.

The LIIXE strongly relates to the effect of surface charge, because the intensity of X-rays is enhanced if the thickness, area and resistivity of specimen increase, i.e., the environment to generating surface charge was enlarged. Thus, we infer that the key mechanism of the enhancement of LIIXE signals could be reasonably attributed to the bombardment of electrons on the target surface. Namely, ion and neutron particles were emitted out from the sample surface by the primary ion beam irradiation, which might bombard the vacuum chamber wall directly and result in the formation of secondary electrons. These secondary electrons with low energy are collected to high potential positions inside the chamber. Strongly charged sample is on a high potential, which might be the same level (higher than several kV) as that of a secondary electron detector attached to scanning electron microscope and FIB instruments. Finally, secondary electrons from the chamber wall are accelerated by such high potential and then attack the sample surfaces, leading to the emission of X-rays. If this assumption is true, LIIXE signals would be influenced by a high voltage of a secondary electron detector located near the charged sample (e.g., electrons would be collected to both the charged sample and detector, in case the high voltage of detector is on).

To confirm this, intensity change of the X-ray signals detected by EDS was monitored when the high voltage (10 kV) of the secondary electron detector was turned on and off. Figure 4 shows the dependency of X-ray spectra on turning on or off the high voltage of the secondary electron detector. Turning off high voltage of the secondary electron detector clearly enhanced the LIIXE signals of both O-K and Si-K. Peak intensity increases more than twice, via accumulating the integral area covered by each peak. This means that approximately half amount

![Figure 3](image-url)  
**Figure 3** Dependency of X-ray yield on the incident Ga ion energy for Al$_2$O$_3$ (a) and SiO$_2$ (b) samples. Samples are square plates of 6×6 mm$^2$ and 0.3 mm in thickness. The total doses for 1 nA beam and 0.25 nA beam were 5×10$^{-8}$ C and 1.25×10$^{-8}$ C, respectively.

![Figure 4](image-url)  
**Figure 4** The change of X-ray spectra from SiO$_2$ between the high voltage of the secondary electron detector on and off.
of the electrons reflected from the chamber wall were collected to the detector and the others could come to the charged sample surface. It is implied that the potential of the sample surface was similar to that of the detector (10 kV) in the present case.

4. Summary

X-ray spectra from both metallic Al and insulating Al₂O₃ samples were measured under irradiation of a 30 kV Ga⁺ focused ion beam with an energy dispersive X-ray spectrometer (EDS), and strong enhancement of low energy X-rays such as Al-K and O-K was observed from Al₂O₃ but not from metallic Al. X-ray yield induced by low energy Ga⁺ ions was independent of the beam current (if the total dose was the same) but increases depending on the beam energy. The change of X-ray signals from SiO₂ between the high voltage of the secondary electron detector on and off suggested that electrons emitted from the chamber wall are accelerated by the high potential of the sample surface and then attack to the sample, followed by X-ray emission. To understand the mechanism of this phenomenon, the more detailed investigation would be necessary.

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