Preliminary Experiment of Total Reflection X-Ray Fluorescence Using Two Glancing X-Ray Beams Excitation

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Preliminary experiment of total reflection x-ray fluorescence using two glancing x-ray beams excitation

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In conventional total reflection x-ray fluorescence (TXRF), one glancing x-ray beam irradiates the sample carrier for excitation of x-ray fluorescence. However, it is also possible to excite x-ray fluorescence by multiple x-ray beams. We performed a preliminary TXRF experiment excited by two independent glancing x-ray beams. The two x-ray beams excitation would be effective to enhance the intensity of x-ray fluorescence and to improve the detection limit in TXRF. X rays from both a rotating Mo anode x-ray tube and a glow discharge x-ray tube with an Fe target irradiated a Cr thin film sample on a quartz glass. The x-ray fluorescence of Cr $K\alpha$ from the sample was slightly enhanced compared with the intensity measured by one x-ray beam excitation. © *1999 American Institute of Physics.* [S0034-6748(99)01103-X]

I. INTRODUCTION

Nowadays, total reflection x-ray fluorescence (TXRF) is recognized as one of the powerful trace elemental analytical methods.[1,2] Since the TXRF method utilizes the x-ray total reflection phenomenon, an optically flat material is used as a sample support, such as Si. There are many TXRF applications on the analysis of silicon wafers. The commercially available TXRF instruments have a good detection limit at the ppt level.[3] However, a lower detection limit has been required. One of the ways to improve the detection limit of TXRF analysis is to use more powerful x-ray sources, such as a rotating anode x-ray generator and synchrotron radiation.[4,5] Synchrotron radiation is a promising source; however, availability is one of the problems.

We propose another approach to enhance the x-ray fluorescence intensity of laboratory TXRF analysis. Normally, one glancing x-ray beam irradiates the sample carrier for the excitation of x-ray fluorescence. However, it is also possible to excite x-ray fluorescence by multiple x-ray beams. The idea of two x-ray beam excitation TXRF is illustrated in Fig. 1. This new TXRF arrangement was considered from analogy of ''the glancing-incidence and glancing-takeoff x-ray fluorescence (GIT-XRF) arrangement.''[6] In the GIT-XRF method, the primary x rays irradiate the sample surface at the glancing incident angle and the x-ray fluorescence is also detected at the glancing exit angle. By changing the path of the measured x-ray fluorescence in the GIT-XRF arrangement with another path of the new irradiating x-ray beam, the new arrangement, (illustrated in Fig. 1), is completed.

Moreover, Tsuji *et al.* have actually attempted ''x-ray fluorescence analysis by using multiple-glancing x-ray beams as an excitation,''[7,8] where the primary x-ray beams irradiate the sample surface from many directions and excite the x-ray fluorescence. As a result, it was concluded that x-ray irradiation from many directions using multiple-x-ray beams is effective to increase the x-ray fluorescence intensity,[7] which would promise highly-sensitive detection of trace elements. Of course, it is also possible to irradiate the sample with the two x-ray beams of different energies. Normally, Mo $K\alpha$ is used for excitation of x-ray fluorescence; however, it is not suitable for the excitation of low-Z elements. The x rays of W $L\beta$ are more effective for excitation of these elements.[9] In the conventional TXRF instrument, we have to change the x-ray target depending on the analyzing element. If both Mo $K\alpha$ and W $L\beta$ x rays simultaneously irradiate the sample surface, the simultaneous trace analysis of multiple elements would be achieved. In this study, we performed preliminary experiments of TXRF excited by two independent x-ray beams, which cross each other on the sample carrier as shown in Fig. 1.

II. EXPERIMENTAL DETAILS

Figures 2(a) and 2(b) show the illustration of an experimental instrument viewed from two directions. One glancing x-ray beam-1 is from a rotating anode x-ray generator using a Mo anode (Rigaku, RU-200, Japan; tube voltage: 40 kV, tube current: 100 mA). The other (x-ray beam-2) is from a glow discharge x-ray tube (GDXT) with a Fe target,[10] which is a Grimm-type glow discharge tube normally used in optical emission analysis. Details of the GDXT have been described elsewhere.[10,11] We applied high voltages of up to 10

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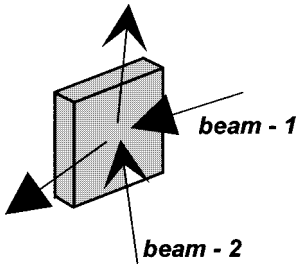

FIG. 1. The idea of TXRF using two glancing x-ray beams.

kV to this GDXT. The electrons, emitted from a cathode surface, bombard the x-ray target, and then x rays pass through a Be window. The GDXT was operated at a discharge voltage of 10 kV, at a discharge current of 14 mA and at a He gas pressure of 0.5 Torr. The distance between the sample and the Mo anode was 300 mm. The x-rays from the rotating anode were collimated with two perpendicular slits of 0.3 and 0.1 mm in width. The distance between the sample and the Be window of the GDXT was 200 mm. The x rays from the GDXT were also collimated with two slits of 0.2 and 0.3 mm in width. Both x-ray beams were not monochromatized in this work.

The sample carrier of quartz glass ($20\times20\,\mathrm{mm}^2$) was perpendicularly attached on a goniometor head, which had two cross rotations and two cross-linear movements. Two incident angles were manually adjusted with this goniometer head. The x-ray fluorescence was detected with a Si(Li)

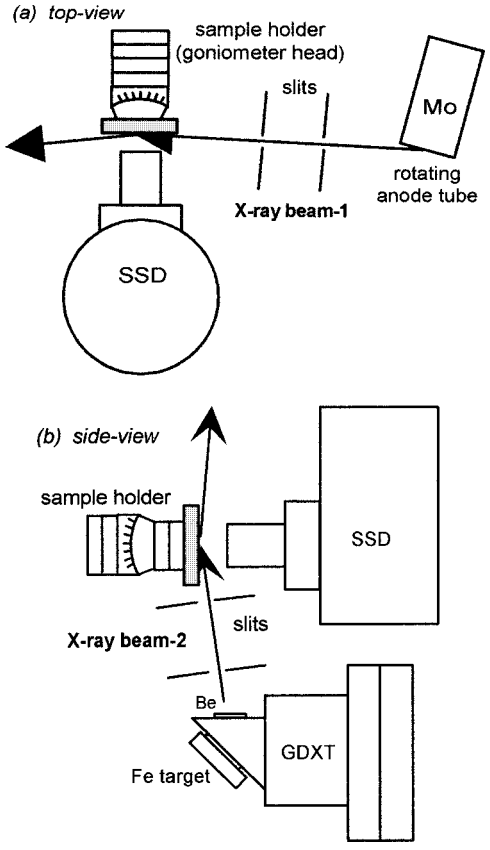

FIG. 2. Illustration of the experimental setup of TXRF apparatus excited by both x rays from a rotating anode (a) and x rays from GDXT (b): (a) top view, (b) side view.

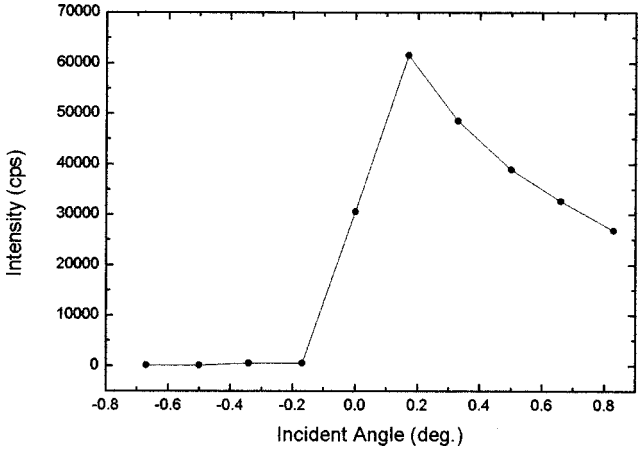

FIG. 3. X-ray intensity of Cr $K\alpha$ as a function of the incident angle.

x-ray detector (Horiba, Japan), which was set in front of the sample carrier at a distance of 10 mm. The sample used was a Cr thin film, which was deposited in a thickness of 40 nm on a quartz glass by a vacuum-evaporation method.

III. RESULTS

Figure 3 shows the detected x-ray intensity of Cr $K\alpha$ as a function of the incident angle. Only the x rays from the rotating Mo anode were used as excitation in this measurement. The Cr $K\alpha$ x-ray intensity showed a peak at approximately 0.18°, which is the critical angle of Mo $K\alpha$ (17.4 keV) on the Cr film. Then, the intensity of the x-ray fluorescence gradually decreased with an increase in the incident angle. The result shown in Fig. 4 is a normal feature in TXRF.

Figure 4 shows a typical x-ray spectrum excited by two x-ray beams emitted from both the rotating anode x-ray tube and the GDXT. The area intensity of Cr $K\alpha$ was 62 300 counts for a counting time of 10 s. Compared with the area intensity that was measured by using only x-rays from the rotating anode tube, it increased by approximately 1000 counts, which corresponds to approximately 1.6% of the total intensity. Since the x rays from GDXT were not originally strong enough, only the difference of 1000 counts could be observed in this preliminary experiment. However,

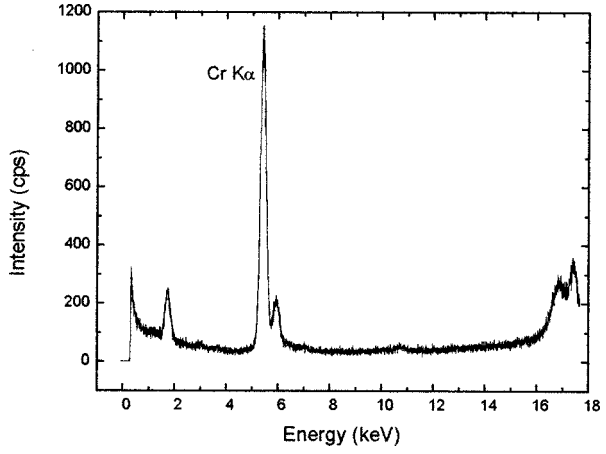

FIG. 4. Typical x-ray spectrum excited by two x-ray beams.

we believe that the excitation using two powerful glancing x-ray beams is effective to increase the x-ray fluorescence and to improve the detection limit in TXRF. We will undertake more strict experiment as a next step using two powerful x-ray sources.

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