Enhancement of TXRF intensity by using a reflector

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We investigated the possibility of enhancing the TXRF intensity by applying an Si reflector. This Si reflector was attached just above the Si sample carrier. The angle between the reflector and the sample carrier was changed by tilting the reflector. We attempted to focus the primary x-rays on the analyzed area by multi-reflection effects between the reflector and the sample carrier. The intensity of Au L\(_{\alpha}\) emitted from a thin layer of Au deposited on an Si sample carrier, was measured as a function of the angle between two Si plates. The observed peaks were explained from simple calculations, suggesting that the reflection of primary x-rays on both the reflector and the sample carrier results in a significant increase in x-ray intensities. In addition, Ar K\(_{\alpha}\) emitted from the air, was reduced substantially by applying the reflector. This indicates that a reflector is also useful for reducing the x-ray background that originates from the air and the sample carrier.

INTRODUCTION

Total reflection x-ray fluorescence (TXRF) is a powerful tool for surface analysis of contaminants on Si wafers. Since the idea of TXRF was first proposed by Yoneda and Horiuchi, many researchers have studied TXRF instruments to improve detection limits. For this purpose, important questions are how to enhance the TXRF intensity and how to reduce the background x-ray intensity. To enhance the TXRF intensity, of course, it is effective to use more powerful x-ray sources such as synchrotron radiation with suitable energy for the analyte.

In this work, we attempted to enhance the TXRF intensity by using an Si reflector. In micro-XRF, capillary optics, such as x-ray poly-capillaries, are used to obtain strong micro-focused x-rays. The primary x-rays are totally reflected on the inner surface of the capillary optics, and are focused to form a narrow, micrometer scale, probe. A similar approach would be useful for the TXRF optics. That is, if the primary x-rays are focused on the analyzed sample area under total reflection conditions, the TXRF intensity would be enhanced, leading to lower detection limits.

The arrangement of our reflector is similar to the 'double-plate sample carrier' suggested by Cheburkin and Shoytk. They used a double-plate sample carrier as a simple TXRF analyzer. Their double-plate sample carrier consists of two glass slides of different length. A sample was deposited on the longer glass plate and another glass plate was placed just above the first. The distance between the two glass plates was about 50 \(\mu\)m. This double-plate sample carrier played the roles of monochromator and collimator. Wobrauschek et al. reported on a double-reflector collimator for TXRF. Normally, this double-reflector is placed between the x-ray source and the sample, and collimated monochromatic x-rays irradiate the surface of the sample. In the case of a double-plate sample carrier, a double-reflector collimator and a sample carrier are combined. Cheburkin and Shoytk emphasized the simplicity of the instrumental design of the double-plate sample carrier, and reported low detection limits for TXRF. Egorov et al. proposed the same idea. They called it 'slitless collimator,' which was formed by two quartz plates mated together. Sanchez et al. also reported low detection limit for TXRF using a double-plate sample carrier.

In each case, two glass plates were attached parallel to each other and the distance between the two plates was fixed. However, we considered that the optimum experimental arrangement would be obtained by adjusting the angle and the distance between the two plates. For this purpose, we developed a simple reflector, which can be tilted to change the angle between the reflector and the fixed sample carrier. Furthermore, we attempted to focus the primary x-rays on the analyzed sample area by multi-reflection between the Si reflector and the Si sample carrier. In this paper, preliminary experimental results obtained using this reflector are presented.

EXPERIMENTAL

A circular thin layer of Au (5 or 10 \(\mu\)m in thickness) was deposited with a diameter of 10 mm on an Si sample carrier (20 \(\times\) 70 mm), as shown in Fig. 1(a). Au Si wafer (30 \(\times\) 80 mm) was pasted on a Cu holder, as shown in Fig. 1(b). This reflector was attached to the Si sample carrier and a vertically moving linear stage (Z-stage), as shown in Fig. 2. The angle (\(\theta\)) between the Si sample carrier and the Si reflector could be changed with the Z-stage, which was driven by a stepping motor (0.5 \(\mu\)m per pulse). Actually, two bars attached to the Z-stage pushed the reflector. The primary x-rays irradiated the sample between these two bars. The distance between the sample carrier and the reflector was...
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Figure 1. (a) Si sample carrier. A thin layer of Au was deposited in a circle with a diameter of 10 mm. (b) Si reflector.

Figure 2. Experimental arrangement of the reflector and the sample carrier.

The TXRF instrument consisted of an anode rotating x-ray generator (RU-200, Rigaku, Japan), a W/C multilayer monochromator and a large goniometer to adjust the incident angle of the primary x-rays. A detailed description of the large goniometer is given elsewhere. Figure 3 shows a schematic overview of this TXRF instrument with the reflector. The x-ray generator was operated with an Mo anode at a tube voltage of 30 kV and a tube current of 90 mA. The primary x-rays, after passing through a slit (0.3 mm in width) placed between the W/C monochromator and the sample, irradiated the sample surface at glancing angles.

RESULTS AND DISCUSSION

The x-ray fluorescence intensities of Au L_α and Si K_α emitted from the Au thin layer (10 nm) on Si were measured as a function of the incident angle of x-rays (\(\phi\)) without the reflector. As shown in Fig. 4, a clear angle dependence was observed. Here, at large incident angles, the Si data plots include the Si K_α x-rays emitted from the Si substrate.
angle dependence of Si K\(\alpha\) and Au L\(\alpha\) at different values of the angle \(\theta\): (a) 0.065°, (b) 0.115° and (c) 0.215°.

underneath the Au layer in addition to the Si K\(\alpha\) x-rays emitted by the Si substrate without the Au layer.

Next, the reflector was attached to the sample and the Z-stage. At fixed different incident angles \(\theta\), the x-ray fluorescence intensities of Au L\(\alpha\) and Si K\(\alpha\) were measured as a function of the reflector angle \(\phi\).

The experimental results obtained at the incident angle values of 0.065, 0.115 and 0.215° are shown in Fig. 5(a), (b) and (c), respectively. These results were obtained using the Al foil spacer (0.03 mm). Initially, x-ray measurements were performed without the Al foil spacer; however, the intensity of x-ray fluorescence was weak, and no clear angle dependence could be obtained. Therefore, the Al foil space was applied to increase the primary x-ray beam. As shown in Fig. 5, a maximum was observed in the angle dependence plots. The angle \(\theta\) at which this maximum occurs depends on the incident angle \(\phi\). The angle corresponding to this maximum in the case of incident angles of 0.065 and 0.115° were 0.16 and 0.22°, respectively. At a large incident angle of 0.215°, it was a broad peak.

We can explain these results using the schematic overview shown in Fig. 6. If the angle \(c\) between the reflector and the x-ray beam is less than the critical angle of total reflection, which is approximately 0.1° for Mo K\(\alpha\) on Si, these x-rays are totally reflected on the surface of the reflector. In this case, the following simple relation is obtained:

\[
c + \theta = 2c + \phi \tag{1}
\]

For the incident angle values \(\phi\) of 0.065, 0.115 and 0.215 and a value for angle \(c\) of 0.1°, the reflector angles \(\theta\) are 0.165, 0.225 and 0.315°, respectively. These calculated values are very close to the measured angles at each peak in Fig. 5. This supports the assumption that the reflected x-rays contribute to the production of the peaks in Fig. 5. If the reflector angle \(\theta\) is reduced much more, the reflector cuts off the primary x-ray beam. In contrast, if the angle \(\theta\) is more than 0.1°, the primary x-rays are absorbed by the reflector. Therefore, a maximum would be observed close to 0.1°. The incident angle \(d\) at which the reflected x-rays irradiate the sample (cf. Fig. 6), can be simply calculated as \(2c + \phi\). In the case where \(c = 0.1°\) and \(\phi = 0.065°\), this incident angle is 0.265°, which is larger than the critical angle of total reflection. Here, we assume that
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From this relation and Eqn (1), the equation \( 2c + \phi = 0.1 \) is obtained. Therefore, when case \( \phi = 0.06^\circ \), \( \theta \) becomes \( 0.0825^\circ \). This indicates that when \( \theta \) is less than \( 0.0825^\circ \), the reflected x-rays are again totally reflected on the sample surface. However, such x-rays were not focused on the sample area, because, in our case, the reflector was placed at a height of 0.03 mm from the sample. Hence the dimensions of the experimental setup should be considered carefully. Anyhow, the idea of focusing the primary x-rays on the sample position under total reflection conditions would be useful for enhancing the x-ray fluorescence intensity.

Figure 7(a) shows an x-ray spectrum recorded at an incident angle \( \phi = 0.06^\circ \) without the reflector. In addition to the characteristic x-ray lines of Au and Si, a strong Ar K\( \alpha \) peak, emitted from air, can be observed. Figure 7(b) is an x-ray spectrum at the same incident angle \( \phi = 0.06^\circ \) and a reflector angle \( \theta = 0.16^\circ \). Compared with the spectrum in Fig. 7(a), the intensity of Si K\( \alpha \) is increased considerably. This is probably because the incident angle of the reflected x-rays was higher than the critical angle, as mentioned before. In addition, Ar K\( \alpha \) was reduced substantially by applying the reflector. This indicates that the reflector is also useful for reducing the x-ray background that originates from air and the sample carrier. That is, the reflector also plays a role of ‘shutter’ for x-rays that is not needed.

Next, another Au thin layer of 5 nm thickness was measured. This Au layer was deposited on an Si substrate in exactly same way as the previous one (the diameter of the Au film was 10 mm). After the incident angle \( \phi \) dependence was determined for the intensities of Au L\( \alpha \) and Si K\( \alpha \), the reflection angle \( \theta \) dependence was investigated. Figure 8 shows the intensities of Au L\( \alpha \) and Si K\( \alpha \) as a function of the reflection angle \( \theta \) at an incident angle \( \phi = 0.04^\circ \). In this case, two peaks were observed for Au L\( \alpha \) at 0.14° (first peak) and 0.06° (second peak). If in Eqn (1) the value of \( c \) is 0.1° and \( \phi = 0.04^\circ \), the reflector angle \( \theta \) is calculated to be 0.14°, which agreed well with the experimental angle of the second peak in Fig. 8. This second peak was not clear in Fig. 5(a) and (b), although a shoulder peak was observed at 0.23° in Fig. 5(c). This is probably because the first peak was very strong owing to the greater thickness (10 nm) of the Au layer, and this first peak would mask the second peak. As shown in Fig. 8, Si K\( \alpha \) intensities were also increased strongly at 0.14°, the same value at which the first peak was observed. However, at 0.06°, the angle at which the second peak was
observed, the Si Kα intensity was weak compared with that of the first peak. This indicates that the incident angle (d) of the reflected x-rays on the reflector was smaller for \( \theta = 0.06^\circ \). This condition would be suitable for the enhancement of the x-ray fluorescence intensities emitted from the sample on the flat substrate under total reflection conditions.

CONCLUSIONS

An Si reflector was attached to the Si sample carrier of a TXRF instrument. The angle \( \theta \) between the reflector and the sample carrier was changed by tilting the reflector. For a thin layer of Au deposited on the Si sample carrier, the Au Lα intensity was measured as a function of \( \theta \). The observed peaks were explained based on the simple calculations, which suggest that the reflection of the primary x-rays on both the reflector and the sample carrier result in significant increases in x-ray intensities. By applying a reflector, the primary x-rays are concentrated on the analyzed sample area. To achieve this focusing under total reflection conditions, it is important to optimize configurational parameters such as the angles \( \theta \) and \( \phi \), the distance between the reflector and the sample carrier, the shape of the reflector, etc. In addition, this type of x-ray optics, consisting of two reflectors, could be used to serve as a waveguide to produce a narrower, more intense x-ray beam (H. J. Sanchez, personal communication, 2001).

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REFERENCES

7. Egorov VK, Zhive AP, Kondratiev OS, Egorov EV. In Abstracts Book of the 8th TXRF 2000 Conference (Vienna, Austria), 2000; 47.