Dependence of Scattered Mn K_α/K_β X-ray Intensity Ratio on the Scatterer Materials

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The K_α/K_β intensity ratio of Mn KX-rays scattered by metallic samples changed remarkably with the geometry between the sample and the ^{55}Fe source-Si(Li) detector system. On the contrary, this intensity ratio changed little in the cases of non-metallic scatterer samples such as lucite or mylar. This difference is interpreted as due to the occurrence of strong or weak interference in the coherent scattering photons.

Key Words: coherent scattering, KX-ray intensity ratio

1. Introduction

In the elemental analysis using the photon-induced X-ray fluorescence method (XRF), the source X-rays scattered by the specimen are detected simultaneously with the characteristic X-rays from the sample. In the determination of unknown elemental concentration embedded in the foil or bulk, it is sometimes estimated from the observed ratio of the X-ray peak count of unknown elements induced by the primary photons to the scattered source X-ray peak count^{1),2)}. For photon energies less than about 10 keV, the scattered X-rays are dominantly resulted from the coherent scattering rather than incoherent scattering such as Compton scattering.

In the course of measurement using Mn K_α and K_β X-rays from ^{55}Fe source, we observed quite different K_α/K_β ratio as function of source-specimen distance. We report here the variations of K_α/K_β ratios for a variety of specimens and the reasons of these variations.

2. Experimental procedure

Experimental apparatus used is shown in Fig. 1. An ^{55}Fe annular source was used as primary X-ray emitter. ^{55}Fe decays through electron capture and emits 5.9 keV (wave length $\lambda=0.21$ nm ($2.1\text{\AA}$)) Mn K_α and 6.5 keV ($\lambda=0.19$ nm ($1.9\text{\AA}$)) Mn K_β X-rays. The K_α/K_β intensity

Fig. 1 Schematic view of experimental arrangement. A: Annular source of ^{55}Fe B: Target C: Collimator
Targets move relative to ^{55}Fe source-Si(Li) detector system. The relation between the scale reading r of the target position and the scattering angle 2θ is shown. The broadness of θ at a fixed r position is tabulated.

ratio is known to be 7.46³⁾.

Planar target materials of Al (1 mm thick), Cu (0.5 mm thick), Ta (0.5 mm thick), W (23 μm thick), blue plate glass (1.5 mm thick), lucite (1 mm thick) and mylar (188 μm thick) were used as scatterers.

Targets were moved along the central axis of an annular source-3 mm thick Si(Li) detector system. The angles 2θ subtended between the

direction of primary and scattered X-rays as function of scale reading r of the target position are shown in Fig. 1. The actual distance between the target and the detector is (scale reading $r+8.7+9.6$) mm, where the 9.6 mm is the distance between the surface of Si(Li) semiconductor and the beryllium window. A whole X-ray spectrum with a blue plate glass target is shown in Fig. 2.

Fig. 2 X-ray energy spectrum obtained with an ^{55}Fe source and a blue plate glass target, 1.5 mm thick. It consists of characteristic X-rays of target elements and scattered Mn KX-rays.

3. Results and discussion

Coherent scattered Mn K_α and K_β X-ray spectra via glass, lucite, Al and W are shown in Fig. 3 for different scale readings. In Fig. 4, after the correction of absorption of X-rays in air, of detection efficiency and of the variation of solid angles of the detector, K_α - K_β X-ray intensity ratios $I(K_\alpha)/I(K_\beta)$ for all the tested target materials are described as function of scale readings. In the figure, the normal K_α - K_β X-ray intensity ratio observed by the direct irradiation of ^{55}Fe source is also indicated. It is seen that the intensity ratios change remarkably with r for metallic scatters, whereas little for glass and organic materials. They are 2.5~76 for Al, 0.6~44 for Cu, 0.3~5.0 for W, 2.6~15 for Ta, 5.5~6.8 for lucite, 6.4~7.3 for glass and 7.0~8.5 for mylar for r between 5 and 15 mm. For glass and organic targets, $[I(K_\alpha)/I(K_\beta)]_{\text{max}}/[I(K_\alpha)/I(K_\beta)]_{\text{min}} \leq 1.4$.

The remarkable contrast between the data of variation of $I(K_\alpha)/I(K_\beta)$ with metallic scatterers and those with non-crystalline or polymer materials as function of r , i.e., θ (the value of

Fig. 3 Coherent scattered Mn K_α and K_β X-ray spectra for non-metallic (glass, lucite) and metallic (Al, W) targets are shown as function of target position (scale reading r defined in Fig. 1).

θ for each r is tabulated in Fig. 1), suggests that this effect be due to the crystal lattice.

According to the well known Laue diffraction, the intensity of scattered photon beam reaches the maximum when the relation

$$2d \sin \theta = \lambda \sqrt{h^2 + k^2 + l^2} \quad (1)$$

is satisfied. Here d is the lattice constant of the scatterer material of cubic unit lattice, h, k and l are integers, λ is the wave length of the primary photon, and θ the angle of reflection. Al and Cu are known to have fcc lattice structure with $d=0.405$ and 0.361 nm (4.05 and $3.61 \text{ \AA}$), respectively⁴⁾. Ta and W are of bcc structure with $d=0.330$ and 0.316 nm (3.30 and $3.16 \text{ \AA}$), res-

Fig. 4 K_α/K_β intensity ratio $I(K_\alpha)/I(K_\beta)$ of coherent scattered Mn K X-rays as function of scale reading r . Note the drastic change of intensity ratios for metallic targets with the variation of r .

pectively. For fcc lattice, the (hkl) reflection takes place if and only if h, k and l are all even or all odd. For bcc lattice, the (hkl) reflection takes place if and only if $h+k+l$ is even.

As was already described, the K_α or K_β X-ray spectra in Fig. 3 were drawn after correcting for the solid angle and the X-ray absorption through the air. Figure 3 indicates that the lowest Mn- K_α X-ray intensity for Al or W target is seen at the scale reading of 15 or 5 mm, respectively. The relative K_α X-ray intensity ratios at other scale readings with respect to such lowest K_α X-ray intensity are calculated and drawn in Figs. 5 (A) (fcc metals) and 6 (A) (bcc metals) in the plane $(\sin \theta, d)$. The relative intensities are classified according to their magnitude of 1~2, 2~4, 4~8 and greater than 8 times. The width of the zone for a fixed scale reading r is related to the broadness of angle θ , and has been determined from the tabulated relation of r and θ in Fig. 1. On the other hand, the lowest Mn- K_β X-ray intensity for Al or W is seen at the scale reading of 10 or 7.5 mm, respectively (see Fig. 3). In the same procedure as the case of K_α X-ray intensity ratio, the re-

Fig. 5 Relative intensity map of scattered Mn- K_α (Fig. 5(A)) and K_β (Fig. 5(B)) X-rays by Al and Cu targets with fcc structure. For each target position (scale reading r) hence scattering angle θ , relative intensity ratio with respect to the observed lowest intensity in Figs. 3 and 4 is classified according to their magnitude. d is the lattice constant of the target material. Solid lines correspond to eq. (1) with (hkl) lattice.

Fig. 6 Relative intensity map of scattered Mn- K_α (Fig. 6(A)) and K_β (Fig. 6(B)) X-rays by Ta and W targets with bcc structure.

lative intensity ratio of K_β X-rays for other scale readings are described in Figs. 5(B) (fcc metals) and 6(B) (bcc metals) in the plane $(\sin \theta, d)$. The solid hyperbolic lines in the figures give the relation (1) for several choices of (hkl) . Diffraction occurs at the angles where the hyperbolic curves given by eq. (1) cross the horizontal lines corresponding to the lattice constants of metallic elements.

From Figs. 5 and 6, it is shown that the observed r —hence θ —dependence of scattered K_α/K_β intensities follows the eq. (1). For example, the highest K_α/K_β intensity ratio in the case of Al target was observed at the scale reading of $r=5$ mm as is seen in Fig. 3 or 4. In this r , Mn- K_α X-rays are strongly enhanced via (311) lattice diffraction (see Fig. 5(A)), whereas the coherent scattering of Mn- K_β X-rays occurs via non-diffraction since the $\sin \theta$ zone corresponding to the scale reading $r=5$ mm (see Fig. 5(B)) is located between the expected diffractions of (222) and (400) lattices. With increasing r , the effect of non-diffraction of K_α X-rays through (311) and (222) lattices suppresses the K_α X-ray intensity, and on the contrary the diffraction through (400) lattice enhances the K_β X-ray intensity. Then, the K_α/K_β intensity ratio decreases with increasing r .

All other examples are interpreted by the same procedure. Thus the remarkable K_α/K_β intensity variation with r or θ is qualitatively reproduced in the following processes: by the enhancement of i) K_α and K_β , ii) either K_α or K_β , iii) neither K_α nor K_β X-rays via diffraction according to the location of metallic targets.

The reason why the intensity of the X-rays scattered by the metallic targets changes so remarkably with r or θ is that the wave lengths λ of the primary Mn KX-rays are comparable to the lattice constants d of scatterer metals, so that the interference between the primary and coherent scattered photons are likely to take place.

In contrast to the case with metallic scatterers, the K_α/K_β intensity ratios change little in the case with non-metallic scatterers such as glass, lucite and mylar as is seen in Figs. 3 and 4. This fact is interpreted to be due either to the lack of crystalline structure, e.g., glass, or to the

large unit of periodic structure, e.g., mylar etc.

It is to be noted that, in the course of XRF elemental analysis using ^{55}Fe source or the primary photons with nearly the same wave length as the metallic lattice constant,

- (1) it is impossible to make quantitative elemental analysis, using the coherent scattered X-ray intensity as standard, and
- (2) it is possible to optimize the ratio (characteristic X-rays of samples)/(coherent scattered primary X-rays) in choosing an optimum scattering angle θ , so as to minimize the effects of pile up of the coherent scattering in the detector and/or of the escape peaks, for such samples as unknown elements are embedded in the metallic foil or bulk.

4. Conclusion

The K_α/K_β intensity ratio of Mn KX-rays from ^{55}Fe source scattered by metallic samples changes remarkably with the scattering angles. On the contrary it changes little when the X-rays are scattered by non-metallic samples.

This difference is interpreted to be due to the occurrence of strong or weak interference in the coherent scattering photons. The wave lengths of Mn KX-rays are comparable to the lattice constants of metallic crystals, whereas because of non-crystalline structure or huge unit lattice, such a kind of diffraction does not take place with non-metallic materials.

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要 旨

散乱した Mn K_{α}/K_{β} X線強度比の散乱体依存性

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^{55}Fe が放出する Mn-K X線のように, X線散乱断面積のうちでコヒーレント散乱が支配的な10 keV 以下のエネルギーをもつX線を試料に照射して, 蛍光X線分析を行うさい, 未知元素の放出するX線収量を, コヒーレント散乱したX線収量を基準にして規格化することは十分な注意を要する。未知元素が, 金属のように局所的に結晶構造をもつ試料の中に含まれている場合, コヒーレント散乱したX線が干渉し合うために, 散乱角すなわち試料と検出器の間の距離によって, 著しくその強度が変化することを実験的に確認した。