

L_{β}/L_{α} Intensity Ratios of Antimony at Different Azimuthal and Polar Angles

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Abstract

L_{β}/L_{α} X-ray intensity ratios of antimony (Sb) were investigated at different azimuthal ($-30^{\circ} \leq \varphi \leq +30^{\circ}$, at intervals of 10°) and polar scattering angles ($85^{\circ} \leq \theta \leq 135^{\circ}$ at intervals of 10°). In the purpose of exciting to Sb and detecting the X-rays emitted from Sb, ^{241}Am point source and Si(Li) detector have been used, respectively. The data was analysed by means of Origin 9 Software and it was determined that L_{β}/L_{α} change by polar scattering angle at a fixed azimuthal scattering angle and by azimuthal scattering angle at a fixed polar scattering angle.

Keywords: Intensity ratios, azimuthal scattering angle, polar scattering angle.

Farklı Azimuthal ve Saçılma Açılarında Antimonun L_{β}/L_{α} Şiddet Oranları

Öz

Sb' nin L_{β}/L_{α} X-ışını şiddet oranları farklı azimuthal ve saçılma açılarında ölçüldü. Sb' yi uyarmak amacıyla ve Sb'den gelen X-ışınların saymak amacıyla ^{241}Am nokta kaynak ve Si(Li) dedektör sırasıyla kullanıldı. Datalar Origin 9 programı aracılığıyla analiz edildi ve L_{β}/L_{α} şiddet oranlarının farklı azimuthal açı ve saçılma açısı değerleriyle değiştiği belirlendi.

Anahtar Kelimeler: Şiddet oranları, azimuthal saçılma açısı, polar saçılma açısı.

1. Introduction

Antimony ($[\text{Kr}] 4d^{10}5s^25p^3$) which exists metallic and non-metallic form is an element in 5A group of periodic table. It founds in nature as free or is obtained from ores of Sb_2S_3 and Sb_2O_3 . The pure form Antimony is used the production of diodes and infrared detectors. The alloyed Antimony and its compounds which have importance for world economy are used in low friction metals, batteries, paints, pottery, make-up and ceramics etc.

The radiation interacts with the matter in different processes. One of the interactions is photoelectric effect. In this interaction, the atom is bombarded by photons and the atom is excited or ionized by moving of any electron of atom to upper energy levels or out of the atom. If this electron ejected is on L sub-shell, electron transitions occur through upper levels to L sub-shell with radiative or nonradiative. The radiation having different frequencies emits during these transitions and they are called L X-rays. The transitions that make L_{α} rays and L_{β} rays occurred are shown in Table 1.

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Table 1. The Siegbahn and IUPAC showing of L X-ray lines.

Siegbahn	IUPAC
$L\alpha_1$	L_3-M_5
$L\alpha_2$	L_3-M_4
$L\beta_1$	L_2-M_4
$L\beta_2$	L_3-N_5
$L\beta_3$	L_1-M_3
$L\beta_4$	L_1-M_2
$L\beta_5$	$L_3-O_{4,5}$
$L\beta_6$	L_3-N_1
$L\beta_7$	L_3-O_1
$L\beta_9$	L_1-M_5
$L\beta_{10}$	L_1-M_4
$L\beta_{15}$	L_3-N_4
$L\beta_{17}$	L_2-M_3

X-rays are characterized by alignment (distortion of the charge cloud) and orientation parameters (the sum-angular momentum or circulation). If there are a vacancy states on K , L_1 , L_2 , M_1 and M_2 shells ($J=1/2$), the transitions to these shells make isotropic and unpolarized emission of X-rays form (Cooper et al., 1969) or on L_3 , M_3 , M_4 and M_5 ($J>1/2$), the transitions to these shells make anisotropic and polarized emission of X-rays form (Flügge et al., 1972). While (L_{β} and L_{γ}) X rays are isotropic, (L_{α} and L_{ℓ}) are anisotropic (Kahlon et al., 1991; Ertuğrul, 1996; Seven and Koçak, 2002; Han et al., 2008 and 2009; Akkuş et al., 2016). The depending on the angular and magnetic field of L_{α} and L_{ℓ} emission (anisotropic) are stronger than L_{β} (isotropic) emission (Demir and Şahin, 2007). Determining of L X-ray intensity correctly is beneficial to test theoretical predictions and in terms of to be used in a wide area like medical area, sample analysis (Doğan et al., 1998) and geological, nuclear and atomic physics (Yalçın et al., 2008). There are lots of studies about L X-rays before. The some of these studies; Doğan et al. (1998) have measured the L shell X-ray

intensity ratios for Ta, W, Re, Au, Hg, Tl, Pb, Bi, Th and U at excitation energies of 59.5 and 122 keV with a Si(Li) detector and observed agreement between the experimental and theoretical values. Yalçın et al. (2008) have determined the measurements of the L X-ray intensity ratio for elements Dy, Ho, Yb, W, Hg, Tl and Pb by photon excitation of ^{241}Am and the radioactive decay of ^{160}Tb , ^{160}Er , ^{173}Lu , ^{182}Re , ^{201}Tl , ^{203}Pb and ^{207}Bi . They have found that the conclusions of the search support the theoretical and other experimental results and L X-ray intensity ratios for samples by radioactive decay of radioisotopes deviated significantly from both experimental and theoretical results in literature. Söğüt et al. (1997) have investigated chemical effects on the L_{β}/L_{α} and L_{γ}/L_{α} X-ray intensity ratios of Hg, Pb and Bi. They have determined that L X-ray intensity ratios are effected by the chemical environment of atoms and L_{β}/L_{α} intensity ratios are affected less than L_{γ}/L_{α} intensity ratios. Cesareo et al. (2009) have studied the depending on the composition and thickness of the layer of X-ray ratios of K_{α}/K_{β} , L_{α}/L_{β} and L_{α}/L_{γ} . They have calculated K_{α}/K_{β} , L_{α}/L_{β} and L_{α}/L_{γ} as a function of material and thickness of the corresponding layer and shown many examples of using these ratios to identify layers and related thicknesses. Wang et al. (2016) have measured the angular distribution of W- L_{α} , L_{β_1} and L_{β_2} X-rays induced by 13.1 keV bremsstrahlung at different emission angles from 110° to 140° at intervals of 10° . They have shown that L_{β_1} X-ray yield shows isotropic emission, while the measured L_{α} and L_{β_2} X-ray yields are anisotropic. They have determined the anisotropy parameters for L_{α} and L_{β_2} X-rays. Kawai has studied chemical effects on the intensity ratio of L_{α} and L_{β} lines of transition metals with an XRF spectrometer and determined to be characterized the bond

covalency of compounds with L_{β}/L_{α} intensity ratio.

In this work, since the changes in L_{β}/L_{α} with respect to angle for Sb have been investigated firstly, this study is important. The aim of the present study is to measure L_{β}/L_{α} intensity ratios of Sb with different polar and azimuthal scattering angles and to observe whether there is a change or not at this value.

2. Materials and Methods

In this study, Sb (powder form) was turned into pellet which radius is with 13 mm by applying pressure. The angle between the γ rays coming from point source and normal of sample was fixed at 45° . The source and

sample were rotated together. The data was obtained at six different polar scattering angles ($85^{\circ} \leq \theta \leq 135^{\circ}$ with steps of 10°) for a fixed azimuthal scattering angle and at seven different azimuthal scattering angles ($-30^{\circ} \leq \varphi \leq +30^{\circ}$, with steps of 10°) for fixed polar scattering angle on the experimental setup (Figure 1). 100 mCi ^{241}Am point source having γ photons with 59.54 keV of energy in order to excite the sample and Si(Li) detector for counting to photons emitted from sample were used. The photons coming to the detector was placed to the channels according to energies via multichannel analyzer. The data was analyzed at Origin 9 computer program. Typical Spectrum is seen for L_{α} and L_{β} rays of Sb in Figure 2.

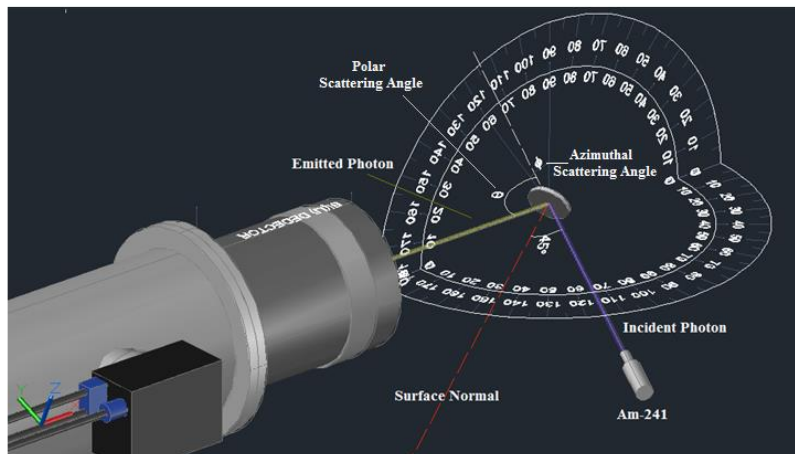


Figure 1. The experimental setup.

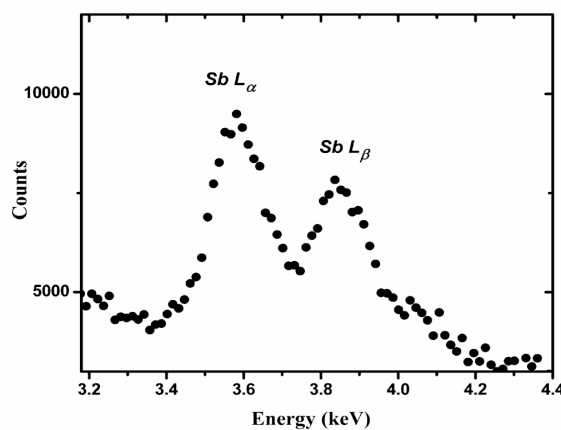


Figure 2. Typical spectrum for L_{α} and L_{β} rays of Sb.

The experimental intensity ratio is given by:

$$\frac{I_{L\beta}}{I_{L\alpha}} = \frac{N_{L\beta}}{N_{L\alpha}} \times \frac{\beta_{L\alpha}}{\beta_{L\beta}} \times \frac{\varepsilon_{L\alpha}}{\varepsilon_{L\beta}} \quad (1)$$

where $N_{L\beta}/N_{L\alpha}$ is the area rate of the L peaks, $\beta_{L\alpha}/\beta_{L\beta}$ is the self-absorption correction ratio of the sample for excited and emitted photons, and $\varepsilon_{L\alpha}/\varepsilon_{L\beta}$ is for the L_{β} and L_{α} the detector yield ratio, respectively. The self-absorption correction value β was determined by the following equation:

$$\beta_{Li} = \frac{1 - \exp[-(\mu/\rho)_i/\cos\theta_1 + (\mu/\rho)_e/\cos\theta_2]t}{((\mu/\rho)_i/\cos\theta_1 + (\mu/\rho)_e/\cos\theta_2)t} \quad (2)$$

$(\mu/\rho)_{i-e}$ is the mass attenuation values (cm^2/g) at exciting energy and emitting energy. θ_1 is the angle between incident photon and surface normal and θ_2 is the angle between emitted photon and surface normal. t represents the target mass thickness in (g/cm^2). The mass attenuation values were taken from (Gerward et al., 2001 and 2004).

The $\varepsilon_{L\alpha}/\varepsilon_{L\beta}$ is obtained from the graph of $I_0G\varepsilon_i$ and $I_0G\varepsilon_j$ versus energy. $I_0G\varepsilon_i$ has

been calculated by measuring the peak areas of characteristic K X-rays of K , Cr , Co , Ni , Cu , Zn , Y , Zr , Nb and Mo . The detector efficiency graph for the polar scattering angle ($\theta = 115^\circ$) is seen in Figure 3. I_0 is γ -ray intensity and G is the geometrical contribution changing with the radioactive element-target array. $I_0G\varepsilon_i$ is given by:

$$I_0G\varepsilon_i = \frac{N_{Ki}}{\sigma_{Ki}\beta_{Ki}t} \quad (3)$$

The theoretical σ_{Ki} (the possibility of K_i X-ray fluorescence) is given by this formula:

$$\sigma_{Ki} = \sigma_K(E) \times \omega_K \times F_{Ki} \quad (4)$$

where $\sigma_K(E)$ was obtained from (Storm and Israel, 1970), ω_K is the fluorescence efficiency of K level and was obtained from (Krause, 1979), $F_{K\alpha}$ and $F_{K\beta}$ are determined as:

$$F_{K\alpha} = (1 + \frac{I_{K\beta}}{I_{K\alpha}})^{-1}, \quad F_{K\beta} = 1 - F_{K\alpha} \quad (5)$$

where $I_{K\beta}/I_{K\alpha}$, K_{β} to K_{α} intensity ratio was taken from (Scofield, 1974).

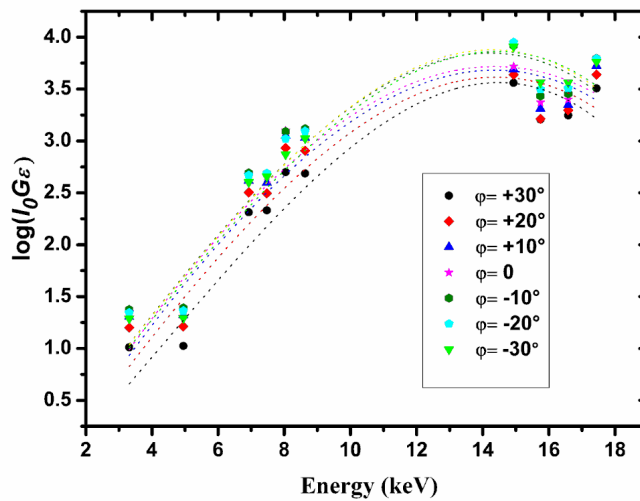


Figure 3. The efficiency graph of the detector for the polar scattering angle ($\theta = 115^\circ$).

To minimize the experimental error, the measurement was taken with and without the sample for different azimuthal and polar scattering angles. The measurement without sample was subtracted from the measurement with sample for all angel values. The overall error in the present measurements is estimated to be 1-3 %.

In this work, experimental error was calculated by using equation following:

$$\Delta \left(\frac{I_{L_{\beta}}}{I_{L_{\alpha}}} \right) = \left[\left(\frac{\Delta N_{L_{\beta}}}{N_{L_{\beta}}} \right)^2 + \left(\frac{\Delta N_{L_{\alpha}}}{N_{L_{\alpha}}} \right)^2 + \left(\frac{\Delta \beta_{L_{\alpha}}}{\beta_{L_{\alpha}}} \right)^2 + \left(\frac{\Delta \beta_{L_{\beta}}}{\beta_{L_{\beta}}} \right)^2 + \left(\frac{\Delta I_0 G \varepsilon_{L_{\alpha}}}{I_0 G \varepsilon_{L_{\alpha}}} \right)^2 + \left(\frac{\Delta I_0 G \varepsilon_{L_{\beta}}}{I_0 G \varepsilon_{L_{\beta}}} \right)^2 \right]^{1/2} \quad (6)$$

where $\Delta N_{L_{\beta}}$, $\Delta N_{L_{\alpha}}$ are counts error of L_{β} and L_{α} X-ray intensity peaks; $\Delta \beta_{L_{\alpha}}$, $\Delta \beta_{L_{\beta}}$ are the β_{Li} errors for L_{β} and L_{α} X-ray photons; $\Delta I_0 G \varepsilon_{L_{\alpha}}$ and $\Delta I_0 G \varepsilon_{L_{\beta}}$ are the effective photon flux errors at L_{α} and L_{β} energies. The total of the uncertainties are sourced from different factors such as the evaluation of peak areas (<0.4%), $I_0 G \varepsilon$ product (<0.5%), absorption correction factor (<0.2%) and experimental geometry (<0.1%).

3. Research Findings

The calculated experimental $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios of Sb by depending on azimuthal (+30°, +20°, +10°, 0°, -10°, -20°, -30°) and polar scattering (85°, 95°, 105°, 115°, 125°, 135°) angles have been shown in Table 2. The variation of $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios with polar and azimuthal scattering angles is seen in Figure 4 and Figure 5. When it is evaluated both Figure 4 and Table 2, it is seen that $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios generally increase with the increment of polar scattering angle (from 85° to 135°) at all azimuthal scattering angles.

These results are compatible with the before studies that; Seven and Koçak (2002) have measured the L_{l} , L_{α} , L_{β} and L_{γ} X-ray production cross-sections in U, Th, Bi, Pb, Tl, Hg and Au using 59.5 keV incident photon energies in the angular range 40–130°. L_{β} and L_{γ} X-rays were found to be angle independent, those for L_{l} and L_{α} X-rays were found to be angle dependent. They have found that $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios increase with the emission angle ($\theta=$ from 40° to 130°, at intervals of 10°) for U, Th, Bi, Pb, Tl, Hg and Au. Ertuğrul (1996) has studied measurement of cross-sections and Coster-Kronig transition effect on L subshell X-rays of some heavy elements in the atomic range $79 \leq Z \leq 92$ at 59.5 keV and has determined that $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios increase with the scatter angle ($\theta=$ 45°, 60°, 75°, 90°, 105°, 120° and 135°) for Pb. Kahlon et al. (1990) have measured the angular distribution and polarization of the L -shell fluorescent X-rays excited by 59.57-keV photons in Th and U and found that $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios increase with emission angle ($\theta=$ from 40° to 120°, at interval of 10°) for U and Th.

Table 2. $I_{L_\beta} / I_{L_\alpha}$ ratios of Sb for different azimuthal and polar scattering angles.

Azimuthal angle (φ)	+30°	+20°	+10°	0°	-10°	-20°	-30°
Scattering angle, θ	I_{L_β}/I_{L_α}	I_{L_β}/I_{L_α}	I_{L_β}/I_{L_α}	I_{L_β}/I_{L_α}	I_{L_β}/I_{L_α}	I_{L_β}/I_{L_α}	I_{L_β}/I_{L_α}
85°	1.2232±0.012	1.2144±0.012	1.2133±0.012	1.2608±0.013	1.2549±0.015	1.2655±0.016	1.2816±0.015
95°	1.2257±0.011	1.2341±0.016	1.2495±0.014	1.3222±0.011	1.3234±0.018	1.3753±0.016	1.3122±0.015
105°	1.3145±0.016	1.2525±0.013	1.3405±0.013	1.3533±0.016	1.4028±0.013	1.4165±0.012	1.4152±0.014
115°	1.3051±0.014	1.3030±0.013	1.3542±0.012	1.4632±0.016	1.4344±0.021	1.4473±0.013	1.4461±0.019
125°	1.3365±0.013	1.3898±0.014	1.4266±0.018	1.4508±0.014	1.4884±0.013	1.5297±0.013	1.5585±0.017
135°	1.3634±0.015	1.4276±0.017	1.4678±0.021	1.5092±0.019	1.4873±0.011	1.6679±0.013	1.6800±0.013

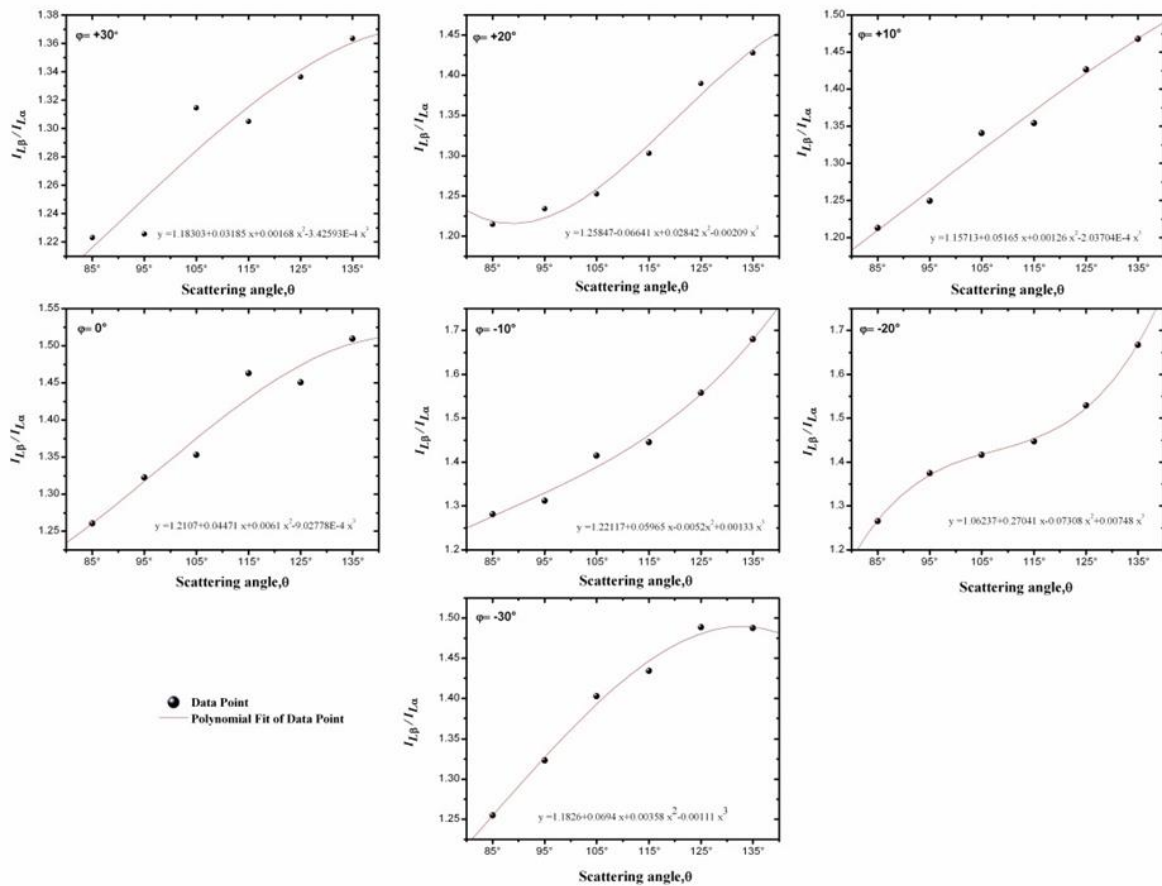


Figure 4. The variation of L_β / L_α with the scattering polar angles (θ) at a fixed azimuthal scattering angle.

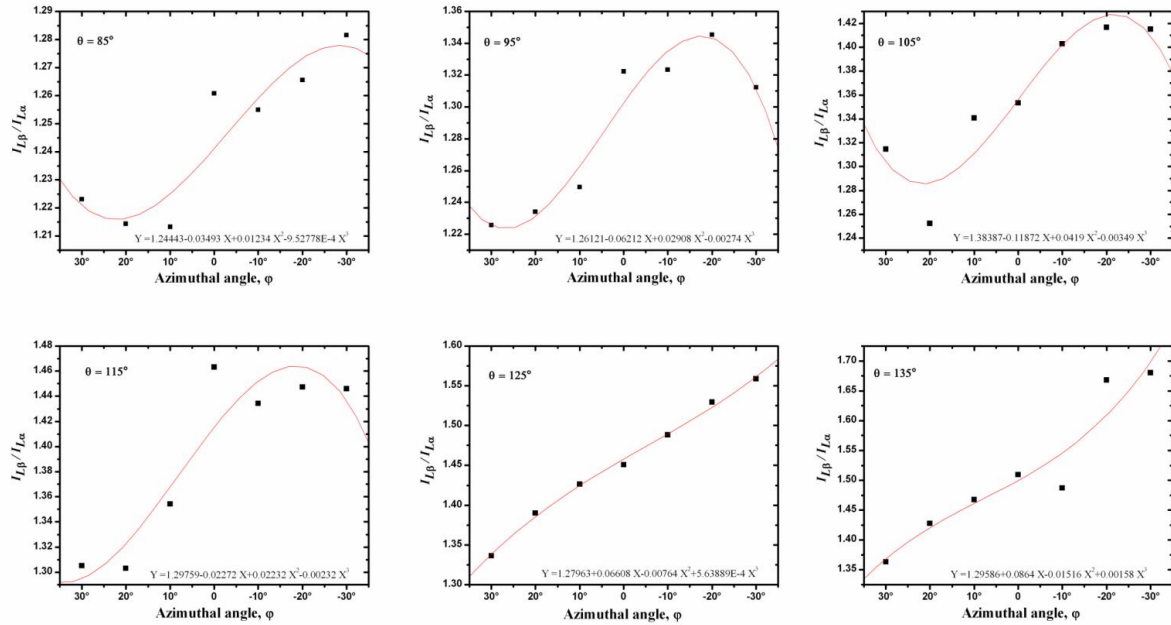


Figure 5. The variation of L_{β}/L_{α} with the azimuthal scattering angles (φ) at a fixed scattering polar angle.

Han et al. (2008) have been studied angular variations of K and L X-ray fluorescence cross sections for some lanthanides and found that $I_{L_{\beta}}/I_{L_{\alpha}}$ increase with emission angle (θ = from 120° to 150° , at intervals of 10°) for Sm, Eu, Gd, Tb, Dy, Ho and Er.

When the results evaluate the before studies, we may arrive the conclusion that since L_{β} X-rays are independent the angle and L_{α} is dependent to the angle, the probability of L_{α} X-rays may decrease the changing of the polar and azimuthal scattering angles. This state may cause of the increasing of $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios.

In the same way, when Figure 5 and Table 2 are examined, it is seen that $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios generally increases with the variation of azimuthal scattering angle (from $+30^{\circ}$ to -30°) at all polar scattering angles. Akkuş et al. (2016) has found that K_{β}/K_{α} intensity ratios are not dependent on azimuthal scattering angle. Han et al. (2008) have found that K_{β}/K_{α} intensity ratios are

constant at all emission angles. Since K_{β}/K_{α} intensity ratio which depends on the physical and chemical environments of the elements in the sample (Raj et al. 1998; Pawloski et al 2002) is constant at all polar and azimuthal scattering angles, it is appropriate for studies made to determine some individual atomic parameters, $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios which change by depending on the angle may not be appropriate for that.

4. Results

In this study, L_{β}/L_{α} X-ray intensity ratios of Sb were investigated at different azimuthal and polar scattering angles. It is seen that $I_{L_{\beta}}/I_{L_{\alpha}}$ ratios change with the polar and azimuthal scattering angles. This value is not same at different angles. This variation should be taken into account at the studies. In terms of confidence of the evaluation, this study may be repeated for different polar and azimuthal scattering angles and elements.

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