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The K-beta/K-alpha intensity ratios of some elements at different azimuthal scattering angles at 59.54 keV

Tuba Akkuş, Yusuf Şahin, Demet Yılmaz, and Fatma Nur Tuzluca

Abstract: In this study K-beta/K-alpha (K_β/K_α) intensity ratios of the elements scandium (Sc), vanadium (V), chromium (Cr), nickel (Ni), copper (Cu), zinc (Zn), bromine (Br), yttrium (Y), zirconium (Zr), and niobium (Nb) have been measured at three azimuthal scattering angles (+30°, 0°, and –30°) for two fixed polar scattering angles (85° and 135°). The elements were excited by 59.54 keV gamma photons emitted from 100 mCi Am^{241} point source. The obtained experimental values were compared with the theoretical values presented in the literature. It is observed that K_β/K_α intensity ratios of the investigated elements do not depend on azimuthal angles at fixed polar angles.

Key words: X-ray intensity ratio, azimuthal dependence, K shell X-ray.

Résumé : À trois angles azimutaux de diffusion (+30°, 0° et –30°) et deux angles polaires de diffusion (85° et 135°), nous mesurons ici les rapports d'intensité K_β/K_α des éléments scandium (Sc), vanadium (V), chrome (Cr), nickel (Ni), cuivre (Cu), zinc (Zn), brome (Br), yttrium (Y), zirconium (Zr) et niobium (Nb). Les éléments sont excités par des photons de 59.54 keV émis par une source ponctuelle de 100 mCi de Am^{241}. Les valeurs expérimentales obtenues sont comparées aux valeurs théoriques présentées dans la littérature. Nous observons que les rapports d'intensité K_β/K_α des éléments étudiés ne dépendent pas de l'angle azimutal à un angle polaire donné. [Traduit par la Rédaction]

Mots-clés : rapport d'intensité de rayons X, dépendance azimutale, rayons X dans la couche K.

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1. Introduction

X-ray intensity ratios are important in explaining the interaction of X-rays with matter. Furthermore the X-ray intensity ratios are used in many fields, such as physics, chemistry, biomedicine, and a variety of other field [1]. Also, the X-ray parameters of the K shell have been extensively investigated both theoretically and experimentally. It is well known that the K_α and K_β X-rays originate from filling of the K shell vacancies and the fluorescent X-rays originating from vacancy states with total angular momentum quantum number 1/2 (j = 1/2) are isotropic. There are many reports on K_β/K_α intensity ratios in the literature at the fixed polar scattering angle but there is no report on various azimuthal scattering angle.

Krause et al. [2] have calculated theoretical K and L X-ray fluorescence cross sections. Puri et al. [3] composed a table of K shell X-ray fluorescence cross sections for some elements in the energy range 1–200 keV. Ertuğrul et al. [4] measured K_β/K_α intensity ratios for elements in the range $22 \le Z \le 69$ at 59.5 keV. Chemical effects on K_β/K_α X-ray intensity ratios of Mo, Ag, Cd, Ba, La, and Ce compounds and total mass attenuation coefficients of Fe and Cu investigated by Söğüt et al. [5]. Bacaksız and Çevik [6] studied K shell fluorescence yields of Cd and Zn in $Cd_{1-x}Zn_xS$ thin films. Yalçın [7] experimentally investigated relative K X-ray intensity ratio following radioactive decay and photoionization. Ertuğrul et al. [8] investigated K_β/K_α X-ray intensity ratios for elements in the range $16 \le Z \le 92$ excited by 5.9, 59.5, and 123.6 keV photons. Çevik et al. [9] theorically and experimentally investigated K_β/K_α intensity ratios for some pure metals at 59.5 and 123.6 keV. K shell intensity ratios for some heavy elements at 123.6 keV measured by Apaydın et al. [10]. Saydam et al. [11] investigated K shell fluorescence cross section and K_β/K_α intensity ratios for Fe, Se, Te, FeSe, FeTe, and TeSe. K X-ray fluorescence parameters in elements with $24 \le Z \le 65$ in an external magnetic field investigated by Demir and Şahin [12]. Önder et al. [13] measured K shell X-rays fluorescence parameters of some elements in atomic range $40 \le Z \le 50$. Turşucu and Demir [14] investigated chemical effect on the K shell X-ray fluorescence parameters of some Ce compounds. Mahalakshmi et al. [15] measured K_β/K_α intensity ratios for elements in the range $51 \le Z \le 67$ at 59.54 keV. Doğan et al. [1] determined chemical effects on K X-ray production cross sections and K_β/K_α intensity ratios of Co, Ni, Cu, and Zn elements in phthalocyanine complexes. Fluorescence parameters of Zr and Sb compounds were studied by Aksoy et al. [16]. Perişanoğlu and Demir [17] determined the effect of external magnetic field on K_β/K_α X-ray intensity ratios of various alloy compositions of Ni–Cr transition metal alloys.

However, as we know there are no experimental results for azimuthal scattering angle dependence of K_β/K_α intensity ratios in the literature. The aim of this study is to verify the dependencies of K_β/K_α intensity ratios of some elements due to the atomic number and azimuthal scattering angles at 59.54 keV exciting photon energy. In the present work K_β/K_α intensity ratios of Sc, V, Cr, Co, Ni, Cu, Zn, Br, Y, Zr, and Nb have been measured at three azimuthal scattering angles (+30°, 0°, and –30°) for two fixed polar scattering angles by using a Si(Li) detector with a resolution of 160 eV at 5.9 keV photon energy.

2. Theory

In the present study K_β/K_α intensity ratios were calculated using the relation [12]

Received 22 August 2016. Accepted 17 November 2016.

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$$\frac{K_\beta}{K_\alpha} = \frac{N_{K_\beta}}{N_{K_\alpha}} \frac{\beta_{K_\alpha}}{\beta_{K_\beta}} \frac{\varepsilon_{K_\alpha}}{\varepsilon_{K_\beta}} \quad (1)$$

where N_{K_β}/N_{K_α} is the ratio of the number of counts under K_β and K_α peaks. β_{K_α} and β_{K_β} are the sample self-absorption correction factors, ε_{K_α} and ε_{K_β} are the efficiencies of detector for K_β and K_α series of X-rays.

The self-absorption correction factors β is calculated for both K_β and K_α separately by using the following expressions:

$$\beta = \frac{1 - \exp\{(-1)[(\mu_i/\cos\theta_1) + (\mu_e/\cos\theta_2)]t\}}{[(\mu_i/\cos\theta_1) + (\mu_e/\cos\theta_2)]t} \quad (2)$$

where μ_i and μ_e are the total mass absorption coefficient of incident photons and emitted characteristic X-rays. The values of μ_i and μ_e are taken from WinxCom [18]. θ_1 and θ_2 are the angles of incident gamma ray and emitted K X-rays with the target normal. t is the mass thicknesses of the sample. The units of μ_i and μ_e are cm^2/g because the units of t are g/cm^2.

The effective incident photon flux factor $I_0 G\varepsilon_{K_i}$ was determined using the following relation [5]:

$$I_0 G\varepsilon K_i = \frac{N_{K_i}}{\beta_{K_i}\sigma_{K_i} t} \quad (3)$$

where N_{K_i} is the net number of counts under the K_β or K_α peaks, σ_{K_i} is the σ_{K_α} or σ_{K_β} fluorescence cross section. I_0 is intensity of incident radiation, G is geometrical factor, ε_{K_i} is the detector efficiency for K_i X-rays. For a defined geometry and constant source of photons, $I_0 G$ is a constant and hence vanishes when intensity ratios are considered.

The theoretical values of σ_{K_α} or σ_{K_β} fluorescence cross sections were calculated using [19]

$$\sigma_{K_\alpha} = \sigma_{K_E} w_K f_{K_\alpha} \quad (4)$$

$$\sigma_{K_\beta} = \sigma_{K_E} w_K f_{K_\beta} \quad (5)$$

where σ_{K_E} is the K shell photoionization cross section for the given elements at the excitation energy E. The values of σ_{K_E} in this work were taken from Scofield [20] and the values of w_K were taken from the report by Hubbell et al. [21]. The fractional emission ratio of the K_α and K_β X-rays, f_{K_α} and f_{K_β}, are defined as [22]

$$f_{K_\alpha} = \left(1 + \frac{I_{K_\beta}}{I_{K_\alpha}}\right)^{-1} \quad (6)$$

$$f_{K_\beta} = \left(1 + \frac{I_{K_\alpha}}{I_{K_\beta}}\right)^{-1} \quad (7)$$

and taken from Scofield [23].

3. Experimental

The experimental setup used in the present study is shown in Fig. 1. The atomic number (Z) dependencies of K_β/K_α intensity ratios are measured at three azimuthal scattering angles (+30°, 0°, and –30°) for two fixed polar scattering angles (85° and 135°). The angles of the sample surface with the incoming photons were fixed at 45° at all measurements while the source and target were together rotated around the z axis. The detector was kept in a fixed position at all measurements. The detector is shielded by a lead collimator to avoid direct exposure of the detector from the radiation incoming from the source and environment.

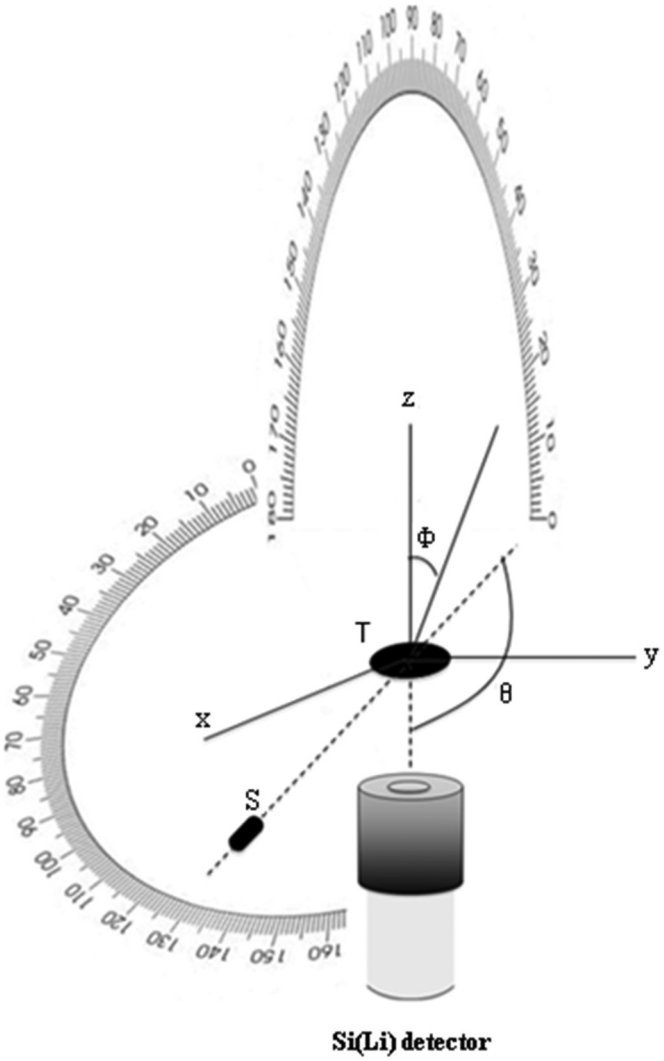


Fig. 1. Schematic representation of the experimental set up. *T*, target; *S*, Am^{241} source; Φ, azimuthal angle; θ, polar angle.

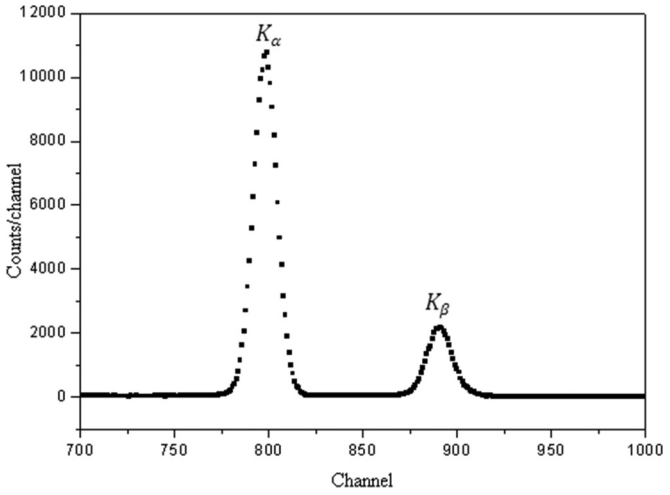
The samples were of thickness ranging from 0.092 to 0.191 g/cm^2 and were prepared from the powders in the form 13 mm diameter cylindrical pellets at a pressure of 7 t. Samples were irradiated by 59.54 keV γ-rays from a 100 mCi Am^{241} radioactive point source. The emitted X-rays were detected by a Si(Li) detector has a resolution of 160 eV at 5.9 keV with active area 120 mm^2, sensitive crystal depth 3 mm and Be window thickness 0.025 mm. The distances from source to target and target to the detector were chosen 2.6 and 3.2 cm, respectively.

The computer program Genie-2000 was used to acquire the spectra and to control the operating parameters of the system. The data were acquired into 4096 channels of the MCA. The peak areas were determined by means of Microcal Origin 7.5 Software program. This program uses least square fit method to determine the net peak areas. Background spectrum was measured for each angle and subtracted from the raw spectrum to correct for background in the peak region. A typical K X-ray spectrum of Br target

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Fig. 2. A typical observed spectrum from Br target at scattering azimuthal angle of 0° and polar scattering angle of 85°.



at azimuthal scattering angle of 0° and polar scattering angle of 85° is shown in Fig. 2.

For each azimuthal scattering angle and polar scattering angle the $I_0G\varepsilon$ values were calculated by using (3). The variation of the factor $\log I_0G\varepsilon$ versus to energy at mentioned scattering angles and their fitting equations are given in Figs. 3 and 4.

4. Results and discussion

In this study it is shown that K_β/K_α intensity ratios are depend on atomic number but independ on the azimuthal scattering angle as well as on the polar scattering angle.

The measured values of K_β/K_α intensity ratio of some elements in the atomic range $21 \le Z \le 42$ for three azimuthal angles (+30°, 0°, –30°) at fixed two scattering polar angles for 85° and 135° are given in Tables 1 and 2. The theoretical K_β/K_α intensity ratios from Scofield are also given in these tables. The experimental values of K_β/K_α intensity ratios in the atomic range $21 \le Z \le 42$ have been compared with theoretical estimates based on the relativistic Hartree model calculated by Scofield [23].

The experimental and theoretical values of K_β/K_α intensity ratios have been plotted comparatively as a function of the atomic number in Figs. 5 and 6. For the samples under investigation, the K_β/K_α intensity ratios increase with increasing atomic number except for the elements Cr and Cu. There is a good third-order polynomial relation between K_β/K_α intensity ratios and atomic number as seen from Fig. 4. Both experimental and theoretical K_β/K_α intensity ratios for each of the elements Cr and Cu are lower than that of previous and later elements in the periodic table. This can be attributed to the unfilled 4s orbitals ($4s^1$) of these two elements, because the outer levels of the atomic ground-state of Cr and Cu are $\ldots 3d^5 4s^1$ and $\ldots 3d^{10} 4s^1$, respectively. Furthermore, for the lower atomic numbers, the inaccuracies in the energies and the transition rates can come from inaccuracies in the description of the interaction between the electrons [24].

As seen from Tables 1 and 2, K_β/K_α intensity ratios are not dependent on azimuthal scattering angle. This is a result from K X-rays originating from filling of the K shell vacancies. Thus, this work demonstrates that K X-ray emissions arising from photon excitations are isotropic.

For the polar scattering angle of 85°, the discrepancies between the present experimental and theoretical values of Scofield are within the range 0.67%–12.69%, 1.64%–14.02%, and 4.70%–18.42% at azimuthal angles of +30°, 0°, and –30°, respectively. Additionally, for the polar scattering angle 135°, the discrepancies between the

Fig. 3. The variation of $I_0G\varepsilon$ as a function of energy for scattering polar angle of 85°. [Colour online.]

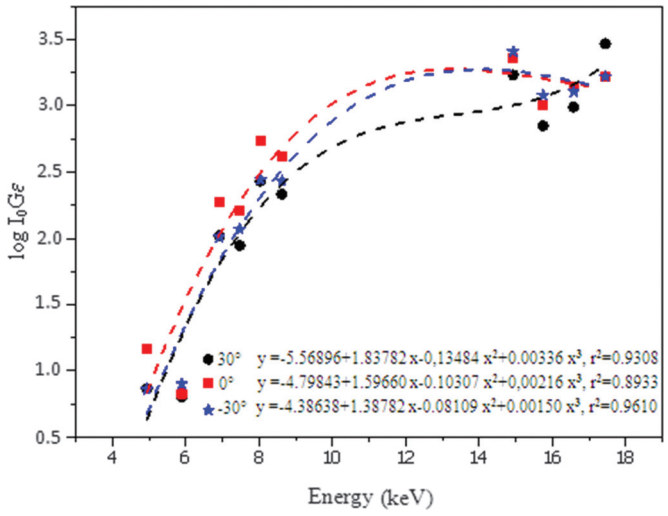
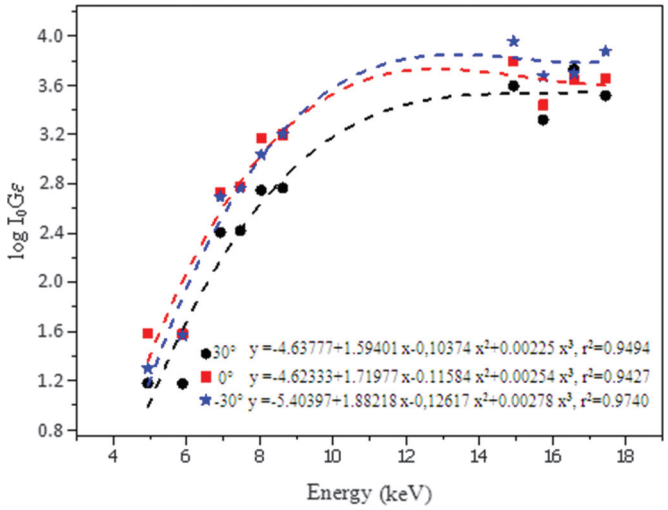


Fig. 4. The variation of $I_0G\varepsilon$ as a function of energy for scattering polar angle of 135°. [Colour online.]



present experimental and theoretical values of Scofield are within the range 0.19%–7.14%, 0.23%–7.51%, and 2.28%–11.27% at azimuthal scattering angles of +30°, 0°, and –30°, respectively. It is clear that the theoretical values are in good agreement with the measured values at polar scattering angle of 135° rather than that of measured values at polar scattering angle of 85°. This means that the detection geometry of 135° is better than the detection geometry of 85° because of the good counting statistics.

The percent errors for the experimental K_β/K_α intensity ratios of the studied elements at polar scattering angle of 85° are estimated in the range 3.47–7.24, 3.59–6.67, and 3.20–6.01 at azimuthal scattering angle of +30°, 0°, and –30°, respectively. Similarly, the percent errors for the experimental K_β/K_α intensity ratios at polar scattering angle of 135° are estimated in the range 3.29–6.12, 3.01–5.77, and 3.16–6.84 at azimuthal scattering angle of +30°, 0°, and –30°, respectively.

The uncertainty calculation for K_β/K_α intensity ratios can be given as

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Table 1. Experimental and theoretical values of K_β/K_α intensity ratios for azimuthal scattering angle of +30°, 0°, –30° at polar scattering angle of $\theta = 85°$.

		K_β/K_α			
		Experimental			Theoretical
Element	Z	$\phi = +30°$	$\phi = 0°$	$\phi = -30°$	[23]
Sc	21	0.1119±0.0060	0.1124±0.0056	0.1220±0.0054	0.1106
V	23	0.1178±0.0076	0.1283±0.0067	0.1334±0.0063	0.1161
Cr	24	0.1161±0.0084	0.1259±0.0073	0.1320±0.0079	0.1153
Co	27	0.1302±0.0087	0.1319±0.0078	0.1377±0.0067	0.1218
Ni	28	0,1314±0.0084	0.1323±0.0085	0.1326±0.0078	0.1227
Cu	29	0.1287±0.0085	0.1289±0.0086	0.1314±0.0079	0.1216
Zn	30	0,1299±0.0061	0.1305±0.0064	0.1307±0.0059	0.1241
Br	35	0.1504±0.0054	0.1521±0.0057	0.1553±0.0052	0.1483
Y	39	0.1815±0.0063	0.1836±0.0066	0.1841±0.0059	0.1696
Zr	40	0.1941±0.0085	0.1981±0.0072	0.1991±0.0075	0.1738
Nb	41	0.1997±0.0073	0.2020±0.0084	0.2098±0.0069	0.1772

Table 2. Experimental and theoretical values of K_β/K_α. intensity ratios for azimuthal scattering angle of +30°, 0°, –30° at polar scattering angle of $\theta = 135°$.

		K_β/K_α			
		Experimental			Theoretical
Element	Z	$\phi = +30°$	$\phi = 0°$	$\phi = -30°$	[23]
Sc	21	0.1127±0.0054	0.1140±0.0061	0.1140±0.0078	0.1106
V	23	0.1229±0.0063	0.1243±0.0059	0.1253±0.0065	0.1161
Cr	24	0.1218±0.0069	0.1222±0.0066	0.1232±0.0076	0.1153
Co	27	0.1225±0.0061	0.1248±0.0072	0.1269±0.0063	0.1218
Ni	28	0.1245±0.0076	0.1254±0.0056	0.1297±0.0062	0.1227
Cu	29	0.1226±0.0075	0.1244±0.0069	0.1244±0.0071	0.1216
Zn	30	0.1243±0.0066	0.1244±0.0062	0.1271±0.0059	0.1241
Br	35	0.1589±0.0057	0.1594±0.0054	0.1650±0.0066	0.1483
Y	39	0.1764±0.0058	0.1763±0.0053	0.1844±0.0062	0.1696
Zr	40	0.1744±0.0082	0.1757±0.0069	0.1853±0.0068	0.1738
Nb	41	0.1788±0.0074	0.1797±0.0059	0.1864±0.0059	0.1772

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

where ΔN_{K_β}, ΔN_{K_α}, $\Delta\beta_{K_\alpha}$, $\Delta\beta_{K_\beta}$, $\Delta I_0 G\varepsilon_{K_\alpha}$, $\Delta I_0 G\varepsilon_{K_\beta}$ are the errors in the intensities N_{K_β} and N_{K_α}, absorpsion correction factors β_{K_α} and β_{K_β}, $I_0 G\varepsilon_{K_\alpha}$, and $I_0 G\varepsilon_{K_\beta}$ product, respectively.

The physical quantities, such as K_α, K_β, N_{K_α}, are measured or semi-empirically calculated. The errors in the individual quantities are calculated from this empirically or semi-empirically obtained quantities with the relative fractional counting error method for single measurement [25].

5. Conclusion

In the present work, K_β/K_α intensity ratios for the elements in the range $21 \le Z \le 41$ have been measured at three azimuthal scattering angles (+30°, 0°, and –30°) and two polar scattering angles (85° and 135°) at 59.54 keV photon energy by using a Si(Li) detector. A filtered Am^{241} point source (100 mCi) emitting 59.54 keV gamma rays was used to excite the samples. The experimental values of K_β/K_α intensity ratios at different polar and azimuthal scattering angles are given in Tables 1 and 2. To the best of our knowledge no experimental data are available for azimuthal dependence of K_β/K_α intensity ratio and the results given in the present paper are the first. A new experimental setup has been designed to set azimuthal and polar angles simultaneously. The experimental results will give comprehensive and sensitive results to the literature. It is important to investigate the angular distribution of X-rays because it gives information about atomic parameters and wave function of the inner shell electrons.

K_β/K_α intensity ratios increase with increasing atomic number. Also, K X-rays originating from vacancy states with $j = 1/2$ are isotropic for both azimuthal and polar scattering angles.

Fig. 5. The variation of experimental and theoretical K_β/K_α intensity ratio values as a function of atomic number for the three values of azimuthal angles (Φ = +30°, 0°, –30°) at scattering polar angle of 85°. [Colour online.]

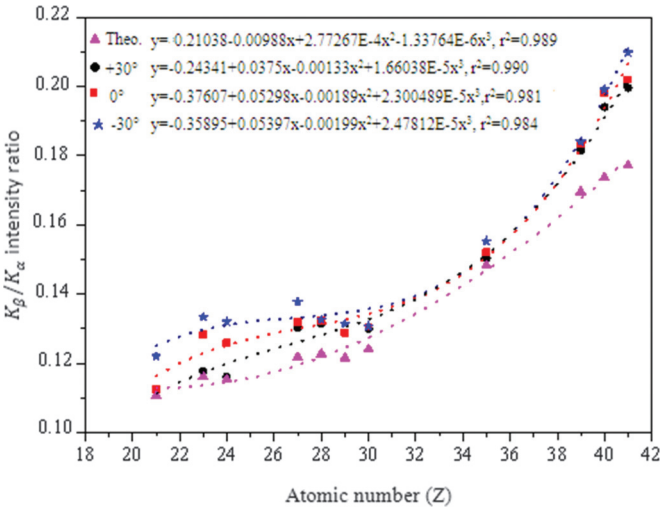
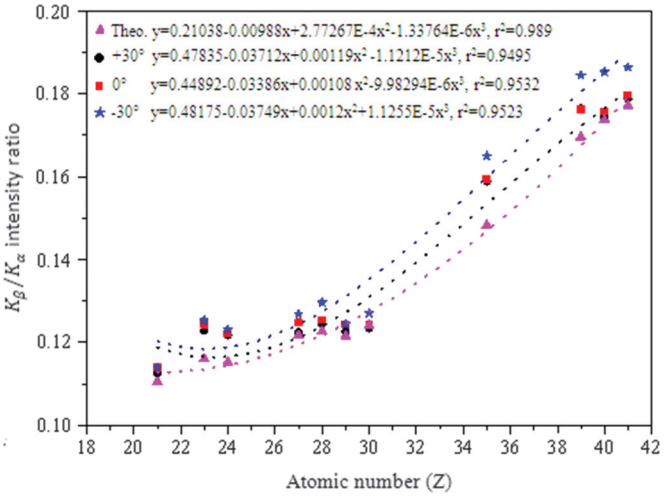


Fig. 6. The variation of experimental and theoretical K_β/K_α intensity ratio values as a function of atomic number for the three values of azimuthal angles (Φ = +30°, 0°, –30°) at scattering polar angle of 135°. [Colour online.]



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