Angular distribution of Au and U *L* x rays induced by 22.6-keV photons

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The angular distribution of the *L* x-ray fluorescent lines from Au and U induced by 22.6-keV x rays from a ^{109}Cd has been measured. A Si(Li) detector having a resolution of 160 eV at 5.90 keV was used to detect these *L* lines over the angular range of 70°–150°. No strong anisotropy was observed as mentioned by some groups. In the case of Au, a maximum anisotropy of 5% was observed while for U it was within experimental errors (2%). From the angular distribution of the *L*1 line of Au, the alignment parameter was obtained and its value was found to be 0.10±0.14.

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I. INTRODUCTION

It is now a well-established fact that when charged particles (protons, electrons, heavy ions) produce vacancies in atoms at energy levels with $J > \frac{1}{2}$, the resulting ions will be aligned. The signature of this alignment of the ions is the anisotropic angular distribution of the ejected Auger electrons, the emitted characteristic radiation, or the degree of polarization of the radiation. This anisotropic behavior is caused by the nonstatistical distributions of different magnetic substates by the ionized atoms. A lot of theoretical studies have been done so far along this line [1–5], and the predictions of these models have been experimentally supported by many workers [6–12].

But in the case of photon-induced ionization, the situation is not very clear. In 1960, Cooper and Zare [13] first put forward their theoretical model advocating that photon-induced ions will not be aligned at all and so the angular distribution of the Auger electrons or of the characteristic radiation subsequent to photoionization will be isotropic. In 1972, Flügge *et al.* [14] predicted that when vacancies are created in states with $J > \frac{1}{2}$, the resulting ions will be aligned. Their predictions were later supported by the theoretical studies of Jacob [15], Oh and Pratt [16], Scofield [17], Berezhko *et al.* [18], Kleiman and Lohmann [19], and recently Kleiman and Becker [20]. After the prediction of Flügge *et al.* [14] Caldwell and Zare [21] first made an experimental investigation of the photon-induced alignment of Cd. By measuring the degree of polarization of the emitted radiation from Cd, they observed the alignment of Cd ions. Since then, many experiments have been done [22–28] to study the alignment of ions by measuring either the angular distribution of the Auger electrons or the degree of polarization of the emitted radiation. All these experiments confirmed a small amount of alignment of the ions after photoionization.

The results of alignment studies *through a measurement of the angular distribution* of the emitted characteristic radiation are still quite confusing. Data from different groups [29–50] show contradictory results. The first such measurement was done by Kahlon *et al.* [29] who observed a very strong angular anisotropy in the *L* x rays emitted from the $J=3/2$ state. Table I shows a list of all such measurements done so far in chronological order and comments on the extent of the anisotropy of the emitted radiation. It is clear from the table that the groups at Patiala, India [29–32,40] and Turkey [34–36,41,43,44,46,50] got a very strong anisotropy while the rest of the groups observed very little anisotropy in their measurements.

Moreover, of all these groups, only Papp and Campbell [33] published the magnitude of the anisotropy and the alignment parameter for the *L* lines of Er. The alignment parameter of the ions of Xe and Au were, respectively, obtained by Küst *et al.* [48] and Yamaoka *et al.* [47]. All the other groups mentioned in Table I showed only the intensity variation of different *L* x-ray lines against the measured angles and from these plots quantitatively inferred the extent of the anisotropy of the respective lines. From these published works, it is observed that the theoretical value of the alignment parameter based on the nonrelativistic calculations of Berezhko *et al.* [18] are 2–3 times higher than the experimental values of Küst *et al.* [48], almost 5 times higher than the value of Papp and Campbell [33], and about half of the experimental values of Yamaoka *et al.* [47].

So the contradiction exists not only in the existence of anisotropy in the emission of *L* x rays but also in the magnitude of the alignment parameter of the ions. Our present investigation is aimed at giving a decisive answer to clear up this contradiction. For this purpose Au and U foils are excited with a ^{109}Cd source and the emitted *L* x rays are then measured at different angles.

II. EXPERIMENT

The geometrical setup of the angular distribution measurement is shown in Fig. 1. A ^{109}Cd point source of strength 30 mCi was used to excite the *L* x rays from the targets. Both the target and the exciting source were held fixed, and the Si(Li) detector used to detect the *L* x rays was placed on a rotatable stand. The angular coverage in this experiment was 70°–150° with respect to the direction of the exciting x rays from the source. The source was kept within a graded collimator made of Al, Cu, and Pb with an exit diameter of 2 mm. It was set at an angle of 60° with respect to the normal to the target. The ORTEC Si(Li) detector used in this

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TABLE I. A summary of all measurements done so far in chronological order on the angular distribution of *L* fluorescence x rays in photon-induced ionization.

Group	Radiation source	Targets	Observed anisotropy
Kahlon *et al.* [29]	^{241}Am (59.6 keV)	Pb (14.5–33 mg/cm^2), U	Strong
Kahlon *et al.* [30]	Same	Pb (14.5–33 mg/cm^2)	Strong
Kahlon *et al.* [31]	Same	Th, U	Strong
Kahlon *et al.* [32]	Same	Au (40 mg/cm^2)	Strong
Papp and Campbell [33]	X-ray tube (8.90 keV)	Er (1.06 mg/cm^2)	~5%
Ertuğrul *et al.* [34]	^{241}Am (59.6 keV)	Au, Hg	Strong
Ertuğrul *et al.* [35]	Same	U, Th, Bi	Strong
Ertuğrul [36]	Same	Au, Hg, Tl, Pb, Bi, Th, U (100–500 μg/cm^2)	Strong
Puri *et al.* [37]	^{109}Cd	ThF_4 (178 μg/cm^2)	Within expt. error
Mehta *et al.* [38]	^{241}Am (59.6 keV), ^{109}Cd (22.6 keV)	$UO_2(CH_3COO)_2.2H_2O$ (12.7 mg/cm^2), UF_4(188 μg/cm^2)	Within expt. error
Kumar *et al.* [39]	Same	Pb (243 μg/cm^2, 65.2 mg/cm^2)	Within expt. error
Sharma and Allawadhi [40]	^{241}Am with secondary targets	Th (0.0517 g/cm^2), U (0.3399 g/cm^2)	Strong
Demir *et al.* [41]	^{55}Fe	Pt, Au, Hg (0.3–0.9 mg/cm^2)	Strong
Kumar *et al.* [42]	^{241}Am with secondary	Pb	Within expt. error
Seven and Kocak [43]	^{241}Am (59.6 keV)	Pt, Re, W, Ta, Hf, Lu, Yb (50–200 μg/cm^2)	Strong
Seven and Koacak [44]	Same	U, Th, Bi, Pb, Tl, Hg, Au (50–200 μg/cm^2)	Strong
Yamaoka *et al.* [45]	Synchrotron (13.172, 14.172, 15.172, 15.672, 16.672 keV)	Pb (266.7 μg/cm^2), Au (483 μ g/cm^2)	Small
Demir *et al.* [46]	^{241}Am (59.6 keV)	Er, Ta, W, Au, Hg, Tl (113 μm)	Strong
Yamaoka *et al.* [47]	Synchrotron (13.002 keV)	Au (0.25 μm)	Small
Küst *et al.* [48]	Synchrotron (4.814, 5.055 keV)	Xe	Small
Tartari *et al.* [49]	^{241}Am (59.54 keV)	Yb, Hf, Ta, W, Pb (67–450 mg/cm^2)	Within expt. error
Seven [50]	Same	Th, U (500 μg/cm^2)	Strong

experiment had an energy resolution of 160 eV at 5.9 keV. All the signals were collected in a PC fitted with an OXFORD MCA card. Two targets—e.g., a Au foil (54 mg/cm^2) and a U foil (194 mg/cm^2)—have been used in this experiment.

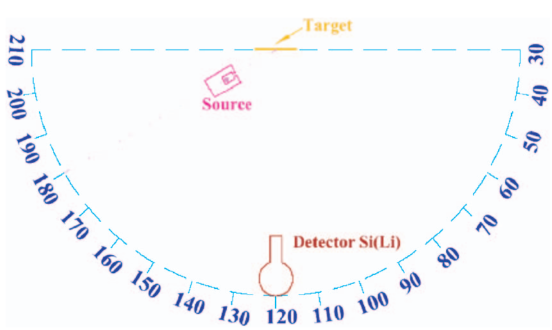

FIG. 1. (Color online) The geometrical setup of the angular distribution measurements.

To check any misalignment of the angular distribution table, the intensity of the $L\alpha$ line of Np (13.94 keV) emitted from an ^{241}Am source of strength of 1 μCi placed at the target position was measured with the Si(Li) detector over the whole angular range. The $L\alpha$ line is supposed to be isotropic. By plotting the line intensities against different angles, the measured distribution is found to be isotropic within 2%. This implies that with our system, we can measure anisotropy parameters greater than 2%.

Target foils were placed on an Al stand placed at the center of the table. Of all the *L* lines, *L*1 is the least intense but it is also a well-resolved singlet emitted from the *L*3 subshell having *J* value equal to 3/2. So alignment of the vacancy states can be best studied by observing the intensity of this line at different angles. Moreover, it has also been reported

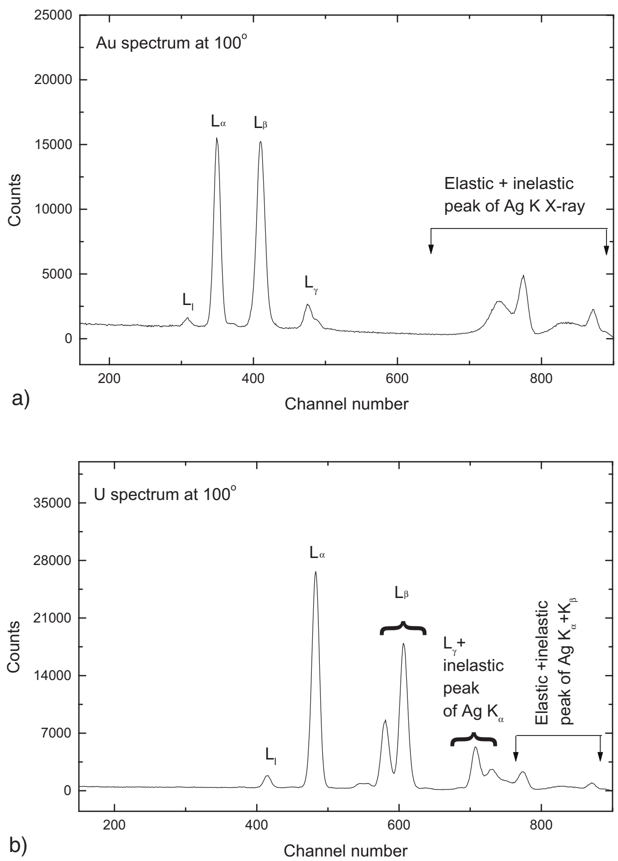

FIG. 2. (a) Au spectrum excited by 22.6-keV x rays and measured at 100° position. (b) same for U.

that the largest anisotropy was observed for the *L*1 line [33]. Data accumulation at each angular position was continued until the net area under the *L*1 line was at least 3000. This was done to make a compromise between the minimization of statistical error and the data accumulation time. Depending on the targets, data accumulation at each angular point varied from 3 to 18 h. Typical spectra of Au and U at 100° are shown in Figs. 2(a) and 2(b), respectively. These spectra show well-separated groups of the *L*1, $L\alpha$, $L\beta$, and $L\gamma$ x rays of Au and U along with the elastic and inelastic peak of Ag *K* X rays coming from the ^{109}Cd source.

III. ANALYSIS

The peak integrals of all the *L* lines were obtained using a peak fitting program called ACT [51]. It is a nonlinear least-squares fitting program which can handle multi-Gaussian peaks with a polynomial background. The widths of the peaks can be made variable, and the asymmetry of the peaks can also be incorporated. The error in the area of the *L*1 line through this fitting procedure for Au and U was ~2%–4% while for the other lines—e.g., $L\alpha$ and $L\beta$—it was <1%.

The line intensities obtained from the fit were corrected for the absorption of x rays inside the sample by dividing the intensity with the absorption correction factor. This factor was calculated numerically following procedures similar to that of Mandal *et al.* [52]. In this calculation, the target is considered to be divided into a large number of layers parallel to its surface. The incident photon beam, while passing through the target, suffers absorption in each of these layers and the interaction point for producing the x rays is considered to be at the middle of each layer. Contributions of the x rays reaching the detector from the midpoint of each layer were then added up, taking into account the absorption in their path. Total x-ray intensities reaching the detector were calculated in two ways: (i) by considering the absorption of x rays in the outgoing path and (ii) without considering the absorption. The ratio of the first to the second quantity gives us the absorption correction factor. All the fundamental parameters like photoelectric cross sections, fluorescence yield, Coster-Kronig transition probabilities, radiative widths, and absorption coefficients were taken from the standard literature [53–56]. The absorption correction factors obtained for the different *L* lines of Au and U are shown in Table II.

TABLE II. Absorption correction factor for different *L* lines of Au and U at different angles.

Target (thickness)	Angle with respect to beam direction	Absorption correction factors		
		*L*1	$L\alpha$	$L\beta$
Au (54 mg/cm^2)	70°	0.29	0.37	0.47
	80°	0.33	0.41	0.51
	90°	0.36	0.44	0.54
	100°	0.38	0.46	0.56
	110°	0.39	0.47	0.58
	120°	0.39	0.47	0.58
U (194 mg/cm^2)	70°	0.47	0.57	0.64
	80°	0.51	0.61	0.68
	90°	0.54	0.64	0.70
	100°	0.56	0.65	0.72
	110°	0.57	0.67	0.73
	120°	0.58	0.67	0.73

The errors in the intensities of the *L*1, $L\alpha$, and $L\beta$ lines were compounded from the fitting errors and errors arising from the absorption correction term. Considering the error in the absorption correction factor to be ~3%, the overall errors in the intensity of the *L*1 line of Au and U were ~4%–5% while for the $L\alpha$ and $L\beta$ lines it was ~3% for both targets.

IV. RESULTS AND DISCUSSION

Assuming that the emission of x rays is like that of an electric dipole, the intensity *I* of a particular line measured at an angle θ can be written as

$$I = (I_0/4\pi)[1 + \beta P_2(\cos\theta)], \quad (1)$$

where β is the anisotropy parameter and is given by

$$\beta = \kappa\alpha A. \quad (2)$$

Here, κ is the correction factor connected with Coster-Kronig transitions, α is the parameter which depends on the

TABLE III. Time normalized intensities of different L lines of Au and U after self-absorption correction.

Target	Angle with respect to beam direction	Time normalized intensity (including absorption correction factor)		
		$L1$	$L\alpha$	$L\beta$
Au	70°	0.30	5.80	5.67
	80°	0.29	4.96	4.88
	90°	0.27	5.65	5.56
	100°	0.26	5.57	5.49
	110°	0.27	5.64	5.60
	120°	0.29	5.42	5.29
	130°	0.27	5.07	5.00
	140°	0.29	5.08	5.01
U	70°	1.00	16.3	10.1
	80°	1.01	15.9	10.1
	90°	0.95	15.6	10.2
	100°	0.96	15.6	10.2
	110°	0.94	15.4	10.2
	120°	0.92	15.4	10.2
	130°	0.95	15.5	10.1
	140°	0.97	15.5	10.1
	150°	0.96	15.5	10.0

final and initial states of the electron involved in the transition, A is the alignment parameter, $P_2(\cos\theta)$ is the second-order Legender polynomial, θ is the angle between the direction of the exciting beam and the detector, and I_0 is the total emitted intensity.

Table III shows our data for the $L1$, $L\alpha$, and $L\beta$ lines of Au and U measured at nine different angles. Different groups [29, 31, 32, 34, 35, 38, 40, 44, 46, 50] have published the cross sections of these lines at different angles. To have a comparison of our data with those of others, a normalization factor was determined by comparing the common angle data point from each of the data sets with that of ours. Such normalized data are shown in Figs. 3 and 4. In this experiment we covered an angular range from 70° to 150° at an interval of 10°. The other groups have covered even greater ranges—e.g., from 40° to 150° with different step intervals. Within this range, the value of $P_2\cos(\theta)$ used in Eq. (1) has the same value at all symmetrical pairs of points about 90° such as at (40°, 140°), (50°, 130°), (60°, 120°), (70°, 110°), and (80°, 100°). This means that the intensity of the x-ray line measured at these symmetrical pair of points should be the same within experimental errors.

From Fig. 3(a) it is observed that the Au $L1$ data sets of Kahlon *et al.*[32], Ertuğrul *et al.* [34], Demir *et al.* [46], and Seven and Koçak [44] show no sign of such symmetrical behavior at (40°, 140°), (50°, 130°), (60°, 120°), (70°, 110°), and (80°, 100°). The intensity of this line, as observed from this figure, decreases very sharply with increasing angle which is the most surprising part of these measurements. Our measured points on this graph do not show any systematic decrease of the intensity with an increase of angle; rather, they show a very slight angular dependence. Figure 4(a) shows similar plots for the U $L1$ line. A sharp decrease of the intensity with an increase of angle was also seen from the data of Kahlon *et al.* [31], Ertuğrul *et al.* [35], Seven [50], and Sharma and Allawadhi [40]. Four data points at angles 70°, 80°, 100°, and 140° of Mehta *et al.* [38] actually coincide with the data of ours where no such strong angular dependence was observed.

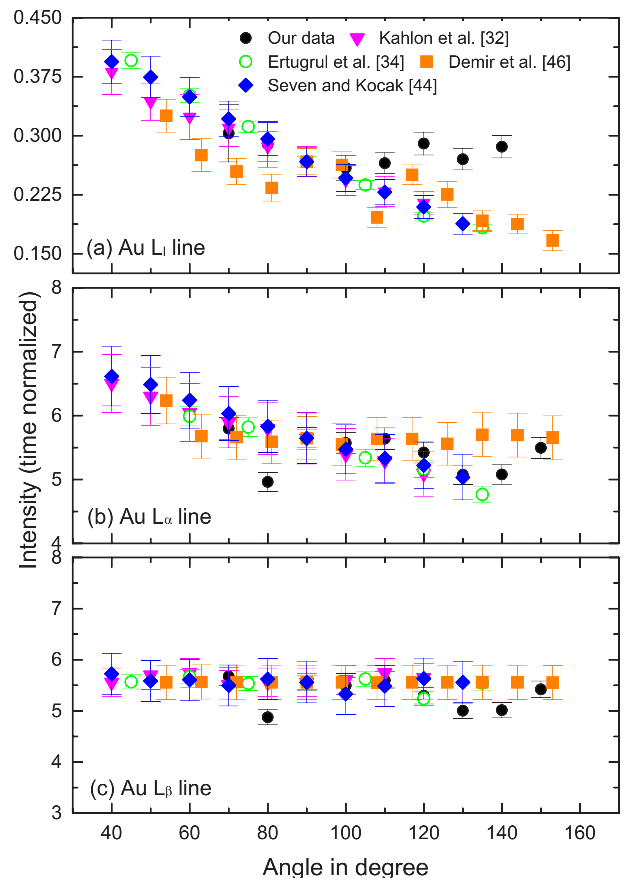

FIG. 3. (Color online) (a) Intensity of the $L1$ line of Au plotted against different angular positions. All the data are normalized at 90°. (b) Same for the $L\alpha$ line. (c) Same for the $L\beta$ line.

Figure 3(b) shows the angular distribution data of the Au $L\alpha$ line. Except for the data points of Demir *et al.* [46], all the other data sets show a similar tendency as was shown by the $L1$ line—i.e., a sharp systematic decrease with an increase of angle. No such systematics was found in our data but a weak dependence on angle may be observed. The angular distribution of the U $L\alpha$ line is shown in Fig. 4(b). Exactly similar behavior—i.e., a decrease of intensity with an increase of angle—was also observed with the data of Kahlon *et al.* [31], Ertuğrul *et al.* [35], Seven [50], and Sharma and Allawadhi [40]. In case of Sharma and Allawadhi [40], their 60° data points went even beyond the scale of the present plot and so this point is not shown in Fig. 4(b). Our data and the data of Mehta *et al.* [38] do not show such behavior. They seem to be isotropic within the experimental errors. Papp and Campbell [33] stated that such a large anisotropy in the $L1$ and $L\alpha$ lines, as observed by the

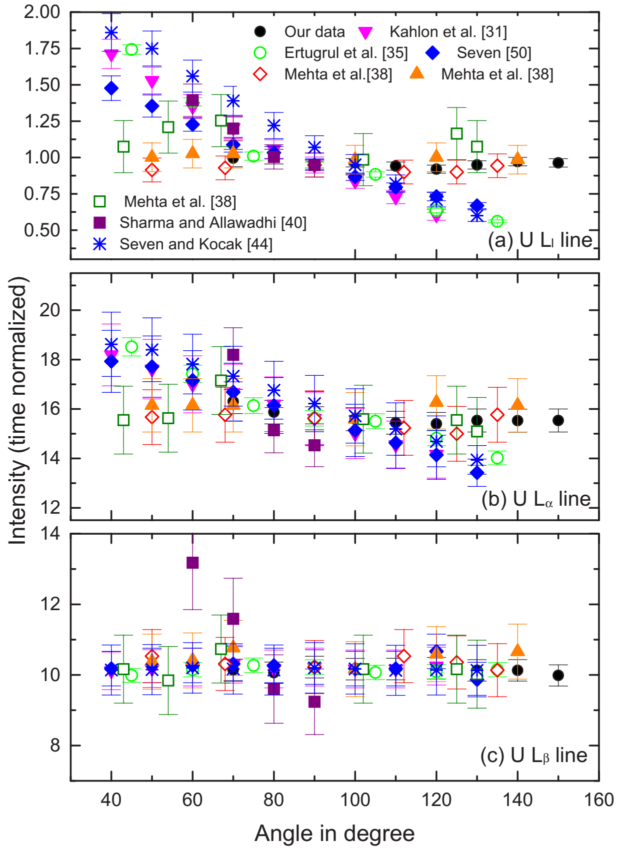

FIG. 4. (Color online) (a) Intensity of the *L*1 line of U plotted against different angular positions. All the data are normalized at 90°. (b) Same for the *L*α line. (c) Same for the *L*β line.

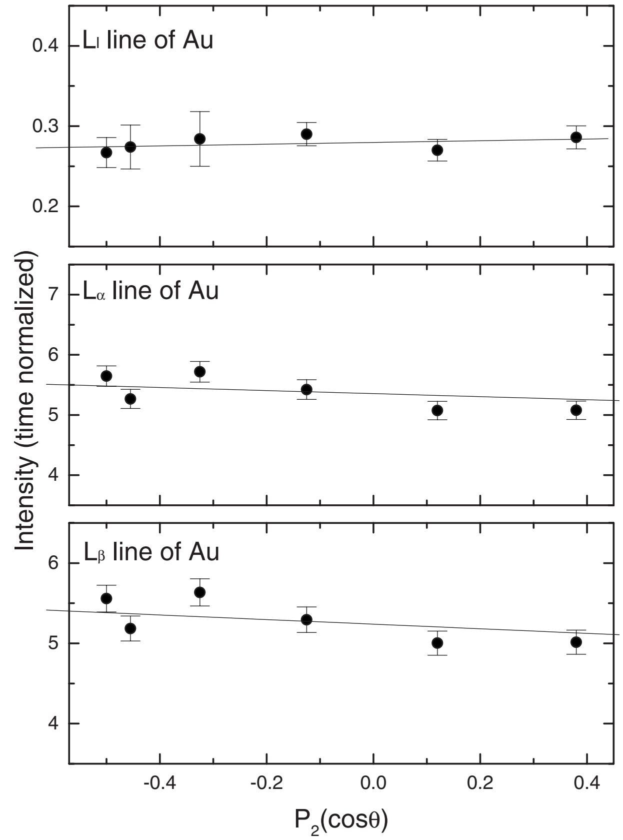

FIG. 5. (a) Intensity of the *L*1 line of Au plotted against $P_2(\cos\theta)$. (b) Same for the *L*α line. (c) Same for the *L*β line.

group [29–32] at Patiala, India and some groups [34–36,43,44,46,50] in Turkey, are connected with the absence of cylindrical symmetry. Kumar *et al.* [42] fitted the data of Kahlon *et al.* [30] with an extra $P_1(\cos\theta)$ term and observed that the value of the coefficient of that term is very high, indicating the existence of large anisotropy.

As far as the angular distribution of the $L\beta$ line is concerned, data from all the groups except the U data of Sharma and Allawadhi [40] show the same behavior—i.e., almost isotropic as is shown in Figs. 3(c) and 4(c).

Of all the photon-induced angular distribution studies, only Papp and Campbell [33] obtained the value of the anisotropy parameter β for the different L lines of Er excited by Cu $K\beta_1$. In order to get a quantitative idea of the anisotropy parameters of the different L lines of Au and U, we plotted our measured intensities against the $P_2(\cos\theta)$ values which are shown in Figs. 5 and 6. Considering the error in each data point, a straight-line fit was made. Our symmetrical data points at (70°, 110°) and (80°, 100°) have been averaged in these graphs. As per Eq. (1), from the intercept and slope of the fitted straight line, the values of the anisotropy parameter β for the $L1$, $L\alpha$, and $L\beta$ were calculated and these are shown in Table IV. Errors in the β values were calculated from the errors in the intercept and slope. As the absolute value of β in this energy domain is very small ($\lesssim 5\%$), the relative error in β is almost 100%. This is true wherever the measured β values are very small [6,33]. It has been mentioned earlier that the geometrical anisotropy of the present setup was ~2%. So the lines showing an anisotropy of 2% or less as observed in the case of U may be considered as isotropic. The values of β for the $L1$, $L\alpha$, and $L\beta_{2,15}$ of Er measured by Papp and Campbell [33] with photons of 8.904 keV are 0.052 ± 0.016, 0.016 ± 0.022, and 0.012 ± 0.015, respectively. Our experimental values of β cannot be compared with those of Papp and Campbell [33] because they used a different target and a different excitation energy.

It may be observed from Eq. (2) that the alignment parameter A is connected to β through two terms κ and α. When the exciting radiation has sufficient energy to excite all the L subshells, some of the vacancies from $L1$ and $L2$ subshells may be transferred to $L3$ via Coster-Kronig transitions. Ions with their vacancies transferred to the $L3$ subshell in this manner will be nonaligned. So the x rays emitted from such ions will dilute the effect of alignment of the ions produced by direct ionization in the $L3$ subshell. Accordingly, the measured alignment parameter should be corrected by a factor κ given by [6]

$$\kappa^{-1} = 1 + f_{23}\sigma_2/\sigma_3 + (f_{13} + f_{12}f_{23})\sigma_1/\sigma_3.$$

Here, f_{12}, f_{13}, f_{23} are the different Coster-Kronig transition probabilities of different L subshells [54] and σ_1, σ_2, and σ_3 are the photoelectric cross sections of the $L1$, $L2$, and $L3$ subshells [53].

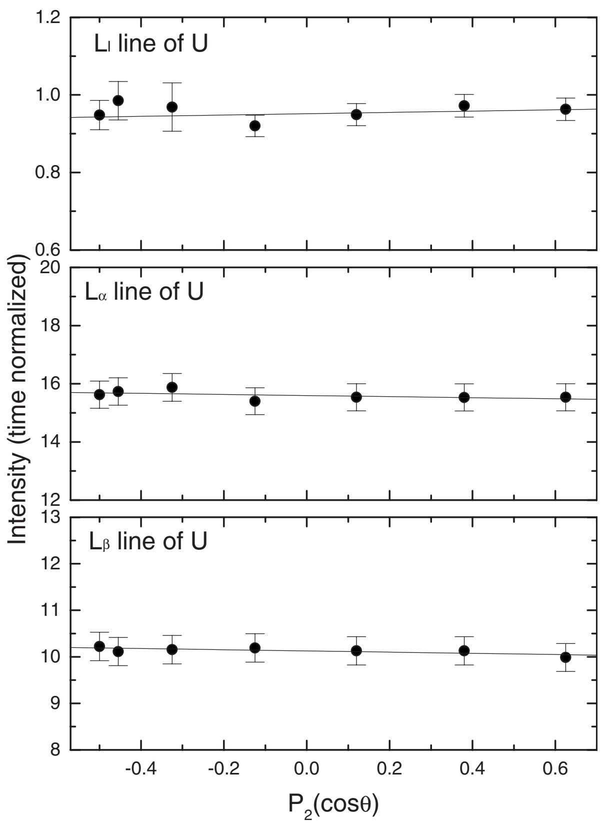

FIG. 6. (a) Intensity of the $L1$ line of U plotted against $P_2(\cos\theta)$. (b) Same for the $L\alpha$ line. (c) Same for the $L\beta$ line.

It is well known that the ^{109}Cd radioactive source has a 4% probability of emitting 88.03-keV γ rays. These γ rays can ionize the K shell of Au, and through the emission of the $K\alpha_1$ line, a K-shell vacancy can be transferred to the $L3$ subshell, thereby producing a negative effect on the alignment. A rough calculation shows that such a transfer of vacancy is about 0.5% compared to the vacancies produced by the direct ionization of the $L3$ subshell of Au by 22.6-keV x rays emitted from the ^{109}Cd source. So no correction for such an effect has been done here. For U, its K-shell binding energy is above 88.03 keV and so such effect does not occur.

The parameter α is defined as

$$\alpha = \sqrt{\frac{3}{2}(2J_1+1)}(-1)^{J_1+J_2+1}\begin{Bmatrix} 1 & J_1 & J_2 \\ J_1 & 1 & 2 \end{Bmatrix},$$

where J_1 and J_2 are the angular momenta of the initial and final states of the electron and the standard notation is the $6j$ symbol. The values of α can be obtained from Berezhko and Kabachnik [3].

Ions which will be aligned due to having a vacancy in the $L3$ subshell ($J=3/2$) can be studied by detecting the lines originating from that subshell—i.e., $L1$ or $L\alpha$ or some of the $L\beta$ lines. But due to the poor resolution of the Si(Li) detector, the components of $L\alpha$ or $L\beta$ cannot be resolved, and so to obtain information on the alignment parameter, the best way would be to concentrate on the analysis of only the $L1$ line which is a singlet. Using the β values of the $L1$ line from Table IV and taking the proper values of κ and α, the final value of the alignment parameter for Au was obtained as 0.10 ± 0.14. As the anisotropy parameter of the L lines of U are 2% or less no effort was made to calculate the alignment parameter of the U ions. Yamaoka *et al.* [47] obtained the values of the A parameter of Au ions as -0.21 ± 0.04 by exciting the target with 13-keV photons. Our excitation energy is different from that of Yamaoka *et al.* [47], and so we are unable to compare our results with their values.

TABLE IV. The values of the anisotropy parameter (β) for the $L1$, $L\alpha$, and $L\beta$ lines of Au and U.

Target	Line	β
Au	$L1$	0.04 ± 0.056
Au	$L\alpha$	-0.05 ± 0.049
Au	$L\beta$	-0.04 ± 0.042
U	$L1$	0.02 ± 0.026
U	$L\alpha$	-0.01 ± 0.008
U	$L\beta$	-0.01 ± 0.004

V. CONCLUSION

By plotting the intensities of the $L1$, $L\alpha$, and $L\beta$ lines of Au and U against different angles ($70° \leq \theta \leq 150°$), it was observed that for $L1$ and $L\alpha$ lines, *no sharp decrease* of intensity of these lines is observed which *contradicts* the findings of the group of Patiala, India [29–32] and of Turkey [34,35,43,44,46,50]. Here, we have observed *a very slight decrease* of the intensities with an increase of the angles which *supports* the observation made by Mehta *et al.* [38], Yamaoka *et al.* [45], and Yamaoka *et al.* [47]. As far as $L\beta$ lines are concerned, an isotropic behavior was observed which was also noticed by other groups except Sharma and Allawadhi [40].

The magnitude of the anisotropy parameters was also obtained by plotting the intensities of the $L1$, $L\alpha$, and $L\beta$ lines against $P_2\cos(\theta)$. The values of the anisotropy parameter (β) of the $L1$, $L\alpha$, and $L\beta$ lines of Au vary from 4% to 5%. For U, these values vary from 1% to 2%. As we have mentioned before, the geometrical anisotropy of our experimental setup was ~2%, U L x-ray lines with a measured anisotropy of 1%–2% may be considered isotropic.

From the the β value of the $L1$ line of Au, the alignment parameter of Au ions was calculated and found to be 0.10 ± 0.14

In conclusion, we can categorically say that the anisotropy of the L x-ray lines of high-Z elements in photon excitation is small ($\leq 5\%$). This is the decisive answer that the present investigation clearly provides.

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