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Anomalous gamma-ray–L-x-ray directional correlations in ^{160}Dy

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An anisotropic directional correlation between gamma rays and L x rays following internal conversion is reported for the first time. Using four detectors, two coaxial and two planar Ge detectors, the spectrum of L x rays was recorded in coincidence with the 1178 keV gamma ray in the decay of ^{160}Tb. The resulting anisotropies are $A(1178\gamma - L1) = -0.03(3)$, $A(1178\gamma - L\alpha) = 0.009(5)$, $A(1178\gamma - L\beta) = 0.003(3)$, and $A(1178\gamma - L\gamma) = 0.00(1)$. The measured anisotropy for the $L\alpha$ peak is in significant disagreement with the theoretical prediction $A(1178\gamma - L\alpha) = -0.003$.

[RADIOACTIVITY ^{160}Tb, measured γ-Lx(θ), deduced anisotropy, Ge(Li) detectors.]

I. INTRODUCTION

The existence, in certain cases, of an anisotropic directional correlation between electromagnetic quanta emitted in two cascading transitions was experimentally established more than 30 years ago. Brady and Deutsch[1] reported anisotropic correlations between coincident gamma quanta emitted in the decays of ^{60}Co and ^{46}Sc. The method of gamma-gamma directional correlations has since become a widely used tool in nuclear spectroscopy.[2] In exact analogy with the case of gamma rays the directional correlation of two x-ray quanta originating from cascading transitions can be anisotropic provided the intermediate atomic state has a spin $J > \frac{1}{2}$ (Ref. 3).

The possibility of anisotropic directional correlations between x rays and gamma rays was first considered by Benoist.[4] Such correlations can be expected if the nuclear and the atomic transitions are cascading, i.e., are causally related through a process that produces changes in both nuclear and atomic states like orbital electron capture or internal electron conversion (Fig. 1). However, a requirement is that the spins of the intermediate nuclear (J_1) and atomic (j_0) states be greater than $\frac{1}{2}$. The lowest atomic state which we have to consider, therefore, is the L_3 subshell ($j_0 = \frac{3}{2}$).

The theory of gamma-L-x-ray directional correlations following orbital electron capture and internal electron conversion was originally formulated by Dolginov.[5] It has later been reinvestigated by Rupnik and Crasemann,[6] Piticu and Vata,[7] and most recently by Carvalho *et al.*[8]

The first experiment designed to detect a possible anisotropy in the directional correlation of gamma rays and L x rays was reported by Halley and Engelkemeir[9] who studied the decay of ^{238}Pu. An upper limit of 2% was assigned to the anisotropy. Then Maesel[10] measuring the directional correlation of L x rays and gamma rays following the allowed electron capture decay of ^{139}Ce reported anisotropy of 0.153(49). This experiment has been repeated by Behar *et al.*[11] using a Si(Li) detector for the detection of x rays. They reported an anisotropy of less than 0.018.

The directional correlation between L x rays and the 570 keV gamma ray in the decay of ^{207}Bi has been measured by Rupnik and Crasemann[6] and by Cambiaggio *et al.*[12] In both cases the correlation was reported isotropic within the experimental limits of error.

In the following we will often refer to the resolved peaks in the L x ray spectrum. The Ll peak corresponds to the transition $L_3 \rightarrow M_1$. The composite $L\alpha$ peak corresponds to the two transitions $L_3 \rightarrow M_4$ and $L_3 \rightarrow M_5$. The highly complex $L\beta(L\gamma)$ peak corresponds to several transitions from the L_1, L_2, and L_3 (L_1 and L_2) subshells to higher lying shells.

II. MEASUREMENTS

A. Source

A difficulty, when trying to detect an anisotropy in the directional correlation between gamma rays and x rays emitted in transitions to the L_3 subshell, is the presence of isotropic x rays due to internal conversion in or electron capture from the K, L_1, or L_2 shells followed by undetected transitions to the L_3 subshell. In order to minimize such a masking of the anisotropy we selected a case to investigate by the following criteria:

(i) The investigated cascade should be fed in a β^--decay.

(ii) One of the coincident nuclear transitions

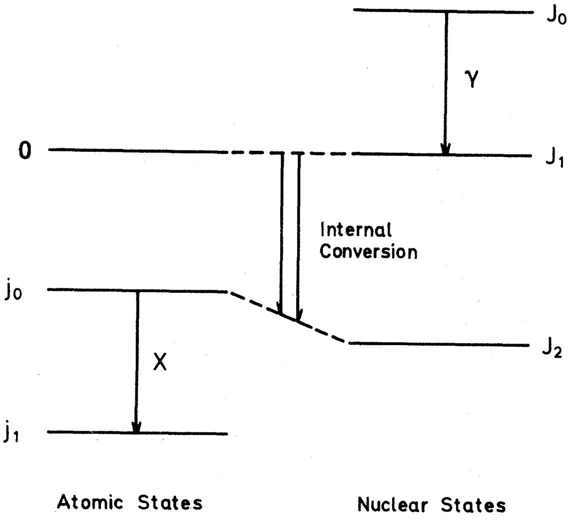

FIG. 1. Related nuclear and atomic transitions.

should be strongly L_3 converted.

(iii) The transition producing the gamma ray should not be coincident (or only weakly coincident) with any other strongly converted transition.

The (2^-) 1178 keV (2^+) 87 keV (0^+) cascade in ^{160}Dy fed in the decay of ^{160}Tb, meets all of the above requirements (Fig. 2). Disadvantages are the comparatively low energy of the L x rays (5.7–9.0 keV) and the long half-life of the intermediate nuclear level [$t_{1/2}$(87 keV) = 2.0 ns].

In order to minimize the self-absorption of L x rays a very thin source of solid $TbCl_3$ was used. Due to hyperfine interactions in the crystalline source in combination with the half-life of the intermediate level the directional correlation of the 1178–87 keV cascade was attenuated by approximately 0.5 (below). Trials were made with different chemical composition of the source, but no more favorable one was discovered.

B. Apparatus

The main part of the measurements was done using a setup where the spectra in two high resolution Ge(Li) detectors (175 and 250 eV full width at half maximum (FWHM), respectively, at 5.9 keV) were recorded in twofold coincidence with two 10% efficiency Ge(Li) detectors (see Fig. 3). The angles between the possible combinations (4) of pairs were 90° and 180°.

The setup included two analog to digital converters set to 1024 channels the outputs of which were routed to different parts of a memory depending on which pair of detectors was involved. This resulted in four spectra of true plus random coincidences and four additional spectra of only random coincidences. Determination of an event being true plus random or random was done by real time analysis of the output from the single time to pulse height converter. A total time window width of 125 ns was used throughout the entire series of measurements. This time window allowed all true coincidences, in the two high resolution Ge(Li) detectors, from less than 5 keV to more than 100 keV, to be recorded with >99 per cent efficiency (See Fig. 4).

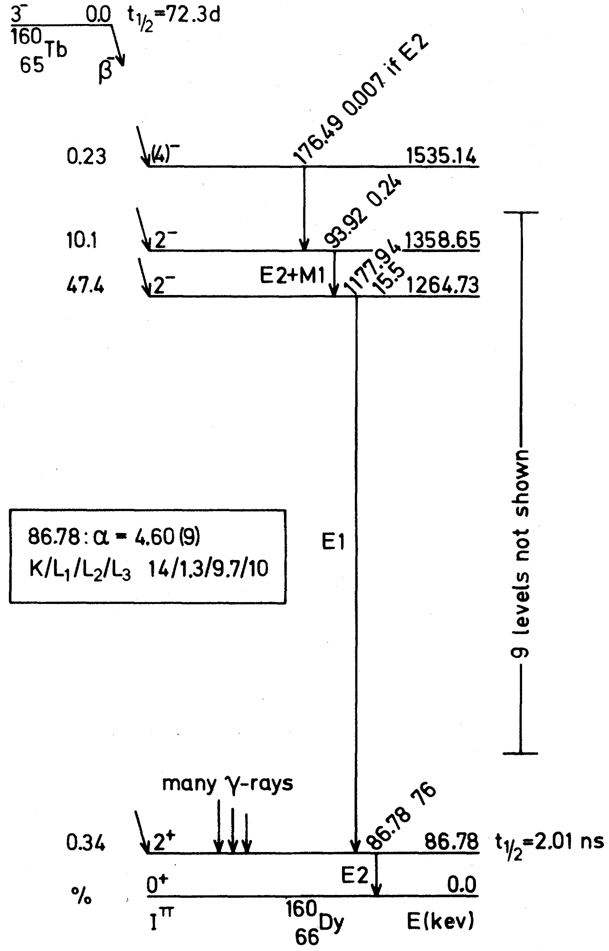

FIG. 2. Simplified decay scheme of ^{160}Tb (Ref. 14).

The source was made by allowing a solution of $TbCl_3$ in hydrochloric acid to dry on the flat end of a plastic rod 2 mm in diameter. The rod was mounted in the plane of the detectors at an angle of 45° to all four detectors and with the source facing the x-ray detectors. The distances between the detectors and the source were chosen such that the effective solid angles of the detectors were equal.

Some of the measurements were done using a two detector setup, with an automatically movable

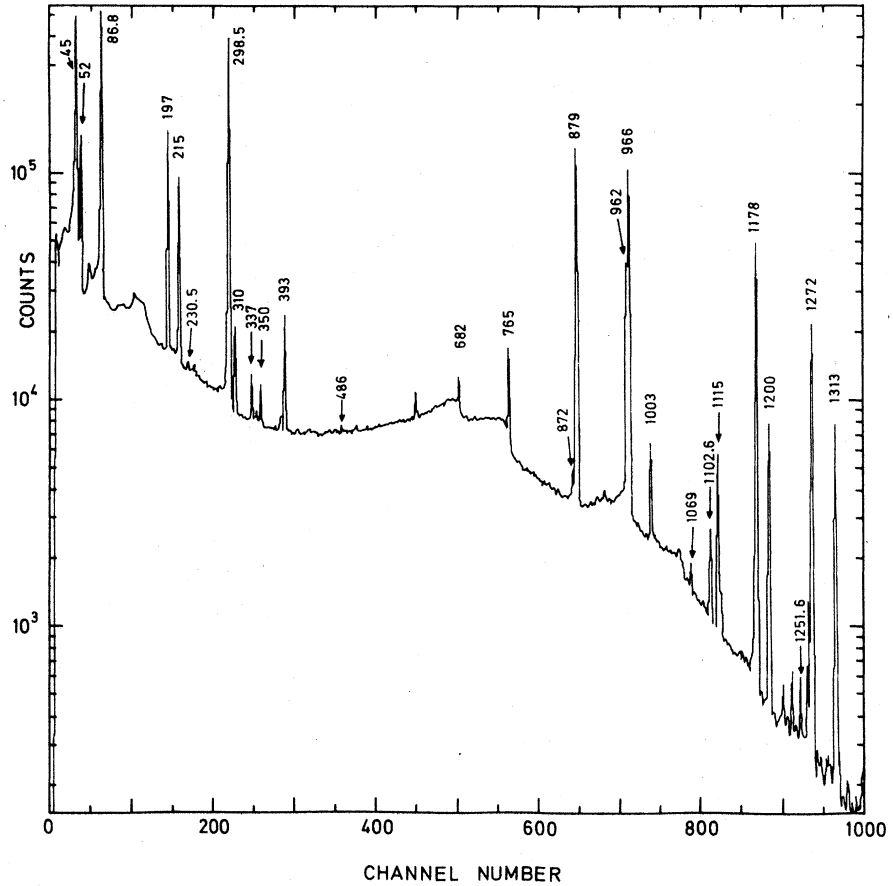

FIG. 3. Gamma-ray spectrum in one of the 10% efficiency detectors. All identified gamma rays belong to the decay of ^{160}Tb.

high resolution Ge(Li) detector in coincidence with a 10% efficiency Ge(Li) detector which was fixed with respect to the source. The high resolution detector was shifted every 2000 s between two positions, 90° and 180°, to minimize possible systematic errors. The corresponding spectra of true plus random and random coincidences were recorded in different memories as in the four detector system. Simultaneously, the singles x-ray spectrum was recorded for normalization purposes.

C. Analysis and results

Assuming the efficiency of the coincidence circuit to be a constant independent of detector pair then the number of coincidences registered between the gamma-ray detector i and the x-ray detector j is given by

$$N(E_\gamma, E_x, \theta)_{ij} = N_0 \epsilon_i(E_\gamma)\epsilon_j(E_x)W(\theta) ,$$

where N_0 is a normalization constant, $\epsilon_i(E_\gamma)$ and $\epsilon_j(E_x)$ are the (energy dependent) detector efficiencies, and the directional correlation function

$$W(\theta) = \sum_{k=\text{even}} A_{kk}P_k(\cos\theta) \qquad (1)$$

is a series of Legendre polynomials where θ is the angle between the detectors. In this expression the detectors are assumed to be pointlike.

The main body of data was obtained with the four detector arrangement described above. Denoting the gamma-ray detectors 1 and 2 and the x-ray detectors 3 and 4 the anisotropy may be written as

$$A \equiv \frac{W(180°)}{W(90°)} - 1 = \left[\frac{N(180°)_{13}N(180°)_{24}}{N(90°)_{14}N(90°)_{23}}\right]^{1/2} - 1 .$$

The advantage with this expression is that the unnormalized coincidence rates can be used directly.

For each run, lasting between one and four weeks, the spectra of random coincidences were first subtracted channel by channel from the corresponding spectra of true plus random coin-

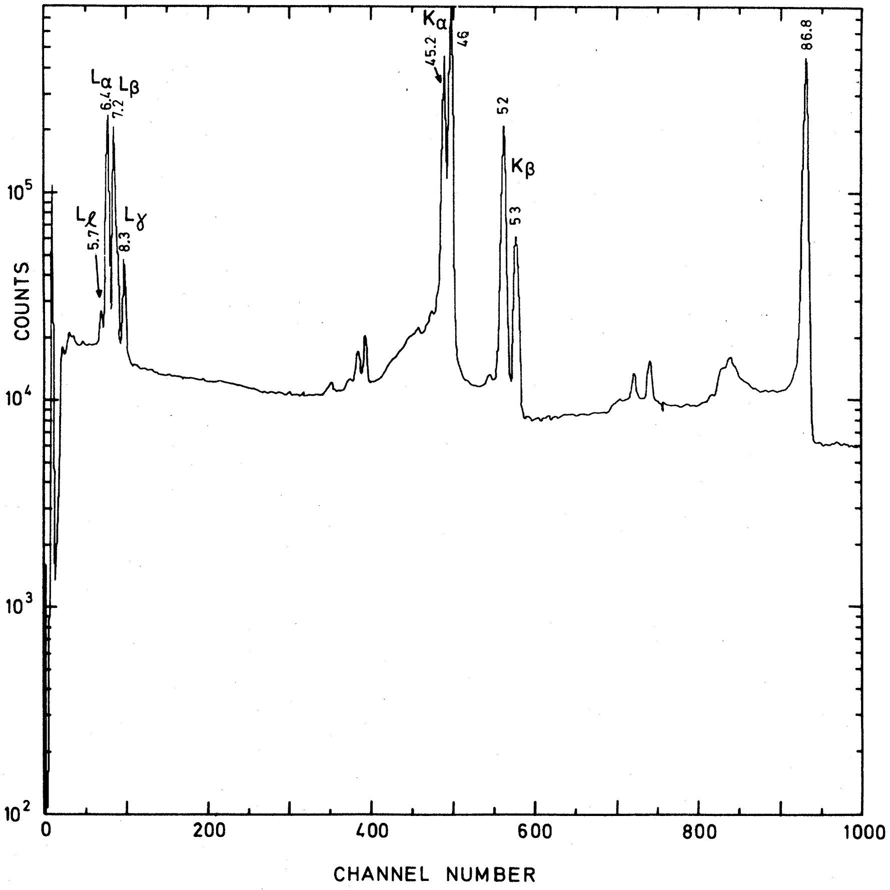

FIG. 4. Low energy spectrum recorded with one of the high resolution detectors.

cidences. Background intervals of about 700 eV width were then chosen immediately below the *Ll* peak and immediately above the *Lγ* peak. An assumed linear background was interpolated between the two intervals and subtracted from underneath the *L*-x-ray peaks. The squared coincidence ratio $N(180°)_{13}N(180°)_{24}/N(90°)_{14}N(90°)_{23}$, was then computed for the *Ll*, *Lα*, *Lβ*, and *Lγ* peaks and for the two background intervals. In order to detect a possible asymmetry in the apparatus the positions of the two gamma-ray detectors were interchanged after 5 out of a total

TABLE I. Anisotropies $[A = W(180°)/W(90°) - 1]$ measured with the four detector apparatus.

	Configuration 1 (5 runs)	Configuration 2 (7 runs)	Average of (12 runs)
$A(1178\gamma\text{-}Ll)$	−0.007(41)	−0.053(46)	−0.029(31)
	$\chi^2/4 = 2.1$	$\chi^2/6 = 1.0$	$\chi^2/11 = 1.3$
$A(1178\gamma\text{-}L\alpha)$	0.0163(48)	0.0072(49)	0.0118(34)
	$\chi^2/4 = 2.4$	$\chi^2/6 = 2.4$	$\chi^2/11 = 2.4$
$A(1178\gamma\text{-}L\beta)$	0.0078(49)	−0.0022(43)	0.0022(32)
	$\chi^2/4 = 0.4$	$\chi^2/6 = 0.7$	$\chi^2/11 = 0.7$
$A(1178\gamma\text{-}L\gamma)$	−0.009(15)	0.010(18)	−0.001(12)
	$\chi^2/4 = 0.7$	$\chi^2/6 = 1.4$	$\chi^2/11 = 1.1$
A(1178γ-bgr left)	0.011(16)	0.016(14)	0.014(11)
	$\chi^2/4 = 1.6$	$\chi^2/6 = 0.6$	$\chi^2/11 = 0.9$
A(1178γ-bgr right)	0.025(16)	0.019(15)	0.022(11)
	$\chi^2/4 = 0.7$	$\chi^2/6 = 1.0$	$\chi^2/11 = 0.8$

of 12 runs. For the last 7 runs

$$A=\left[\frac{N(180°)_{14}N(180°)_{23}}{N(90°)_{13}N(90°)_{24}}\right]^{1/2}-1\,.$$

For each of the configurations the weighted average of the squared coincidence ratios was calculated for each of the six intervals and from these the corresponding anisotropies. Using as weights the inverse statistical variances the results in Table I were obtained. The slight positive anisotropy of the background (bgr) is probably due to the Compton distribution from 87 keV quanta.

From the independent results of the two sets of runs no large asymmetry is observed. However, the somewhat large value of the reduced chi square of the data for the $L\alpha$ peak, could indicate the presence of a fluctuating source of error other than purely statistical. We therefore multiply the errors quoted in the last column of Table I by $(\chi^2/11)^{1/2}$ wherever this quantity is >1. Now adding to this set of data the results of four runs with the two detector set up (above) where the coincidence spectra were normalized with respect to the corresponding singles spectra in the moving x-ray detector, we obtain as a final result the values in the second column of Table II. The results are based on a total of approximately one million coincidences in the peaks.

In the four detector apparatus the spectrum was registered up to approximately 90 keV in one of the x-ray detectors. Normalizing the number of coincidences in the peak corresponding to the 87 keV gamma ray to the $K\alpha$ peak (assumed to be isotropic) the anisotropy of the 1178-87 keV gamma-gamma cascade could be measured simultaneously. The average obtained after 15 runs was $A(1178\gamma-87\gamma)=0.161(3)$, $\chi^2/14=1.0$.

III. THEORY

In the formulation of Carvalho *et al.*[8] the directional correlation coefficients A_{kk} in (1) are given by

$$A_{kk}=A_k(L_\gamma,L'_\gamma,J_0,J_1)A_k(L_x,L'_x,j_1,j_0)\times U_k(L_{IC},L'_{IC},J_1,J_2,j_0)\,.$$

Here $A_k(L_\gamma,L'_\gamma,J_0,J_1)$ is defined in the usual way[2] in terms of the multipole mixing ratio δ and $A_k(L_x,L'_x,j_1,j_0)$ is the same quantity for the x-ray transition. (The notation is the same as in Fig. 1.) The entire dependence on the internal conversion process, coupling the nuclear and atomic systems, is contained in the factor $U_k(L_{IC},L'_{IC},J_1,J_2,j_0)$ which involves radial integrals of the electron wave function. To compute the factor U_k we used expressions (12) and (13) in the work of Carvalho *et al.*, interchanging J_1 and J_2 on the right-hand side in (13). [In their formula (13) Carvalho *et al.* have erroneously interchanged J_1 and J_2. This formula, as it stands, is therefore appropriate for the case where the converted transition proceeding between nuclear spins J_1 and J_2 precedes the gamma transition.] The electron wave functions and the radial integrals were calculated using a computer code by Pauli and Raff.[13] Our result was $U_2=0.395$.

TABLE II. Measured anisotropies (final result) and theoretical predictions.

	Experiment	Theory
$A(1178\gamma\text{-}Ll)$	−0.03(3)	−0.027
$A(1178\gamma\text{-}L\alpha)$	0.009(5)	−0.0026
$A(1178\gamma\text{-}L\beta)$	0.003(3)	−0.0008
$A(1178\gamma\text{-}L\gamma)$	0.00(1)	0

The comparatively long lifetime of the intermediate 87 keV level resulted in an attenuation of the directional correlation of the 1178-87 keV cascade. The attenuation is caused by hyperfine interactions leading to a loss of alignment in the intermediate state. The time integrated attenuation can be expressed by attenuation factors G_k multiplying the gamma-gamma directional correlation coefficients.[2] In our case the atomic K and L shells are fully occupied while the nuclear system is in the intermediate state. The effect on the gamma–x-ray directional correlation should therefore be a similar attenuation caused by the same loss of nuclear alignment and consequently should be described by the same attenuation factors.

For the attenuated directional correlation of the 1178-87 keV cascade

$$W(\theta)=1+G_2Q_2(87)A_2(87)Q_2(1178)A_2(1178)P_2(\cos\theta)\,,$$

where we have included the factors correcting for the finite solid angle of the detectors Q. The term with $k=4$ is small and is not included.[14] Assuming the 87 keV transition to be of pure $E2$ multipolarity $A_2(87)=-0.5976$. From our measured anisotropy, $A(1178\text{-}87)=0.161(3)$, we compute

$$G_2Q_2(87)Q_2(1178)A_2(1178)=-0.171(4)\,.$$

Now assuming that the attenuation is the same for the gamma–L-x-ray cascade and because the effective solid angles of the x-ray detectors are very nearly independent of energy we rewrite expression (1) for the directional correlation we expect to observe

$$W(\theta)=1+G_2Q_2(1178)A_2(1178)Q_2(X)A_2(X)U_2P_2(\cos\theta)\,,$$
$$Q_2(X)=Q_2(87) \tag{2}$$

where all factors are determined except $A_2(X)$.

The $L\alpha$ and $L\beta$ peaks (Fig. 5) are composite and contain several components corresponding to transitions to the L_3 subshell with different final spin j_1. Weighting the individual $A_2(X)$ coefficients with theoretical x-ray intensities[8,15] and including the effect of isotropic contributions following internal conversion in the K, L_1, and L_2 shells[3,8] we obtain $A_2(Ll)=0.27$, $A_2(L\alpha)=0.027$, $A_2(L\beta)=0.008$, and $A_2(L\gamma)=0$.

We are now in a position to evaluate expression (2) and obtain the anisotropies expected from theory for the directional correlation of 1178 keV gamma rays and the spectrum of L x rays. The expected anisotropies are compared to the ones measured in Table II.

IV. DISCUSSION

There is a discrepancy, established at a 96% level of confidence, between the measured anisotropy for the $L\alpha$ peak and the theoretically predicted value. The small anisotropy measured for the $L\beta$ peak provides a strong argument against an explanation for the discrepancy in terms of scattering between the detectors or some other systematic error introduced by the measuring equipment. We have analyzed our coincidence data by normalizing the number of coincidences in the Ll, $L\alpha$, and $L\gamma$ peaks to the number of coincidences in the $L\beta$ peak. The resulting anisotropies should be compared to the difference between the anisotropies predicted for the corresponding peaks and should be free of any systematic bias. Such a comparison is made in Table III. The discrepancy between the measured and the predicted difference in anisotropy for the $L\alpha$ and $L\beta$ peak is established at a 95% level of confidence. The low value for the reduced chi square as compared to those in Table I indeed shows that there are some correlated fluctuations in the coincidence data for the $L\alpha$ and $L\beta$ peaks. However, there is no evidence for a systematic bias in the original data.

In a special run with the four detector apparatus the gate was set to accept the background immediately below the 1178 keV peak (in energy). The results rule out the presence of a coincident

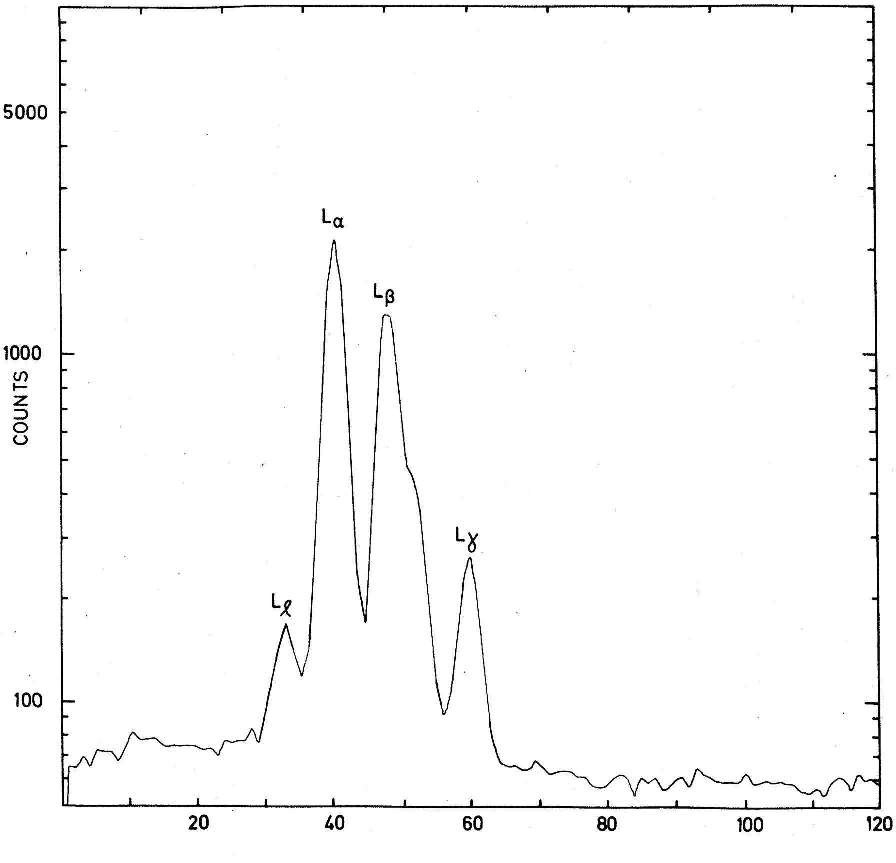

FIG. 5. Spectrum of L x rays in coincidence with 1178 keV gamma rays.

TABLE III. Measured anisotropies for the Ll, $L\alpha$, and $L\gamma$ peaks normalized to the $L\beta$ peak in coincidence with the 1178 keV gamma transtion and the theoretical predictions.

	Experiment	Theory
$A(Ll)$-$A(L\beta)$	$-0.02(3)$ $\chi^2/28=1.1$	-0.026
$A(L\alpha)$-$A(L\beta)$	$0.006(4)$ $\chi^2/32=0.8$	-0.0018
$A(L\gamma)$-$A(L\beta)$	$0.00(1)$ $\chi^2/32=1.1$	0.0008

background under the peak large enough to account for the measured anisotropy.

There is nothing in the appearance of the coincident $L\alpha$ peak to suggest the presence of an unresolved gamma ray (c.f. Fig. 5). However, this possibility was investigated somewhat further by computing the anisotropy channel by channel over the $L\alpha$ peak. No undue fluctuation or trend in the anisotropy was detected, thus a strongly anisotropic gamma ray, if present, must lie within less than 200 eV of the $L\alpha$ energy. Also, the relative intensities of the L x rays in coincidence with the 1178 keV transition were compared to the corresponding intensities in coincidence with the $K\alpha$ x rays. The observed change in intensities between the two gates followed theoretical predictions well. A measurement of the anisotropy of the $K\alpha$ x-Lx directional correlations, using the two detector system gave as a result $A(K\alpha - Ll) = 0.29(7)$, $A(K\alpha - L\alpha) = 0.031(7)$ and $A(K\alpha - L\beta) = 0.003(8)$ in close agreement with the results reported by Catz and Macias[16] for Tb atoms. These values are corrected for the finite solid angle of the detectors.

If we are unable to find anything in the experiment to account for the rather large discrepancy between the measured anisotropy for the $L\alpha$ peak and the theoretical prediction, then what about the latter? The measured anisotropy for the Ll peak agrees with the theoretical prediction although the experimental uncertainty is large. The ratio of the A_{22}-coefficients of the Ll and $L\alpha$ peaks should depend only on the ratio of the respective $A_2(X)$-coefficients in (2). As both peaks correspond to transitions only to the L_3 subshell the damping due to the presence of isotropic x rays should also be the same. For small anisotropies the ratio is therefore expected to be given by

$$\frac{A(1178\gamma - Ll)}{A(1178\gamma - L\alpha)} \simeq \frac{A_2(Ll)[P(L\alpha_1)+P(L\alpha_2)]}{A_2(L\alpha_1)P(L\alpha_1)+A_2(L\alpha_2)P(L\alpha_2)} = \frac{0.5[P(L\alpha_1)+P(L\alpha_2)]}{0.1P(L\alpha_1)-0.4P(L\alpha_2)}$$

Inserting theoretical transition probabilities[15] we predict $A(Ll)/A(L\alpha)=10$ to be compared to a measured ratio of $-3(4)$. This difference may indicate that it is the relative transition probabilities assumed for the individual transitions composing the $L\alpha$ peak which are at fault. Theoretically predicted relative strengths of the $L\alpha_1$ and $L\alpha_2$ transitions agree well with experimental results obtained with radioactive sources or sources ionized by photon or electron bombardment.[17] To our knowledge, however, the present experiment is the first to determine at least indirectly the $L\alpha_1/L\alpha_2$ ratio following internal electron conversion in the L shell. Our measured value for the anisotropy of the $K\alpha - L\alpha$ x-ray cascade agrees with the predicted value. It would thus seem as if the relative intensity of the $L\alpha_1(L_3 \rightarrow M_5)$ and $L\alpha_2(L_3 \rightarrow M_4)$ transitions depends on whether the L_3 vacancy is created directly by internal electron conversion or not. To test this unexpected possibility an experiment is being prepared at this laboratory aimed at measuring the directional correlation between $87L$ conversion electrons and L x rays.

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