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Directional correlation between α particles and L x rays in the decay of ^{238}Pu and ^{244}Cm

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Anisotropy in the directional correlation of nuclear radiations and L x rays has been clearly identified for the first time. L_3 x-ray groups, L_l and $L\alpha$, are observed to be directionally correlated with α particles in the decays of ^{238}Pu and ^{244}Cm. The ratio of anisotropy for L_l and $L\alpha$ is consistent with the recent observation that L_l has a much greater admixture of $M2$ than predicted by relativistic calculations.

Several attempts to demonstrate directional correlation between nuclear radiations and L x rays have been reported [1-8]. Anisotropy has not been clearly established in any of these studies. Most recently, Bargholtz *et al.* [8] reported statistical evidence of a very small directional correlation between γ rays and $L\alpha$ in the decay of ^{160}Tb. The ratio of directional correlation of the L_l and $L\alpha$ groups was interpreted as being anomalous. The results are inconclusive and difficult to explain.

Directional correlations between nuclear radiations in cascade in nuclear deexcitation (e.g., radioactive decay) are well understood [9], as are the directional correlations between x rays in cascade in atomic deexcitation [10]. The directional correlation between nuclear radiations and x rays in cascade has received less attention. Dolginov [11] gave a theoretical description of the correlation between nuclear radiations and x rays; however, with little experimental data available for comparison, the theory remains largely untested.

Only cascades in which the intermediate states have total angular momentum $J > \frac{1}{2}$ allow anisotropic directional correlation. Consequently the most tightly bound subshell that is expected to support anisotropic directional correlation is the $L_3(2p_{3/2})$ subshell. The L x-ray spectrum cannot be fully resolved, but falls into four groups of lines that can be resolved. L_l and $L\alpha$ groups are due to the transitions $L_3 \rightarrow M_1$ and $L_3 \rightarrow M_{4,5}$, respectively, and therefore are expected to show if directional correlation is present. $L\beta$ is due to transitions from all subshells thus diluting any directional correlation, while $L\gamma$ is due to transitions from the L_1 $(2s_{1/2})$ and L_2 $(2p_{1/2})$ subshells and should be isotropically distributed relative to nuclear emissions as should x-rays from the K $(1s_{1/2})$ shell. However, Khalil [12] has described $J > 1$ admixtures into the K $(1s_{1/2})$ shell of atoms with highly deformed nuclei which contribute to anisotropic directional correlation between γ rays and K x rays in the decay of ^{181}Ta.

The anisotropic angular distribution of L_3 x rays of Th and U following proton ionization has recently been used by Papp *et al.* [13] to demonstrate deficiencies in the theoretical description of Scofield [14] of the multipolarity of L_l.

In the present work, the directional correlation between α particles and L x rays in the decays of ^{238}Pu and ^{244}Cm arises predominantly from the triple cascade; α decay to the first excited state of the daughter nucleus followed by internal conversion of an $E2$ electromagnetic transition and x-ray emission:

$$\begin{array}{ll} \text{nuclear angular momentum} & 0^+ \xrightarrow{\alpha} 2^+ \xrightarrow{e^-} 0^+ \\ & \qquad\quad | \\ \text{atomic angular momentum} & \qquad\ \ 0 \longrightarrow 3/2 \xrightarrow{x} I \end{array}$$

where the final atomic states are $I = \frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$. Other decay cascades are of negligible strength in comparison. Conversion of the 43 keV $E2$ electromagnetic transition in these decays goes to the L subshells in the percentage ratios [15] L_1:L_2:L_3:α decays::0.4:11.0:9.7:100 for ^{238}Pu and L_1:L_2:L_3:α decays::0.3:8.8:7.6:100 for ^{244}Cm. The number of L_3 vacancies deriving from Coster-Kronig transitions amount to only 15% in the decay of ^{238}Pu and 22% in the decay of ^{244}Cm [15]. These L_3 vacancies follow an intermediate state of $J = \frac{1}{2}$, which no anisotropy is expected to follow.

The L x-ray spectra of ^{238}Pu and ^{244}Cm gated by α particles was obtained at $\theta = 90°$ and $180°$. The coincidence system was a conventional slow coincidence network gating the x-ray spectrum. The coincidence resolving time used was 250 ns. The α-particle gated spectrum of L x rays from the decay of ^{238}Pu in the 90° position is shown in Fig. 1.

A Princeton Gamma-Tech Si(Li) x-ray detector with an active diameter of 6 mm, active depth of 3 mm, a 30-μm Be window and a resolution (full width at half maximum, FWHM) of 150 eV at 5.9 keV (Mn $K\alpha$) was used. The α-particle detector was an Ortec silicon surface barrier detector, 100 μm thick, 100-mm^2 active area with a FWHM resolution of 16 keV. Because the α-particle resolution was not important, a degraded spectrum was acceptable and the experiment was conducted in air. The ^{238}Pu and ^{244}Cm sources were prepared by electrodeposition for high-resolution x-ray and α-particle spectrometry [16]. The ^{238}Pu source was approximately 24 kBq, while the ^{244}Cm source was approximately 13 kBq. Both sources were 7 mm in diameter deposited on a vacuum deposited Cu layer on 127-μm thick Kapton.

The arrangement of source and detectors is shown in Fig. 2. The source was mounted in a fixed position relative to the x-ray detector while the α-particle detector was placed at $\theta = 90°$ or $180°$. The x-ray detector viewed the source through the source backing which effectively stopped charged particles and M x rays. The x-ray and

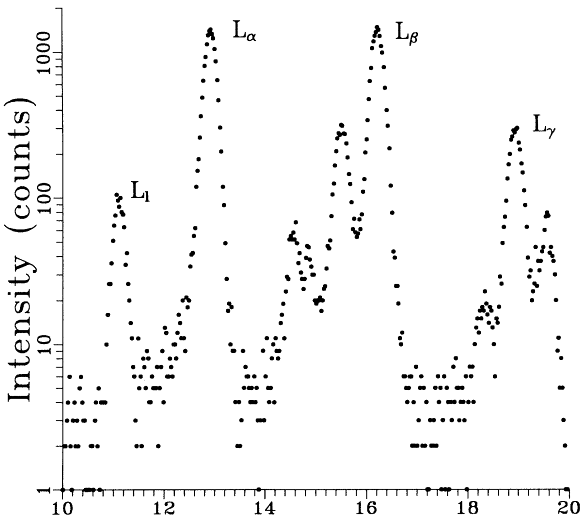

FIG. 1. *L* x-ray spectrum gated by α particles in the decay of ^{238}Pu at the 90° position.

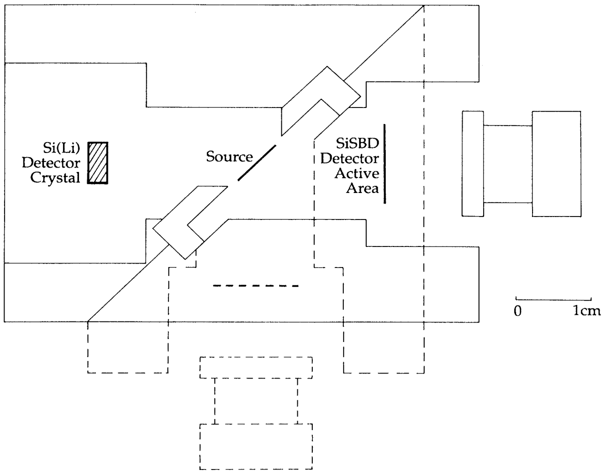

FIG. 2. Experimental directional correlation apparatus in the 180° position showing the source and the active volumes of the detectors. The apparatus in the 90° position is also indicated by the dashed outlines.

α-particle detectors were always at 45° to the normal of the source surface.

The minimum number of angles at which measurements need to be made is the same as the number of terms in the equation $W(\theta)=1+\sum A_{kk}P_k(\cos\theta)$. In the current experiment only one term of the summation is expected, i.e., $k=2$, so that only two angles need be measured: $\theta=90°$ and 180°. However, it is usual to make measurements at $\theta=270°$ as well as 90° to check if source nonuniformities are important. This was not possible in the present work, and in order to simulate the same effect the source was remeasured after rotation by 180° in the source holder.

For each source four sets of approximately five spectra were taken. The first set in the 180° position, followed by a second set in the 90° position. The source was then rotated through 180° in the source holder, a third set in the 90° position and then the fourth set in the 180° position. Basic data from the measurements is given in Table I.

The source and the detectors have finite size. This has the following two effects: (i) The direction between the nuclear and atomic radiations is not always exactly 90° or 180°, and, accordingly, (ii) a bias may be introduced into the experiment. While each detector is geometrically related to the source in an identical manner at 90° and 180°, the geometrical relationship in coincidence is

TABLE I. Experimental data from directional correlation experiment. $E-n$ denotes $\times 10^{-n}$.

	Angular position and source orientation			
			Source rotated	
	180°	90°	180°	90°
^{238}Pu source				
No. spectra	6	5	4	5
L_l (counts/s)	4.44$E-3$	5.21$E-3$	4.37$E-3$	5.21$E-3$
$L\alpha$ (counts/s)	7.53$E-2$	8.03$E-2$	7.62$E-2$	8.01$E-2$
$L\beta$ (counts/s)	1.07$E-1$	1.11$E-1$	1.08$E-1$	1.12$E-1$
$L\gamma$ (counts/s)	2.54$E-2$	2.57$E-2$	2.49$E-2$	2.58$E-2$
α rate (counts/s)	577.2	574.9	577.0	575.5
Count time (s)	1 021 082	682 310	501 639	598 396
^{244}Cm source				
No. spectra	4	5	5	4
L_l (counts/s)	2.29$E-3$	2.26$E-3$	1.98$E-3$	2.48$E-3$
$L\alpha$ (counts/s)	3.83$E-2$	3.94$E-2$	3.73$E-2$	3.99$E-2$
$L\beta$ (counts/s)	4.69$E-2$	4.76$E-2$	4.63$E-2$	4.85$E-2$
$L\gamma$ (counts/s)	8.97$E-3$	9.04$E-3$	9.00$E-3$	9.46$E-3$
α rate (counts/s)	316.4	312.5	312.0	316.0
Count time (s)	488 206	595 273	1 012 727	1 019 386

different in the two positions. A correction is required for the finite sizes.

To compute the correction, assume that the source and the detectors can be represented by disks. Consider an area $d\mathbf{S}_s$ at $\mathbf{r}_s$ on the source; the probability of an emitted x ray striking an area $d\mathbf{S}_X$ at $\mathbf{r}_X$ on the x-ray detector is then $d\mathbf{S}_X\cdot(\mathbf{r}_s-\mathbf{r}_X)/4\pi|\mathbf{r}_s-\mathbf{r}_X|^3$. Thus the average solid angle for x rays striking the x-ray detector is (normalized to the area of the source equivalent to 1)

$$\frac{1}{4\pi}\int_{\mathbf{S}_s}\int_{\mathbf{S}_X}|\mathbf{r}_s-\mathbf{r}_X|^{-3}d\mathbf{S}_X\cdot(\mathbf{r}_s-\mathbf{r}_X)d\mathbf{S}_s\,. \qquad (1)$$

The average geometric efficiency for coincident detection of α particles and L x rays having an isotropic distribution is

$$\varepsilon_g=\frac{1}{16\pi^2}\int_{\mathbf{S}_s}\int_{\mathbf{S}_X}\int_{\mathbf{S}_\alpha}|\mathbf{r}_s-\mathbf{r}_X|^{-3}|\mathbf{r}_s-\mathbf{r}_\alpha|^{-3}\times d\mathbf{S}_\alpha\cdot(\mathbf{r}_s-\mathbf{r}_\alpha)d\mathbf{S}_X\cdot(\mathbf{r}_s-\mathbf{r}_X)d\mathbf{S}_s\,. \qquad (2)$$

TABLE II. Anisotropy of L x-ray groups in the decays of ^{238}Pu and ^{244}Cm.

L x-ray group	^{238}Pu A_{22} $\sigma(A_{22})$	^{244}Cm A_{22} $\sigma(A_{22})$
L_l	-0.104 ± 0.019	-0.072 ± 0.025
$L\alpha$	-0.024 ± 0.004	-0.014 ± 0.005
$L\beta$	-0.006 ± 0.003	$+0.000\pm0.004$
$L\gamma$	-0.001 ± 0.006	$+0.001\pm0.010$

In the current situation the angular distribution is expected to be $W(\theta)=1+A_{22}P_2(\cos\theta)$. Therefore the average value of $P_2(\cos\theta)=(3\cos^2\theta-1)/2$, for each of the experimental positions, needs to be computed. To calculate this average, the average value of $\cos^2\theta$ is to be evaluated:

$$\langle\cos^2\theta\rangle_{\rm av}=\frac{1}{16\pi^2\varepsilon_g}\int_{\mathbf{S}_s}\int_{\mathbf{S}_X}\int_{\mathbf{S}_\alpha}[(\mathbf{r}_s-\mathbf{r}_X)\cdot(\mathbf{r}_s-\mathbf{r}_\alpha)]^2|\mathbf{r}_s-\mathbf{r}_X|^{-5}|\mathbf{r}_s-\mathbf{r}_\alpha|^{-5}d\mathbf{S}_\alpha\cdot(\mathbf{r}_s-\mathbf{r}_\alpha)d\mathbf{S}_X\cdot(\mathbf{r}_s-\mathbf{r}_X)d\mathbf{S}_s\,. \qquad (3)$$

These integrals have been evaluated numerically by dividing each of the three disks into 100 regions of equal area: $\varepsilon_g(180°)/\varepsilon_g(90°)=0.9688$, while $\langle\cos^2\theta\rangle_{\rm av}(180°)=0.8995$ and $\langle\cos^2\theta\rangle_{\rm av}(90°)=0.04544$.

X-ray spectra were analyzed using the trapezoid method. Count rates of L x-ray groups were corrected for the bias $[\varepsilon_g(180°)/\varepsilon_g(90°)]$ introduced by the measurement geometry. Source nonuniformity was estimated from the difference between α-particle count rates in two source orientations rotated through 180° and a correction was applied for the difference in α-particle gating rate. These rates were then fitted using linear least squares weighted by counting statistics to derive the number of x rays detected per α decay in the 90° position, $W(90°)$ and the corrected rate difference in the 180° position $[W(180°)-W(90°)]$. These yield the experimental anisotropies, $A=[W(180°)-W(90°)]/W(90°)$. Using the values for $\langle\cos^2\theta\rangle_{\rm av}$ at 90° and 180°, A can be expressed as

$$A=1.281A_{22}/(1-0.432A_{22})\,. \qquad (4)$$

Solutions for A_{22} are given in Table II together with their estimated standard errors. These were estimated from the standard errors estimated in the least-squares fitting procedure and by the method of propagation of errors. There were insufficient counting statistics to perform fitting to individual lines in the $L\beta$ group.

The present work demonstrates that α particles and L x rays are directionally correlated in the decays of ^{238}Pu and ^{244}Cm. No evidence of anisotropy for intermediate states having $J=\frac{1}{2}$ is observed in the decays of these heavily deformed nuclei, the theoretical expectation of isotropy for $L\gamma$ is confirmed.

Papp *et al.* [13] have observed that the ratio of anisotropy of L_l and $L\alpha$ is $\beta L_l/\beta L\alpha=6.2$ following proton ionization. The ratio of anisotropy should be independent of the ionization mode. The current work gives slightly lower ratios in general agreement with the work of Papp *et al.* Calculations by Scofield [14] indicate that $\beta L_l/\beta L\alpha$ should be approximately 10. This difference can be explained by a greater $M2$ admixture in L_l than that predicted by Scofield.

No evidence is found of any anomalous directional correlation of the type reported by Bargholtz *et al.* [8].

The present work has established anisotropy of nuclear and L x rays in the heavy even-even nuclei that were first studied for this effect [1-3]. Earlier work with low-resolution x-ray spectrometers is consistent with results of the present study in that the anisotropy over the entire L x-ray spectrum is less than could be measured in those studies.

A comparison of the present results with the theory of Dolginov [11] is in progress.

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