Communications

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Search for an anisotropy between Ll x rays and γ rays in the decay of ^{243}Cm†

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The anisotropy of Ll x rays and γ rays of ^{239}Pu following the decay of ^{243}Cm has been investigated. A Ge(Li) crystal with 250 eV resolution at 5.9 keV was used to detect Ll x rays. A NaI(Tl) crystal was used to detect six γ rays between 210 and 285 keV. The measured anisotropy is $A = (3 \pm 6) \times 10^{-3}$. This is less than 2% of the maximum predicted anisotropy assuming that the magnetic substate alignment has been completely retained in the coupling of cascade γ rays and x rays.

[RADIOACTIVITY ^{243}Cm; measured Ll x-$\gamma(\theta)$. Ge(Li) and NaI(Tl) detectors.]

Recently Rupnik and Crasemann[1] reviewed, simplified, and extended Dolginov's[2] theory of directional correlations between x rays and γ rays emitted after electron capture and internal conversion. Rupnik and Crasemann also reviewed the results of previous angular correlation measurements between L x rays and nuclear radiation and reported the results of a measurement of the anisotropy between the 570-keV γ ray and x rays emitted from the L_3 shell ($Ll, L\alpha, L\beta$) created in electron capture and internal conversion decay of ^{207}Bi. This measurement was hampered by the fact that over 98% of all 570-keV γ ray-L_3 x ray coincidence events are expected to be isotropic. The measured anisotropy was found to be $A = (2 \pm 2) \times 10^{-3}$. However, due to the large isotropic component in the cascade measured, the theoretically expected asymmetry could be as small as $A = -1.4 \times 10^{-4}$. Therefore, this experiment did not substantiate or contradict the prediction from Dolginov's theory of a finite γ ray-L_3 x ray anisotropy. Because of the relatively large directional correlation expected for the $Ll(L_3\text{-}M_1)$ transition, Rupnik and Crasemann suggest that a measurement of the directional correlation between Ll x rays and γ rays might give a more definitive test of the theory. Following that suggestion, the present work on the directional correlation between Ll x rays and γ rays following the decay of ^{243}Cm was undertaken.

The 32-y ^{243}Cm source was chosen because of the large amount of L_3 subshell internal conversion of several excited states of ^{239}Pu populated in the α decay of ^{243}Cm. Specifically, 64.3% of the composite 210-, 228-, 254-, 273-, 277-, and 285-keV γ-ray–Ll x-ray cascade results from direct L_3 internal conversion and thus may be anisotropic. The Ll x rays in the remaining 35.7% of this cascade result from either L_1 or L_2 internal conversion followed by Coster-Kronig transitions to the L_3 subshell and therefore are expected[1] to be isotropic because they originate from subshells of spin $\frac{1}{2}$. There is no K conversion of the internally converted transitions in these cascades. Although this composite cascade consists of ten different components, nearly $\frac{3}{4}$ of total coincidence intensity is due to only two cascades. The portion of the ^{243}Cm decay scheme[3] relevant to this discussion is shown in Fig. 1. It should be noted that there are several higher energy γ transitions in ^{243}Cm decay not shown on Fig. 1.

The source consisted of 100 μCi of ^{243}Cm deposited as a 1-mm diam spot of $^{243}Cm(NO_3)_3$ on a 0.5-mm thick aluminum disk covered by 2.5-mm beryllium. The source was placed in the center of an angular correlation table where it was viewed by two detectors. The experimental system used to measure the anisotropy between the six γ rays in the range of 210–285 keV and Ll x rays following internal conversion in the decay of ^{243}Cm is similar to a system described previously.[4] The L x rays were detected with a 10-mm planar Ge(Li) crystal with energy resolution of 250 eV (full width at half-maximum) at 5.9 keV. This detector was

capable of completely resolving the Ll x rays from the more intense $L\alpha_{1+2}$ peak as shown in Fig. 2. γ rays were detected in a 5 cm×5 cm NaI(Tl) scintillation detector capable of resolving the six γ rays from all other γ rays following the decay of ^{243}Cm (see Fig. 2). The NaI(Tl) crystal was shielded by a tapered lead collimator and the Ge(Li) crystal was shielded laterally by a tapered collimator of aluminum-brass-lead-brass-aluminum construction. The purpose of these collimators was to minimize the contribution of erroneous coincidence events due to scattering of radiation from one detector to the other. Coincidence spectra were obtained with a standard fast-slow coincidence system employing crossover timing with resolving time $2\tau = 40$ nsec.

The γ-ray spectrum in coincidence with Ll x rays was measured at 90°, 180°, and 270°. A gate was set on the Ll x rays detected in the Ge(Li) crystal because the Ll x ray was well resolved with this detector and had a high peak-to-background ratio. The γ ray spectrum detected in the NaI(Tl) crystal was stored in the multichannel analyzer because of the poor definition of the γ ray peaks and the moderately high background contribution. The spectrum at each angle was accumulated for approximately 20 h. A single data set consisted of one measurement each at 90° and 270° and two measurements at 180°. Measurements at 90° and 270° were added together for data analysis. A total of three data sets were accumulated in this experiment. These data were normalized to the number of Ll x rays detected in the Ge(Li) crystal at each angle in order to remove the effect of source decentering (<0.5%). Background, determined in regions of the spectrum where there were no γ-ray peaks, was subtracted from the composite γ-ray peak. The contribution from random coincidences was determined by taking long measurements with an additional 500 nsec relative delay between the output of the two detectors. The data were corrected for these random coincidences which were found to be angle independent. The ex-

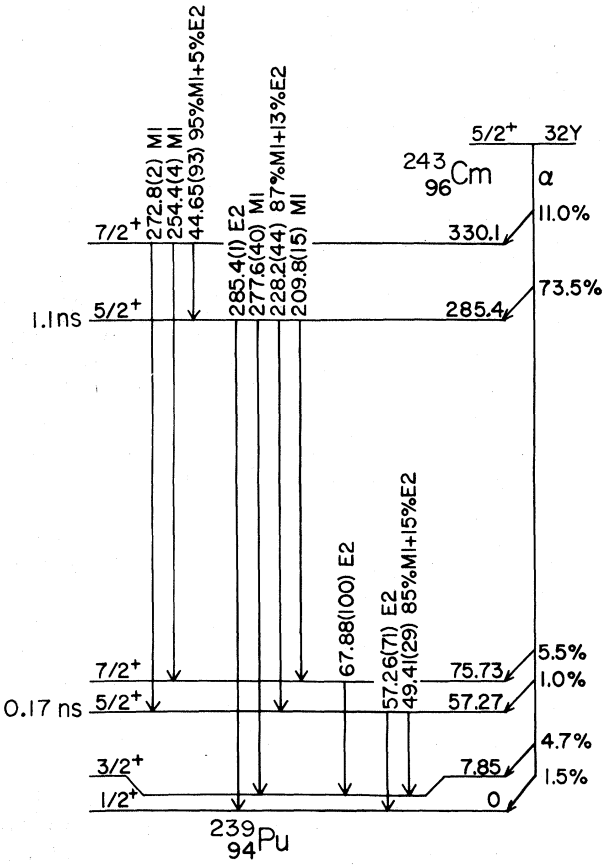

FIG. 1. Partial decay scheme of ^{243}Cm. The numbers in parentheses are the total transition intensities. γ-ray multipolarities are taken from the compilation of Ref. 3.

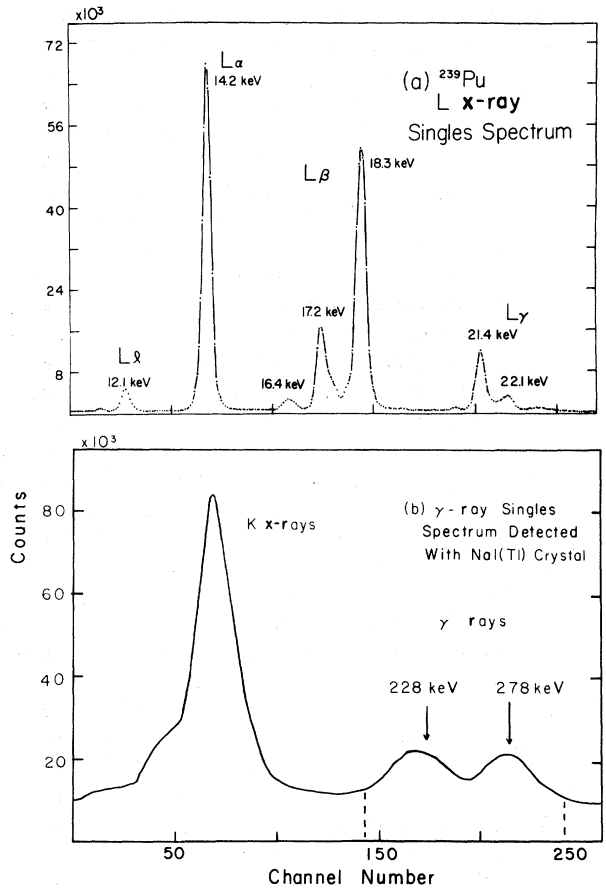

FIG. 2. (a) Spectrum of Pu L x rays detected in the Ge(Li) crystal. (b) Spectrum of γ rays following ^{243}Cm decay detected in the NaI(Tl) crystal.

tent of scattering of x rays and γ rays between detectors was determined by placing a 0.50-mm copper absorber on the face of the radioactive source toward the L x-ray detector. Contributions to the coincidence spectrum due to scattering were found to be negligible at all angles. The effect of the finite solid angle subtended by the Ge(Li) detector was determined by extrapolation from the data of Camp and Van Lehn[5] and found to be negligible. The solid angle correction factor for the NaI(Tl) detector was determined from Yates[6] and used to correct the coincidence data.

The anisotropy was calculated with the corrected coincidence data using the following relation:

$$A = \frac{[W(\pi) - W(\frac{1}{2}\pi)]}{W(\frac{1}{2}\pi)} , \quad (1)$$

where $W(\theta)$ is the number of γ rays coincident with L x rays at angle θ. No measurable anisotropy was determined within experimental error in the composite γ-ray–L x-ray cascade. The measured anisotropy is

$$A = (3 \pm 6) \times 10^{-3} . \quad (2)$$

The theoretical prediction of the maximum possible angular correlation coefficient A_{22}, assuming that the nuclear and atomic states are strongly coupled was calculated as follows:

$$A_{22} = \sum_i C(i) n_{L3}(i) A_{22}(i) , \quad (3)$$

where $C(i)$ is the coincidence intensity of γ-ray–Ll x-ray cascade i relative to the total coincidence intensity of the composite γ-ray–Ll x-ray cascade; $n_{L3}(i)$ is the number of primary vacancies in the L_3 subshell relative to the total number of L_3 vacancies created which are in coincidence with the observed γ ray of cascade i, i.e., the fraction of Ll x rays in the cascade which are expected to be anisotropic; and $A_{22}(i)$ is the predicted maximum angular correlation coefficient for cascade i.[7] The anisotropy of the composite cascade is then

$$A = 3A_{22}/(2 - A_{22}) . \quad (4)$$

The maximum anisotropy predicted for the measured composite γ-ray–Ll x-ray cascade following ^{243}Cm decay calculated from Eqs. (3) and (4) and using the data in Tables I and II was found to be either -0.221 or -0.076, depending on the sign of the mixing ratio for the 228.2 keV transition. In this calculation it has been assumed that the magnetic substate alignment has been completely retained in the coupling of the cascade γ rays and x rays following internal conversion. The anisotropy measured in this work is less than 2% of the maxi-

TABLE I. Summary of L electron intensities and conversion coefficients for the low energy transitions in ^{243}Cm decay [data taken from A. Artna-Cohen, Nucl. Data B6, 577 (1971)].

Transition energy (keV)	α_L	α_T	L_1	L_2	L_3	n^i_{L3} [a]
44.7	74.2	99.5	0.60	0.23	0.17	0.292
49.4	81.7	109	0.32	0.37	0.31	0.528
57.3	167	223	0.02	0.53	0.45	0.771
67.9	74.0	98.4	0.02	0.57	0.40	0.741

[a] Calculated as follows:

$$n_{L_3} = \frac{L_3}{L_1 f_{13} + L_2 f_{23} + L_3} ,$$

assuming $f_{13} = 0.6$ [extrapolated from W. Bambynek, B. Crasemann, R. W. Fink, H.-U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. Venugopala Rao, Rev. Mod. Phys. 44, 716 (1972)], $f_{23} = 0.23$ [M. R. Zalutsky and E. S. Macias, Phys. Rev. A 11, 71 (1975)] for $_{94}$Pu.

TABLE II. Data used for predicting the anisotropy of the composite γ-ray–Ll x-ray cascade following ^{243}Cm decay [data taken from A. Artna-Cohen, Nucl. Data B6, 577 (1971)].

Cascade (keV) T_1-T_2	$C(i)$ [a]	$N_{L3}(i)$ [b]	$A_{22}(i)$ [c]	$C(i)N_{L3}(i)A_{22}(i)$ [d]
1. 273-57	0.0041	0.771	0.0676	0.002
2. 273-49	0.0017	0.528	0.0676	0.0001
3. 254-68	0.0075	0.741	−0.2207	−0.0012
4. 285-45	0.0053	0.292	−0.2703	−0.0004
5. 278-45	0.1061	0.292	0.1892	0.0059
6. 228-45	0.0920	0.292	−0.2162	−0.0058
7. 210-45	0.0028	0.292	0.0676	0.0001
8. 210-68	0.0216	0.741	0.1655	0.0026
9. 228-57	0.5384	0.771	−0.3018	−0.1254
			−0.1012	−0.0420
10. 228-49	0.2203	0.528	−0.3018	−0.0351
			−0.1012	−0.0118

[a] Cascade i coincidence intensity relative to the total intensity of γ-Ll cascades.

[b] The number of primary vacancies in the L_3 subshell relative to the total number of L_3 vacancies created in transition T_2.

[c] Angular correlation coefficient for cascade i calculated assuming the observed γ-ray transition T_1 is the multipolarity listed in Fig. 1, and $A_2(Ll) = 0.5057$ [J. H. Scofield, UCRL Report No. UCRL-51231, 1972 (unpublished)]. The γ-ray multipolarities are from the compilation of Ref. 3. Two values given for the same cascade are due to the uncertainty in the sign of the mixing ratio (δ).

[d] The sum of the entries in this column is the predicted experimental angular correlation coefficient. The anisotropy A is calculated from Eq. (2) in the text.

mum predicted anisotropy. A more complete interpretation of the present measurement requires a detailed calculation of the anisotropy for the measured composite cascade using a method such as that developed by Ferguson and Geiger.[8]

A larger observable anisotropy may result if the anisotropy of a cascade involving an *Ll* x ray and single γ ray were measured. This experiment would be best performed by detecting the γ rays with a Ge(Li) crystal.

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[8]A. J. Ferguson and J. S. Geiger (private communication)