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GAMMA-GAMMA DIRECTIONAL CORRELATIONS IN ^{140}Ce

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Abstract: Directional correlation measurements have been performed on six γ-ray pairs occurring in the de-excitation of ^{140}Ce. Of these, two are 1–3 correlations with an unobserved intermediate γ-ray. Both NaI(Tl)-Ge(Li) and Ge(Li)-Ge(Li) detector systems were used. The following mixing ratios were deduced for the 329, 433, 487, 752, 816 and 868 keV transitions:

$$\delta(329; \mathrm{D, Q}) = 0.018 \pm 0.007,$$
$$\delta(433; \mathrm{D, Q}) = 0.36 \pm 0.02 \text{ or } -0.14 \pm 0.03,$$
$$\delta(487; \mathrm{Q, O}) = -0.002 \pm 0.007,$$
$$\delta(752; \mathrm{D, Q}) = -0.33 \pm 0.03,$$
$$\delta(816; \mathrm{D, Q}) = 0.031 \pm 0.005,$$
$$-0.09 < \delta(868; \mathrm{D, Q}) < 0.10.$$

E RADIOACTIVITY ^{140}La [from ^{139}La(n, γ)]; measured $\gamma\gamma(\theta)$. ^{140}Ce deduced γ-mixing. Ge(Li) and NaI(Tl) spectrometers.

1. Introduction

The level structure of the semi-magic nucleus ^{140}Ce has been extensively studied through the β^- decay of ^{140}La. A summary of work prior to 1968, as well as numerous references, may be found in the work of Baer *et al.* [1]). The essential features of the ^{140}Ce decay scheme proposed by Baer *et al.* [1]) can be considered to be well established. More recent investigations of ^{140}Ce through the β^- decay of ^{140}La [refs. [2-5])], the β^+, EC decay of ^{140}Pr [refs. [5-7])] and nuclear reactions [8]) have failed to change any major feature of this decay scheme. However, the multipolarities of many of the transitions are at best poorly known. Most of the current information is based on conversion electron studies and consequently only very broad limits have been established for the extent of multipole mixing in most of the transitions. A few γ-γ directional correlation studies have been made [9,10]), but the poor resolution of NaI(Tl) detectors has greatly limited the number of cascades that could be investigated. With the advent of large-volume Ge(Li) detectors it has become possible to investigate many correlations which previously were impossible to measure.

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The present investigation was undertaken in order to obtain precise measurements of the multipole mixing in γ-ray transitions following the decay of ^{140}La ($T_{\frac{1}{2}} = 40.2$ h) to levels in ^{140}Ce. In fig. 1 we reproduce those features of the ^{140}Ce level scheme proposed by Baer *et al.* [1]) which are relevant to this study. A pure E2 transition from the first excited state (1596.6 keV) to the ground state was assumed and the multipolarities of the other transitions shown were then deduced.

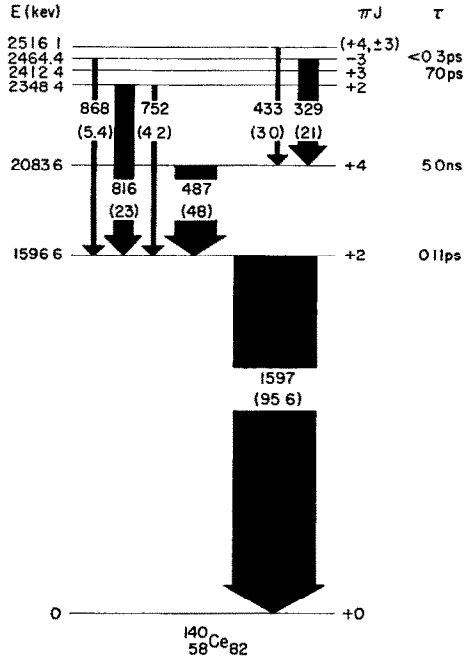

Fig. 1. Partial level scheme of ^{140}Ce populated in the decay of ^{140}La. Abstracted from ref. [1]).

2. Apparatus and procedure

Data were obtained with three automated angular correlation systems. The first used a Harshaw 7.6 cm × 7.6 cm NaI(Tl) crystal (integral mount) in one channel and an Ortec 17 cm^3 Ge(Li) detector in the other. In using this system the 1597 keV γ-ray was gated with the NaI(Tl) detector and the coincident Ge(Li) spectrum was accumulated with a multichannel analyzer. The second apparatus was similar to this with the following exceptions: a larger volume Ge(Li) detector (Ortec, 29 cm^3) and a computer controlled analyzer were utilized. Again, the NaI(Tl) detector was used to gate the 1597 keV γ-ray. For the measurement of the 329 keV–487 keV correlation the NaI(Tl) detector in the second system was replaced by an Ortec 32 cm^3 Ge(Li) detector.

Source material was obtained by irradiating $La(NO_3)_3$ in the UM reactor and

dissolving this material in water. Sources were made by placing a drop of this solution in a thin-walled lucite cylinder.

Since each run lasted between one and two half-lives it was necessary to correct the data for the decay of the source. Following this time correction procedure, chance coincidences were subtracted from the measured coincidences at each angle. The resulting true coincidences were then added to give a net number of true coincidences at each of the seven angles 90°, 105°, 120°, 135°, 150°, 165° and 180°. A computer program was used to perform a least-squares fit of the function

$$W(\theta) = A_0 + A_2 P_2(\cos\theta) + A_4 P_4(\cos\theta)$$

to these data following the method of Rose [11]). The ratios A_2/A_0 and A_4/A_0 and their associated uncertainties [12]) were obtained from the fit.

The ratios A_2/A_0 and A_4/A_0 were corrected for the finite solid angles subtended by the detectors by dividing each, respectively, by the geometric corrections Q_2 and Q_4 given by Camp and Van Lehn [13]) as

$$Q_K = \frac{J_K^A(\gamma_1)}{J_0^A(\gamma_1)} \frac{J_K^B(\gamma_2)}{J_0^B(\gamma_2)} \left(\frac{R}{1+R}\right) + \frac{J_K^A(\gamma_2)}{J_0^A(\gamma_2)} \frac{J_K^B(\gamma_1)}{J_0^B(\gamma_1)} \left(\frac{1}{1+R}\right), \quad (K = 2, 4)$$

where

$$R = \frac{\varepsilon_A(\gamma_1)}{\varepsilon_A(\gamma_2)} \frac{\varepsilon_B(\gamma_2)}{\varepsilon_B(\gamma_1)}$$

and $\varepsilon_A(\gamma)$ and $\varepsilon_B(\gamma)$ denote detector photopeak efficiencies. The subscripts A and B refer to the two detectors. The ratios J_K/J_0 for the 17 cm^3 and 32 cm^3 Ge(Li) detectors were determined in this laboratory by means of a collimated beam experiment [14]). For the 29 cm^3 Ge(Li) detector these ratios were obtained from a Monte Carlo calculation by Camp and Van Lehn [15]). We have shown [14]) for three Ge(Li) detectors (17 cm^3, 32 cm^3 and 38 cm^3) that there is agreement between the calculated values of Camp and Van Lehn and our experimental values. The calculated values for J_K/J_0 given by Yates [16]) were used for the NaI(Tl) detectors. Data given by Yates [16]) were used to determine the efficiency factors $\varepsilon(\gamma)$ for the NaI(Tl) detectors. Experimental values of $\varepsilon(\gamma)$ were determined in this laboratory for the 17 cm^3 and 32 cm^3 Ge(Li) detectors. Calculated values of $\varepsilon(\gamma)$ obtained from the computer program of Aubin *et al.* [17]) were used for the 29 cm^3 Ge(Li) detector. We have shown [18]) that there is excellent agreement between the relative photopeak efficiencies obtained in this laboratory for Ge(Li) detectors and the efficiency values given by the program of Aubin *et al.* [17]).

In several of the runs in which the 752 keV–1597 keV and 816 keV–1597 keV correlations were measured, a lead absorber having a thickness of 0.64 cm was placed in front of the Ge(Li) detector in order to reduce the flux of lower-energy γ-rays incident upon the detector and thereby improve the resolution for the peaks of interest. This necessitated a determination of Q_2 and Q_4 for these special circumstances. Data

from the collimated beam experiment were used to recalculate Q_2 and Q_4 incorporating the effect of the absorber. The quantities Q_2 and Q_4 were each changed by less than 0.05 % for all energies of interest.

For five of the six correlations measured the 1597 keV γ-ray was gated and no interfering correlation was present. In obtaining the 329 keV–487 keV correlation we gated the 487 keV peak and it was necessary to correct for an interfering correlation arising from coincidences between the Compton distribution of the 1597 keV γ-ray and the 329 keV γ-ray.

Each of the five correlations in which the 1597 keV γ-ray was gated was measured with both NaI(Tl)–Ge(Li) systems. In every case satisfactory agreement was obtained between the two measurements.

For each correlation measured our value of A_2/A_0 was used to determine a mixing ratio, δ. Our A_4/A_0 value was used to eliminate one of the two values of δ consistent with the A_2/A_0 value. In figs. 2 and 3 we show graphical determinations of δ from our measured values of A_2/A_0 and A_4/A_0.

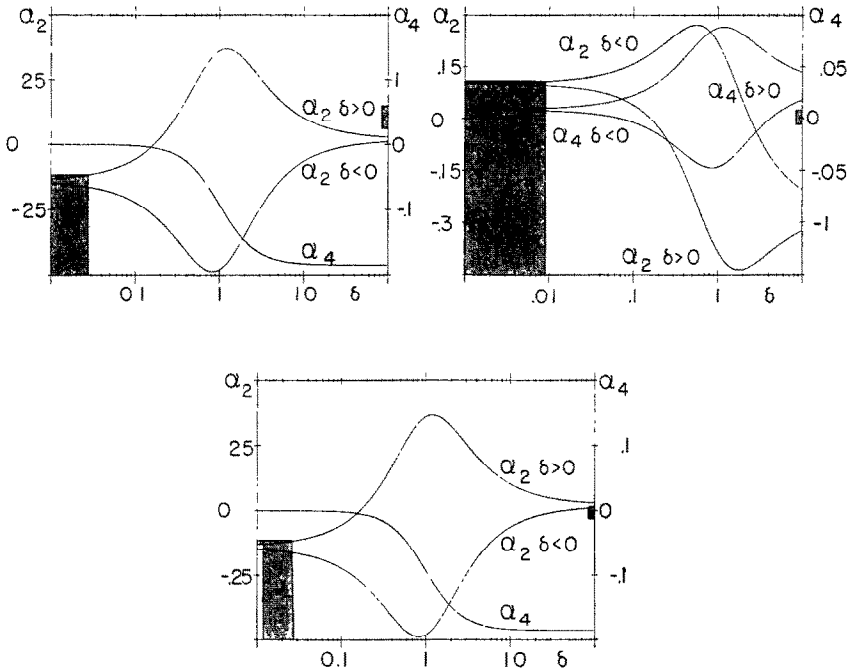

Fig. 2. Graphs of mixing ratio (δ) versus correlation function coefficients ($\alpha_k = A_k/A_0$; $k = 2, 4$). In each case the mixing ratio refers to the first transition in the cascade. Shaded bands show a graphical determination of δ from our measured values of α_2 and their associated uncertainties; truncated bands show our experimental limits on α_4. Upper left: 329 keV–487 keV correlation. Spin-multipolarity sequence is 3(D, Q)4(Q)2. Upper right: 487 keV–1597 keV correlation. Spin-multipolarity sequence is 4(Q, O)2(Q)0. Lower: 329 keV–1597 keV 1–3 correlation. Spin-multipolarity sequence is 3(D, Q)4(Q)2(Q)0.

There has been some confusion concerning the sign of δ in γ-γ directional correlations. In particular, if one analyzes the data using the formulas of Biedenharn and Rose [19]) one finds that the sign of δ depends on whether the transition is the first or second in the cascade. In discussing this ambiguity Dzhelepov *et al.* [20]) suggest the adoption of a convention in which the sign of δ given by the formulas of Bieden-

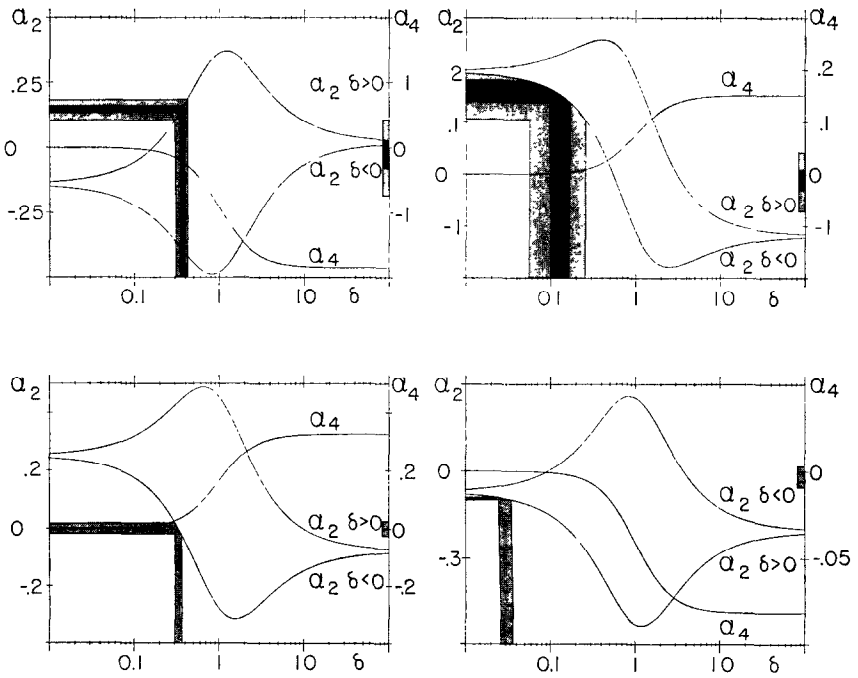

Fig. 3. Graphs of mixing ratio (δ) versus correlation function coefficients ($\alpha_k = A_k/A_0$; $k = 2, 4$). In each case the mixing ratio refers to the first transition in the cascade. Shaded bands show a graphical determination of δ from our measured values of α_4 and their associated uncertainties. Where two bands are shown, two independent determinations of α_2 were made. Truncated bands show our experimental limits on α_4. Upper left: 433 keV–1597 keV 1–3 correlation. A spin-multipolarity sequence of 3(D, Q)4(Q)2(Q)0 was assumed. Upper right: 433 keV–1597 keV 1–3 correlation. A spin-multipolarity sequence of 4(D, Q)4(Q)2(Q)0 was assumed. Lower left: 752 keV–1597 keV correlation. Spin-multipolarity sequence is 2(D, Q)2(Q)0. Lower right: 816 keV–1597 keV correlation. Spin-multipolarity sequence is 3(D, Q)2(Q)0.

harn and Rose [19]) is reversed if the transition is the second in the cascade and otherwise is left unchanged. We have chosen to adopt this convention. In the present investigation data for each cascade measured were used to calculate the mixing ratio of the first transition, thus our mixing ratios are the same as those calculated from the formulas of Biedenharn and Rose [19]). It should be noted that Krane and Steffen [ref. [21])] and other authors adopt the opposite convention for the sign of δ.

3. Results

The 329 keV–487 keV–1597 keV *cascade.* The three angular correlations, 329 keV–487 keV, 487 keV–1597 keV and 329 keV–1597 keV (487 keV unobserved) were each measured. The results of these measurements provided an internal check on the experiment since two independent values of the mixing ratio δ could be deduced from the data. These two values were in good agreement.

The 329 keV–487 keV correlation was obtained using the Ge(Li)–Ge(Li) system by gating the 487 keV γ-ray and displaying the spectrum in coincidence with it. In addition, the energy region just above 487 keV was simultaneously gated and its coincidence spectrum displayed. In this way it was possible to correct for an interfering correlation arising from coincidences between the 329 keV γ-ray and the Compton distribution of the 1597 keV γ-ray. More than 2.5×10^3 true coincidences were obtained at each angle. Of these less than 6 % were due to the interfering correlation. We obtain the correlation coefficients given in table 1.

A spin-multipolarity sequence 3(D,Q)4(Q)2 has been assumed for the 329 keV–487 keV cascade. The ratio A_2/A_0 was used (see fig. 2) to obtain $\delta(329) = 0.010 \pm 0.018$. The ratio A_4/A_0 precludes the other value of δ which is consistent with our A_2/A_0 measurement.

The 487 keV–1597 keV correlation was measured using the two NaI(Tl)–Ge(Li) systems. From a least-squares fit based on more than 10^5 true coincidences at each angle we obtain the correlation coefficients given in table 1.

These results are consistent with a 4(Q)2(Q)0 spin-multipolarity sequence for the 487 keV–1597 keV cascade. However if we assume a small M3 contribution to the 487 keV transition we obtain

$$\delta(487) = -0.002 \pm 0.007,$$

based on a 4(Q,O)2(Q)0 sequence. This value allows us to set an upper bound of 0.008 % on the octupole content of the 487 keV transition.

The 329 keV–1597 keV 1–3 correlation was also measured using the NaI(Tl)–Ge(Li) systems. More than 3.5×10^4 true coincidences at each angle were utilized to obtain the correlation coefficients given in table 1.

Based on a 3(D,Q)4(Q)2(Q)0 spin multipolarity sequence for the 329 keV–487 keV–1597 keV cascade we obtain, by the method of Arfken *et al.* [22]), $\delta(329) = 0.019 \pm 0.007$. If we allow the possibility of a mixture in the 487 keV transition and use the value for $\delta(487)$ reported above, the value which we calculate for $\delta(329)$ is unchanged.

We have formed a weighted average of our two values for $\delta(329)$ by using the reciprocals of the variances as weights. We obtain

$$\delta(329) = 0.018 \pm 0.007.$$

The 433 keV–1597 keV *correlation.* This 1–3 correlation was measured using the

two NaI(Tl)–Ge(Li) systems. The 1597 keV peak was gated in the NaI(Tl) channel and the 433 keV peak was obtained from the coincidence spectrum in the Ge(Li) channel. For this correlation the numbers of true coincidences obtained from the two systems were comparable (more than 2×10^3 true coincidences at each angle for system 1 and more than 6×10^3 true coincidences at each angle for system 2). The two sets of correlation coefficients obtained are given in table 1.

TABLE 1
Correlation coefficients obtained in the present investigation

Cascade	A_2/A_0	A_4/A_0
329 keV– 487 keV	-0.132 ± 0.015	0.042 ± 0.018
487 keV–1597 keV	0.103 ± 0.004	0.001 ± 0.007
329 keV–1597 keV	-0.124 ± 0.006	-0.004 ± 0.009
433 keV–1597 keV	0.142 ± 0.037 [a] 0.150 ± 0.013 [b]	-0.015 ± 0.057 [a] -0.011 ± 0.019 [b]
752 keV–1597 keV	-0.001 ± 0.017	0.001 ± 0.026
816 keV–1597 keV	-0.096 ± 0.004	-0.003 ± 0.006

[a]) System 1.
[b]) System 2.

Examination of the ^{140}Ce level scheme (see fig. 1) shows that for the 2516.1 keV level the spin assignment is uncertain, but the possible values of J are limited to 3 and 4. For each of these cases we have calculated the two values of $\delta(433)$ corresponding to our two measurements of A_2/A_0. Assuming a spin-multipolarity sequence of 3(D,Q)4(Q)2(Q)0 for the 433 keV–487 keV–1597 keV cascade we obtain

$$\delta(433) = 0.36\pm0.02.$$

If a 4(D,Q)4(Q)2(Q)0 sequence is assumed we obtain

$$\delta(433) = -0.14\pm0.03.$$

These values are weighted averages of the values obtained from the two systems.

The 752 keV–1597 keV *correlation.* This directional correlation was obtained by gating the 1597 keV peak in the NaI(Tl) channel and displaying the 752 keV peak in the coincidence spectrum of the Ge(Li) channel. A least-squares fit which utilized more than 4×10^3 true coincidences at each of the seven angles was made to the data. We obtain the correlation coefficients given in table 1.

Based on a 2(D,Q)2(Q)0 spin-multipolarity sequence for the 752 keV–1597 keV cascade we calculate

$$\delta(752) = -0.33\pm0.03.$$

The 816 keV–1597 keV *correlation.* This correlation was also measured by gating the 1597 keV γ-ray with the NaI(Tl) detector and displaying the 816 keV peak in

coincidence with it in the Ge(Li) channel. The data consisted of more than 3.5×10^4 true coincidences at each of the seven angles. We obtain the correlation coefficients given in table 1.

The spin-multipolarity sequence for the 816 keV–1597 keV cascade is 3(D,Q)2(Q)0. For this sequence the measured value of A_2/A_0 gives

$$\delta(816) = 0.031 \pm 0.005.$$

4. Summary

In table 2 we show the mixing ratios deduced from the present investigation. Our data are consistent with the assignment of the 487 keV γ-ray as a pure E2 transition. For both the 329 keV and the 816 keV γ-rays our data do not support pure M1 transi-

TABLE 2

Multipolarity determinations for transitions in ^{140}Ce

Transition energy (keV)	Correlation measured	Spin sequence assumed	Mixing ratio, δ	Quadrupole content, Q % [a]
329	329–487	3(D, Q)4(Q)2	0.018 ± 0.007 [b]	$0.032^{+0.030}_{-0.020}$
	329–(487)–1597	3(D, Q)4(Q)2(Q)0		
433	433–(487)–1597	3(D, Q)4(Q)2(Q)0	0.36 ± 0.02	$11.7^{+1.2}_{-1.0}$
		4(D, Q)4(Q)2(Q)0	-0.14 ± 0.03	$1.9^{+1.0}_{-0.7}$
487	487–1597	4(Q, O)2(Q)0	-0.002 ± 0.007	> 99.992 [c]
752	752–1597	2(D, Q)2(Q)0	-0.33 ± 0.03	$9.9^{+0.4}_{-0.5}$
816	816–1597	3(D, Q)2(Q)0	0.031 ± 0.005	$0.10^{+0.04}_{-0.03}$
868	868–1597	3(D, Q)2(Q)0	$-0.09 < \delta < 0.10$ [d]	> 1 [d]

[a]) For dipole-quadrupole mixtures the quadrupole content is obtained from the formula $Q = \delta^2/(1+\delta^2)$.

[b]) Value reported is a weighted average of values obtained from the 329–487 and 329–(487)–1597 correlations.

[c]) For this quadrupole-octupole mixture the quadrupole content is obtained from the formula $Q = 1/(1+\delta^2)$.

[d]) Data on the 868 keV–1597 keV correlation do not permit an accurate determination of a correlation function but do allow us to place these limits on the mixing ratio for the 868 keV transition. The limits of $0 \leqq Q \leqq 5$ % deduced from K-shell conversion coefficients by Baer *et al.* [1]) were used to eliminate one of the two ranges of δ consistent with our data.

tions but indicate slight E2 admixtures in each. The mixing ratio which we have deduced for the 329 keV transition is in good agreement with that reported by Letissier *et al.* [10]). However, the 433 keV–1597 keV correlation function which we measure is not in agreement with that reported by Cords [9]). Our data do not allow us to exclude either of the possible values given by Baer *et al.* [1]) for the spin of the 2516.1 keV level. We have deduced mixing ratios based on each of the two possible spin-multipolarity sequences. Our mixing ratio for the 752 keV transition is in agreement with that deduced from K-shell conversion coefficients by Baer *et al.* [1]).

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