Directional Correlation of the Gamma Rays of La^{140} and Ce^{140}*

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The directional correlation of coincident gamma rays in La^{140} and Ce^{140} has been reinvestigated. Using a coincidence circuit having a resolving time of $\tau=10$ nsec, the gamma rays in coincidence with a given gamma ray, as seen by a single counter, were displayed on a multichannel analyzer. Analysis of the intensity of the lines in the coincidence spectrum was made with the help of many calibration lines and a least-squares reduction technique using an electronic computer. In Ce^{140} the following coincidences have been investigated: 923–1597, 815–1597, 487–1597, 329–1597, and 329–487 keV. In La^{140} the coincidence 162–304 keV was investigated. A discussion of the data in light of the disintegration schemes is given.

1. INTRODUCTION

MANY studies have been made on the disintegration $Ba^{140}\rightarrow La^{140}\rightarrow Ce^{140}$. The level schemes are given in the *Nuclear Data Cards*.[1] The directional correlation of some gamma rays in the decay has been measured by various authors. Bolotin, Pruett, Roggenkamp, and Wilkinson[2] have measured the directional correlation of the gamma rays (815–1597), (487–1597), (329–1597), and (329–487 keV) in Ce^{140} while Kelley and Wiedenbeck[3] have made similar measurements on the same gamma-rays of Ce^{140} and, in addition, on the pair (304–162-keV) in La^{140}. Coleman[4] was able to separate a gamma ray of energy 923 keV in the decay of La^{140} from that formerly found at 815 keV and measure its directional correlation with the gamma ray at 1597 keV. Bishop and Perez y Jorba,[5] have measured the 487–1597 and 815–1597 keV correlation. Bannerman, Lewis, and Curran[6] have also measured the 815–1597-keV correlation. More recently, the correlations concerning the 329-keV transition, the 329–1597 and 329–487 correlations, have been measured in connection with the g-factor measurements of the 2.08 MeV level of Ce^{140}.[7–9] Due to the complexity of the spectra under consideration, the values of A_2 and A_4 determined from the directional correlation experiments of various authors are not in good agreement. The experiments described above were done using scintillation coincidence sets and single-channel analyzers and, in many cases, using wide windows so that several gamma rays may have been included in one window setting.

It is the purpose of the present study to try to obtain improved results using a multichannel analyzer in order to study simultaneously the directional correlation of several gamma rays with a given gamma ray. Since scintillation spectrometers suffer from poor resolution, a new "stripping" technique has been worked out with the help of an electronic computer.

2. APPARATUS AND EXPERIMENTAL METHOD

The apparatus consists of a conventional fast-slow coincidence counting set.[10] Figure 1 shows a block diagram of the apparatus. NaI(Tl) crystals (cylinders, $1\frac{1}{2}$ in.$\times 1\frac{1}{2}$ in.) were mounted on RCA 6810A photomultiplier tubes with the pulses from the anode being fed directly to limiters using 404A tubes. The pulses, whose amplitudes had been limited, were then clipped by a delay line and fed to a fast coincidence circuit employing a 1N23D diode. The pulses from the 1N23D diode enter an amplifier and discriminator unit. In the slow circuitry the dynode pulse of one phototube was fed to the usual linear amplifier and single-channel analyzer. The pulse from the dynode of the other photomultiplier was fed directly to a 200-channel analyzer. The output of the single-channel analyzer and that of the fast-coincidence circuit gate a slow-double-coincidence circuit which in turn gates the multichannel analyzer. Thus, all gamma rays in coincidence with those selected by the single-channel analyzer are displayed, allowing several correlations to be measured simultaneously.

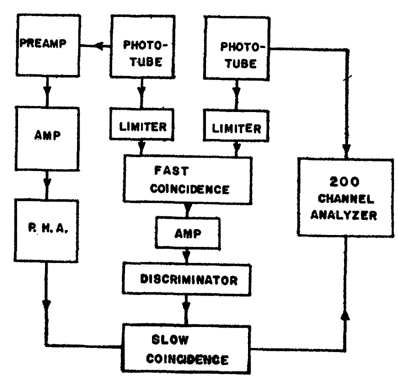

FIG. 1. Block diagram of the apparatus.

* Supported by the U. S. Office of Naval Research.

[1] *Nuclear Data Cards* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington, 5, D. C.)
[2] H. H. Bolotin, C. H. Pruett, P. L. Roggenkamp, and R. G. Wilkinson, Phys. Rev. **99**, 62 (1955).
[3] W. H. Kelley and M. L. Wiedenbeck, Phys. Rev. **102**, 1130 (1956).
[4] C. F. Coleman, Phil. Mag. **46**, 1132 (1955).
[5] G. R. Bishop and J. P. Perez y Jorba, Phys. Rev. **98**, 89 (1955).
[6] R. C. Bannerman, G. W. Lewis, and S. C. Curran, Phil. Mag. **42**, 1097 (1951).
[7] N. Kaplan, S. Ofer, and B. Rosner, Phys. Letters **3**, 391 (1963).
[8] R. M. Levy and D. A. Shirley, Phys. Letters **3**, 46 (1962).
[9] H. J. Körner, E. Gerdau, C. Gunther, K. Auerbach, G. Mielken, G. Strube, and E. Bodenstedt, Z. Physik **173**, 203 (1963).
[10] R. E. Bell, R. L. Graham, and H. E. Petch, Can. J. Phys. **30**, 35 (1952).

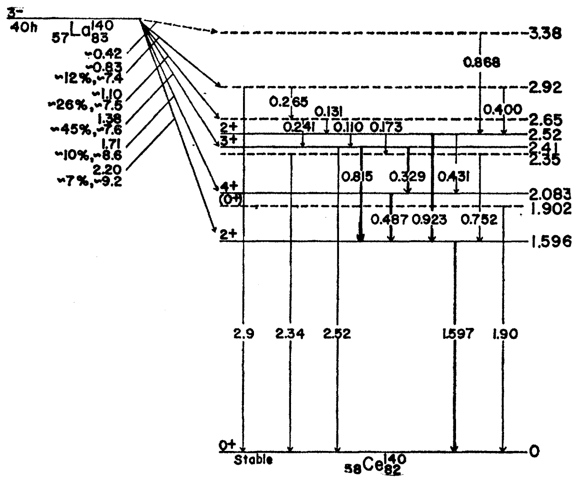

FIG. 2. Disintegration scheme of $La^{140} \rightarrow Ce^{140}$. Energies given in MeV.

The resolving time of the fast-coincidence circuit was measured by counting chance coincidences from two separate sources shielded from each other. Provisions were also made to measure the resolving time by inserting a delay in one branch of the fast circuitry. The resolving time was found to be $2\tau = 20$ nsec.

Ba^{140}–La^{140} equilibrium sources were used for the study of the gamma rays of Ce^{140}, and Ba^{140} sources for the study of the gamma rays of La^{140}. The separations were accomplished by standard procedures. The sources were solutions of the chlorides contained in cylindrically shaped polyethylene holders. The crystals were shielded from each other by lead cones. The source-to-crystal distance was 8.25 cm. The sources were centered to 1% or less and data were taken at 15° intervals with a random sequence of angles being employed.

3. METHOD OF DATA ANALYSIS

The analysis of the complex gamma-ray spectra involved in this work was carried out with the help of a computer. The main program used separates a complex spectrum into its individual gamma-ray components or separated spectra. The central idea used in this program is similar to that put forth by Heath.[11] If one considers a complex spectrum to be the sum of a group of monoenergetic spectral distributions, then the number of counts in a given channel, K, is given by

$$N_K = \sum_{J=1}^{n} N_{JK}, \qquad (1)$$

where N_{JK} is the number of counts contributed to the Kth channel by the Jth gamma ray of a spectrum having n gamma rays. Now if the shape is known, one can write

$$N_{JK} = I_J S_{JK}, \qquad (2)$$

where I_J is a multiplying constant, S is the shape, and S_{JK} is the probability of a gamma ray of energy E_J giving a count in the Kth channel. Thus, $\sum_K S_{JK} = 1$. The total number of counts emitted by the Jth gamma ray of energy E_J is

$$\sum_K N_{JK} = \sum_K I_J S_{JK} = I_J. \qquad (3)$$

Thus, I_J is the intensity, i.e., the total number of counts in the spectrum arising from the Jth gamma ray. Combining Eqs. (1) and (2) the number of counts recorded in the Kth channel can be represented by

$$N_K = \sum_{J=1}^{n} I_J S_{JK}.$$

With the shapes known, it only remains to calculate the intensities. In general, K will run over a much larger range of values than J, causing the problem to be greatly overdetermined. Since the problem is overdetermined one can apply least-squares methods to obtain n equations which are then solved simultaneously by a matrix inversion subroutine in the program. The I_J's are the solutions and once they are found all necessary information related to the separated spectra can be generated.

There are several practical problems encountered in carrying through the calculation just described. One of these is to obtain shapes for gamma rays of arbitrary energy. Heath[11] discusses this problem in detail. In the present work no attempt was made to write a program that would generate shapes for gamma rays of arbitrary energy. All the gamma-ray transitions of Ce^{140} and La^{140} in the regions of interest to this investigation fall near energies of isotopes emitting monoenergetic gamma rays. The shapes used were the spectra of K^{42}, Mn^{54}, Be^7, Nb^{95}, Cr^{51}, and Ce^{141}.

Another problem is the contribution of sum coincidence rates to the spectrum to be analyzed. A program was written that calculates the shape of the sum spectra and makes a reasonably accurate calculation of the intensity. This program is restricted to cascades of two gamma rays and does not take into account the effects of angular correlations on the intensity or the adjustment for branching at any of the transition levels involved. The decay of the Ba^{140} does not have any sums of strong enough intensity to interfere with the measurements of this investigation. The decay of La^{140} does have the possibility of interfering sum peaks at 815 keV from the 329–487 keV cascade. Calculations show that the intensity of the 815-keV sum peak is lower by about three orders of magnitude than the 329- or 487-keV photopeaks. Therefore, the effect of sum coincidences was considered to be negligible for the 815–1597-keV correlation.

Once the results from the main program are obtained and normalized, two other programs are used. One makes a least-squares fit of the data to the usual expression

$$W(\theta) = \sum_K A_K P_K(\cos\theta), \quad K = 0, 2, 4$$

[11] R. L. Heath, IRE Trans. Nucl. Sci. 9, 294 (1962).

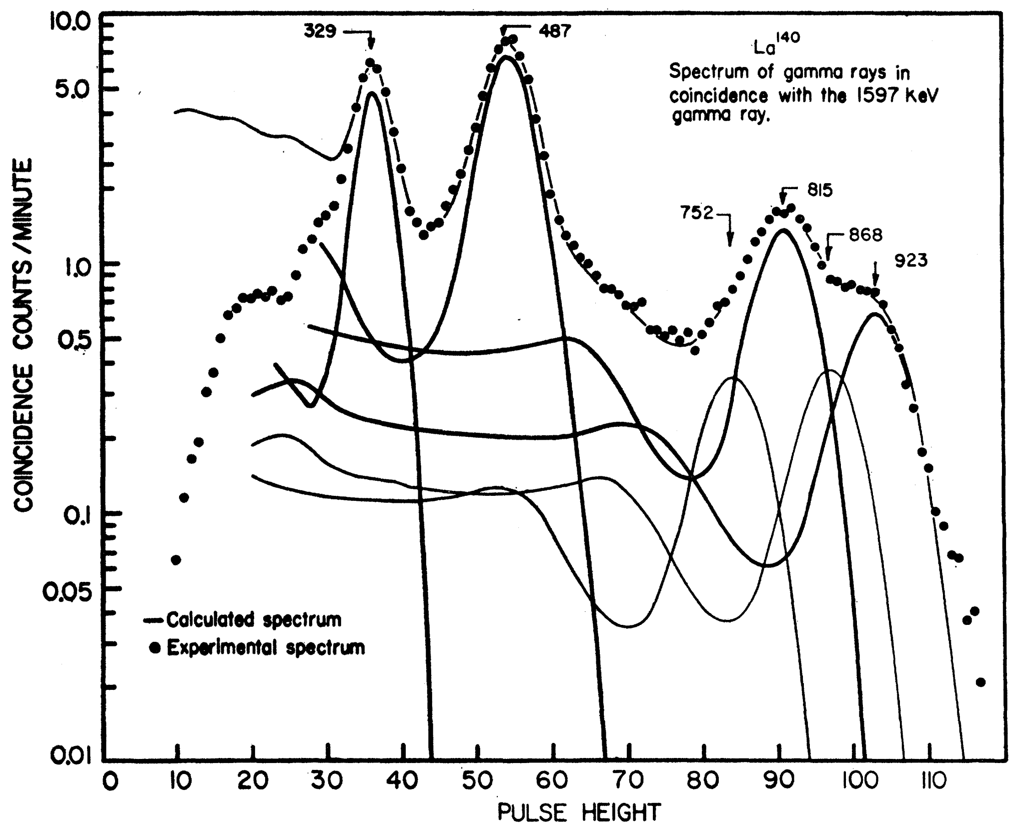

FIG. 3. Gamma rays in coincidence with the 1597-keV gamma ray of Ce^{140}. The actual data is represented by the dots and the solid lines are the individual gamma-ray components calculated by the computer.

while the other calculates the quadrupole content, where applicable, from the values of A_2.

Equation (3) of the previous section demonstrates that it is easy to obtain the total number of counts in a separated spectrum. It is more difficult to obtain the counts under the photopeak of a given gamma ray. For this reason, a sample of the data was analyzed for directional correlation using both the total number of counts in the spectrum and the counts under the photopeak. Both gave the same result. Thus, all results stated here were obtained by using the total number of counts in the spectrum.

The calculation of errors was carried out differently for the two isotopes reported here. In the case of La^{140} a large number of counts was collected in each coincidence spectrum. Thus, each 90- to 180-deg pass could be fitted by the least-squares procedure. Since a large number of coincidence counts was collected for each gamma-ray energy (approximately 300 000 in the case of the 487-keV gamma ray), it was felt that the main source of error would arise from the stripping process and instrumental errors. For this reason, the errors quoted for the correlations of Ce^{140} are simply the average deviation of individual measurements from that value found when all the data were combined. However, in the case of Ba^{140} the growth of the La^{140} gamma rays into the spectrum necessitated much shorter running times at each angle. Thus, the errors quoted for the correlations of La^{140} are calculated by the usual method of taking into account only statistical deviations.[12] The errors assigned to the quadrupole content are just the result of propagating the errors of A_2 through the mixing calculation.

4. DIRECTIONAL CORRELATION EXPERIMENTS ON THE GAMMA RAYS OF Ce^{140}

Figure 2 shows the presently accepted decay scheme of La^{140} as published in the *Nuclear Data Cards*.[1] It should be noted that the 1597-keV gamma-ray transition from the first excited state is in coincidence with all other gamma rays of lower energy. For this reason, gating on the 1597-keV line allows one to measure simultaneously all correlations of interest except that between the 329–487-keV lines. This correlation was measured by obtaining spectra in coincidence with a gate at 487 keV.

Figure 3 shows a typical spectrum in coincidence with the 1597-keV line. The dots represent the actual data. These data are then broken down by the program into distributions for the individual gamma rays as shown by the solid lines. The sum of all individual spectra is then given by the solid curve joining the dots. In this case, the analysis was made from about 300 keV out.

[12] M. E. Rose, Phys. Rev. **91**, 610 (1953).

TABLE I. Theoretical and measured correlation coefficients for the gamma rays of Ce^{140}.

Correlation (keV)	Spin sequence in order of descending energy	Theoretical coefficients		Measured coefficients and quadrupole content for the dipole transitions			Reference
		A_2	A_4	A_2	A_4	Q	
487–1597	4–2–0	0.102	0.009	0.097 ±0.005	0.016 ±0.009		A[a]
				0.107 ±0.005	−0.006 ±0.009		3
				0.106 ±0.024	−0.0013±0.0019		2
				0.083 ±0.015	0.021 ±0.019		5
923–1597	2–2–0	0.250	0.0	0.24 ±0.01	−0.005 ±0.018	<0.001	A
				0.200 ±0.035	−0.002 ±0.043	0.001 −0.01	4
815–1597	3–2–0	−0.0714	0.0	−0.099 ±0.009	−0.008 ±0.017	$0.001^{+0.0010}_{-0.0005}$	A
				−0.0094±0.0090	0.007 ±0.012		3
				0.0037±0.0054	−0.025 ±0.020		2
				−0.075 ±0.019	0.013 ±0.024		5
				0.111	0.0088		6
329–1597	3–4–2–0	−0.140	0.0	−0.104 ±0.015	−0.005 ±0.028	$0.007^{+0.006}_{-0.005}$	A
				−0.095 ±0.005	0.016 ±0.010		3
				−0.081 ±0.003			2
				−0.100 ±0.005		0.005	7
329–487	3–4–2	−0.140	0.0	−0.094 ±0.014	−0.044 ±0.007	$0.003^{+0.002}_{-0.001}$	A
				−0.103 ±0.006	0.025 ±0.008		3
				−0.092 ±0.020			2
				−0.105 ±0.003	−0.001 ±0.002	0.0017±0.0003	9
				−0.097 ±0.004	0.001 ±0.004	0.005	7
				−0.10 ±0.03		<0.005	8

[a] A denotes the results of the present investigation.

That the actual data fell below the calculated points in a region below 300 keV arises from the fact that the limiters did not work below this energy. The 700–900-keV region is of particular interest because of the wide divergence of values found by previous investigations for the 815–1597-keV correlation. In the present investigation it has been possible to separate the 815–1597- and the 923–1597-keV correlations.

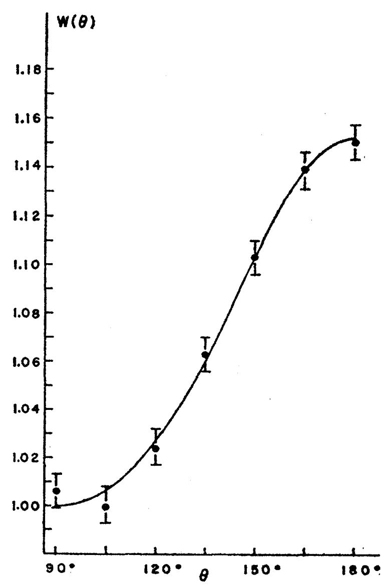

FIG. 4. Directional correlation of the 487–1597-keV cascade of Ce^{140}.

A. Results for Coincidences Using the 1597-keV Gate

In this series of experiments, five sets of data were taken at each angle. The computer analysis attempted to separate lines at energies of 923, 868, 815, 752, 487, 431, and 329 keV. No difficulties in spectrum analysis were encountered in the 700–950-keV region of the spectrum, which was the region of primary interest in this investigation. The line at 431 keV is so weak that it makes very little difference whether it is considered or not. The value of the ratio of the intensity of the 487-keV line to that at 431 keV (∼17) is in agreement with the work of Prikhodtseva and Khol'nov.[13] As mentioned before, the coincidence efficiency of the limiters began to drop at approximately 300 keV for this experiment. For this reason, correlations involving the 329-keV transition have larger errors than would be expected. No effort was made to improve the accuracy of this measurement since the correlations have been measured accurately by others[7–9] and the present measurements are in agreement with their results. Data of acceptable accuracy were obtained for the 329–1597, 487–1597, 815–1597, and 923–1597-keV correlations. The results are given in Table I. The 487–1597-keV correlation plays an important role in this investigation in that it was considered to be an internal check on the

[13] V. P. Prikhodtseva and Iu V. Khol'nov, Izv. Akad. Nauk SSSR Ser. Fiz. 22, 176 (1958).

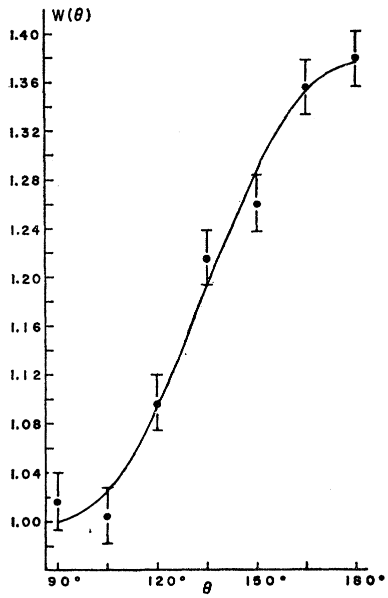

FIG. 5. Directional correlation of the 923–1597-keV cascade of Ce^{140}.

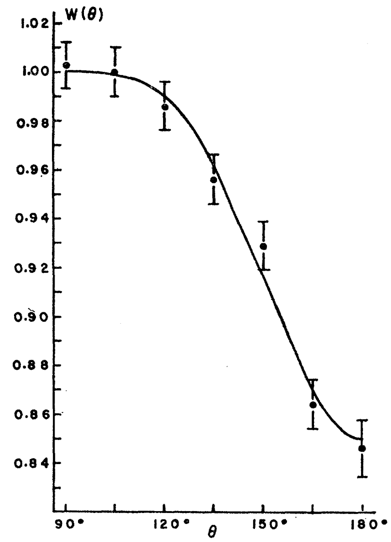

FIG. 6. Directional correlation of the 815–1597-keV cascade of Ce^{140}.

present method of analysis, since the spins of the states involved are known and the several previous determinations of the correlation coefficients agree among themselves and with the present experiment. The result for the 923–1597-keV correlation definitely indicates a spin of 2 for the 2520-keV state. A least-squares fit to the data can also be obtained assuming that $A_4=0$ which gives the same value of A_2 as is shown in Table I, indicating that this is a $2(D)\ 2(Q)0$ cascade. Calculations for the mixing of the 815- and 329-keV transitions were made assuming the spin assignment of the 2.41-MeV level to be 3, as given in the *Nuclear Data Sheets*.[1] The directional correlation curves for the 487–1597, 923–1597, 815–1597, and 329–1597-keV pairs are shown in Figs. 4–7, respectively.

B. Results for Coincidences Using the 487 keV Gate

This gate provided the data necessary to measure the 329–487-keV correlation. Unlike the 1597-keV gate, it accepts Compton events from higher energy transitions, the majority of which are due to the 1597-keV line. The method of analysis used here allows for accurate calculation of these Compton contributions. This is done by submitting the spectra from the 1597-keV gate as one of the shapes to be separated. Since these 1597-keV gate spectra demonstrate a correlation, a separate shape spectrum is needed for each angle. From this point, the analysis is analogous to the previous case and gives the results stated in Table I and the correlation curve of Fig. 8. This gate also provides more data for the 487–1597-keV correlation giving correlation coefficients that agree well with those from the 487–1597-keV data of the 1597-keV gate. Quadrupole mixing of the 329-keV transition can be calculated from both the 329–1597- and 329–487-keV correlations and the two are in agreement within errors.

C. Discussion of the Results

The present work has been able to resolve the difficulties in measuring the directional correlations of the 815–1597- and the 923–1597-keV pairs of gamma rays. As mentioned earlier the present experiment gives a value of $A_2=0.24\pm0.01$ which indicates a spin and parity of 2^+ for the state of energy 2.52 MeV.

The previous results of the directional correlation of the 815–1597-keV pair disagree both in regard to magnitude and sign. The present experiment separates the 815-keV line from its neighbors and, therefore, should give a more reliable result. Unfortunately, this correlation does not give a clear-cut result for the

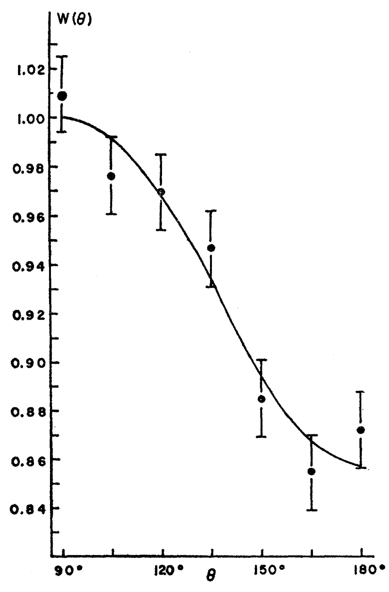

FIG. 7. 329–1597-keV skip correlation of Ce^{140}.

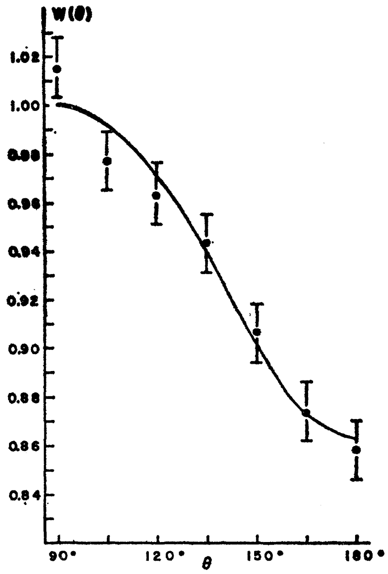

FIG. 8. Directional correlation of the 329–487-keV cascade of Ce^{140}.

character of the 2.41-MeV level on account of quadrupole mixing.

5. DIRECTIONAL CORRELATION MEASUREMENTS OF La^{140}

Figure 9 shows the decay of Ba^{140} as given in *Nuclear Data Cards*.[1] Although the limiters were redesigned for this experiment and functioned with maximum efficiency down to approximately 100 keV, the only feasible correlation is that of the 162–304-keV cascade. All data were taken with the gate on the 304-keV line. As before everything in coincidence with the gate was displayed on a multichannel analyzer. If the source were pure Ba^{140}, only coincidences with the 162- and 30-keV gamma rays would be expected. However, the daughter, La^{140}, grows in after separation, which results in coincidences being collected from the gamma rays of La^{140}. Since the 329-keV gamma ray of La^{140} grows into the gate, it was necessary to make frequent chemical separations. One pass, from 90 to 180 deg, was made with each source. This took about $5\frac{1}{2}$ h with data being collected at each angle for 30 min. The angles were still taken in a random sequence, but the sequence was taken in reverse order for alternate sources. In all, a total of nine passes were made resulting in 63 coincidence spectra. For these spectra the main computer program was used to remove the La^{140} contributions. To correct for the effect of La^{140} growing into the source, coincidence spectra were taken with exactly the same discriminator and window settings for the gate but using a pure La^{140} source. Thus, a coincidence spectrum of this type was taken at every angle so that its shape would display the correlation of the underlying La^{140} contribution to the Ba^{140} data. These spectra were then submitted to the program as shapes for calculating the intensity of the La^{140} contribution. After removal of the La^{140} contributions, there was a total of about 20 000 counts collected at each angle in the 162-keV spectrum giving the correlation curve shown in Fig. 10. The counting rates contributed by the 132-keV gamma ray were considered to be negligible.

Analysis of the data resulted in the following correlation for the 162–304-keV cascade:

$$W(\theta)=1+(0.062\pm0.005)P_2(\cos\theta)+(0.015\pm0.008)P_4(\cos\theta),$$

which does not agree well with the only other measurement by Kelley and Wiedenbeck[3] who find

$$W(\theta)=1+(0.0937\pm0.008)P_2(\cos\theta)-(0.056\pm0.010)P_4(\cos\theta).$$

On the basis of their data, they consider several possible spin sequences for the cascade, but conclude that the most favored is $1(Q)3(D,Q)4$ in descending order of energy. However, since the time of their investigation the ground-state spin of La^{140} has been measured by Petersen and Shugart,[14] using the atomic beam magnetic resonance method, and found to be $I=3$. There have been several investigations of the beta decay of Ba^{140} and the level scheme of La^{140}. Beach, Peacock, and Wilkinson[15] observed beta groups with end points at 480 and 1022 keV. Foster and Peacock[16] observed beta groups with end points at 515, 586, and 1022 keV. Kelley and Wiedenbeck[3] propose a level scheme for La^{140} using gamma-ray summing data obtained from a well crystal of NaI(Tl). Their scheme is in agreement with that of Fig. 9 with the exception that they propose that the 537-keV gamma ray terminates at the ground state. That portion of the scheme in Fig. 9 marked by heavier lines was proposed independently by Silant'ev,[17] and

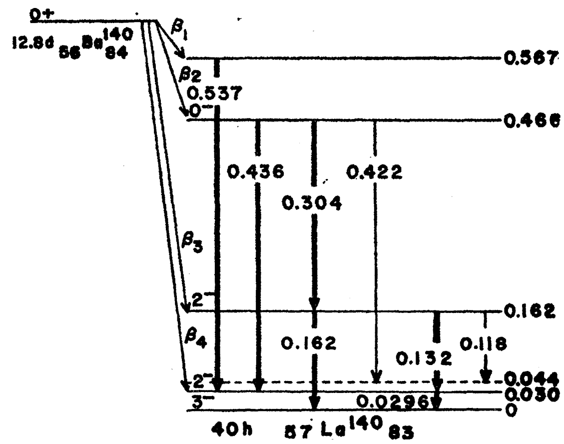

FIG. 9. Disintegration scheme of $Ba^{140} \rightarrow La^{140}$. Energies given in MeV.

[14] F. R. Petersen and H. A. Shugart, Bull. Am. Phys. Soc. 5, 343 (1960).
[15] L. A. Beach, C. L. Peacock, and R. G. Wilkinson, Phys. Rev. 76, 1624 (1949).
[16] C. C. Foster and C. L. Peacock (private communication).
[17] A. N. Silant'ev, Zh. Eksperim. i Teor. Fiz. 34, 569 (1958) [translation: Soviet Phys.—JETP 7, 394 (1958)].

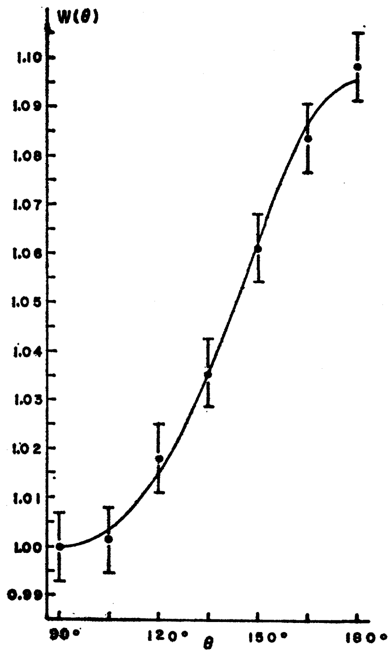

FIG. 10. Directional correlation of 162–304-keV cascade of La^{140}.

Boskma and DeWaard.[18] Silvant'ev used beta-gamma and gamma-gamma scintillation coincidence techniques, while Boskma and DeWaard used beta-gamma and gamma-gamma scintillation coincidence techniques and a coincidence magnetic spectrometer. Boskma and DeWaard observed a fourth beta group, β_3, not reported by the other investigators with an end-point energy of 830±50 keV. Table II lists the end-point energies and log *ft* values found by the above-mentioned authors.

TABLE II. End-point energies and log *ft* values for the beta transitions from Ba^{140} to La^{140}.

Group	End-point energy (keV)	log *ft*	Reference
β_1	480	6.9	17
	480		14
	515	6.5	15
β_2	586	7.3	15
	590	7.1	17
β_3	830	8.2	17
β_4	1020	8.6	17
	1022	7.7	15
	1022		14

[18] P. Boskma and H. Dewaard, Nucl. Phys. **14**, 145 (1959).

TABLE III. Possible correlation coefficients for the 162–304-keV gamma-ray cascade of La^{140}.

Spin sequence in descending order of energy	A_2	A_4
1(*D*)1(*Q*)3	0.0357	0
1(*D*)2(*D*)3	0.04999	0
2(*D*)2(*D*)3	−0.04999	0
2(*D*)1(*Q*)3	−0.0071	0
0(*D*)1(*Q*)3	−0.0714	0
0(*Q*)2(*D*)3	−0.0714	0

From the values of log *ft* for the transition β_3 the 162 keV level could be assigned a character 1^- or 2^-, the value 0^- being ruled out by internal conversion coefficient data.[18] On similar grounds the 466-keV level could be assigned characters 0^-, 1^- or 2^-. Table III shows the theoretical correlation coefficients for these possible spin sequences. Since the present measured value of A_2 does not agree with any of the values in Table III and the value of A_4 is definitely finite, one can only conclude that mixing must be present in at least one of the gamma transitions. In the spin sequences with the intermediate spin of 1, one would expect the 162-keV transition to be $E2$ with no mixing and the 304-keV transition to be predominantly $M1$ with some $E2$ mixing. However, the F_4[19] coefficient for a 1(*Q*)3 transition is zero. Thus, even with mixing, the sequence $J(D)1(Q)3$ would result in an A_4 identically equal to zero. Since the measured value of A_4 is finite, a spin of 1^- for the 162-keV level must be ruled out, leading to the conclusion that the 162-keV level must have a spin of 2^-. The value of the log *ft* for β_3 supports this conclusion. This leaves three possible spin sequences to be examined in Table III. If the 466-keV level has a spin of 0^-, one would expect the cascade to be described by $0(Q)2(D,Q)3$. However, a plot of A_2 and A_4 versus the quadrupole content reveals that the measured value of A_4 is inconsistent with any possible value of mixing, thus ruling out a spin of 0^- for the 466-keV level. Unfortunately, the correlation data cannot distinguish between the final two spin possibilities 1^- and 2^- for this level, since there is a possibility of mixing in both transitions. However, the log *ft* value for β_2 would indicate that this spin is probably either 0^- or 1^-. Therefore, it is felt that the most probable spin sequence is $1^- \to 2^- \to 3^-$.

[19] L. C. Biedenharn and M. E. Rose, Rev. Mod. Phys. **25**, 729 (1953).