

Directional Correlation of the Gamma Rays of La^{140} , Ce^{140} , Pt^{192} , and As^{75}

W. H. KELLY* AND M. L. WIEDENBECK

Department of Physics, University of Michigan, Ann Arbor, Michigan

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The decay scheme of La^{140} was investigated by using summing techniques. This scheme was found to be compatible with a modification of the schemes proposed earlier by other investigators. The 537-kev gamma ray was found to parallel the other transitions. Directional correlation measurements made in the 162-kev to 304-kev cascade do not allow definite spin and multipole order assignment. The following spin sequence (in the order of decreasing energy) appears to be the most probable $1(Q)3(D,Q)4$.

Ce^{140} was found to have energy levels with spins 0, 2, 4, and 3 in the order of increasing energy. The multipole orders were found to be: 1.600 Mev-quadrupole; 0.490 Mev-quadrupole; 0.328 Mev-dipole with some quadrupole admixture; and 0.815 Mev-dipole with some quadrupole admixture. These assignments are in agreement with those made by other investigators.

It was found that the energy levels of Pt^{192} have spins 0, 2, 2, 4 (3 cannot be definitely excluded) in the order of increasing energy.

The spins of the fourth and fifth excited states could not be assigned unambiguously but were found to lie in the ranges 2 to 5 and 3 to 5, respectively. Multipole orders of the transitions were assigned as follows: 316 kev-quadrupole; 468 kev-quadrupole (some dipole admixture cannot be definitely excluded); 612 kev-quadrupole; and 296 kev-dipole-quadrupole mixture. These assignments are in rough agreement with those deduced from internal conversion measurements made by other investigators and with previous directional correlation results obtained by other investigators.

The results obtained for As^{75} did not allow definite spin and multipole order assignments. However, the measurements permit limits to be placed on the spin assignments. The 281-kev level has spin 3/2 or 5/2 with 3/2 being preferred. The 269-kev level probably has spin 1/2 or 3/2 and the 405-kev state has a spin in the range 1/2 to 5/2 with 3/2 being preferred.

APPARATUS

THE instrument used consisted of a conventional type fast-slow coincidence spectrometer with an effective resolving time of 3×10^{-8} second. Amplification in the fast channel was accomplished by two three-tube feedback loops in cascade with pulse amplitudes limited between stages by biased crystal diodes. The remainder of the apparatus is similar to that described by Stewart, Scharenberg, and Wiedenbeck.¹

NaI(Tl) crystals, $1\frac{1}{2}$ in. diameter by 1 in. height, obtained from the Harshaw Chemical Corporation were coupled to Dumont 6292 photomultipliers. Each crystal had 12 mm of lead as lateral absorber and $\frac{1}{2}$ in. of aluminum on the front in addition to the 32 mils of aluminum and $\frac{3}{32}$ in. of packed magnesium oxide commercial mounting.

LANTHANUM-140

Previous Investigations

The decay scheme of lanthanum-140 has been the subject of several investigations.²⁻⁴ Cork *et al.*⁴ proposed the decay scheme shown in Fig. 1(A) based on relative intensities and energies. Figure 1(B) shows the decay scheme proposed by Beach *et al.*³ based on similar measurements. Figure 1(C) shows the scheme that has been suggested by Roggenkamp *et al.* and reported in the Hollander, Perlman, and Seaborg table of isotopes.²

It has been shown³ that there are no beta transitions between the ground states of La^{140} and Ce^{140} . This is

interpreted as meaning the ground state of La^{140} has a spin greater than or equal to 3 with a negative parity since the ground state of Ce^{140} has a spin 0^+ .

The internal conversion coefficient has been measured for the 540-kev gamma ray by Maerker and Birkhoff⁵ and Rohr and Birkhoff.⁶ They found that the $K/L+M$ ratio was 6 and 5.2, respectively. The K -shell conversion coefficient was determined to be $(5.6 \pm 1.9) \times 10^{-3}$. This leads to the classification of the 540-kev gamma ray as $E2$.

Present Investigation

The radioactive material obtained from Oak Ridge was BaCl_2 in a HCl solution. Figure 2 shows the spectrum of the equilibrium mixture of Ba^{140} and La^{140} .

Lanthanum chloride in dilute HCl was added as a carrier and the lanthanum then precipitated as the hydroxide by adding concentrated NH_4OH . After centrifuging, the liquid $\text{Ba}(\text{OH})_2$ solution was used as a source for the correlation measurements. This procedure was repeated every 4 to 5 hours. The maximum amount of lanthanum present during the directional correlation measurements was about 6 parts in 100.

Figure 3 shows a comparison of the spectrum taken within a half-hour after the chemical separation with that taken about six hours later. It is seen that the correlation measured in this time will be affected relatively little (less than 1% on the average) by the buildup of the La^{140} activity.

Decay Scheme

A study of the decay scheme was made using the gamma summing method. A 2-in. \times 2-in. NaI(Tl) crystal with a $\frac{1}{4}$ -in. hole drilled to its center was used. The

* Eastman Kodak predoctoral fellow. Now at Michigan State University, East Lansing, Michigan.

¹ Stewart, Scharenberg, and Wiedenbeck, *Phys. Rev.* **99**, 691 (1955).

² Hollander, Perlman, and Seaborg, *Revs. Modern Phys.* **25**, 469 (1953).

³ Beach, Peacock, and Wilkinson, *Phys. Rev.* **76**, 1624 (1949).

⁴ Cork, LeBlanc, Stoddard, Martin, Branyan, and Childs, *Phys. Rev.* **83**, 856 (1951).

⁵ R. E. Maerker and R. D. Birkhoff, *Phys. Rev.* **89**, 1159 (1953).

⁶ R. C. Rohr and R. D. Birkhoff, *Phys. Rev.* **98**, 1266 (1955).

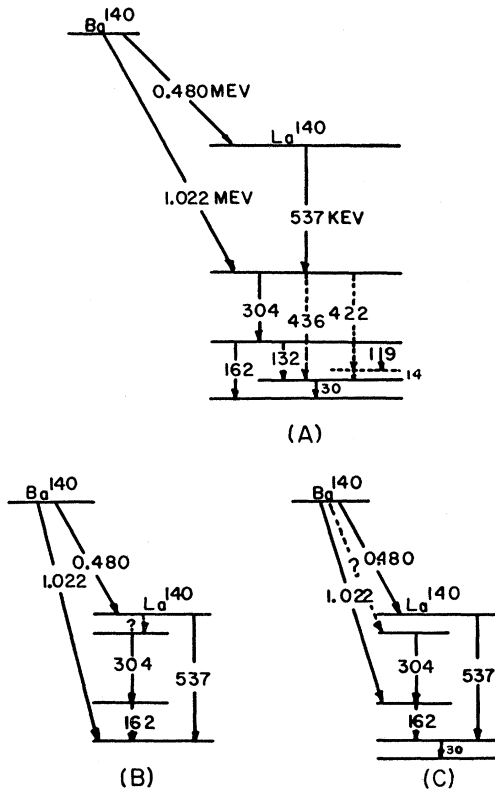


FIG. 1. Previously proposed decay schemes of La^{140} . (A) proposed by Cork *et al.*⁴ (B) proposed by Beach *et al.*³ (C) proposed by Roggenkamp *et al.*²

summing spectrum was measured with the source inside this hole. The outside spectrum was obtained with the source about $2\frac{1}{2}$ in. above the crystal. Figure 4 shows the spectra obtained.

Resolved peaks appear in the normal spectrum (source outside) at positions corresponding to gamma-ray energies of 162 kev, 304 kev, 436 kev, and 537 kev. The presence of a 30-kev gamma ray is indicated. This line appears at the lower limit of the spectrometer where the instrument is nonlinear. An unresolved gamma ray appears at a position corresponding to about 132 kev.

The summing spectrum shows strong diminishing of the 30-kev, 132-kev, 162-kev, and 304-kev lines. Sum lines appear at 436 kev and 466 kev. These lines are relatively weak and are not resolved. The 537-kev gamma ray does not appear to participate in the summing.

These data are seen to be consistent with the decay scheme which is also shown in Fig. 4. This decay scheme is essentially a combination of those presented in Fig. 1. It is suggested that the high-energy beta transition is to the 30-kev state because of the necessity of a high spin ground state. The existence of a beta transition with end-point energy at about 530 kev has to be postulated to account for the strong gamma transitions at inter-

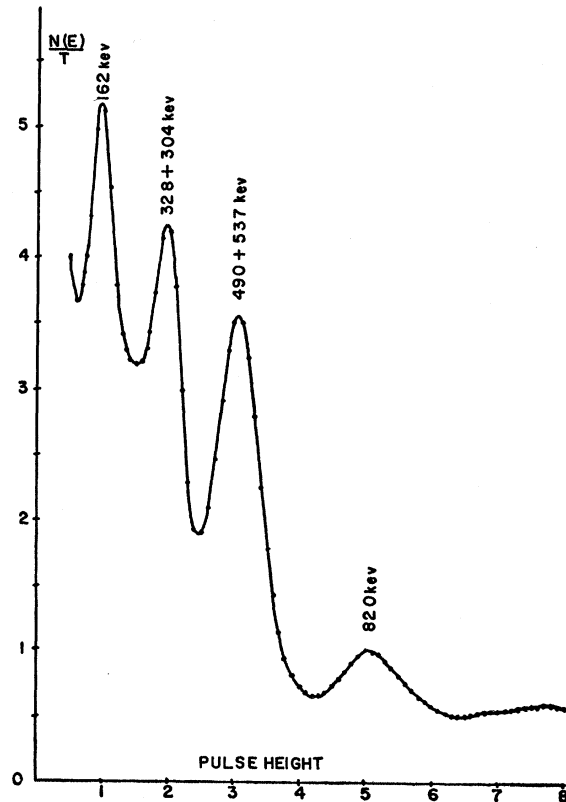


FIG. 2. Spectrum of the equilibrium mixture of Ba^{140} and La^{140} .

mediate energies. This beta ray has not yet been observed.

Directional Correlation Measurements of La^{140}

The differential discriminators were adjusted to positions corresponding to the 162-kev and 304-kev lines. The window widths corresponded to the approximate width of these lines at half-maxima. No corrections were made for the small interference 304 kev-132 kev correlation since the 132-kev gamma ray is very weak.

Figure 5 shows the directional correlation data that were obtained. These data can be represented by the least squares curve adjusted for finite geometry

$$W(\theta) = 1 + (0.0937 \pm 0.008)P_2(\cos\theta) - (0.056 \pm 0.010)P_4(\cos\theta).$$

This result cannot be explained using pure transitions. The existence of the large $P_4(\cos\theta)$ term indicates that the intermediate state must have a spin ≥ 2 .

It was found that the combinations (in the order of decreasing energy) $1(Q)3(D,Q)4$ and $1(O)4(D,Q)4$ could be used to explain the data. Figures 6 and 7 show the coefficients A_2 and A_4 plotted *versus* the mixture ratio δ for these combinations. The indicated regions show the values of δ consistent with the A_2 and A_4 values

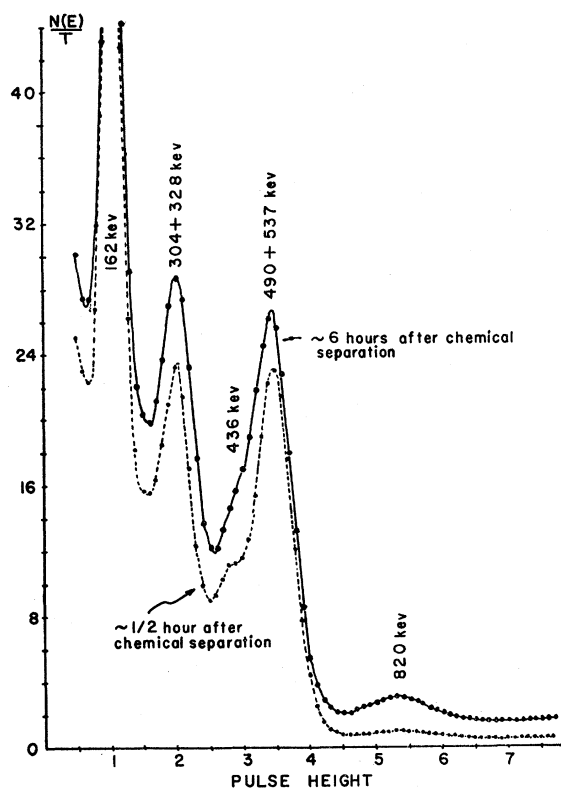


FIG. 3. Spectra of La^{140} taken approximately $\frac{1}{2}$ hour and approximately 6 hours after chemical separation.

obtained from the experiment. Figure 8 shows a similar curve for the $0(Q)2(D,Q)3$ combination. Although this gives δ values corresponding to the A_2 and A_4 that are not consistent, this case cannot be absolutely excluded because the mixture ratio values are so close to each other.

The double mixture combinations $1(D,Q)2(D,Q)2$ and $2(D,Q)3(D,Q)3$ cannot be definitely excluded. Explicit calculations were not carried out here. The presence of the negative A_4 eliminated the other double mixture combinations that were considered.

Discussion

The 1.022-Mev beta transition to the 30-keV state suggests that the 30-keV state has a spin equal to or less than 2. A similar statement can be made about the 466-keV state. Thus the 436-keV transition is probably at most quadrupole. From the spectrum, the 304-keV transition appears to be more probable than the 436-keV transition, and hence the 304-keV transition would not be expected to be octupole. From these evidences (and from the absence of a 466-keV crossover transition), $1(O)4(D,Q)4$ spin sequence is unlikely.

Internal conversion measurements⁶ made on the 537-keV gamma ray indicates that it is an $E2$ transition. The presence of the beta transition to the 537-keV state indicates that this state has a low spin value. It

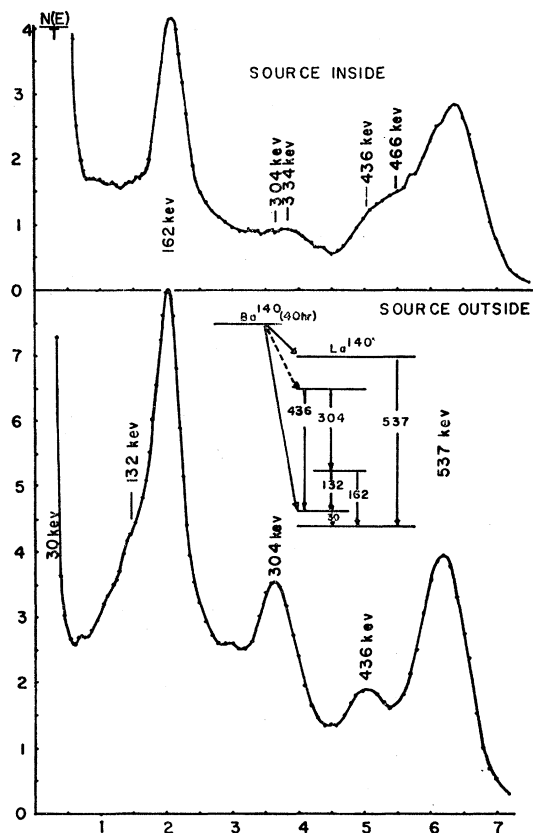


FIG. 4. Summing spectra and proposed decay scheme of La^{140} .

is therefore difficult to understand the absence of a transition between the 537-keV and 466-keV levels.

On the basis of these arguments, the $1(Q)3(D,Q)4$ sequence appears to be most favored for the 304-keV to 162-keV cascade.

CERIUM-140

Result of Previous Investigations

Lanthanum-140 decays by complex beta emission to cerium-140. The essential features of this decay scheme are shown with the spectrum in Fig. 9.

Both directional correlation and polarization correlation measurements have been made previously on the gamma rays of Ce^{140} ,⁷⁻⁹ with some agreement between investigators. Table I summarizes their results and interpretations. The parities were assigned on the basis of polarization correlation⁷ and internal conversion measurements.⁸ It is seen from this table that fair agreement is obtained for the 1.60 Mev-0.490 Mev correlation and definite disagreement on the 1.60 Mev-0.815 Mev correlation. From the decay scheme and spectrum, it would be expected that the best agreement

⁷ G. R. Bishop and J. P. Perez y Jorba, Phys. Rev. **98**, 89 (1955).

⁸ Bolotin, Pruett, Roggenkamp, and Wilkinson, Phys. Rev. **99**, 62 (1955).

⁹ B. L. Robinson and L. Madansky, Phys. Rev. **84**, 1067 (1951).

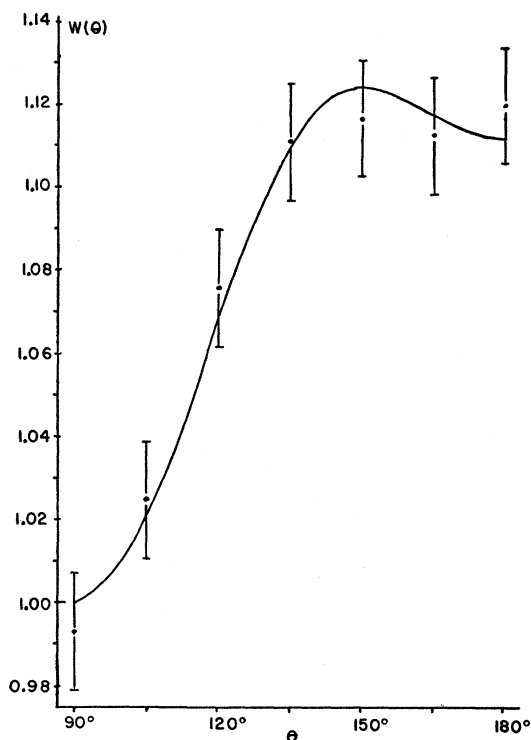


FIG. 5. Directional correlation measurements of the 304 keV-162 keV gamma cascade in La^{140} .

would be obtained for the latter correlation since smaller interference effects are expected. Although there is some agreement on the spin assignment, there is a disagreement on the magnitude of the correlation and hence the multipole order of the 0.815-Mev gamma ray.

The internal conversion coefficients have been measured by Bolotin *et al.*⁸ and Maerker and Birkhoff,⁵ and Rohr and Birkhoff.⁶

Results of the Present Investigation

The lanthanum-140 was obtained from Oak Ridge as lanthanum oxide. This was dissolved in concentrated

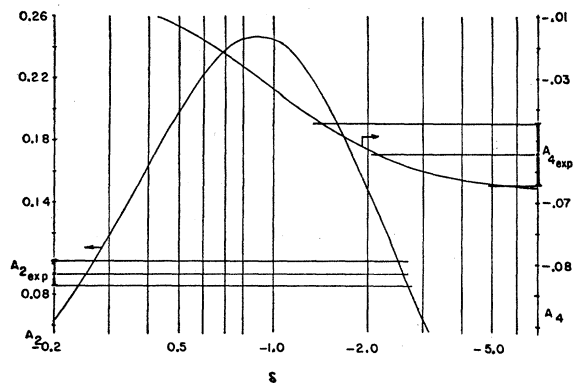


FIG. 6. A_2 and A_4 versus δ for the $1(Q)3(D,Q)4$ spin sequence in La^{140} .

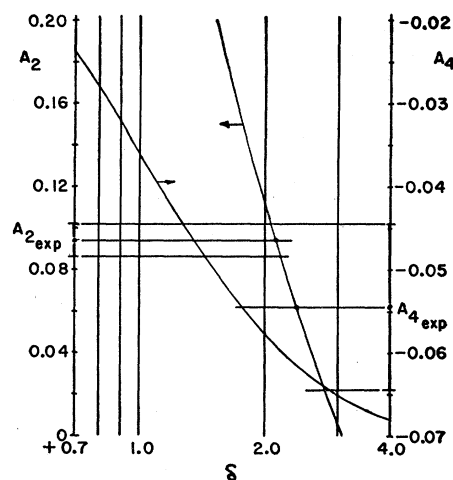


FIG. 7. A_2 and A_4 versus δ for the $1(O)4(D,Q)4$ spin sequence in La^{140} .

hydrochloric acid and the solution was used for the sources for the directional correlation measurements.

Directional Correlation of 1.600 Mev-0.815 Mev

Figure 10 shows the results of the directional correlation measurements made on the 1.600-Mev to 0.815-Mev gamma rays. Figure 10(A) shows the correlation measured with the discriminators set integral above 0.700 Mev and the detectors at 0.9% solid angle. The broken line shows, for comparison, the theoretical directional correlation for a $3(D)2(Q)0$ cascade modified for finite geometry. Least squares analysis and corrections for solid angle yields the equation

$$W(\theta) = 1 - (0.0094 \pm 0.009)P_2(\cos\theta) + (0.0068 \pm 0.012)P_4(\cos\theta).$$

This result is not in agreement with the curves obtained by Bolotin *et al.*⁸ and Bishop *et al.*⁷ (See Table I.)

It is possible that summing of the strong low-energy lines enhanced by crystal to crystal scattering can account for the difference between the results obtained

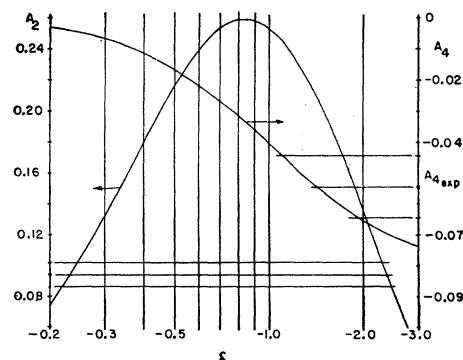


FIG. 8. A_2 and A_4 versus δ for the $3(D,Q)2(Q)0$ spin sequence in La^{140} .

here and those obtained by others. Summing could be appreciable here as well as in the measurements of Bolotin. Rough estimates indicate that this could be of the order of 10-20%. Much less summing would occur in the measurements of Bishop because of his small solid angle. On the other hand, crystal-to-crystal scattering may enhance a negative correlation. From these possible explanations, it would be expected that the initial measurements reported here might lie between those of Bishop and Bolotin since fairly large solid angles (0.9%) were used and lead shielding was used only on the sides of the crystals. The discriminator settings were approximately the same as those used by the other investigators.

5 mm of lead was placed on the front of each detector and the correlation measured again using the same discriminator settings. This lead should reduce scattering, if present, to a point such that the anisotropy was changed by less than 0.1% and summing was reduced by a factor of about 20. Figure 10(B) shows the directional correlation obtained with this arrangement. The least squares curve corrected for finite geometry is

$$W(\theta) = 1 - (0.0329 \pm 0.014)P_2(\cos\theta) + (0.0087 \pm 0.013)P_4(\cos\theta).$$

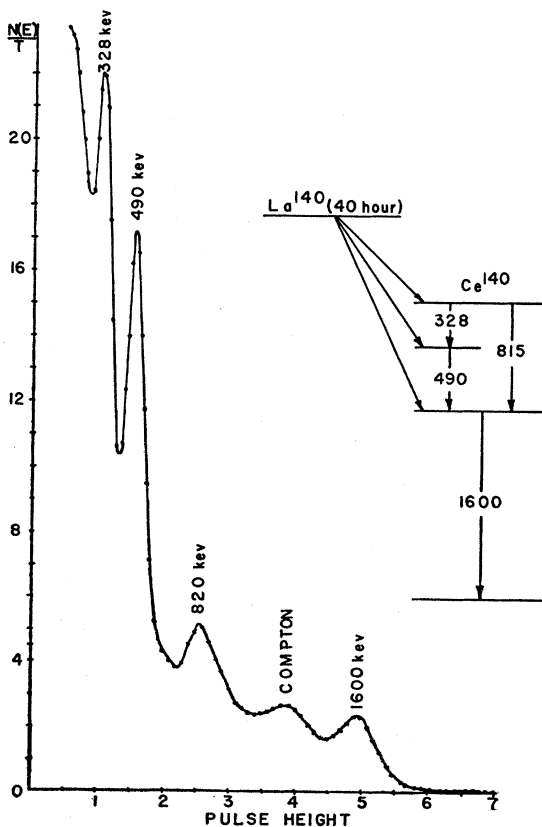


FIG. 9. Decay scheme and spectrum of ^{140}Ce .

As another check against the presence of summing and crystal-to-crystal scattering, one of the discriminators was adjusted to accept differentially only the high energy side of the 0.815-Mev photopeak and the other set integrally above about 1.40 Mev. With these settings, crystal-to-crystal scattering would be energetically eliminated and summing would be reduced by a factor of at least 8. The result is shown in Fig. 10(C). The solid-angle corrected least squares curve is given by

$$W(\theta) = 1 - (0.017 \pm 0.007)P_2(\cos\theta) + (0.012 \pm 0.009)P_4(\cos\theta).$$

The results of the three experiments reported here are reasonably close, indicating that summing and crystal-to-crystal scattering are small effects here. It should be noted that the errors quoted on the data presented above take into account instrument errors as well as statistical errors.

Perturbation effects are expected to be very small because of the estimated short lifetime of the intermediate state and the fact the source was in solution. In addition, data for the 1.600 Mev-0.490 Mev correlation exhibit the full theoretical correlation. Since these two correlations have the same intermediate state, it

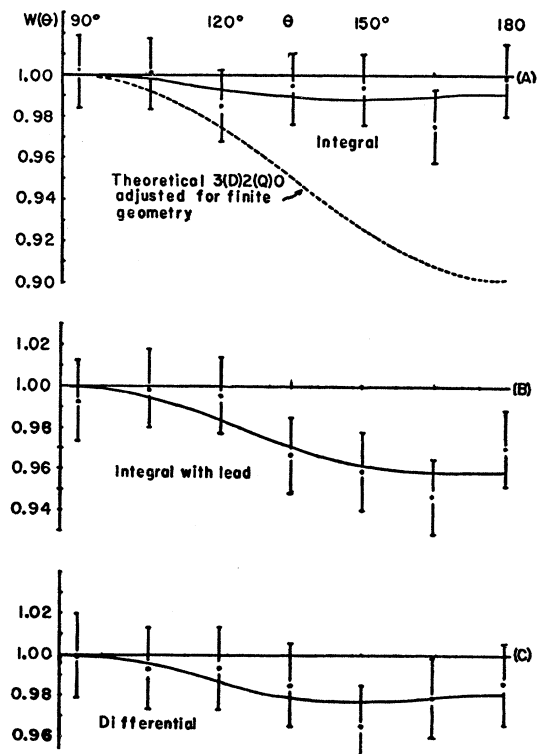


FIG. 10. Directional correlation measurements of 0.815 Mev-1.60 Mev. (A) discriminators were set integral above 700 kev. (B) Same discriminator settings, 5 mm of lead on the front of the detectors to reduce summing and crystal-to-crystal scattering. (C) One discriminator set differentially on the high-energy side of the 0.815-Mev line and the other integral above 1.40 Mev.

TABLE I. Results of angular correlation measurement on Ce^{140} .

Cascade	Measured correlation	Interpretation (in order of increasing energy)	Reference
1.60 Mev-0.815 Mev	$1+0.1111P_2+0.00879P_4$	0-2-4	9
	$1-(0.0745\pm 0.019)P_2+(0.013\pm 0.024)P_4$	$0^+-2^+-3^-$	7
	$1+(0.0367\pm 0.0054)P_2-(0.025\pm 0.020)P_4$	$0^+(Q)2^+(OD,Q)3^+$ $0^+(Q)2^+(OD,Q)4^+$ not excluded	8
1.60 Mev-0.490 Mev	$1+(0.083\pm 0.015)P_2+(0.021\pm 0.019)P_4$	$0^+-2^+-4^+$	7
	$1+(0.106\pm 0.0054)P_2-(0.0013\pm 0.0019)P_4$	$0^+-2^+-4^+$	8
1.60 Mev-0.328 Mev (1-3 correlation)	$1-(0.081\pm 0.03)P_2$	0-2-4 (3 or 4)	8
0.328 Mev-0.490 Mev	$1-(0.092\pm 0.020)P_2$	2-4 (3 or 4)	8

can be concluded that the small correlation observed is not due to perturbation effects.

The above correlation data can be interpreted by using the spin assignment $3(D,Q)2(Q)0$ with a mixture in the 0.815-Mev transition. Figure 11 shows the coefficient A_2 plotted versus the mixture ratio δ . The experimentally determined values of A_2 are indicated with their errors along with the range of δ consistent with these coefficients. The values of A_2 used here are from the data presented in Fig. 10. For this spin assignment, A_4 is negative for all real values of δ . However, it is small (~ -0.0005) for the ranges of δ indicated in Fig. 11. Even though the experimental values of A_4 are positive, they are within the experimental error of the coefficient determined from the mixture ratio.

The above data can also be fitted using a $2(D,Q)2(Q)0$ or a $4(Q,Q)2(Q)0$ spin sequence. For reasons to be presented in the discussion of the 0.328 Mev-1.600 Mev correlation, the 2-2-0 assignment is unlikely. Because of the relatively long lifetime of an octupole transition, as compared to a quadrupole transition, an octupole-quadrupole mixture ($\sim 40\%$ octupole, $\sim 60\%$ quadrupole) is unlikely.

The $3(D,Q)2(Q)0$ spin sequence is in best agreement with the internal conversion data^{5,6,8} which indicates that the 0.815-Mev gamma ray corresponds to an $E2+M1$ transition and the 1.600-Mev transition is $E2$.

Directional Correlation of 1.600 Mev-0.490 Mev

The least squares curve for the 1.600 Mev-0.490 Mev directional correlation corrected for solid angle is described by the equation

$$W(\theta) = 1 + (0.107 \pm 0.005)P_2(\cos\theta) - (0.00055 \pm 0.009)P_4(\cos\theta)$$

which compares well with the basic 2^2-2^2 theoretical value

$$W(\theta) = 1 + 0.1020P_2(\cos\theta) + 0.0091P_4(\cos\theta).$$

Summing and interfering Comptons detected with the 0.490-Mev photopeak contribute to the correlation here. Rough estimates show that summing is less than 1% of the observed coincidence rate. The interfering Compton electrons are due to the 0.815 Mev-1.600 Mev correlation and since this correlation is small, it is expected

that the effect would be to decrease slightly the observed anisotropy. These results are in agreement with the internal conversion data,^{5,6,8} and the correlation results obtained by others.

Directional Correlation of 1.600 Mev-0.328 Mev

This is the 1-3 correlation. The discriminators were set on the peaks with window widths corresponding to the widths of the lines at half-maximum. Least squares analysis and solid angle correction gives for $\nu_{\max} = 4$

$$W(\theta) = 1 - (0.0954 \pm 0.005)P_2(\cos\theta) + (0.016 \pm 0.01)P_4(\cos\theta)$$

and for $\nu_{\max} = 2$,

$$W(\theta) = 1 - (0.0897 \pm 0.008)P_2(\cos\theta),$$

in essential agreement with the result obtained by Bolotin *et al.*⁸ The theoretical curve for a $3(D)4 \cdots 2(Q)0$ is (the intermediate radiation is unobserved)

$$W(\theta) = 1 - 0.140P_2(\cos\theta).$$

Notice that the measured anisotropy is less negative than the theoretical value. This is in the direction to be expected if interfering Compton electrons of the 0.815-Mev and 0.490-Mev gamma rays are present. Part of this difference between theory and experiment can be due to a small dipole-quadrupole mixture in the 0.328-Mev transition. The $P_4(\cos\theta)$ term is needed if mixtures are present.

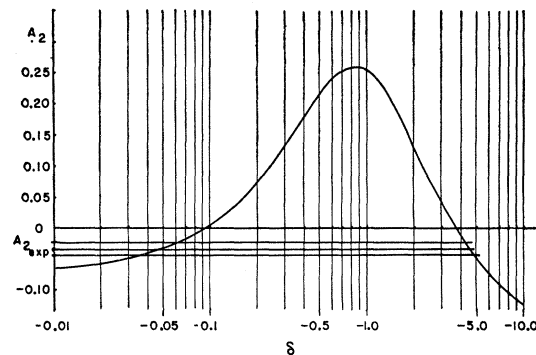


FIG. 11. A_2 versus δ for a $3(D,Q)2(Q)0$ assignment to the 0.815 Mev-1.60-Mev cascade of Ce^{140} .

A spin assignment of 2 to the top state (2.45-Mev level) would be unlikely since a quadrupole-octupole mixture would be needed to explain the negative correlation measured.

The spin sequence (3-4-2-0) determined by this cascade is in agreement with the sequences determined by the other correlations discussed above and with the internal conversion data.

Directional Correlation of 0.328 Mev-0.490 Mev

For this correlation, the differential discriminators were set to positions corresponding to the peaks of these lines and the window widths were adjusted to correspond to the width of the lines at half-maximum. Interfering radiations enter via both detectors here and so good quantitative data cannot be obtained from this correlation. The least squares curve corrected for finite geometry is given by

$$W(\theta) = 1 - (0.103 \pm 0.006)P_2(\cos\theta) + (0.0249 \pm 0.008)P_4(\cos\theta).$$

Theoretically, this should be the same as the 1.60 Mev-0.328 Mev correlation. The agreement is fair and serves to corroborate the spin assignments.

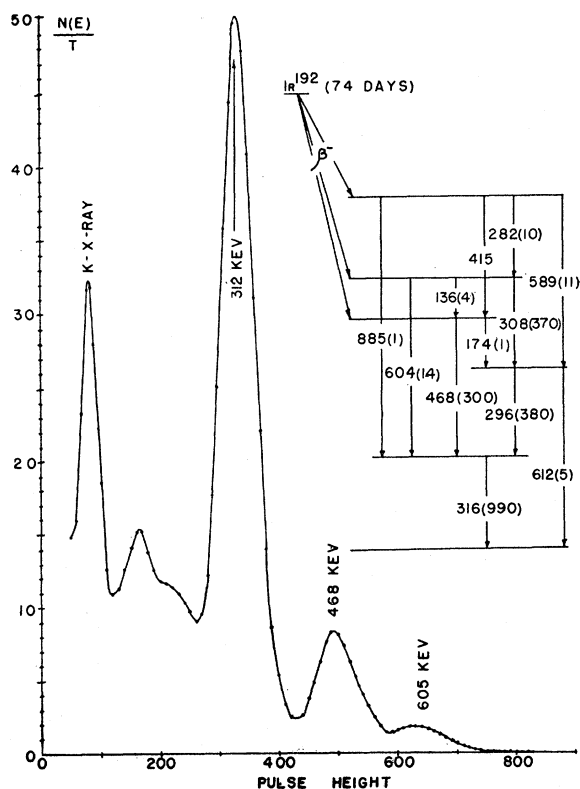


FIG. 12. Pt^{192} decay scheme and spectrum. The numbers in parentheses are the relative intensities as reported by Muller *et al.*¹⁶

Summary

The directional correlation data presented above are in good agreement with the spin assignments 0-2-4-3 for the energy levels of Ce^{140} in the order of increasing energy. These assignments are consistent with the internal conversion data^{5,6,8} and most of the angular correlation measurements⁷⁻⁹ made by the other investigators.

The measured correlation function for the cascade of 0.815 Mev-1.600 Mev was shown to be in disagreement with that measured by other investigators. Summing and crystal-to-crystal scattering (demonstrated to be small here) are postulated as possible explanations for the differences.

PLATINUM-192

Introduction

The decay scheme of Pt^{192} has been the subject of many investigations.¹⁰⁻¹⁷ The presently accepted decay scheme was proposed by Cork *et al.*¹⁰ and later modified by Pringle *et al.*¹¹ and Johns *et al.*¹²

TABLE II. Internal conversion data for Pt^{192} .^a

Gamma ray (kev)	K/L	Interpretation ^b
207	1.1	E2
296	2.2	E2+M1 or E2
309	2.2	E2+M1 or E2
316	1.9	E2
468	3.5	E2
590	6.4	E2+M1
605	5.6	E2+M1
613	5.6	E2
880	4.0	E3

^a See reference 13.

^b See reference 18.

Bashilov *et al.*¹³ have measured the internal conversion coefficients using a "ketron" spectrometer with a finely powdered source mounted on paper. The K/L ratios shown in Table II are computed from their data. The interpretations were made by using the empirical curves of Goldhaber and Sunyar.¹⁸

On the basis of $\log(ft)$ values and the observed pattern of gamma intensities, Johns *et al.*¹² assign a spin 4 to the ground state of Ir^{192} .

Since this investigation was completed, Taylor and Pringle¹⁹ have published the results of their directional

¹⁰ Cork, LeBlanc, Stoddard, Childs, Branyan, and Martin, *Phys. Rev.* **82**, 258 (1951).

¹¹ Pringle, Turchinets, and Taylor, *Phys. Rev.* **95**, 115 (1954).

¹² M. W. Johns and S. V. Nablo, *Phys. Rev.* **96**, 1599 (1954).

¹³ Bashilov, Anton'eva, Dzhelepov, *Izvest. Akad. Nauk Ser. Fiz. S.S.S.R.* **16**, 264 (1952).

¹⁴ J. L. Wolfson, *Proc. Roy. Soc. Canada* **44**, 193(A) (1950).

¹⁵ Muller, Hoyt, Klein, and DuMond, *Phys. Rev.* **88**, 775 (1952).

¹⁶ K. I. Roulston and R. W. Pringle, *Phys. Rev.* **87**, 930 (1952).

¹⁷ D. C. Lu and M. L. Wiedenbeck, *Phys. Rev.* **94**, 501 (1954).

¹⁸ M. Goldhaber and A. W. Sunyar, *Phys. Rev.* **83**, 906 (1951).

¹⁹ H. W. Taylor and R. W. Pringle, *Phys. Rev.* **99**, 1345 (1955).

correlation measurements. They propose a spin sequence 0,2,2,3(4),4 for the ground state and 316, 613, 784, and 921-keV levels. A spin of 3 is assigned to the 784-keV level since they have seen the 784-keV cross-over. Spin 4 was not definitely excluded, however.

Results

The source was finely powdered iridium metal obtained from Oak Ridge.

Figure 12 shows the spectrum obtained and the essential features of the decay scheme. The numbers in parentheses are relative intensities as measured by Muller *et al.*¹⁵

Pt¹⁹² 468 keV-316 keV Correlation

The differential discriminators were set to positions corresponding to the peak of the 468-keV gamma ray

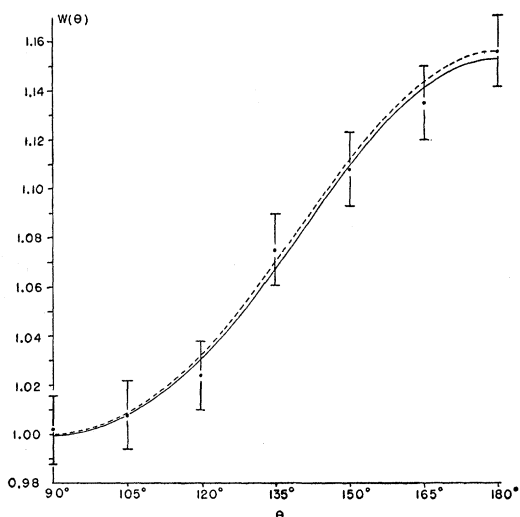


FIG. 13. Pt^{192} 468 keV-316 keV directional correlation. The solid curve is the least-squares curve and the broken curve the theoretical correlation for a 2^2-2^2 basic correlation modified for finite geometry.

316-keV composite peak. The window widths were adjusted to correspond to the widths of the measured lines at half-maximum. Figure 13 shows the least squares fit to the data (solid line) with the flags indicating the rms statistical errors, and the theoretical curve (broken line) for a basic 2^2-2^2 correlation modified for finite geometry. The least squares curve corrected for solid angle²⁰ was found to be

$$W(\theta) = (0.0992 \pm 0.006)P_2(\cos\theta) + (0.0085 \pm 0.007)P_4(\cos\theta),$$

which agrees well with the theoretical expression

$$W(\theta) = 1 + 0.102P_2(\cos\theta) + 0.0091P_4(\cos\theta)$$

²⁰ Annihilation radiation was used to obtain the angular resolution correction.

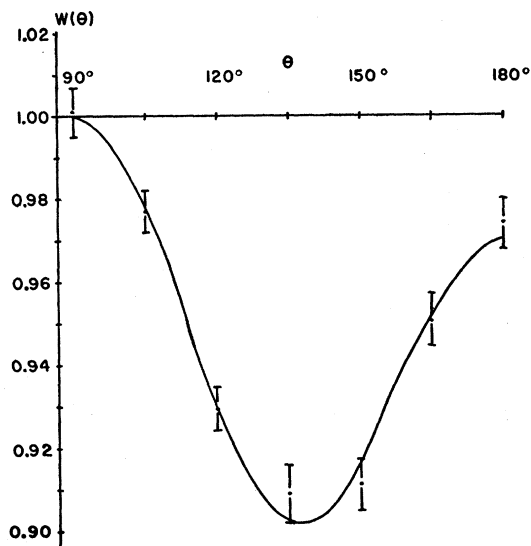


FIG. 14. Pt^{192} 306 keV-296 keV-316 keV "double" directional correlation.

for the basic 2^2-2^2 correlation. Since the measured coefficient of the P_4 term is small, it is conceivable that the correlation could be explained by a $3(D,Q)2(Q)0$ spin sequence as suggested by Taylor *et al.*¹⁹ However, in view of the good agreement between the above data and the theoretical curve for a $4(Q)2(Q)0$ sequence, we feel that the $3(D,Q)2(Q)0$ sequence appears unlikely.

Interference correlations arising from other cascades (e.g., summing of lower-energy photoelectrons with Compton electrons, etc.) are small and no corrections were made.

Pt¹⁹² 306 keV-296 keV-316 keV Correlations

Both counters were set on the composite peak corresponding to the gamma rays of 306 keV+296 keV+316 keV with window widths approximately equal to the width of the photoelectron peak at the base. The data, essentially a combination of four correlations (i.e., 306-296, 296-316, 306-316, and 468 Compton-316), is shown in Fig. 14 with the least squares curve and root mean square statistical errors indicated. This curve, when corrected for finite geometry, can be represented by

$$W(\theta) = 1 - (0.0565 \pm 0.004)P_2(\cos\theta) + (0.0929 \pm 0.005)P_4(\cos\theta).$$

The presence of the relatively large positive coefficient of the $P_4(\cos\theta)$ term and the existence of the 612-keV cross-over gamma transition indicate that the second excited state must have a spin of at least $J_1=2$.

Different combinations of J_0 and J_1 (spins of the fourth and second excited states, respectively) for $0 \leq J_0 \leq 6$ and $2 \leq J_1 \leq 4$ in the sequence J_0-J_1-2-0 were used in calculations in an attempt to interpret the above result. For $J_1=2$ and a dipole-quadrupole mix-

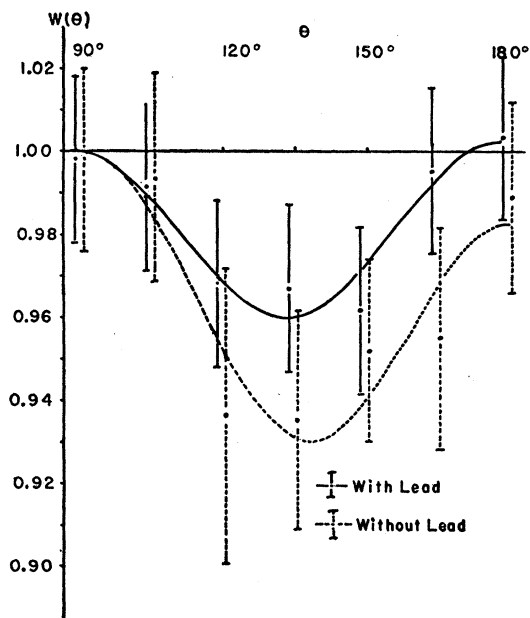


FIG. 15. Pt^{192} 589 keV–612 keV directional correlation measured with (solid curve) and without (dashed curve) 7.5 mm of lead absorbers to reduce summing.

ture in the intermediate transition the following method of calculation was used. The correlation function was assumed to be a function of two adjustable parameters β and δ , where δ is the mixture ratio

$$\delta^2 = I(L+1)/I(L)$$

and β is the ratio of the intensity of the triple 306–296–316 keV cascade to the intensity of the double 296–316 keV cascade. From the decay scheme and relative intensity measurements it was expected that $\beta \sim 1$. The coefficients of the Legendre polynomials were equated and the resulting fourth-order equations for δ were solved for $J_0=0, 1, 3, 4, 5$, and 6. The intensity ratios β were computed for the real values of δ . These results are presented in Table III. The best values of β are obtained for $J_0=3, 4$, and 5. Since the relative intensities of the gamma rays are not accurately known, corrections for the interfering 4–2–0 correlation could not be made.

For $J_0=J_1=2$, there is a high probability that a mixture can occur in two transitions. The data can be interpreted with such a spin choice.

Making similar calculations it was found that no combinations of $J_0=0, 1, 2, 3, 4, 5, 6$ and $J_1=3, 4$ could be used to fit the above results. (Mixtures were considered in only one of the transitions at a time.)

Pt^{192} 589–612 keV Correlation

Because of the relatively low intensities of these two lines and the high intensities of the low-energy lines, it is possible to obtain a large interference due to

accidental summing of the low-energy lines in the crystal detectors, e.g., 4 summing coincidences to one 589 keV–612 keV coincidence. Imposing 7.5 mm of lead between the source and the crystals allows an improvement by a factor of approximately 8 in the good/bad ratio of coincidences. Figure 15 shows the measured correlations with and without lead. It seems reasonable on the basis of these data to conclude that the correlation function for the 589–612 keV cascade lies above the correlation shown. This means that the 1201-keV state can have a spin 2, 3, 4, or 5.

Summary and Conclusions

From the above data, we see that the spin sequence in the order of increasing energy 0, 2, 2, 4(3), (2, 3, 4, 5), (2, 3, 4, 5) is consistent with the directional correlation data.

A comparison of the above data and the internal conversion results of Bashilov¹³ (Table II) shows that the agreement for the 468-, 316-, and 612-keV transitions is good. The conversion data implies that the parities are all positive for the first three excited states.

From the information presented, it is seen that a mixture of $\sim 95\%$ quadrupole and $\sim 5\%$ dipole for the 296-keV data is consistent with the angular correlation data. This is in approximate agreement with the K/L internal conversion ratio. This helps to confirm the spin assignment of a 2^+ to the second excited state. Unfortunately, neither of the two types of data are good enough to enable us to differentiate definitely between the spins 2, 3, 4, 5 of the fourth excited state. The best agreement between the internal conversion data and the directional correlation data is obtained for a spin assignment of 2^+ or 3^+ to the fourth excited state and a spin 5 to the fifth. The ground state of Ir^{192} has been assigned a 4^- and the $\log ft$ values determined by Johns¹² indicate that the two higher-energy beta transitions can be classed as first forbidden and the lower-energy transitions as second forbidden. We see the spin assignment of 3^+ to the fourth excited state and a negative parity for the fifth is reasonable.²¹

TABLE III. Summary of calculations made for triple cascade using δ and β as parameters.

Spin sequence	δ	β	Remarks
0–2–2–0	Complex	—	
1–2–2–0	0.880	0.267	
	0.644	0.015	
3–2–2–0	-4.01 ± 0.7	1.16 ± 0.7	Possible
	-0.350	< 0	
4–2–2–0	-13.9 ± 4.8	1.28 ± 0.8	Possible
	-0.344	< 0	
5–2–2–0	-3.91 ± 0.7	1.15 ± 0.7	Possible
	-0.318	< 0	
6–2–2–0	-0.726	0.111	
	-0.334	< 0	

²¹ Baggerly, Marmier, Boehm, and DuMond, Phys. Rev. **100**, 1364 (1955).

ARSENIC-75

The gamma-gamma directional correlations of the unresolved 124 keV–281 keV and the 138 keV–269 keV cascades of As^{75} were studied previous to the recent paper by Schardt and Welker.²² The results obtained and the methods used are in essential agreement with theirs and so will be described very briefly here.

Figure 16 gives the decay scheme suggested by Lu *et al.*²³ and the spectrum obtained.²⁴ The correlations were measured for three different positionings of the single-channel discriminators.

(a) Both discriminators set symmetric on the two pairs of unresolved lines with the window widths adjusted to correspond to the approximate width of these lines at their base. The least-squares curve after correction for finite geometry (Rose correction²⁵) is

$$W(\theta) = 1 - (0.103 \pm 0.005)P_2(\cos\theta) - (0.0100 \pm 0.009)P_4(\cos\theta).$$

For the case of two coefficients, $W(\theta) = 1 - (0.107 \pm 0.006)P_2(\cos\theta)$.

(b) The higher energy discriminator setting was increased 10% and the low-energy position decreased 10% (i.e., to accept proportionately more of the 281 keV–124 keV cascade). The window widths here and in case (c) were about a third as wide as they were in case (a). The least squares curve after correction for finite geometry is

$$W(\theta) = 1 - (0.278 \pm 0.008)P_2(\cos\theta) - (0.0072 \pm 0.009)P_4(\cos\theta).$$

For the case of two coefficients, $W(\theta) = 1 - (0.281 \pm 0.009)P_2(\cos\theta)$.

(c) In like fashion, the equipment was adjusted so that more of the 269 keV–138 keV cascade was accepted. The curve obtained after correction for angular resolution is

$$W(\theta) = 1 - (0.0166 \pm 0.007)P_2(\cos\theta) + (0.0080 \pm 0.009)P_4(\cos\theta).$$

For the case of two coefficients, $W(\theta) = 1 - (0.0136 \pm 0.007)P_2(\cos\theta)$.

It should be pointed out that in each of the above cases the coefficient of the $P_4(\cos\theta)$ term is comparable to the error and its existence is therefore questioned.

Since the measured correlation functions $W(\theta)$ have been normalized by dividing through by A_0 , the correlation function for two interfering cascades can be written

$$W(\theta) = [nW_s(\theta) + W_a(\theta)] / (n+1),$$

where $W_s(\theta)$ and $W_a(\theta)$ are the correlation functions for the cascades of 138 keV–269 keV and 124 keV–281 keV

²² A. W. Schardt and J. P. Welker, *Phys. Rev.* **99**, 810 (1955).

²³ Lu, Kelly, and Wiedenbeck, *Phys. Rev.* **97**, 139 (1955).

²⁴ A. W. Schardt, *Bull. Am. Phys. Soc. Ser. II*, **1**, 85 (1956).

²⁵ M. E. Rose, *Phys. Rev.* **91**, 610 (1953).

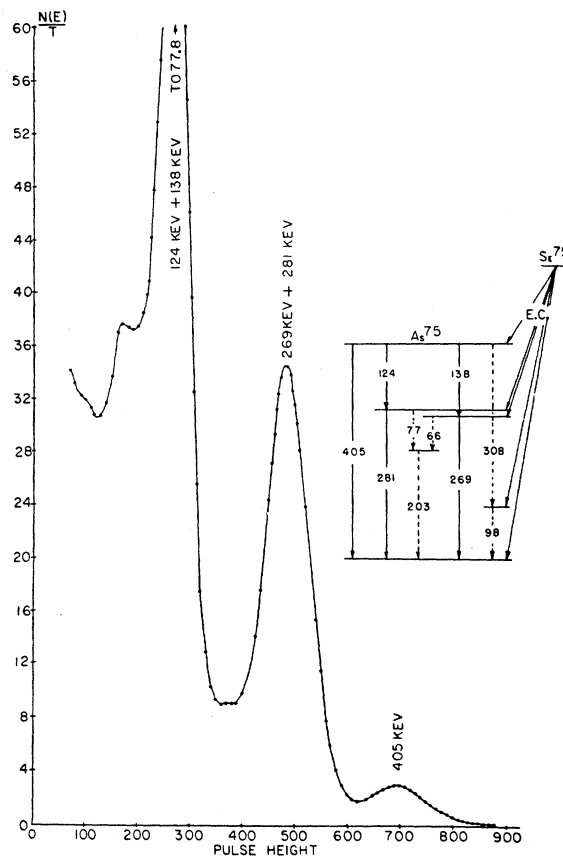


FIG. 16. Decay scheme and spectrum of As^{75} .

keV, respectively, and

$$n = \frac{N_s E(138)E(269)}{N_a E(124)E(281)}$$

N_s/N_a is the ratio of the total number of nuclei that decay per second through the 138 keV–269 keV mode to the number that decay through the 124 keV–281 keV mode. $E(138)$ is the efficiency of the apparatus for the 138-keV gamma ray and includes the solid angle, etc. These efficiencies can be evaluated from known line shapes and equipment parameters. The three quantities N_s/N_a , $W_s(\theta)$, and $W_a(\theta)$ can be evaluated from the above data. The following results were obtained:

$$W_s(\theta) = 1 + (0.016 \pm 0.03)P_2(\cos\theta)$$

for the 138 keV–269 keV correlation,

$$W_a(\theta) = 1 - (0.41 \pm 0.03)P_2(\cos\theta)$$

for the 124 keV–281 keV correlation, and

$$N_s/N_a = 1.8 \pm 1.$$

These results are similar to those obtained by Schardt and Welker²¹ who used a slightly different method of

analysis that necessitated the determination of N_s/N_a in a separate experiment.

Conclusions

Neither of the above correlation functions can be obtained using unmixed transitions. Comparison of these results with the internal conversion data of Schardt and Welker²¹ and the agreement between the two different directional correlation data lead us to essentially the same conclusions put forth by them: namely, the 269-keV state probably has a spin 1/2 although a spin 3/2 cannot be definitely excluded; the 281-keV state probably has a spin 3/2 with a 5/2 spin

not being excluded. We found that the assignment of spins 1/2, 3/2, or 5/2 to the 405-keV level is consistent with the correlation data. The $\log ft$ values, the internal conversion data, and the decay spectrum of Ge^{75} presented by Schardt and Welker²² seem to indicate that the 3/2 assignment to this state is more likely.

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Neutron-Capture Cross Section of Actinium-227†

H. W. KIRBY, G. R. GROVE, AND D. L. TIMMA*

Mound Laboratory, ‡ Monsanto Chemical Company, Miamisburg, Ohio

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In the production of actinium-227 by thermal neutron irradiation of radium-226, thorium-228 has been recovered as a by-product. The reactions involved are: $\text{Ra}^{226}(n,\gamma)\text{Ra}^{227}\rightarrow\text{Ac}^{227}+\beta^-$, and $\text{Ac}^{227}(n,\gamma)\text{Ac}^{228}\rightarrow\text{Th}^{228}+\beta^-$. The thermal neutron capture cross section of actinium-227 is 495 ± 35 barns, as determined by the pile oscillation method. The half-life of thorium-228, determined by daily alpha counting for two years, is 697.8 ± 0.7 days or 1.910 ± 0.002 year.

INTRODUCTION

IN his announcement of the isolation of actinium-227 from neutron-irradiated radium-226, Hagemann¹ noted the presence of an undetermined, but large, amount of lead-212, indicating the presence of thorium-228. The possibility that thorium-228 was formed as a result of second-order neutron capture made it desirable to determine the capture cross section of actinium.

The radium which had been used by Hagemann for his actinium studies and from which the actinium and thorium isotopes had been separated was subsequently irradiated again in a thermal neutron reactor. The radium was adsorbed on a Dowex-50 ion exchanger, and the radium and actinium isotopes were eluted in two and four normal nitric acid, respectively. Thorium isotopes could be expected to remain on the resin.²

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* Present address: Monsanto Chemical Company, Inorganic Chemicals Division, Everett Station, Boston, Massachusetts.

‡ Mound Laboratory is operated by Monsanto Chemical Company for the U. S. Atomic Energy Commission under Contract Number AT33-1-GEN-53.

¹ F. Hagemann, *J. Am. Chem. Soc.* **72**, 768 (1950).

² F. T. Hagemann, *The Actinide Elements* (McGraw-Hill Book Company, Inc., New York, 1954), National Nuclear Energy Series, Plutonium Project Record, Vol. 14A, Div. IV, Chap. 2, p. 27.

EXPERIMENTAL

A preliminary estimate of the cross section of actinium was made by an alpha-counting technique. Several small portions of the radium- and actinium-depleted resin were boiled with concentrated nitric acid and filtered. The filtrates were purified of radium isotopes by several barium nitrate precipitations, and portions of the solution containing thorium isotopes were alpha-counted daily. Other portions of the filtrates were analyzed for actinium-227 by extraction with thenoyltrifluoroacetone.¹ No actinium was detected.

The radium fraction recovered from the ion-exchange column was analyzed calorimetrically and the radium-226 content was found to agree with the sample weight to within less than one percent. Thus, radium-228 was not present in any significant amount.

The growth and decay of the thorium-228/thorium-227 isotopes was analyzed by the method of differential decay,³ and the ratio of their activities was converted to a thorium-228/actinium-227 weight ratio as of the time of removal from the reactor.

The integrated neutron flux could be estimated with about 10% uncertainty, and the total time in the reactor was known. The neutron capture cross section of ac-

³ H. W. Kirby, *Anal. Chem.* **26**, 1063 (1954).