Gyromagnetic Ratio of an Excited State and Other Angular Correlation Measurements for Pb^{204m}†

V. E. KROHN AND S. RABOY
Argonne National Laboratory, Lemont, Illinois
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The spin assignment, 9–4–2–0, for the levels of the triple cascade of Pb^{204m} has been found consistent with angular correlation measurements provided the first and second gamma rays are multipole mixtures. The first gamma ray must be $E5$ with a one percent mixture of $M6$, and the second gamma ray must be $E2$ with 0.5 percent $M3$. The gyromagnetic ratio of the 2.6×10^{-7} sec state was measured by angular correlation techniques, and the result, $g=+0.054\pm0.005$ nuclear units, was obtained. If one assumes the validity of the spin 4 assignment for this state, the magnetic moment is 0.22 ± 0.02 nuclear magnetons.

An attempt to observe time-dependent attenuation of the angular correlation in an HNO_3 medium gave an upper limit of 10 percent attenuation with variation of the delay time from 1.85×10^{-7} to 5.5×10^{-7} sec.

INTRODUCTION

IN some cases it is possible to use angular correlation techniques to determine the gyromagnetic ratio of an excited state of a nucleus which decays through this state by the successive emission of nuclear radiations. Up to the present time, measurements of this type have been restricted to gamma-gamma cascades. For such measurements it is necessary that there be an angular correlation pattern other than isotropy between a pair of gamma rays having one member of the pair before and one after the state of interest. Also, present techniques require that the state of interest have a lifetime in the range of 10^{-5} to 10^{-9} sec. The method of measurement involves studying the angular correlation pattern of the gamma rays as a function of a magnetic field applied perpendicularly to the plane of the two gamma-ray counters. The precession of the pattern depends on the gyromagnetic ratio of the intermediate state of the gamma-gamma cascade, i.e., on the interaction of the applied field with the magnetic moment of this state.[1,2] A state of Cd^{111} has been measured at Zurich by this method,[3] and the present authors have reported a measurement for an excited state of Ta^{181}.[4] The existing data on 1.1-hour Pb^{204m} with a 2.6×10^{-7} sec intermediate state indicated that it would be suitable for a similar measurement.[5] During the course of the present work a preliminary result for Pb^{204m} has been published,[6] and a group at the University of Illinois have been studying the same isomer.[7]

Apparatus

The detecting equipment consisted of a pair of NaI(Tl) crystals and 5819 photomultiplier tubes. The signals from the photomultipliers were received by a fast-coincidence circuit (10^{-7} sec) acting in parallel with pulse-height analyzers (Fig. 1). The outputs from the two analyzers together with the output from the fast-coincidence circuit were then fed into a relatively slow triple-coincidence circuit ($\sim3\times10^{-6}$ sec). The pulse-height analyzers could be used as discriminators or as analyzers with window widths up to 25 volts in an analyzer range of 100 volts. For delayed coincidence work, various lengths of 1500-ohm delay-line cable were inserted at the appropriate input to the fast-coincidence circuit. The resolving time of the system was obtained by measurement of chance coincidences at frequent intervals during the course of the experiment and was found to be independent of counting rate and pulse height throughout the ranges used.

The NaI(Tl) crystals were right cylinders, 1.5 inches in diameter and 1 inch long, packaged by the Harshaw Chemical Company. Lucite light pipes, 5 inches long and 1.75 inches in diameter, were used to reduce magnetic field effects on the photomultipliers. The Lucite was optically bonded to the NaI(Tl) package with Dow Corning No. 200 Fluid. The Lucite light pipes and photomultipliers were packaged by Moenich.[8] In order to minimize the effect of the magnetic field on the photomultipliers, μ-metal shields were used together with three concentric cylinders of soft iron.

The magnet was made of Armco iron, and fields up to 23 000 oersteds were obtained. Conical pole pieces with a full angle of 120° and flat ends were used. The gap height was 0.25 inch, and the gap diameter was 0.5 inch. The sources were contained in small teflon holders placed in the magnet gap. The holders were cylinders with an inner height and diameter of 0.125 inch, and the walls were 0.06 inch thick. Lead collimators were used to protect the NaI(Tl) crystals from scattered radiation and to define the solid angles which had a half-angle of 10°.

In order to study scattering from the magnet and magnetic field effects, control experiments were per-

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[1] G. Goertzel, Phys. Rev. **70**, 897 (1946).
[2] K. Alder, Helv. Phys. Acta **25**, 235 (1952).
[3] Aeppli, Albers-Schonberg, Frauenfelder, and Scherrer, Helv. Phys. Acta **25**, 339 (1952).
[4] S. Raboy and V. E. Krohn, Phys. Rev. **95**, 1689 (1954).
[5] Sunyar, Alburger, Friedlander, Goldhaber, and Scharff-Goldhaber, Phys. Rev. **79**, 181 (1950).
[6] V. Krohn and S. Raboy, Phys. Rev. **95**, 608 (1954).
[7] Frauenfelder, Lawson, and Jentschke, Phys. Rev. **93**, 1126 (1954).
[8] R. K. Swank and J. S. Moenich, Nucleonics **12** No. 8, 41 (1954).

TABLE I. Measurements with Co^{60}.

Integral bias level (kev) Counter 1	Counter 2	Magnetic field (oersteds)	$W(180°)/W(90°)$
510	200	0	1.125±0.010
575	575	0	1.154±0.010
575	575	19 600	1.148±0.013

TABLE II. Measurements with Co^{60}.

Integral bias level (kev) Counter 1	Counter 2	Magnetic field (oersteds)	$W(225°)/W(135°)$
510	200	0	0.997±0.011
510	200	8400	1.012±0.023
200	200	19 600	0.99 ±0.025

formed with the gamma rays of a Co^{60} source. The intermediate lifetime of this cascade is so short that the magnetic field would not be expected to distort the angular correlation pattern. The ratio of coincidences at 180° to the coincidences at 90° was measured for the conditions shown in Table I.

In addition, the ratio of coincidences at 225° to coincidences at 135° was studied for different fields. These angular positions are symmetrical, and the ratio expected is unity. The results are tabulated in Table II.

The data of Tables I and II demonstrate the lack of magnetic effects on the performance of the apparatus. The results of Table I are not corrected for solid angle, and the last two measurements are considered consistent with existing results[9–11] for the Co^{60} cascade. The first entry of this table shows the effect of scattering from the magnet.

Pb^{204m} Decay Scheme

During the course of the present experiment it became evident that the Pb^{204m} decay scheme which appeared in the survey literature was incomplete, and evidence for a third gamma ray (899 kev) was reported in a previous publication.[12] The proposed decay scheme is presented in Fig. 2. This result has now been confirmed by groups in Stockholm[13] and Amsterdam[14] who have reinvestigated the internal conversion spectrum of Pb^{204m}. The energy values in the decay scheme were taken from the results of the Amsterdam group,[14,15] who report a precision of ±1 kev for each gamma-ray energy. The order of the last two gamma rays in the decay scheme has not been determined experimentally, but there are several arguments in favor of the order given in Fig. 2, among which are the following: (1) If Pb^{204} had a first excited state at 375 kev, one would expect the main branch in Tl^{204} beta decay to proceed via this state,[16] but no 375-kev gamma rays have been observed in the Tl^{204} decay; and (2) Both transitions following the 2.6×10^{-7} sec state of Pb^{204} have been identified as $E2$[13–15] for which the Weiskopf formula[17] predicts that the 899-kev transition should be 100 times faster than the 375-kev transition which is itself anomalously slow by a factor of 10^3.

The spin assignments of Fig. 2 are suggested by the internal conversion measurements[13–15] and are consistent with the angular correlation measurements described in the next section.

Angular Correlation Measurements

A series of measurements were made to determine the anisotropy of the angular correlation patterns of the 912–375-kev and 912–899-kev gamma-ray cascades. For these measurements the Lucite light pipes were not used, the resolving time of the apparatus was increased to 2.0×10^{-7} sec, and the half-angle subtended by the detectors was increased to 12°. The sources used were thallium foils irradiated with 22-Mev deuterons and dissolved in concentrated nitric acid.

In order to select the 912–899-kev cascade, 0.125 inch of lead and 0.015 inch of tantalum absorber was used on each counter. The analyzers were set to accept pulses above 500 kev, and the input to one side of the fast coincidence unit was delayed 3.5×10^{-7} sec. For selecting the 912–375-kev cascade, the lead absorber was removed from one counter, this counter's discriminator was set to accept pulses from 290 to 460 kev, and the output from the other counter was delayed 3.5×10^{-7}

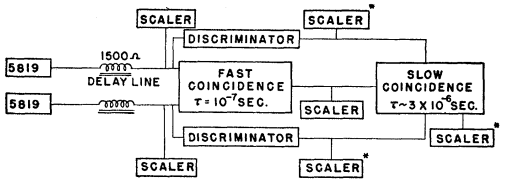

FIG. 1. Block diagram of the counting circuits with amplifiers omitted. All data of the present report came from the scalers marked with asterisks.

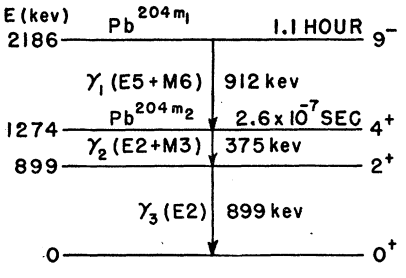

FIG. 2. Proposed decay scheme of Pb^{204m}.

[9] E. L. Brady and M. Deutsch, Phys. Rev. **78**, 558 (1950).
[10] J. S. Lawson, Jr. and H. Frauenfelder, Phys. Rev. **91**, 649 (1953).
[11] E. D. Klema and F. K. McGowan, Phys. Rev. **91**, 616 (1953).
[12] V. E. Krohn and S. Raboy, Phys. Rev. **95**, 1354 (1954).
[13] I. Bergstrom *et al.* (private communication).
[14] Wapstra, Nijgh, and Ornstein (private communication).
[15] Maeder, Wapstra, Nijgh, and Ornstein, Physica **20**, 521 (1954).
[16] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952) and Yuasa, Laberrigue-Frolow, and Feuvrais, Compt. rend. **238**, 1505 (1954).
[17] M. Goldhaber and A. Sunyar, Phys. Rev. **83**, 906 (1951).

sec. With the latter arrangement a small contribution from the 912–899-kev cascade is present. This contribution can be determined by changing the delay from one input to the other as the change does not alter the efficiency of the system for detection of the 912–899-kev cascade. It was found that 16 percent of the coincidences detected came from the 912–899-kev cascade. The measurement of the 912–375-kev cascade was corrected for the 16 percent contribution of the 912–899-kev cascade. After solid angle corrections, the measured anisotropies, $[W(180°)/W(90°)]-1$, were 0.44 ± 0.02 for the 912–899-kev cascade and 0.34 ± 0.02 for the 912–375-kev cascade.

In order to obtain calculated values in agreement with the measured anisotropies it was necessary to consider multipole mixing of both the first and second gamma rays. The formulas used in the calculation are outlined in the following paragraphs. We are indebted to Professor Fritz Coester[18] for this extension of the work of Biedenharn and Rose.[19]

The A_k in the angular correlation formula,

$$W(\theta)=\sum_k A_k P_k(\cos\theta), \tag{1}$$

are given by

$$A_k=A_k(ab)A_k(cb) \tag{2}$$

for two successive gamma transitions; and by

$$A_k=A_k(ab)\bar{A}_k(bc)A_k(dc) \tag{3}$$

for the case where the second gamma ray of a triple cascade is not observed. The $A_k(ab)$ and $A_k(bc)$ are given by

$$A_k(ab)=F_k(L_1L_1 j_a j_b)+\delta_1^2 F_k(L_1+1\ L_1+1\ j_a j_b) +2\delta_1 F_k(L_1\ L_1+1\ j_a j_b), \tag{4}$$

and $\bar{A}_k(bc)$ is given by

$$\bar{A}_k(bc)=[W(j_b L_2 k j_c;\ j_c j_b)+\delta_2^2 W(j_b\ L_2+1\ k j_c;\ j_c j_b)] \times(2j_b+1)^{\frac{1}{2}}(2j_c+1)^{\frac{1}{2}}, \tag{5}$$

where j_a, j_b, etc., designate the spins of the nuclear states; L_1, L_2, etc., indicate the lowest allowed multipolarity of the gamma rays; δ_1, δ_2, etc., are the mixing parameters of Biedenharn and Rose; and the W's are Racah coefficients. The indices a, b, etc., and 1, 2, etc., start at the top of the cascade. Note that

$$\bar{A}_k(bc)=\bar{A}_k(cb). \tag{6}$$

The F coefficients of Eq. (4) are identified by

$$F_k(L\ L\ j_a j_b)=F_k(L\ j_a j_b), \tag{7}$$

$$F_k(L\ L+1\ j_a j_b)=(-)^{j_b-j_a-1}(2j_b+1)^{\frac{1}{2}} \times(2L+1)^{\frac{1}{2}}(2L+3)^{\frac{1}{2}}G_k(L j_a j_b), \tag{8}$$

where the terms on the right involve the F and G coefficients of Biedenharn and Rose.[19] An extensive table of F coefficients as defined above is being prepared at this laboratory,[20] but for the present calculation, values involving spins less than 4 were obtained from the tables of Biedenharn and Rose,[19] and the values indicated in Table III were calculated from the formulas of the same authors.

TABLE III. Values of some F coefficients.

	$k=2$	$k=4$	$k=6$
F_k (5594)	−0.5523	0.0963	−0.0037
F_k (6694)	0.1481	−0.3997	0.0647
F_k (5694)	0.2957	−0.2319	0.0412

For δ's less than unity and the 9–4–2–0 spin assignment, only one solution gave calculated anisotropies equal to the measured values. This was $\delta_1=-0.09\pm0.06$ with $\delta_2=-0.07\pm0.02$. The errors indicate the range of the δ's corresponding to the uncertainty in the measured anisotropies.

The expected values of the K shell conversion coefficients can be calculated with the mixing parameters obtained from the anisotropy measurements. The pure electric- and magnetic-multipole conversion coefficients were obtained by interpolation from tables.[21] The expected conversion coefficients were then calculated and are given in Table IV together with measurements of the Amsterdam group.[14]

TABLE IV. Calculated and experimental values of K conversion coefficients and relative intensities of K conversion lines of Pb^{204m}.

δ_1	δ_2	K_{375}/K_{912}	K_{899}/K_{912}	$\alpha_K(912)$	$\alpha_K(375)$
−0.09	−0.07	0.78	0.11	0.063	0.049
0	0	0.69	0.12	0.059	0.040
Experiment[a]		0.75 ± 0.03	0.135 ± 0.02		

[a] Preliminary results from reference 14.

We conclude that the 9–4–2–0 spin assignment with mixing parameters in the neighborhood of $\delta_1=-0.12$; $\delta_2=-0.07$ is consistent with the existing data. The 912-kev gamma ray is then $E5$ with a one percent mixture of $M6$, and the 375-kev gamma ray is $E2$ with a one-half percent mixture of $M3$. The values of the A_k were calculated for this spin and mixing assignment and are presented in Table V. These values for A_k

TABLE V. Calculated values of the angular correlation coefficients for the 9–4–2–0 spin assignments and $\delta_1=-0.12$; $\delta_2=-0.07$.

Gamma-ray cascade kev	A_0	A_2	A_4	A_6
912–899	1.013	0.271	−0.041	0
912–375	1.013	0.227	−0.058	0.00004
375–899	1.005	0.143	0.000	0

[18] F. Coester, Argonne National Laboratory Report ANL-5316 (unpublished).

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

[20] M. Ferentz and N. Rosenzweig, Argonne National Laboratory Report ANL-5324 (unpublished).

[21] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023 (unpublished).

correspond to an anisotropy of 0.23 for the 375–899-kev cascade, and an experimental measurement of this quantity would be worthwhile. This measurement is rather difficult with deuteron-irradiated thallium sources because of prompt coincidences from Pb^{202m}. However, either fast neutron irradiation of Pb^{204} (separated isotope) or "milking" of a Bi^{204} solution[5] should provide better sources.

It should be noted that the difference in the measured anisotropies leads directly to mixing of the second gamma of the Pb^{204m} cascade if the $E2$ assignment for the last two gamma rays is accepted. An extension[22] of a theorem of Weneser and Hamilton[23] requires that these anisotropies be equal if neither of the $E2$ gamma rays is mixed. As the ground state of Pb^{204} has spin 0, there can be no mixing of the third gamma ray.

Note added in proof.—The spin schemes, 8-3-2-0 and 7-3-2-0, have been considered and found inconsistent with the existing data for Pb^{204m}.

Gyromagnetic Ratio of the 2.6×10^{-7} sec State of Pb^{204}

One method used to measure the gyromagnetic ratio of the 2.6×10^{-7} sec state of Pb^{204} involved measuring the ratio of coincidences at 135° to coincidences at 225° as a function of the magnetic field. This method has the advantage that both the sign and magnitude of g are obtained. Counter 1 was biased to count pulses above 510 kev while counter 2 was biased to count everything above 200 kev. The output from counter 1 was delayed 3.5×10^{-7} sec. The NaI(Tl) crystal of counter 1 was covered with a lead absorber 0.125 inch thick and 0.015 inch of tantalum foil. Counter 2 was covered with 0.015 inch of tantalum foil. The sources were deuteron-irradiated thallium foils dissolved in concentrated nitric acid.

Coincidences were obtained when counter 1 detected the 912-kev gamma ray in delayed coincidence with either the 899-kev gamma ray or the 375-kev gamma ray. A variable magnetic field was applied perpendicularly to the plane of the two counters. Under these circumstances the angular distribution is given by

$$W(\theta,\omega)=\int_{t_1}^{t_2}\sum_k A_kP_k[\cos(\theta+\omega t)]e^{-t/\tau}dt, \qquad (9)$$

where τ is the mean life of the intermediate state, t_1 is given by the delay minus the coincidence resolving time, t_2 is the delay plus the resolving time, and ω is the angular velocity of the Larmor precession and is given by

$$\omega=g(e/2Mc)H. \qquad (10)$$

The ratio, $W(135°,\omega)/W(225°,\omega)$, is unity for zero field, rises to a maximum, and then falls to values below

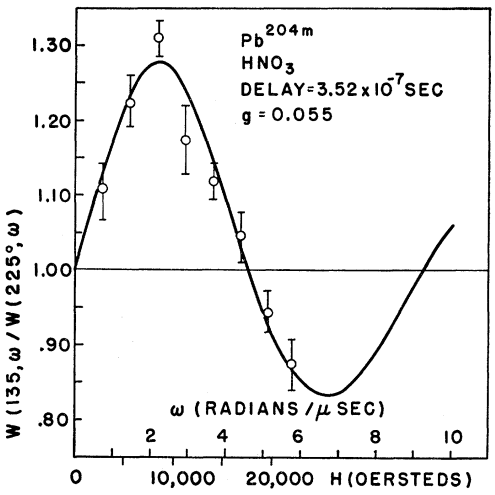

FIG. 3. $W(135°)/W(225°)$ calculated as a function of ω (smooth curve) and fitted to the experimental points obtained as a function of magnetic field.

one (Fig. 3). The return to unity corresponds to a rotation of the pattern of approximately 90° during the delay time. If the resolving time were negligible, the return to unity would correspond to a rotation of exactly 90°. The resolving time of 1.0×10^{-7} sec together with the exponential decay of the intermediate state leads to a smear of various angles of rotation so that the oscillations of $W(135°,\omega)/W(225°,\omega)$ are rapidly damped.

One can calculate $W(135°,\omega)/W(225°,\omega)$ as a function of ω by using Eq. (9). The solid curve in Fig. 3 is the result of such a calculation. The magnetic field scale and the ω scale were adjusted to obtain a fit of the experimental data to the calculated curve in the vicinity of the region where the curve returns to unity.

In calculating the curve, we assumed that the angular correlation pattern contains only a $P_2(\cos\theta)$ term. The asymmetry, i.e., $W(180°)/W(90°)$, measured in the presence of the magnet at zero field was used to calculate an A_2. This value of A_2 was used in Eq. (9) to calculate $W(135°,\omega)/W(225°,\omega)$. It is easily shown that the nonzero values of ω which give unity for $W(135°,\omega)/W(225°,\omega)$ are independent of A_2 and A_4 provided there is no time-dependent attenuation[24,25] of the angular correlation pattern. The neglect of A_4, therefore, does not contribute error to the gyromagnetic ratio determination. The coefficients of terms higher than $P_4(\cos\theta)$ are negligible for the Pb^{204m} decay scheme.

The measurements made in the presence of the magnet are not corrected for solid angle. The entire experiment was performed with the same solid angles, and proof that the solid angle correction is independent of magnetic field is given in the Appendix.

From the adjustment of the ω and H scales of Fig. 3 we obtain $g=+0.055\pm0.003$ nuclear units.

[22] S. Raboy and V. E. Krohn (to be published).
[23] J. Weneser and D. R. Hamilton, Phys. Rev. **92**, 321 (1953).
[24] A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).
[25] F. Coester, Phys. Rev. **93**, 1304 (1954).

An additional measurement was made with the irradiated thallium foils dissolved in concentrated sulfuric acid. The 912-kev gamma-ray pulse was delayed 2.4×10^{-7} sec. Inasmuch as the magnetic field was not adequate to rotate the pattern 90° during this shorter delay time, a different measurement was made. The ratio of coincidences at 180° and 90° was measured as a function of magnetic field. This ratio starts from a maximum at zero magnetic field and reaches unity when the pattern has been rotated 45° during the delay time. Figure 4 shows the experimental results. The smooth curve is $W(180°,\omega)/W(90°,\omega)$ as a function of ω, calculated from Eq. (9).

Values of ω which give unity for $W(180°,\omega)/W(90°,\omega)$ are independent of A_2 and A_4, so for purposes of calculation, we assumed only a $P_2(\cos\theta)$ term and adjusted the abscissa scales to get the gyromagnetic ratio. The sulfuric acid curve gives $g=0.050\pm0.006$ nuclear units, which is in good agreement with the value obtained with nitric acid.

The agreement of the values obtained for g with different delay conditions indicates that the interaction of the magnetic moment of the first excited state of Pb^{204m} with the magnetic field does not make a large contribution to the results. The effect of this interaction would be determined by the lifetime and gyromagnetic ratio of the first excited state and would be independent of the delay (for lifetimes as short as the longest which would be reasonable for this state). However, the calculated curves of Figs. 3 and 4 require that the observed precession be roughly proportional to the delay time if consistent values for g are to be obtained.

Time-Dependent Attenuation of the Angular Correlation Pattern

In the interpretation of the angular correlation data and the determination of the gyromagnetic ratio it was assumed that there was no time-dependent attenuation of the angular correlation pattern.[24,25] Such attenuation could cause errors in the spin and mixing assignments and in the g-factor determination. For this reason, plus interest in the attenuation problem for its own sake, an attempt was made to observe time-dependent attenuation in the nitric acid medium.

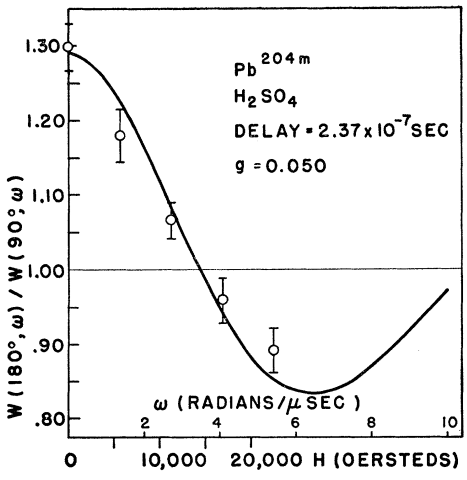

FIG. 4. $W(180°)/W(90°)$ calculated as a function of ω (smooth curve) and fitted to the experimental points obtained as a function of magnetic field.

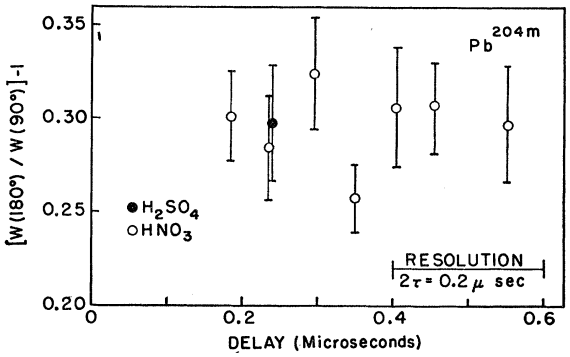

FIG. 5. Anisotropy as a function of delay. The results are for an unknown combination of the 912–375-kev and 912–899-kev cascades and are not corrected for solid angles or scattering.

The anisotropy, $[W(180°)/W(90°)]-1$, was measured as a function of the delay time which was varied from 1.85×10^{-7} to 5.5×10^{-7} sec. The results are shown in Fig. 5. The theory of time-dependent attenuation in liquid media[24,25] predicts that each term in the angular correlation formulae [e.g., the terms in the sum of Eq. (9)] will be attenuated by a factor of the form $G_k=\exp(-\lambda_k t)$. The results of Fig. 5 do not show evidence of such attenuation, but a ten percent attenuation of the anisotropy during the interval of time investigated would be consistent with the data. If a ten percent attenuation were present, it would cause less than one percent error in the gyromagnetic ratios measured under the conditions of the present experiment.

From Fig. 5 an anisotropy of 0.289 and, hence, $A_2=0.176$ was taken for the calculation of the curves in Figs. 3 and 4. These values are uncorrected for solid angle and scattering by the magnet and apply for an unknown combination of the 912–375-kev and 912–899-kev cascades. Hence, they are considered consistent with the anisotropy measurements previously reported.

The Illinois group[26] have reported that Pb^{204m} anisotropies measured in nitric and sulfuric acid did not give reproducible results and fell below measurements made with metallic melts. We are unable to reconcile their results with the absence of attenuation found in the present measurements.

The best value obtained for the gyromagnetic ratio of the 2.6×10^{-7} sec state of Pb^{204m} is

$$g=+0.054\pm0.005 \text{ nuclear units.}$$

[26] Frauenfelder, Lawson, Jentschke, and DePasquali, Phys. Rev. 92, 1241 (1953).

If the spin of this state is 4, the magnetic moment is

$$\mu=0.22\pm0.02 \text{ nuclear magnetons.}$$

The uncertainties quoted for the last values of g and μ include an attempt to allow for systematic errors.

ACKNOWLEDGMENTS

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APPENDIX

In order to demonstrate that the solid angle correction to the angular correlation expression is independent of magnetic field and can be applied to Eq. (9) of the text, we start with Eq. (127) of Biedenharn and Rose[19] from which Eq. (9) of the text is derived for the case of detectors with negligible solid angle

$$W_d=\frac{4\pi}{\tau}e^{-t/\tau}\sum_\nu\frac{A_\nu}{2\nu+1}\times\sum_{M=-\nu}^{\nu}Y_\nu^M(\theta_1,\varphi_1)Y_\nu^{M*}(\theta_2,\varphi_2)e^{-iM\omega t}. \quad \text{(A1)}$$

This is the relative probability per unit solid angle for emission of a gamma ray with direction θ_2, ϕ_2 at time t after emission of a gamma ray with direction θ_1, ϕ_1. τ is the nuclear lifetime, ω is the angular velocity of the Larmor precession, and the Y_ν^M are the spherical harmonics. The magnetic field direction is along the z axis.

We take the y axis along the axis of crystal 1 and define θ as the angle between the two crystals in the xy plane.

With detectors of finite solid angle, the number of coincidences observed is proportional to

$$W_d'=\frac{4\pi}{\tau}e^{-t/\tau}\sum_\nu\frac{A_\nu}{2\nu+1}\sum_{M=-\nu}^{\nu}\int_{\Omega_1}\int_{\Omega_2}\times d\Omega_1 d\Omega_2 Y_\nu^M(\theta_1,\varphi_1)Y_\nu^{M*}(\theta_2,\varphi_2)e^{-iM\omega t}, \quad \text{(A2)}$$

where $d\Omega_1$ and $d\Omega_2$ are elements of solid angle in crystal 1 and crystal 2, respectively, and the indicated integrations are over the faces of the crystal detectors.

Let β_1 be the polar angle of the first gamma ray with respect to the axis of crystal 1, and α_1 be the azimuthal angle of this gamma ray measured in the surface of crystal 1. Let β_2 and α_2 play similar roles for the second gamma ray with respect to crystal 2. We can expand the Y_ν^M as follows:

$$Y_\nu^M(\theta_1,\varphi_1)=\sum_{M'}D_{MM'}^{\nu*}(R_1)Y_\nu^{M'}(\beta_1,\alpha_1),$$

and

$$Y_\nu^{M*}(\theta_2,\varphi_2)=\sum_{M''}D_{MM''}^{\nu}(R_2)Y_\nu^{M''}(\beta_2,\alpha_2), \quad \text{(A3)}$$

where R_1 is that rotation which brings the initial z axis coincident with the axis of crystal 1, i.e., $R_1=(0,-\frac{1}{2}\pi,0)$ in terms of the Euler angles. R_2 is the rotation which brings the initial z axis into coincidence with the axis of crystal 2, i.e., R_2 is $(-\theta,-\frac{1}{2}\pi,0)$. The $D_{MM'}^{\nu}$ and $D_{MM''}^{\nu}$ are the elements of the νth irreducible representation of the rotation group.[27]

Substituting (A3) into (A2) we obtain

$$W_d'=\frac{4\pi}{\tau}e^{-t/\tau}\sum_\nu\frac{A_\nu}{2\nu+1}\sum_{M=-\nu}^{\nu}\times\int_0^{2\pi}\int_0^{2\pi}\int_0^{\beta_{01}}\int_0^{\beta_{02}}d\alpha_1 d\alpha_2 d\beta_1 d\beta_2\times\{\sin\beta_1\sin\beta_2\sum_{M'',M'}D_{MM'}^{\nu*}(R_1)D_{MM''}^{\nu}(R_2)\times Y_\nu^{M'}(\beta_1,\alpha_1)Y_\nu^{M''}(\beta_2,\alpha_2)e^{-iM\omega t}\}. \quad \text{(A4)}$$

The integration over α_1 reduces the sum over M' to one term, $M'=0$, and similarly the sum over M'' is reduced to one term by the integration over α_2. (A4) becomes

$$W_d'=\frac{4\pi}{\tau}e^{-t/\tau}\sum_\nu\frac{A_\nu'}{2\nu+1}\times\sum_{M=-\nu}^{\nu}D_{M0}^{\nu*}(R_1)D_{M0}^{\nu}(R_2)e^{-iM\omega t}, \quad \text{(A5)}$$

where

$$A_\nu'=A_\nu 4\pi^2\int_0^{\beta_{01}}P_\nu(\cos\beta_1)\sin\beta_1 d\beta_1\times\int_0^{\beta_{02}}P_\nu(\cos\beta_2)\sin\beta_2 d\beta_2. \quad \text{(A6)}$$

The A_k of Eq. (9) are related to the A_k' measured with finite solid angle by Eq. (A6) and an arbitrary normalization factor. It is evident that the solid angle correction factor, A_k/A_k', is independent of magnetic field, i.e., ω, and in fact is the usual correction for angular correlation experiments.[28] If the detector efficiencies are functions of β_1 and β_2, the efficiency functions appear in Eq. (A6) and the proof still holds.

[27] E. Wigner, *Gruppentheorie und ihre Anwendung auf die Quantenmechanik der Atomspektren* (Friedrich Vieweg & Sohn, Breunschweig, Germany, 1931), Chap. 15.

[28] S. Frankel, Phys. Rev. **83**, 673 (1951).