Magnetic Moments of the First Excited 2+ States in Sm^{152}, Gd^{154}, and Gd^{156}†

R. W. BAUER AND M. DEUTSCH
Department of Physics and Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, Massachusetts

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The gyromagnetic ratios g of the 122-keV state of Sm^{152}, of the 123-keV state of Gd^{154}, and of the 89-keV state of Gd^{156} have been measured to be 0.35 ± 0.03, 0.367 ± 0.03, and 0.32 ± 0.03, respectively, by the precession of the angular correlation of gamma-ray cascades proceeding through these states in an external magnetic field. The perturbations of the angular correlations for sources both in aqueous solution and in molten anhydrous chlorides have been studied. The attenuation in a transverse magnetic field was used to determine the hyperfine structure constant of the 89-keV state of Gd^{156} in the $^8S_{7/2}$ ground state of Gd^{3+} in aqueous solution as $a=17\pm3$ Mc/sec. The perturbations in molten Gd sources were found to be much weaker than in solution and to be unaffected by magnetic fields.

1. INTRODUCTION

THE study of the gyromagnetic ratio g of the so-called rotational nuclear states, especially of the first 2+ excited state of highly deformed even-even nuclei, has been the subject of considerable theoretical and experimental interest during the last years. These magnetic moment measurements provide additional information to the investigation of the collective behavior of nuclei.

Recent calculations by Nilsson and Prior[1] and by Yang Hu and Villars[2] make specific predictions for the deformed nuclei between Sm and Os, and above Ra. The theoretical gyromagnetic ratios from both calculations are below the value of Z/A predicted by the simplest quasi-hydrodynamic model.[3]

Experimental determination of g in the deformed region have been accumulated rapidly during the past few years (see Sec. 6). The experiments seem to agree with the more refined theory, but most of them are subject to considerable corrections involving the influence of extranuclear fields on nuclear orientation.[4–8]

We have investigated the properties of the first excited 2+ states of Sm^{152}, Gd^{154}, and Gd^{156} using the method of gamma-gamma angular correlation. These states, whose lifetimes are of the order of 2 nsec, are fed from the beta decay of Eu^{152}, Eu^{154}, and Eu^{156}, respectively. The relevant portions of the decay schemes for the three isotopes are shown in Fig. 1. By means of an investigation of the attenuations of the angular correlations from sources both in aqueous solutions and in anhydrous chloride melts at high temperatures, with and without externally applied magnetic fields, we believe that we can evaluate the necessary corrections and have obtained values of g for the three states and of the hfs splitting in the case of Gd^{156} in the $^8S_{7/2}$ ground state of the Gd^{3+} ion in aqueous solution.

Our method follows closely the ones used by Steining and Deutsch[8] for Gd^{154}. The reader is referred to this paper for a more detailed discussion.

We write the angular correlations in the form

$$W(\theta,t)=1+G_2(t)A_2P_2(\cos\theta)+G_4(t)A_4P_4(\cos\theta), \quad (1)$$

$$W(\theta)=1+G_2A_2P_2(\cos\theta)+G_4A_4P_4(\cos\theta). \quad (2)$$

No terms higher than the fourth order of the Legendre polynomials $P_k(\cos\theta)$ were necessary to be included in the expansions. The first of the equations gives the correlation as a function of delay time t between formation and decay of the 2+ state. In the usual approximation $G_k(t)=e^{-t/\tau_k}$. This defines the attenuation time constant τ_k. The second equation gives the correlation including all gamma rays, regardless of delay. For exponential attenuation $G_k=(1+\tau_N/\tau_k)^{-1}$ where τ_N is the mean life of the 2+ state.

In the presence of a transverse magnetic field H (applied in a direction perpendicular to the plane con-

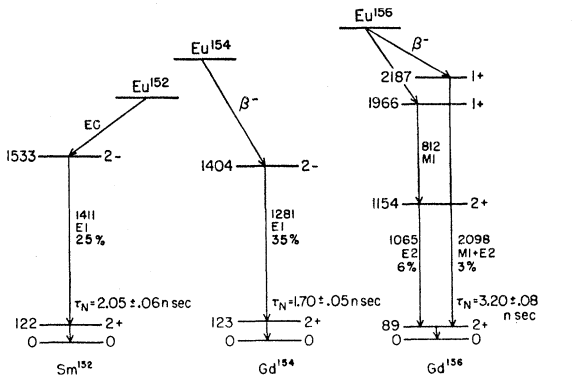

FIG. 1. Relevant features of the radioactive decay schemes of the Eu isotopes used in the experiments. The energies are given in keV.

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[1] S. G. Nilsson and O. Prior, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **32**, No. 16 (1961).

[2] C. Y. Yang Hu, Ph.D. thesis, Massachusetts Institute of Technology, 1962 (unpublished); C. Y. Yang Hu and F. Villars (to be published).

[3] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat-fys. Medd. **27**, No. 16 (1953).

[4] A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).

[5] R. M. Steffen, Suppl. Phil. Mag. **4**, 293 (1955).

[6] K. Alder, Helv. Phys. Acta **25**, 235 (1952).

[7] E. Heer and T. B. Novey, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1959), Vol. 9, p. 199.

[8] R. Stiening and M. Deutsch, Phys. Rev. **121**, 1484 (1961).

taining the two radiations) the angular correlation pattern, in general, rotates through an angle determined by the Larmor precession frequency. Equation (1) gives for this case

$$W(\theta, t, \pm H) = 1 + G_2(t) A_2 P_2[\cos(\theta \pm \omega_L t)] + G_4(t) A_4 P_4[\cos(\theta \pm \omega_L t)], \quad (3)$$

where $\omega_L = g\mu_N H_{\text{eff}}/\hbar$ is the precession frequency in terms of the nuclear gyromagnetic ratio g, the nuclear magnetic moment μ_N, and the effective magnetic field H_{eff} at the nucleus.

In experiments where the interaction has cylindrical symmetry, as in the case of the precession experiment and of the transverse magnetic decoupling (Secs. 4, 5) the appropriate form of the angular correlation $W(\theta)$ is

$$W(\theta) = 1 + \bar{G}_2 B_2 \cos 2\theta + \bar{G}_4 B_4 \cos 4\theta, \quad (4)$$

rather than in terms of Legendre polynomials. The relations between the two expressions are given in references 5 and 8. For exponential attenuation of $G_k(t)$, Eq. (3) gives an integral correlation of the form

$$W(\theta, \pm H) = 1 + [\bar{G}_2 B_2 - (4/7)\bar{G}_4 B_4] \frac{\cos 2(\theta \pm \Delta\theta_{22})}{[1 + (2G_2\omega_L\tau_N)^2]^{1/2}} + \bar{G}_4 B_4 \frac{\cos 4(\theta \pm \Delta\theta_{44})}{[1 + (4G_4\omega_L\tau_N)^2]^{1/2}} + \left(\frac{4}{7}\right)\bar{G}_4 B_4 \frac{\cos 2(\theta \pm \Delta\theta_{24})}{[1 + (2G_4\omega_L\tau_N)^2]^{1/2}}, \quad (5)$$

where $\Delta\theta_{22} = \frac{1}{2}\tan^{-1}(2G_2\omega_L\tau_N)$, $\Delta\theta_{24} = \frac{1}{2}\tan^{-1}(2G_4\omega_L\tau_N)$, $\Delta\theta_{44} = \frac{1}{4}\tan^{-1}(4G_4\omega_L\tau_N)$. For the case of no attenuation ($G_2 = G_4 = 1$), Eq. (5) reduces to the relations given in references 5 and 7.

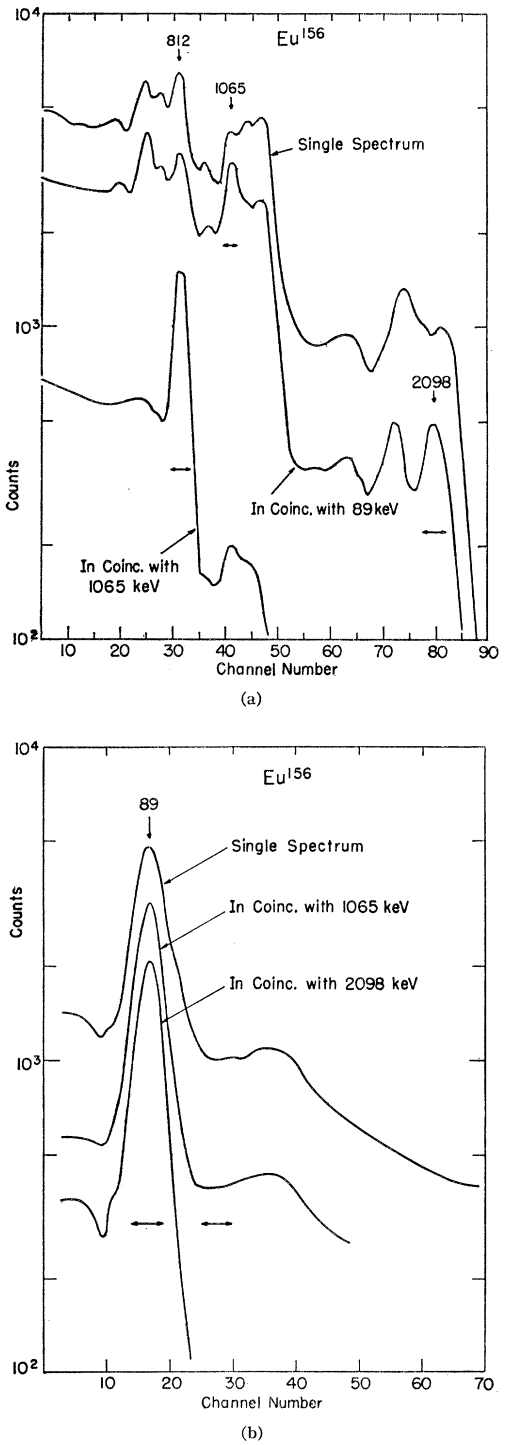

FIG. 2. Gamma-ray pulse-height distributions. The horizontal arrows indicate the pulse-height intervals used in the coincidence experiments. (a): Spectra observed with the 3-×3-in. NaI(Tl) crystal. (b): Spectra observed with the nonactivated NaI crystal.

2. EXPERIMENTAL ARRANGEMENT AND PROCEDURE

The measurements of the gyromagnetic ratios consisted of two steps: (1) The sources were investigated for their attenuation of the angular correlation by means of a measurement of $G_k(t)$ as a function of time. This step is essential, we feel, to reach an understanding of the attenuation mechanisms involved in order to be able to evaluate the necessary corrections in the g measurements. (2) The same sources were used in a transverse magnetic field, where both the magnitude of the angular correlation and its precession were measured as a function of magnetic intensity H. The precession measurements consisted in a determination of the change in coincidence counting rate R at one or several fixed angles for a reversal of the applied field. We define the quantity R given by

$$R = 2[W(\theta, +H) - W(\theta, -H)]/[W(\theta, +H) + W(\theta, -H)]. \quad (6)$$

This precession method has been described repeatedly in the literature.[5,7] For an angular correlation, where A_4 and higher terms can be neglected and where R is

measured at 135°, we get, by combining Eqs. (5) and (6), after some simple manipulation,

$$R(3\pi/4)=4\bar{G}_2B_2\omega_LG_2\tau_N/[1+(2\omega_LG_2\tau_N)^2]. \quad (7)$$

It is important that the proper value of the angular correlation is used in evaluating g. This point has been discussed in detail by Stiening and Deutsch.[8] In our measurements, the angular correlation in the magnet, with zero field, has been determined at frequent intervals between the R measurements. The experimentally observed correlation coefficients in the magnet were found to be slightly lower (about 2 to 5%) compared to measurements in a scatter-free geometry. In each case under study, we experimentally determined the effective angular correlation in the magnet and have used the thus obtained empirical coefficients of the correlation expansion to evaluate g. It is easily seen that a distortion of the angular correlation is produced by Compton scattering of quanta with initially higher energy than the radiations investigated. This effect has to be corrected for in the study of Eu^{156}. This, however, is not the case in Eu^{152} and Eu^{154} where the cascades under study are only single gamma cascades and the high-energy gamma rays of the respective cascades are the most energetic gamma rays in coincidence with the respective 122- and 123-keV gamma rays. Lower energy gamma rays are easily separated from this by pulse-height selection. We believe that the elastic scattering of the 122- and 123-keV gamma rays is therefore the main cause in the reduction of the angular correlation in Eu^{152} and Eu^{154}.

The electronic circuits consisted of a generally conventional fast-slow coincidence arrangement with the usual single-channel and multichannel pulse-height selectors. Details of the time-to-pulse height converter and of the stabilizing circuits have been given by Stiening and Deutsch.[8] As a protection against slight timing and pulse-height shifts in the apparatus, the data for each experiment were taken automatically according to a suitable program in which counter position and/or magnetic field directions were changed at intervals of 5 to 10 min.

For the measurement of the time dependence of the angular correlations (step 1 described above) a pure (nonactivated) NaI scintillator cooled to liquid N_2 temperature was used to detect the low-energy gamma rays (120 and 89 keV). For integral angular correlation measurements $W(\theta)$ where the fast response of the pure NaI scintillator was not needed, a crystal of NaI(Tl) of similar dimensions was used. The high-energy gamma rays were detected by a 3×3-in. NaI(Tl) crystal. The arrangement has been described by Stiening and Deutsch.[8] The pulse height and time resolution of the equipment is illustrated in Figs. 2 and 3.

The sources Eu^{152} and Eu^{154} with a specific activity of about 0.5 mC/mg were prepared from isotopically enriched Eu^{151} and Eu^{153} oxides, respectively, by slow-

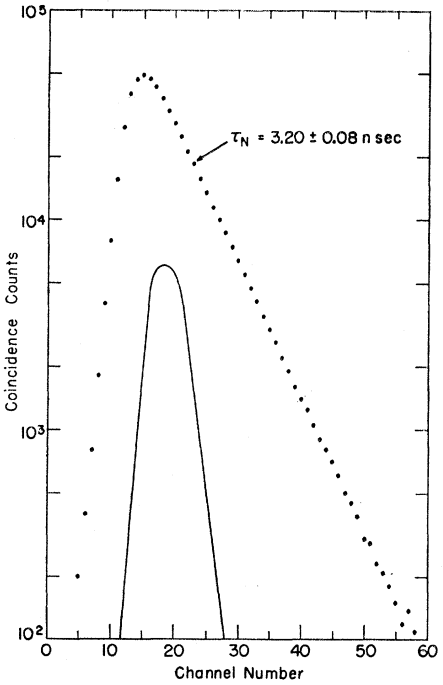

FIG. 3. Decay curve of the 89-keV state in Gd^{156} observed with the time-to-pulse-height converter using the nonactivated NaI crystal for the detection of the 89-keV gamma radiation. The solid line illustrates the time resolution for prompt coincidences between gamma rays of the same energies.

neutron bombardment. The sources were used without chemical purification. Eu^{156} was prepared by the Oak Ridge National Laboratory by exposure of enriched Sm^{154} oxide to a neutron flux of 1.7×10^{14} n/cm^2 sec for three days by the process $Sm^{154}+n\rightarrow Sm^{155}\rightarrow Eu^{155}+\beta$, $Eu^{155}+n\rightarrow Eu^{156}$. Europium was separated from samarium and other rare earths either by ion exchange or by reduction of the sulphate on zinc amalgam. Sources with a specific activity similar to the value quoted above were obtained. Utilizing the high neutron flux the Eu^{156} to Eu^{155} ratio was high enough to allow measurements with the Eu^{156} sources up to three months after bombardment. In general, a source strength of about 50 μC was used for the coincidence measurements.

Aqueous solution sources were prepared as chlorides in excess hydrochloric acid and sealed in very thin Pyrex tubes of about 1-mm diameter. The concentrations of the $GdCl_3$ solution (containing Eu^{154} or Eu^{156}) and of the $SmCl_3$ solution (containing Eu^{152}) were approximately 50 g/liter. Anhydrous chloride melt sources were prepared in similar quartz tubes by drying a hydrochloric acid solution to which about 2 mg of $GdCl_3$ (for the Eu^{154} and Eu^{156} sources) or 2 mg of $SmCl_3$ (for the Eu^{152} sources) had been added, in vacuum. The source capsules were heated in vacuum by means of a 0.001-in.-thick niobium ribbon which completely surrounded the source.

A conventional C-yoke electromagnet producing fields up to 26 kG at a power dissipation of 500 W was used

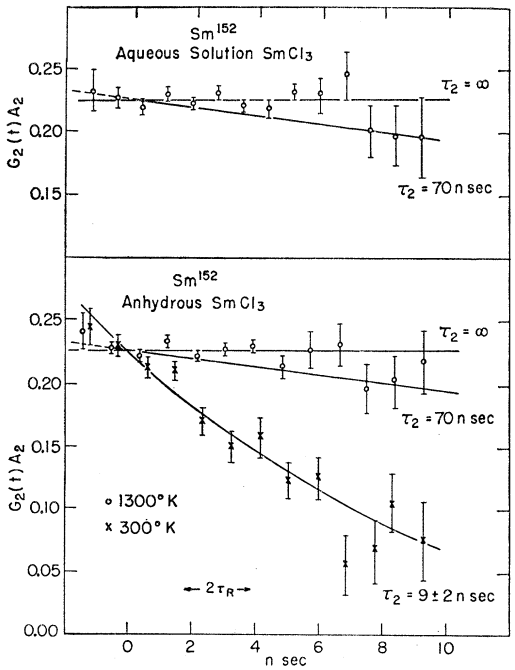

FIG. 4. Time dependence of the angular correlation of the 1411-keV gamma–122-keV gamma cascade in Sm^{152} for aqueous solution and anhydrous chloride sources, the latter measured above and below the melting point. The time resolution is given by $2\tau_R$. $\tau_2=70$ nsec gives the lower limit of the attenuation time constant for the solution and melt sources.

for the precession and decoupling experiments. The stability, uniformity, and calibration of the magnetic field was better than 2%.

The angular correlation data have been corrected for the finite angular resolution of the counters according to the procedure of Rose[9] and, where necessary, for the Compton background underneath a photopeak due to the presence of higher energy gamma rays. The horizontal arrows in Fig. 2 indicate the pulse-height intervals used in the coincidence experiments.

3. EXPERIMENTAL RESULTS ON Sm^{152}

The gamma-gamma cascade used in our measurements on the 122 keV state of Sm^{152} was the 1411–122 keV cascade, through which about 25% of the total disintegrations of Eu^{152} take place. The portion of the decay scheme relevant to this experiment is shown in Fig. 1.

A preliminary report on the Sm^{152} experiments has been presented previously.[10] The following describes the results in more detail.

We have measured the time dependence of the angular correlation $W(\theta,t)$ of the cascade for both aqueous solution sources and anhydrous chloride melts. The results for the coefficients of $P_2(\cos\theta)$ are presented in Fig. 4. Within experimental accuracy, the aqueous solution source shows no attenuation ($\tau_2>70$ nsec) which is in agreement with the results previously reported by Stiening and Deutsch[8] and Goldring and Scharenberg.[11] Also, for the anhydrous chloride source, no attenuation was found (within the accuracy of the experiment) above the melting point (above 1000°K) where again as limit we can set $\tau_2>70$ nsec. Below the melting point the correlation is obviously perturbed. From our measmeasurements, we obtain an angular correlation coefficient $A_2=0.225\pm0.007$ in agreement with previously reported results.[12–15] The correlation shows only a very weak A_4 term ($\leqslant 0.02$). This correlation uniquely determines the spin assignments and multipolarities of the gamma rays as given in Fig. 1. For the g measurement, we have used the unperturbed sources: aqueous solution at room temperature, and the anhydrous chloride above its melting point. This procedure has eliminated very considerable corrections in the evaluation of the g measurement which have introduced large errors in previously reported experiments[15,16] where strongly attenuated sources with G_2 of the order of 0.60 had been used.

The ion Sm^{3+} is known to be strongly paramagnetic. As a consequence of this, the externally applied magnetic field H_{ex} is appreciably modified by the atomic structure. Expressing the effective magnetic field H_{eff} at the nucleus in terms of $H_{eff}=\beta H_{ex}$ we can define a g_{eff}, which has not been corrected for paramagnetic effects, in terms of

$$g_{eff}=\beta g=\omega_L\hbar/\mu_N H_{ex}, \tag{8}$$

where g is the value corrected for the paramagnetic effects. The other quantities are defined below Eq. (3). In the case of Sm^{3+} the temperature dependence of the coefficient β cannot be evaluated using Curie's law as has been attempted in other rare earths (see reference 17). An explicit calculation by Kanamori and Sugimoto[17] predicts a value of 1.15 for β at room temperature, decreasing to about 1.04 between 1000 and 1500°K.

To investigate the temperature dependence of the paramagnetic correction, we measured g_{eff} at four temperatures: 300°K (unperturbed aqueous solution source), 1100, 1300, and 1500°K (unperturbed melt source for the three latter temperatures). Measurements below 1100°K were not carried out since anhydrous $SmCl_3$ sources below the melting point showed appreciable attenuation. The upper temperature limit of 1500°K could not be exceeded because of the melting point of the quartz capsule enclosing the source. The

[9] M. E. Rose, Phys. Rev. **91**, 61 (1953).
[10] R. W. Bauer and M. Deutsch, Bull. Am. Phys. Soc. **6**, 224 (1961).
[11] G. Goldring and R. P. Scharenberg, Phys. Rev. **110**, 701 (1958).
[12] B. Hartmann and T. Weidling, Arkiv Fysik **10**, 355 (1956).
[13] S. Ofer, Nuclear Phys. **4**, 477 (1957).
[14] G. Manning and J. Rogers, Nuclear Phys. **19**, 675 (1960).
[15] P. Debrunner and W. Kündig, Helv. Phys. Acta **33**, 392 (1960).
[16] K. Sugimoto, J. Phys. Soc. Japan **13**, 240 (1958).
[17] J. Kanamori and K. Sugimoto, J. Phys. Soc. Japan **13**, 754 (1958).

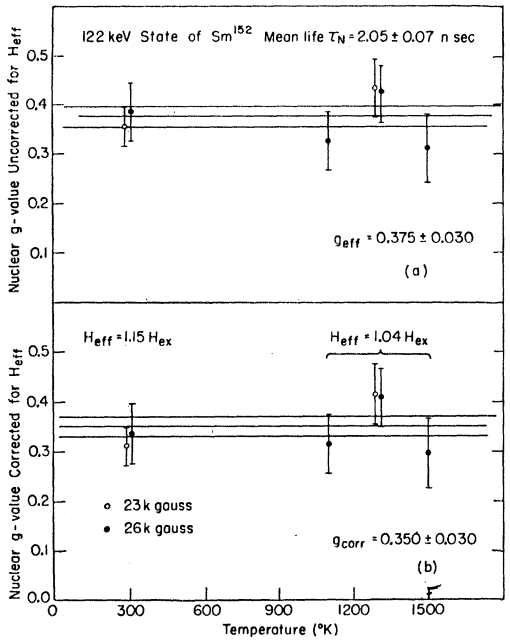

FIG. 5. Results of the angular correlation precession measurement of the gyromagnetic ratio g of the 122-keV state of Sm^{152} as a function of temperature using unperturbed liquid sources. (a) Results for g_{eff} defined in Eq. (8). (b) Results for g after paramagnetic correction. The horizontal lines represent the weighted means of the measurements and the estimated uncertainties.

measurements were carried out at an angle of 135° using fields of 23 and 26 kG.

The observed fractional coincidence counting rate difference R under field reversal [see Eq. (7)] when combined with Eq. (8) gave the effective values g_{eff} shown in Fig. 5(a). Each individual point represents a measurement of R (which is of the order of 0.05) determined to a statistical accuracy of about 5%.

To evaluate the results of our measurements, it was necessary to know with precision the angular correlation under the conditions of the experiments, the attenuation $G_2(t)$ and the nuclear lifetime τ_N.

The mean life of the 2+ state has been reported by Sunyar[18] and by Birk[19] to be 2.02±0.14 and 2.09±0.08 nsec, respectively. We have carefully remeasured this quantity by the delayed coincidence technique. The result is $\tau_N=2.05\pm0.06$ nsec where the error is primarily nonstatistical and represents uncertainties of the time calibration.

For both the aqueous solution and melt sources, the experimentally determined attenuation time τ_2 is larger than 70 nsec, assuming exponential attenuation, thus giving $\tau_2/\tau_N>34$. The integral attenuation is therefore $G=1.00_{-0.03}{}^{+0.00}$, i.e., less than 3% per mean life for both sources.

[18] A. Sunyar, Phys. Rev. **98**, 653 (1955).
[19] M. Birk, G. Goldring, and Y. Wolfson, Phys. Rev. **116**, 730 (1959).

The observed angular correlation in the magnet with zero field, measured at frequent intervals between the R measurements, when corrected for random coincidences, but not for the finite solid angle subtended, showed a coefficient of $\bar{G}_2B_2=0.135\pm0.003$, with $\bar{G}_4B_4$ less than 0.01. Within statistical accuracy (about 2%) the angular correlation coefficient $\bar{G}_2B_2$ was not detected to be reduced by the applied magnetic field.

Without a paramagnetic correction, the weighted mean of our measurements is $g_{eff}=+0.375\pm0.03$ which is in agreement with the results obtained by Goldring and Scharenberg[8] and Debrunner and Kündig[15] using unattenuated aqueous solution sources at room temperature.

Our results are consistent with a temperature independent coefficient β. They are almost in equally good agreement with the temperature dependence predicted by Kanamori and Sugimoto.[17] It would, therefore, seem that their theoretical values of β are close to the true values. Measurements of g_{eff} as a function of temperature in crystalline sources by Debrunner and Kündig[15] showed radically different behavior. These results probably are characteristic of the coupling to the crystal lattice rather than the Sm^{3+} ion.

With the paramagnetic corrections as calculated by Kanamori and Sugimoto,[17] the weighted mean of our measurements is $g=+0.350\pm0.03$. The corrected results are presented in Fig. 5(b).

4. EXPERIMENTAL RESULTS ON Gd^{154}

The gamma-gamma cascade used in our measurements on the 123 keV state of Gd^{154} was the 1281–123 keV cascade, through which about 35% of the total disintegrations of Eu^{154} take place. The portion of the decay scheme relevant to this experiment is shown in Fig. 1.

This 2+ state in Gd^{154} has been studied in detail by Stiening and Deutsch.[8] Our measurement of the time dependence of the angular correlation $W(\theta,t)$ of the cascade for aqueous solution sources is in agreement with Stiening's result. Our experiment [see Fig. 6(b)] shows a perturbation of $\tau_2=8.0\pm1.0$ μsec. Using the result of the nuclear mean lifetime measurement by Stiening of $\tau_N=1.70\pm0.05$, we get $\tau_2/\tau_N=4.7\pm0.6$; thus the integral attenuation is $G_2=0.83\pm0.02$. Stiening and Deutsch[8] report that they have succeeded in removing most of this perturbation by applying a magnetic field of about 3 kG in the direction of one of the radiations (longitudinal decoupling), obtaining $G_2=0.96\pm0.07$. These authors also observed a transverse decoupling effect in which the attenuation G_2 is reduced from about 0.82 to 0.50 ± 0.06 by applying a magnetic field of about 25 kG perpendicular to the plane of the counters. Assuming that at these magnetic fields the magnetic hfs interaction is fully decoupled, they could calculate the hfs constant a from the saturation value of G_2. These experiments show that the attenuation is

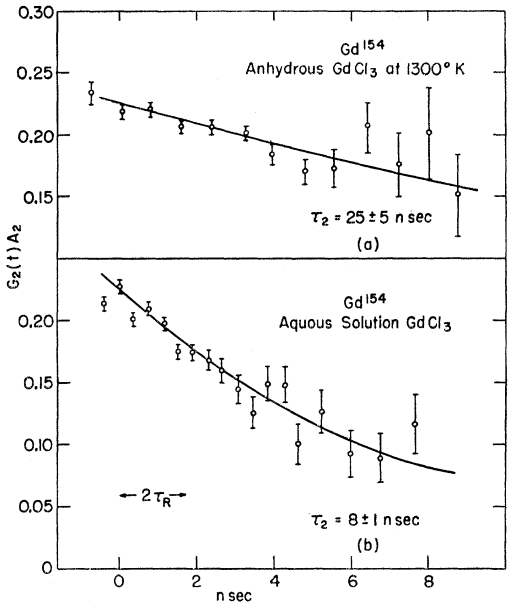

FIG. 6. Time dependence of the angular correlation of the 1281-keV gamma–123-keV gamma cascade in Gd^{154} for anhydrous chloride and aqueous solution sources.

predominantly due to magnetic interaction, but a small amount of electric quadrupole perturbation cannot be excluded. The aqueous solution sources, however, are unsuitable for direct g-value measurements because of the magnetic field-dependent nuclear spin relaxation mechanism.

Similar to the method employed by Stiening and Deutsch,[8] we have used sources of anhydrous $GdCl_3$ at about 1300°K for the precession type measurement of g. It is believed that in the ionic melt sources the strong interactions, which exist in ionic crystals, fluctuate sufficiently rapidly to produce a short electronic relaxation time under all conditions. Our measurement of the time dependence of the angular correlation $W(\theta,t)$ shows a weak attenuation with $\tau_2=25\pm5$ nsec [see Fig. 6(a)], giving $\tau_2/\tau_N=14.5\pm3$ and $G_2=0.93\pm0.01$. Within statistical accuracy (about 2%) the value of G_2 for the ionic melt did not change in the applied magnetic field of 26 kG. The angular correlation coefficient for this cascade was found to be $A_2=0.226\pm0.007$ in agreement with previous results,[8] giving a value for $G_2A_2=0.210\pm0.007$. Within the accuracy of the experiment no G_4A_4 term could be detected ($G_4A_4<0.01$). This correlation uniquely determines the spin assignments and multipolarities of the gamma rays as given in Fig. 1.

The measurements of g were carried out at an angle of 135° using a magnetic field of 26 kG. The observed fractional coincidence counting rate difference R under field reversal was 0.0379 ± 0.002, corrected for random coincidences. The observed angular correlation in the magnet with zero field, measured at frequent intervals between the R measurement, when corrected for random coincidences only, had a value of $\bar{G}_2B_2=0.128\pm0.003$ with $\bar{G}_4B_4$ less than 0.01. Combining Eqs. (6) and (7) we get for the precession frequency

$$\omega_L=g\mu_N H_{\rm eff}/\hbar=(4.80\pm0.40)\times10^7 \text{ rad/sec},$$

using $\tau_N=1.70\pm0.05$ nsec and $G_2=0.93\pm0.01$. With an effective field $H_{\rm eff}=1.042\ H_{\rm ex}$ at 1300°K for the paramagnetic and diamagnetic corrections (see reference 8) we obtain a g value of $+0.37\pm0.04$. This result is in excellent agreement with the g value of $+0.36\pm0.06$ measured by Stiening and Deutsch. The weighted mean of the two measurements is $g=+0.367\pm0.03$.

5. EXPERIMENTAL RESULTS ON Gd^{156}

The decay of Eu^{156} is known to be relatively complex compared to the decay of the other two Eu isotopes studied. The decay of Eu^{156} to Gd^{156} has been investigated extensively by Cline and Heath,[20] and by Ewan, Graham, and Geiger[21] who observed at least 24 gamma transitions of which at least 8 transitions populate the 89-keV state directly, and an equal number indirectly via an intermediate gamma transition.

In our measurements on the 89-keV state we used primarily the 1065–89-keV and the 2098–89-keV cascades. The portion of the decay scheme relevant to our experiments is shown in Fig. 1.

The 2098–89-keV cascade, though weak (only about 3% of total disintegrations), shows a large A_2 term. It fulfills the requirements of being a single gamma cascade, i.e., the only gamma ray in coincidence with the 2098-keV gamma ray is the 89-keV ray and, on the other hand, the most energetic gamma ray in coincidence with the 89-keV ray is the 2098-keV ray. These ideal conditions existed also in the cascades used in the Sm^{152} and Gd^{154} experiments. Because of the low intensity of this cascade and the high internal conversion of the 89-keV gamma ray[21] ($\alpha_K=1.4$) we employed additional cascades to improve the statistical accuracy. The other cascade used extensively (1065–89 keV), through which about 6% of the total disintegrations take place, shows a large A_4 term. This cascade, however, is part of a multiple gamma cascade and therefore less well suited for our measurements. The 1154-keV state is known not to be directly populated by a beta transition, but predominantly by a 812-keV gamma transition.[21] We have confirmed these results experimentally [see Fig. 2(a), spectrum in coincidence with 1065 keV]. We, furthermore, found in the low-energy pulse-height spectrum [see Fig. 2(b), spectrum in coincidence with 1065 keV] a Compton continuum below the 89-keV photopeak which was due to the 812-keV gamma transition. This Compton background in the coincidence spectrum appeared as "prompt coincidences" in the time spectrum gated by the 1065- and 89-keV photopeaks. This allows an estimate of the upper limit for the lifetime of the 1154-keV state as 0.5 nsec. To correct for the unwanted

[20] J. E. Cline and R. L. Heath, Nuclear Phys. **22**, 598 (1961).
[21] G. T. Ewan, R. L. Graham, and J. S. Geiger, Nuclear Phys. **29**, 153 (1962).

coincidences we simultaneously measured the 1065–89 keV gamma correlations together with the correlation of the 1065-keV ray with gamma rays from the Compton continuum selected by a window set slightly above the 89-keV photopeak [see Fig. 2(b)]. This procedure enabled us to use the stronger 1065–89-keV cascade and to compare the results of the precession and decoupling experiment with those independently obtained with the weaker, but more ideal 2098–89-keV cascade measured simultaneously with the former cascade. No difference in the results was detected within statistical accuracy.

To evaluate the results of our measurements, it was necessary to know the nuclear lifetime τ_N of the 89-keV state with precision. The mean-life of this state has been reported by Nathan[22] and Bell *et al.*[23] to be 2.74±0.14 and 3.16±0.10 nsec, respectively. We have carefully remeasured this quantity by the delayed coincidence technique. Figure 3 illustrates a decay curve of the 2098–89-keV gamma cascade observed with the time-to-height converter. The ordinate represents $\log[0.6W(\pi/2)+0.8W(3\pi/4)+0.1W(\pi)]$ which should be independent of the angular correlation since no terms higher than A_4 appear in $W(\theta)$. Our result from several measurements and from the two different cascades is $\tau_N=3.20\pm0.08$ nsec where the error is primarily non-statistical and represents uncertainties of the time calibration.

We have measured the time dependence of the angular correlation $W(\theta,t)$ for the 2098–89-keV and 1065–89-keV cascades for aqueous solution sources. The results for the coefficients of $P_2(\cos\theta)$ for the former cascade and of $P_4(\cos\theta)$ for the latter cascade are presented in Fig. 7. Our experiments show a perturbation of $\tau_2=17.5\pm2.0$ nsec and $\tau_4=11.5\pm1.5$ nsec, which is equal to $\tau_2/\tau_N=5.5\pm0.6$ and $\tau_4/\tau_N=3.6\pm0.5$. With exponential attenuation, we get the integral coefficients $G_2=0.84\pm0.02$ and $G_4=0.78\pm0.02$. It is to be noted that for the aqueous solution sources the experimental ratio τ_2/τ_4 is of the order of 1.5. In the limit of rapid relaxation the ratio τ_2/τ_4 should be 3.34 for magnetic dipole interaction, and 0.59 for electric quadrupole interaction. Since the hyperfine coupling frequency, the electron relaxation time and the nuclear mean life are all of comparable order of magnitude the usual approximations[4] are not valid. Therefore, we cannot reach definite conclusions concerning the nature of the perturbing interaction until calculations for this case have been carried out. For the same reason, no conclusions concerning the ratio of the nuclear magnetic moments $\mu(156)/\mu(154)$ can be reached from the ratio of the attenuations. Presently calculations are being carried out by Steffen *et al.*[24] which consider the influence of a combined magnetic and electric interaction. Results of this investigation will be available in the near future.[25]

[22] O. Nathan, Nuclear Phys. **5**, 401 (1958).
[23] R. E. Bell and M. H. Jorgensen, Nuclear Phys. **12**, 413 (1959).
[24] K. Alder, E. Matthias, W. Schneider, and R. M. Steffen, Bull. Am. Phys. Soc. **7**, 19 (1962).
[25] R. M. Steffen (private communication).

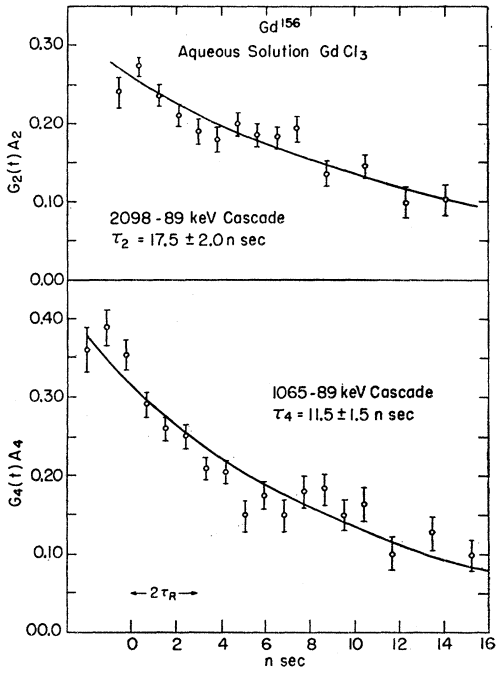

FIG. 7. Time dependence of the angular correlation in Gd^{156} for aqueous solution sources.

After correcting for counter solid angles and, where necessary, for the prompt Compton background underneath the photopeak due to higher energy gamma rays, we obtained the following gamma-gamma angular correlations with aqueous solution sources:

(1) For the 2098–89-keV cascade,

$$W(\theta)=1+(0.23\pm0.02)P_2(\cos\theta)+(-0.12\pm0.02)P_4(\cos\theta),$$

giving, after correction for the attenuation, $A_2=0.27\pm0.02$ and $A_4=-0.15\pm0.02$, which, together with the internal conversion measurements by Ewan *et al.*,[21] determine the spin and parity of the 2187-keV state to be 1^+ and the 2098-keV gamma transition to be a 80% $M1$–20% $E2$ mixture (amplitude mixing ratio $\delta=+0.5\pm0.1$).

(2) For the 1065–89 keV cascade,

$$W(\theta)=1+(0.05\pm0.02)P_2(\cos\theta)+(0.24\pm0.02)P_4(\cos\theta),$$

giving $A_2=0.06\pm0.02$ and $A_4=0.31\pm0.02$, which, together with the internal conversion data, determines the spin and parity of the 1154 keV state to be 2^+. The analysis of this γ-vibrational level has been discussed in detail by Gregers Hansen *et al.*[26] The 1065-keV gamma transition is essentially all $E2$, as expected from K selection rules; only an upper limit for the $M1$ admixture can be set (about 5%), which corresponds to a mixing ratio of $\delta>+4$, since it is known[21] that the 1154-keV state is also fed by a weak 1049-keV gamma transition

[26] P. Gregers Hansen, O. B. Nielsen, and R. K. Sheline, Nuclear Phys. **12**, 389 (1959).

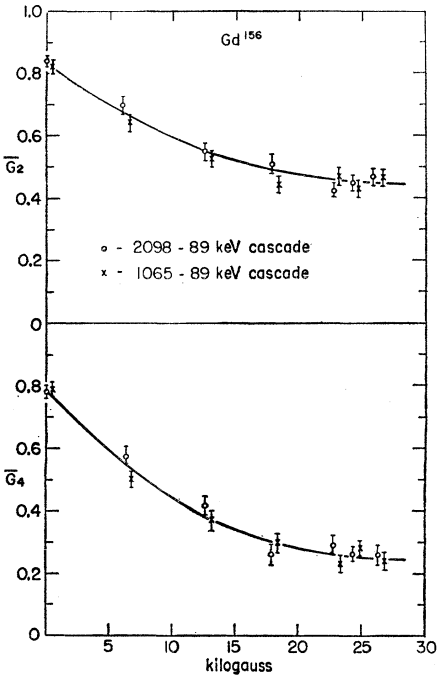

FIG. 8. Effect of the magnetic transverse decoupling field on the angular correlation.

(not shown in Fig. 1) which, of course, can distort the 1065–89-keV angular correlation slightly. This 1049-keV gamma ray, however, will not influence our precession and decoupling experiments.

(3) For the 812–1065-keV cascade (see Fig. 1),

$$W(\theta)=1+(-0.03\pm0.02)P_2(\cos\theta)+(0.00\pm0.02)P_4(\cos\theta).$$

(4) For the 812–1154-keV cascade (where the 1154-keV transition is the crossover from the second excited 2+ state to the ground state, not shown in Fig. 1),

$$W(\theta)=1+(-0.18\pm0.02)P_2(\cos\theta)+(-0.01+0.02)P_4(\cos\theta).$$

Since the lifetime of the intermediate state of the last two cascades has been estimated to be less than 0.5 nsec (see above), the attenuation of the two correlations is expected to be less than 5% per mean life, which is within the uncertainties quoted for the correlation coefficients. The two correlations, together with internal conversion data, determine the spin and parity of the 1966-keV state to be 1^+ and the 812-keV gamma transition to be essentially pure $M1$ ($E2$ admixture $\leqslant 2\%$).

Using aqueous solution sources, we have carried out a transverse decoupling experiment by measuring the time-integrated angular correlation as a function of the magnetic field. Figure 8 shows the dependence of $\bar{G}_2$ and $\bar{G}_4$ on a transverse field. The experimental points for both the 2098–89-keV and the 1065–89-keV cascades are given. For sufficiently large magnetic fields, $\bar{G}_2$ approaches 0.45 compared with 0.83 in zero field, and $\bar{G}_4$ approaches 0.25 compared with 0.78 in zero field. The field required to reach saturation is of the order of 25 kG.

Again as in the case of Gd^{154}, the saturation values of $\bar{G}_2$ and $\bar{G}_4$ in Fig. 8 presumably correspond to the effect of a Paschen-Back decoupled hfs interaction, for the direct effect of the magnetic field on $\bar{G}_2$ and $\bar{G}_4$ is negligible. This transverse decoupling effect of the electron spin from the relaxation interaction, to date only detected with Gd isotopes, has been discussed in detail by Stiening and Deutsch.[8]

Assuming that the saturation values of $\bar{G}_2=0.45\pm0.05$ and $\bar{G}_4=0.25\pm0.05$ represent the effect of a fully decoupled hfs interaction, we can calculate the hfs constant a (see Appendix I of reference 8). Figure 9 gives the results of the calculations, together with the experimentally determined saturation values. It is to be noted that $\bar{G}_2$ and $\bar{G}_4$ give two independent values of $a\tau_N$, which agree with each other exceedingly well. This appears to indicate that the interaction due to the strong transverse magnetic field overrides the time-dependent electric interaction which seems to be present in aqueous solutions. The result for the magnetic hfs constant is

$$a\tau_N=0.34\pm0.05 \text{ radius}.$$

Using $\tau_N=3.20\pm0.08$ nsec, we find $a=17\pm3$ Mc/sec. This compares with 26.4 ± 5 Mc/sec for the 123-keV state of Gd^{154} as determined by Stiening and Deutsch.[8] If we use the evaluation of the hyperfine magnetic field by these authors, we obtain for the gyromagnetic ratio of the 89-keV state of Gd^{156} the value $g=0.26\pm0.06$.

Similar to the method employed in the Gd^{154} experiment, we have used sources of anhydrous $GdCl_3$ at about 1300°K for the precession type measurement of g. Our measurement of the time dependence of the angular correlations $W(\theta,t)$ of the 2098–89-keV and the 1065–89-keV cascades showed weak attenuations: $\tau_2=26\pm5$ nsec, giving $\tau_2/\tau_N=8.1\pm1.6$ and $G_2=0.89\pm0.02$;

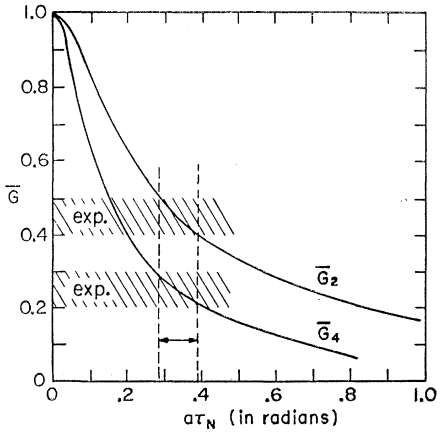

FIG. 9. Effect of the magnetic hfs on the time-integrated angular correlation for Gd^{156} (electronic spin $J=7/2$, nuclear spin $I=2$) in the case of a strong transverse magnetic field. The experimental saturation values for $\bar{G}_2$ and $\bar{G}_4$ with their uncertainties are included in the figure.

$\tau_4=18\pm3$ nsec, giving $\tau_4/\tau_N=5.6\pm0.9$ and $G_4=0.85\pm0.02$. These results are presented in Fig. 10. Within statistical accuracy (about 2%) the values of G_2 and G_4 did not change in the applied magnetic field.

The measurements of g were carried out at angles of 112.5° and 157.5° using a magnetic field of 26 kG. Field reversals and angular position changes were performed automatically according to a suitable schedule. The angular correlation function in the magnet with zero field was again measured at frequent intervals between the R measurements. Both the 2098–89-keV and the 1065–89-keV cascades were used in the measurement. A graphical solution of Eqs. (5) and (6) representing the evaluation of the 1065–89-keV cascade at 157.5° is illustrated in Fig. 11, giving a value $\omega_L\tau_N=0.134\pm0.005$. A similar analysis of the simultaneously measured 2098–89-keV cascade gives $\omega_L\tau_N=0.132\pm0.007$. The two values of $\omega_L\tau_N$ are in close agreement. The possibility that the measurement using the 1065–89-keV cascade, being part of a triple cascade, is influenced by the 1154-keV excited state is shown to be negligible.

The weighted mean of our results is $\omega_L\tau_N=0.133\pm0.005$ at a field of 26 kG. With an effective field $H_{\text{eff}}=1.042H_{\text{ex}}$ at 1300°K (see Sec. 4), using $\tau_N=3.20\pm0.08$ nsec, we obtain a g value of $+0.320\pm0.03$.

6. DISCUSSION OF RESULTS

The gyromagnetic ratios g of 0.350 ± 0.03 for the first excited state of $_{62}Sm_{90}^{152}$ and of 0.367 ± 0.03 for $_{64}Gd_{90}^{154}$ obtained from the precession measurements are of the same magnitude within statistical accuracy. This indicates that no appreciable difference in the magnetic moment is caused in the two nuclei by adding two protons. The g value of 0.320 ± 0.03 for $_{64}Gd_{92}^{156}$ appears to be about 10–20% below that of $_{64}Gd_{90}^{154}$, indicating the general trend within isotopes of the same element predicted by the more refined theories using pairing correlation corrections.[1,2] The value of 0.26 ± 0.06 for the g value of Gd^{156} obtained from the hyperfine structure measurement agrees within the accuracies of the decoupling experiment with the g value obtained by the direct measurement using the precession method.

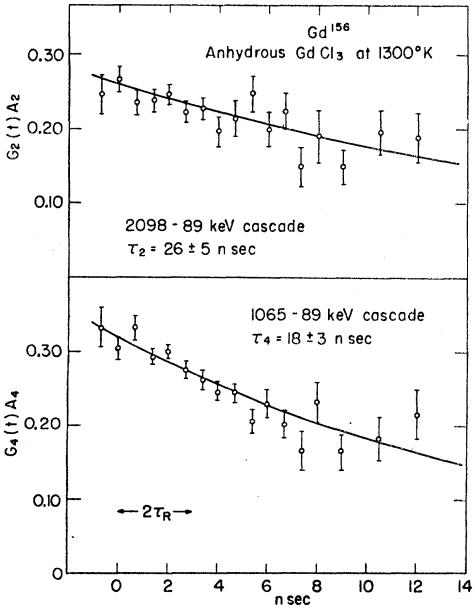

FIG. 10. Time dependence of the angular correlation in Gd^{156} for anhydrous chloride sources.

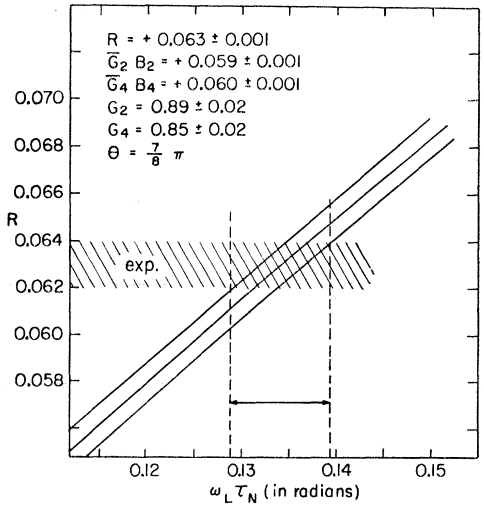

FIG. 11. The measured fractional coincidence counting rate difference R for a reversal of the magnetic field, is plotted against the quantity $\omega_L\tau_N$ giving the precession of the angular correlation pattern. The curve is obtained from Eqs. (5) and (6) using the measured values of the angular correlation and attenuation coefficients.

It is to be noted that the g value for Gd^{156} obtained from the decoupling experiment may appear to be somewhat low compared with the directly measured value using the precession method, just opposite from what seems to be the case in Gd^{154} as found by Stiening and Deutsch.[8] Though we believe that the interaction due to the transverse magnetic field is strong enough to override the time-dependent electric interaction apparently present in aqueous solution sources of Gd^{156}, we cannot be absolutely certain that the magnetic field of 26 kG is strong enough to produce complete decoupling in view of the fact that the lifetime in Gd^{156} is about twice as long as that in Gd^{154}.

The g values determined by the precession measurements have uncertainties of the order of 10%, of which about half is due to the statistical counting error. The other 5% of the errors quoted are due to the uncertain nature of the residual attenuations, uncertainties concerning the ionic state during the initial fraction of a nsec after beta decay and electron capture, the difficulties of measuring τ_N with great precision and of determining the effective field H_{eff} at the nucleus with high accuracy. These errors, we feel, make it extremely difficult to reduce the experimental uncertainties much below 10%.

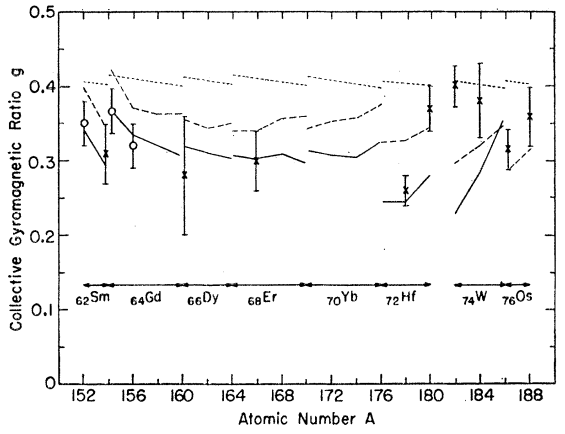

FIG. 12. The collective gyromagnetic ratios of even-even nuclei in the deformed region from Sm to Os. The open circles represent our experimental results, the crosses those of other investigators (for references see text). The vertical lines indicate experimental errors. The theoretical predictions are represented as follows: dotted lines for Z/A, dashed lines for results by Yang Hu *et al.* (reference 2) using deformation parameters calculated from experimental intrinsic quadrupole moments, solid lines for results by Nilsson *et al.* (reference 1) for "Case A". The lines are drawn for neighboring isotopes.

Our experimental results of the gyromagnetic ratios for the first excited states of Sm^{152}, Gd^{154}, and Gd^{156} are presented in Fig. 12. To allow a comparison with recent theories the following information has been added to Fig. 12: (1) values of Z/A predicted by the simplest quasi-hydrodynamic "homogeneous flow" model due to Bohr and Mottleson[3]; (2) theoretical results of the calculations by Nilsson and Prior[1] based on Inglis' cranking model,[27] with Belyaev's pairing correction[28]; (3) theoretical results by Yang Hu and Villars[2] obtained from an extension of the treatment of the moment of inertia problem due to Peierls and Yoccoz,[29] with pairing corrections included. Our experimental results agree with Nilsson's theory, while the calculation by Yang Hu predicts slightly larger values which, however, are generally still below Z/A.

In Fig. 12, for reasons of systematics, we have also included the experimental results of gyromagnetic ratios of even-even nuclei between Sm and Os reported in the literature to date. These experimental values are: Sm^{154} by Goldring *et al.*,[11] after applying the paramagnetic correction by Kanamori *et al.*,[17] Dy^{160} by Manning *et al.*[14] (not included are the results by the Zurich group[30,31] which give values of 0.18 ± 0.08 and 0.46 ± 0.05 for measurements on the same isotope). Er^{166} by Manning *et al.*,[14] Kündig,[31] and by Bodenstedt *et al.*[32] The results of the three groups agree within the uncertainties quoted. The weighted mean of the three results is given in Fig. 12. Hf^{178} by Karlsson *et al.*,[33] Hf^{180} by Bodenstedt *et al.*,[34] W^{182} by Kegel,[35] W^{184} by Bodenstedt *et al.*,[36] Os^{186} weighted mean of the results by Bodenstedt *et al.*,[37] and Lerjefors *et al.*,[38] Os^{188} by Karlsson *et al.*[39]

Almost all the presently available data on the gyromagnetic ratios of even-even nuclei in the deformed region under study seem to suggest that the pairing correlation plays an important role in the collective properties of nuclei. Below the atomic number 180 the experimental results appear to be lower than the value of Z/A, in close agreement with Nilsson's theoretical values. Toward the upper end of the deformed region, however, the distinction is less clear and the experimental points scatter between the values predicted by the simple homogeneous flow model and the results from the pairing correlation evaluations. But as a whole, the experimental results are not in disagreement with the more refined theories using pair correlation corrections.

ACKNOWLEDGMENTS

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[27] D. R. Inglis, Phys. Rev. **96**, 1059 (1954); **97**, 701 (1955).
[28] S. T. Belyaev, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd **31**, No. 11 (1959).
[29] R. E. Peierls and J. Yoccoz, Proc. Phys. Soc. (London) **A70**, 381, 388 (1957).
[30] P. Debrunner, W. Kündig, J. Sunier, and P. Scherrer, Helv. Phys. Acta **31**, 326 (1958).
[31] W. Kündig, Helv. Phys. Acta **34**, 125 (1961).
[32] E. Bodenstedt, H. J. Körner, C. Günther, and J. Radeloff, Nuclear Phys. **22**, 145 (1961).
[33] E. Karlsson, E. Matthias, and S. Ogaza, Arkiv Fysik **22**, 257 (1962). *Note added in proof.* The gyromagnetic ratio of Hf 178 has been corrected to $+0.29\pm0.02$. Figure 12 shows a previously communicated result. We are grateful to Dr. Karlsson for informing us of this correction.
[34] E. Bodenstedt, H. J. Körner, E. Gerdau, J. Radeloff, C. Günther, and G. Strube, Z. Physik **165**, 57 (1961).
[35] G. H. R. Kegel, Ph.D. thesis, Massachusetts Institute of Technology, 1961 (unpublished).
[36] E. Bodenstedt, E. Matthias, H. J. Körner, E. Gerdau, F. Frisius, and D. Hovestadt, Nuclear Phys. **15**, 239 (1960).
[37] E. Bodenstedt, H. J. Körner, G. Strube, C. Günther, J. Radeloff, and E. Gerdau, Z. Physik **163**, 1 (1961).
[38] C. A. Lerjefors, E. Matthias, and E. Karlsson, Nuclear Phys. **25**, 404 (1961).
[39] E. Karlsson, C. A. Lerjefors, and E. Matthias, Nuclear Phys. **25**, 385 (1961).