Magnetic Moment and Hyperfine Structure Coupling of the First 2+ State in Gd^{154}†

RAE STIENING* AND MARTIN DEUTSCH
Department of Physics and Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, Massachusetts
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The g value of the 0.123-Mev 2+ state of Gd^{154} is found to be $g=0.36\pm0.06$. The hyperfine structure constant of the same nuclear state in the ${}^8S_{7/2}$ ground state of Gd^{3+} in aqueous solution is determined as $a=26.4\pm5$ Mc/sec. The perturbation of the angular correlation of gamma-ray cascades proceeding through this state was studied. It is found that the perturbation in aqueous solution can be decoupled by magnetic fields either parallel to one of the gamma rays or perpendicular to the plane of observation. The perturbation in molten $GdCl_3$ is found to be much weaker than in solution and to be unaffected by magnetic fields. Auxiliary measurements involving the first 2+ states in Sm^{152} and Gd^{156} are described.

1. INTRODUCTION

THE measurement of the gyromagnetic ratio g of the so-called rotational nuclear states, especially the first 2+ state of highly deformed nuclei, has recently attracted considerable interest since it provides an important additional parameter to test the more refined theories of collective excitation. The simplest quasihydrodynamic model[1] necessarily predicts $g=Z/A$ or possibly $g=Z'/A'$, where Z' and A' are the number of protons and the total number of nucleons outside closed shells, respectively. A recent calculation considering pairing correlations[2] makes specific predictions for the deformed nuclei between Sm and Os, generally indicating $g<Z/A$.

Several experimental determinations have been published[3–5] or are in process of publication.[6,7] Their results seem to agree at least roughly with expectations but all of them are subject to very considerable corrections involving the influence of atomic electrons on the nuclear orientation. The mechanisms by which angular correlations of nuclear radiations are reduced by extranuclear effects have been discussed by several authors.[8–10] It is not obvious that the assumption of correlation coefficients decreasing exponentially with time is sufficiently accurate in the case of large attenuations to which it has been applied. The present work* shows that it may not even be automatically assumed that this attenuation is independent of an applied magnetic field $H_\perp$ required for the measurement of g.

In the case of nuclei forming paramagnetic ions the effective value of $H_\perp$ must be calculated with a correction involving the hfs (hyperfine structure) interaction and the paramagnetic susceptibility. Heretofore there has been no experimental basis for the use of the properties of the ground state of the daughter ion in a radioactive decay for these calculations.

We have studied these questions in some detail for the case of the first 2+ state of Gd^{154}, and we believe that we have arrived at an adequate understanding of the mechanisms involved and have obtained values of g and of the hfs splitting in the Gd^{3+} ion. Our experiments measure the angular correlation between gamma rays involving the 2+ intermediate state following the beta decay of Eu^{154} and, in some auxiliary measurements, of Eu^{156} and Eu^{152}. The relevant features of the decay schemes are shown in Fig. 1. Most of the measurements concerned the 1.28–0.123 Mev cascade in Gd^{154} and the subsequent discussion refers to this unless otherwise indicated.

We shall write[9] for the angular correlations

$$W(\theta,t)=1+G_2(t)A_2P_2(\cos\theta)+G_4(t)A_4P_4(\cos\theta),$$
$$W(\theta)=1+G_2A_2P_2(\cos\theta)+G_4A_4P_4(\cos\theta).$$

The first of these 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 τ_K. The second equation gives the correlation including all gamma rays, regardless of delay. For exponential attenuation $G_K=1/(1+\tau_N/\tau_K)$, where τ_N is the mean life of the 2+ state. For the Sm^{152} cascade and the 1.28-Mev Gd^{154} cascade $A_4\simeq0$ and we have $A_2=[W(\pi)-W(\pi/2)]/[W(\pi/2)+\frac{1}{2}W(\pi)]$. Most of our results are presented in terms of this quantity.

Angular correlations from aqueous solution sources have been known to be markedly perturbed ($G<1$) for Gd^{154},[3] but believed to be unperturbed for Sm^{152}.[3,11] This is confirmed by our experiments described in Sec. 3. The difference in behavior of two very similar nuclear

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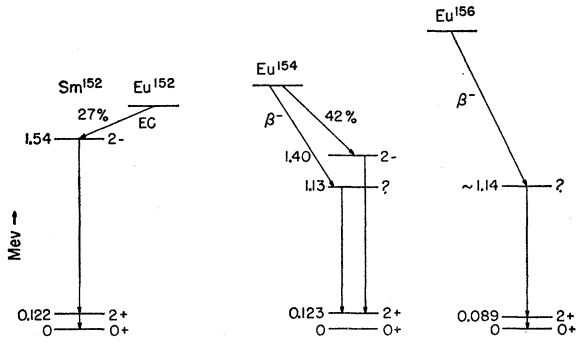

FIG. 1. Relevant features of the radioactive decay schemes involved in the experiments.

states must be due to the different electronic structure of the ions. Goldring and Scharenberg[3] called attention to the fact that the $^8S_{7/2}$ ground state of the Gd^{3+} ion with a spherical half-filled $4f$ shell shows an abnormally long spin relaxation time so that the magnetic hfs coupling, although quite weak, may be sufficient to reorient the nuclear spin direction. This conjecture is proved correct in Sec. 4, where we show that a magnetic field $H_{\parallel}$ of a few hundred gauss applied in the direction of one of the gamma rays substantially increases the angular correlation. A field $H_{\parallel}=3300$ gauss raises G_2 to 0.964 ± 0.077 from its zero-field value of 0.82 ± 0.02 for a water solution source.

From the fact that we have achieved substantially complete decoupling together with the observation that in the Gd^{156} cascade $G_4(t)$ falls nearly to zero in about 5 nanoseconds (nsec), we conclude that after beta decay the great majority of the atoms reach the ground state of the Gd^{3+} ion in a time considerably less than τ_N.

In another type of magnetic decoupling experiment described in Sec. 5, we find that a magnetic field $H_{\perp}$ perpendicular to the plane of the counters reduces G_2 below its zero-field value. For fields $H_{\perp}\geq 15\,000$ gauss $G_2\simeq 0.5$. This is interpreted as indicating that the relaxation mechanism in water ceases to be effective when the separation between electronic m states due to the external magnetic field is sufficiently large. The hfs coupling constant for the 2+ state may then be calculated from this value of G_2. The calculations are presented in Appendix I. The result is $a=26.4\pm5$ Mc/sec. The unperturbed Sm^{152} cascade resulting from the decay of Eu^{152} showed no effects due to magnetic decoupling.

Since the rapid, field-dependent, nuclear spin relaxation in aqueous solutions makes such sources unsuitable for direct g-value measurements we searched for another, more suitable medium. As shown in Sec. 6, a melt of anhydrous $GdCl_3$ at about 1000°C shows only relatively weak ($G_2=0.93\pm0.01$) and field-independent attenuation. With such a source a measurement of g by the method of the rotation of the angular correlation pattern in a transverse field $H_{\perp}=15\,000$ gauss (Sec. 7) yielded $g=0.36\pm0.06$. This is in good agreement with the value $g=0.41\pm0.08$ derived from the hfs coupling (Sec. 5) and spectroscopic estimate[12,13] of the internal magnetic field.

2. APPARATUS AND PROCEDURE

(a) Source Preparation

Eu^{154} and Eu^{152} (12.5 yr) sources with a specific activity of about 0.5 mC/mg were prepared from isotopically highly enriched oxides by slow-neutron bombardment. These sources were used without chemical purification. Eu^{156} was prepared by exposure of enriched Sm^{154} to a high neutron flux 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 by reduction of the sulphate on zinc amalgam.

Aqueous solution sources were sealed in very thin Pyrex tubes of about 1-mm diameter. Melt sources were prepared in similar "Vycor" tubes by drying a hydrochloric acid solution to which about 0.5 mg of $GdCl_3$ had been added, in vacuum. The solid chloride appeared somewhat dark after heating, perhaps indicating a change in composition. The source capsules were heated in vacuum by means of a 0.001-inch thick niobium ribbon which completely surrounded the source.

(b) Magnets

Longitudinal decoupling fields $H_{\parallel}$ (Sec. 4) were provided by Alnico horse-shoe magnets as shown in Fig. 2. The pole-piece and source assembly was held fixed and various field strengths were obtained by interchanging magnets. A brass mockup was used for zero-field data.

Conventional C-yoke electromagnets were used for the transverse field $H_{\perp}$ (Sec. 6). In all cases sufficient magnetic shielding was provided for the counters to

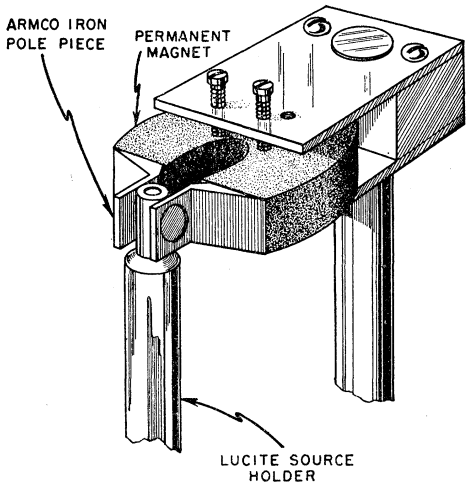

FIG. 2. Magnet for longitudinal decoupling experiment. One of the gamma rays reaches the counter through the thinned-down area in one pole piece.

[12] W. Low and D. Shaltiel, J. Phys. Chem. Solids **6**, 315 (1958).
[13] D. R. Speck, Phys. Rev. **101**, 1725 (1956).

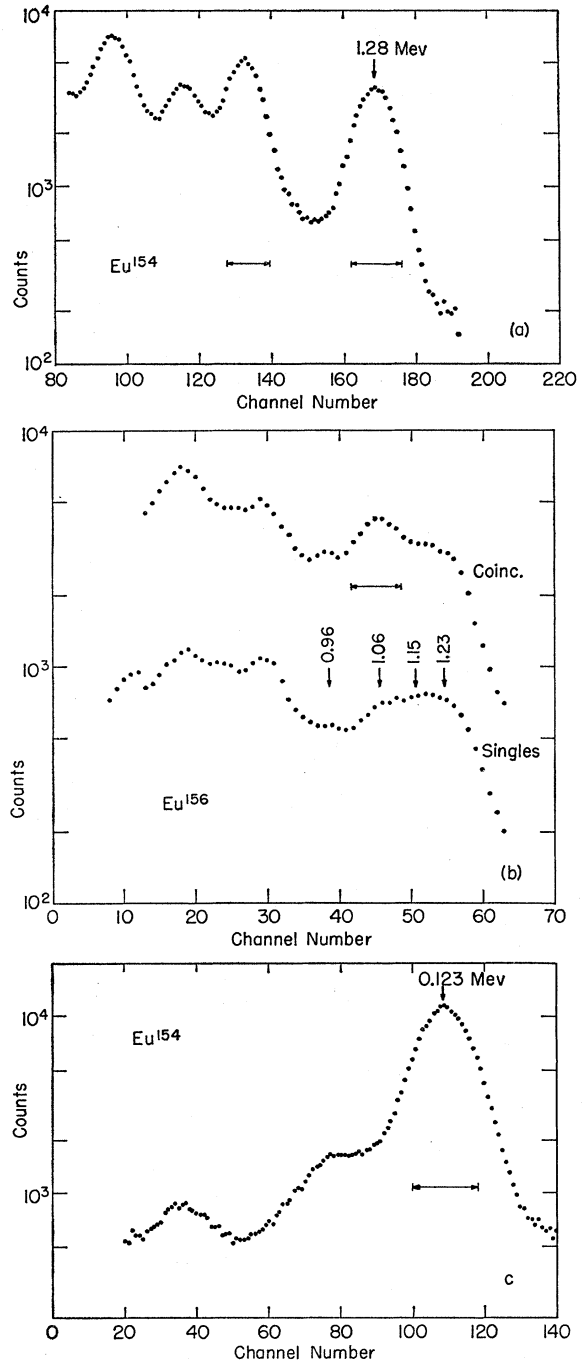

FIG. 3. Pulse-height spectra. The horizontal arrows indicate the pulse-height intervals used in the coincidence experiments. (a) and (b): Spectra observed with the 3-in.×3-in. crystal. (c): Spectrum observed with the nonactivated NaI crystal.

reduce magnetic field effects on either pulse height or timing below detectable levels. Small counting rate changes (of the order of 0.1%) introduced by mechanical motion of the magnet and the heating filament were accounted for in the usual fashion by interpreting them as a change in counter efficiency.

(c) Electronics and Detectors

The only major departure of our instrumentation from conventional designs is the use of a pure (nonactivated) NaI scintillator cooled to liquid N_2 temperature, to detect the 0.12-Mev and 0.089-Mev radiations in the delayed-coincidence experiments. A $\frac{1}{4}$-in. thick by 1-in. diameter crystal was coupled to a quartz-faced RCA 14-stage photomultiplier with *S*-20 response cathode by means of a four-inch long quartz light pipe in an arrangement very similar to that described by Beghian *et al.*[14] When the faster response of the pure iodide was not needed, a crystal of NaI(Tl) of similar dimensions was used with a 7265 RCA photomultiplier. The high-energy gamma rays were detected by a 3-in.×3-in. crystal mounted on an RCA 7046 photomultiplier. The pulse height and time resolution of the arrangement is illustrated in Figs. 3 and 4.

The electronic circuits consisted of a generally conventional fast-slow coincidence arrangement with the usual single-channel and multichannel pulse-height selectors. A time-to-pulse-height converter based on the design of Bell and Graham[15] was used with some modifications utilizing solid-state electronic components.

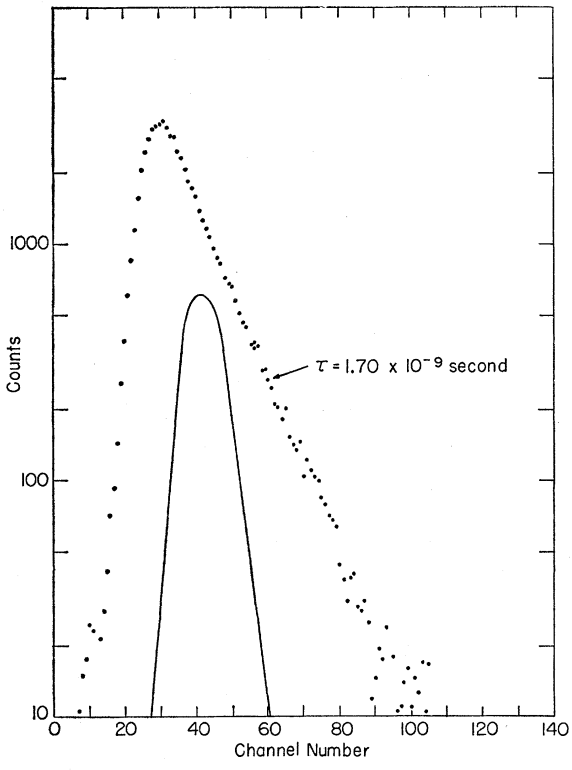

FIG. 4. Decay curve of the 0.123-Mev state in Gd^{154} observed with the time-to-pulse-height converter. The solid line illustrates the time resolution for prompt coincidences between gamma rays of the same energies.

[14] L. E. Beghian, G. H. R. Kegel, and R. P. Scharenberg, Rev. Sci. Instr. **29**, 753 (1958).

[15] R. E. Bell, R. L. Graham, and H. E. Petch, Can. J. Phys. **30**, 35 (1952).

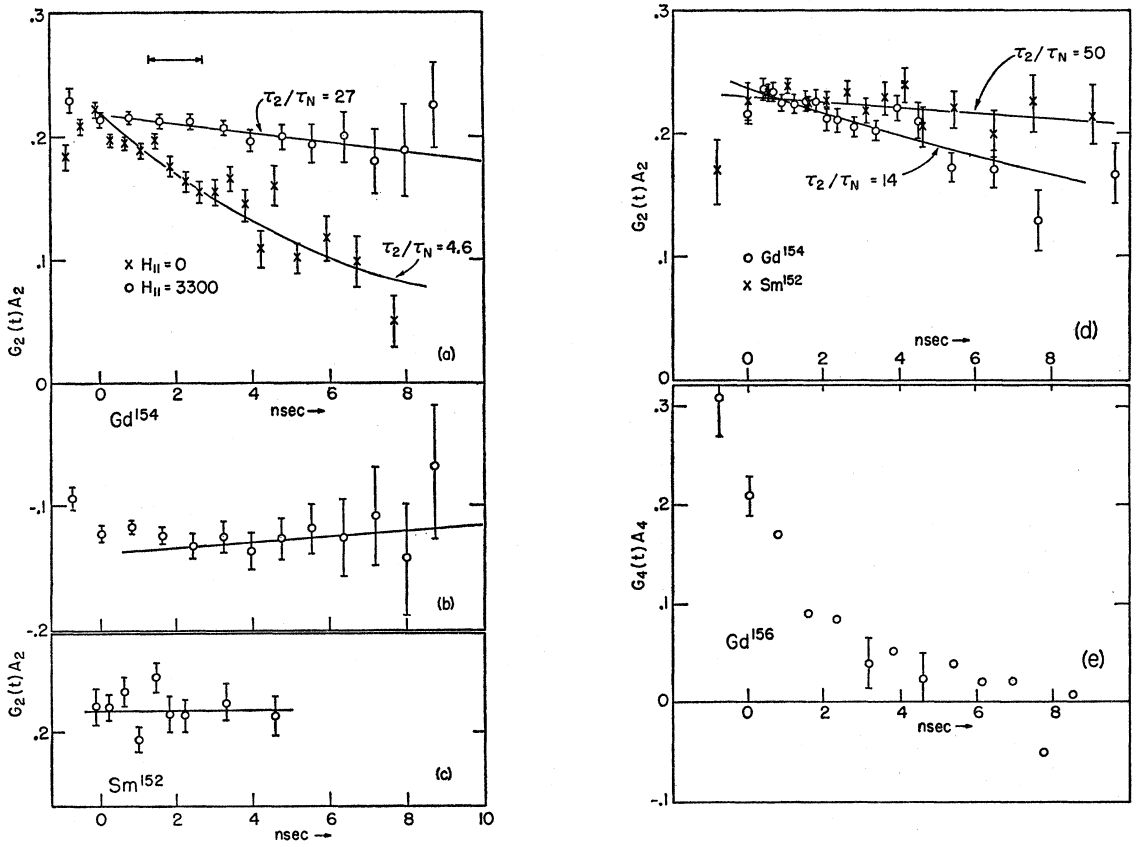

FIG. 5. Dependence of the angular correlation on delay time. The horizontal arrow indicates the instrumental time resolution. (a): 1.28 Mev–0.123-Mev cascade in Gd^{154}. Aqueous solution source with and without longitudinal decoupling field. (b): 1.03 Mev–0.123-Mev cascade in Gd^{154} with decoupling field. (c): Sm^{152} cascade without field. (d): Molten chloride sources at 1000°C. (e): Gd^{156} aqueous solution source.

The gain of the pulse-height selecting "slow" channels was stabilized by a device similar to that published by deWaard[16] but the feedback was applied to the slow amplifier gain rather than the photomultiplier voltage supply in order to avoid changes in electron transit time. As a further protection against drifts in the apparatus, the data for each experiment were taken automatically according to a suitable schedule in which counter positions or magnetic field directions were changed at intervals of ten or twenty minutes.

3. PERTURBATION IN AQUEOUS SOLUTIONS

In none of the gamma-ray cascades studied is the unperturbed angular correlation known *a priori*. The only method of determining the attenuation G and the unperturbed correlation coefficients A is a measurement of $W(\theta,t)$, i.e., the delay-time dependence of the angular correlation.

Figure 4 illustrates a decay curve of the Gd^{154} cascade observed with the time-to-height converter. The ordinate represents $\log[W(\pi/2)+\frac{1}{2}W(\pi)]$ which should be independent of the angular correlation since no terms higher than A_2 appear in $W(\theta)$. At all values of t for which the decay curve in Fig. 4 is a straight line, the slope of an exponential $G_2(t)A_2$ should be correctly given by the data. The effect of the finite time resolution is only a constant shift of the time axis. Figures 5(a) and (c) show G_2A_2 as a function of delay time for aqueous solution sources of Eu^{152} and Eu^{154}. A correction for finite angular resolution of the counters was applied to the data according to the procedure of Rose.[17] The Sm^{152} state is seen to be unperturbed within the accuracy of our measurement, i.e., $\tau_2/\tau_N > 20$ or $G_2 > 0.95$. This confirms Ofer's[11] conclusion that there is an admixture of $M2$ radiation to the $2^- \rightarrow 2^+$ transition.

The Gd^{154} correlation is obviously perturbed.

Figure 5(e) shows $G_4(t)A_4$ for the correlation in Gd^{156} under the same conditions as Figs. 5(a) and (c). A_2 in this case was too small to yield significant data. Here the attenuation is even more marked. We note that neither Fig. 5(a) nor 5(e) shows evidence of the oscillatory behavior of $G_k(t)A_k$ expected from the hfs coupling to a free ion. On the other hand, the ratio of the attenuation rates for A_2 and A_4 is quite plausible for a magnetic interaction[9] (assuming g to be similar in Gd^{154} and Gd^{156}) but not for an electric quadrupole interaction. The fact

[16] H. deWaard, Nucleonics **13**, 36 (1955).

[17] M. E. Rose, Phys. Rev. **91**, 61 (1953).

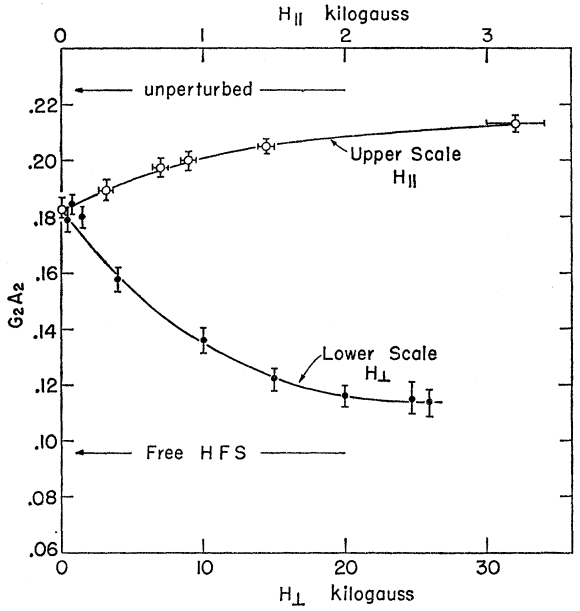

FIG. 6. Effect of longitudinal and transverse decoupling fields on the angular correlation.

that $G_4(t)$ falls to zero in Fig. 5(e) indicates that all of the atoms have reached a "perturbed" ionic state—presumably the ground state of Gd^{3+} in a time short compared with τ_N. The nonperiodic behavior of $G(t)$ is probably due to electronic spin relaxation. In fact, a measurement[18] of the linewidth at 9 kMc/sec of the Gd^{3+} paramagnetic resonance in the source used to obtain Fig. 5(a) gave a result consistent with a relaxation time of the order of 10^{-10} sec. The effects entering into the resonant linewidth are not quite the same as those relevant to our experiment but the order of magnitude is clearly correct to cause an exponential behavior of $G(t)$. We cannot at this point rule out the possibility that different Gd^{3+} ions reach the ground states at different times after formation within, say, the first nanosecond. This also would tend to destroy the periodicity of $G(t)$. Finally, there may be a contribution from electric field gradients either due to charges associated with the solvent or due to polarization of the ion. The evidence presented in the following sections indicates that the two last-named effects do not contribute in an important manner. It must be admitted that none of our experiments would detect a true "impulse" perturbation, i.e., loss of nuclear orientation within a fraction of a nanosecond. There is no indication, experimental or theoretical, for the existence of such perturbation, however.

Assuming, then, an exponential attenuation, we find from Fig. 5(a) $\tau_2=(7.8\pm1.0)\times10^{-9}$ sec, $\tau_2/\tau_N=4.6\pm0.6$. This yields $G_2(154)=0.82\pm0.02$. The time integral correlation coefficient for this source is $A_2G_2=0.184\pm0.005$ so that we deduce $A_2=0.224\pm0.018$. Combining this determination with a similar value obtained from the curve in Fig. 5(d), we arrive at the value $A_2=0.227\pm0.006$.

The source used in Fig. 5(a) was a strong solution of $Gd(NO_3)_3$. Other sources containing, instead, $EuCl_3$ in concentrations ranging from 5×10^{-4} mole/liter to 0.05 mole/liter gave the same values of G_2 within about two percent. The same was true, with lesser statistical accuracy, for sources containing $HoCl_3$ or concentrated HNO_3. The ionic relaxation phenomenon thus seems to be primarily a function of the solvent.

4. MAGNETIC DECOUPLING OF THE PERTURBATION

It is well known theoretically[9] that the perturbation of an angular correlation by the hfs interaction with a free paramagnetic atom or ion can be removed by applying a magnetic field in the direction of one of the radiations but this phenomenon does not seem heretofore to have been observed experimentally.

Figures 5(a) and (b) show $G_2(t)$ for a Eu^{154} source in aqueous solution in the magnet shown in Fig. 2 with $H_{11}=3300$ gauss. Points are shown for both gamma cascades indicated in Fig. 1. The solid lines are drawn for $\tau_2/\tau_N=27\pm7$ which gives the best fit to the data.

Figure 6 shows the value of G_2 for various values of H_{11}. No measurable effect of H_{11} was observed in the case of Eu^{152} sources or in the case of solid crystalline Eu^{154} sources. If the attenuation of the correlation were due to hfs interaction in free Gd^{3+} ions, the magnitude of H_{11} required for reaching substantially $G_2=1$ would be of the order of $10\ a/\mu_0$ gauss, where a is the hfs coupling constant defined by

$$E(\text{hfs})=a\mathbf{I}\cdot\mathbf{J}.$$

We know from nuclear magnetic resonance measurements[12] that for the expected g value of the 0.123-Mev level, the upper curve in Fig. 6 should reach its saturation value for $H_{11}\simeq100$ gauss. In fact, a value about ten times larger is required. This is undoubtedly due to the fact that the electron spin must be decoupled from the perturbing interaction. The condition for longitudinal decoupling is a sufficiently rapid averaging of the transverse component of the hfs field. For a free ion this implies simply that the electronic Larmor frequency be much greater than the hfs constant a. No calculation has ever been made of the form which the curve in Fig. 6 should take in the case of relaxation times comparable with the reciprocal hfs frequency. The nature of the relaxation mechanism probably plays a role in this. For a relaxation by collisions capable of transferring an energy large compared with $\mu_J H_{11}$, the decoupling condition is probably $\mu_c H_{11}\gg\hbar/t_c$, where t_c is the mean time between collisions. Although the order of magnitude of the relaxation time is correct in our case to fit Fig. 6, we shall see in Sec. 5 that the true relaxation mechanism

[18] We are indebted to Professor M. W. P. Strandberg for this measurement.

probably involves interactions with a definite cutoff in the energy-transfer spectrum.

It should be kept in mind that the value of G_2 which has to be brought to unity by the decoupling field is not the zero-field value in Fig. 6, but the value indicated by the lower arrow in the figure. This value corresponds to G_2 for free Gd^{3+} ions as deduced from the experiments described in Sec. 5. The small residual attenuation in a 3300-gauss field is not fully understood. Some of it may be an experimental artifact. In view of the small dimensions of the pole pieces (Fig. 2), we cannot be sure that H_{11} is truly parallel to one counter axis for all parts of the source. It is, however, also possible that the lack of complete saturation represents the spectrum of the relaxation interaction.

The fact that substantially complete saturation can be reached shows that none of the ions spend an appreciable fraction of τ_N in any state with either much stronger hfs interaction, much longer relaxation time or much smaller electronic g value than the Gd^{3+} ground state. The behavior of the attenuation with $H_{11}=0$, on the other hand, shows also that there is not any appreciable time interval in which a significant fraction of the nuclear spins are unperturbed. We conclude therefore that the ionic ground state is reached in less than 1 nanosecond by the great majority of the ions.

5. TRANSVERSE DECOUPLING

The lower curve in Fig. 6 shows the dependence of G_2A_2 for an aqueous solution on a transverse field $H_\perp$. For sufficiently large $H_\perp$, G_2 approaches 0.5 compared with 0.82 in zero field. The field $H_\perp$ required to reach saturation is almost one order of magnitude larger than H_{11}.

The saturation value of G_2 in Fig. 6 presumably corresponds to the effect of a Paschen-Back decoupled hfs interaction for the direct effect of $H_\perp$ on the magnitude of G_2 is negligible. If the ionic spin relaxation time were long even in zero field, the effect of $H_\perp$ would be expected to be quite small (see Appendix I). We must therefore assume that the effect of the field is here again as in Sec. 4 a decoupling of the electron spin from the relaxation interaction The difference in field strength required in the two cases is somewhat analogous to the transverse and longitudinal relaxation times T_1 and T_2 in nuclear magnetic resonance experiments. In order to reach saturation (Fig. 6), it is presumably necessary that transitions between electronic m states be slow compared with the nuclear lifetime, which is a much more stringent condition than that for "longitudinal" decoupling. Thus the separation between the Zeeman components in the strong field $H_\perp$ is greater than any abundant frequency in the perturbing interaction spectrum.

Assuming that the saturation value of $G_2=0.50\pm0.06$ represents the effect of a fully decoupled hfs interaction, we can calculate the hfs constant a (see Appendix I). The result is

$$a\tau_N=0.28\pm0.05.$$

Using $\tau_N=(1.70\pm0.05)\times10^{-9}$ sec (see Sec. 7), we find $a=26.4\pm5$ Mc/sec. This compares with 11.9 ± 0.4 Mc/sec and 16.0 ± 0.3 Mc/sec, for the ground states of Gd^{155} and Gd^{157}, respectively.[12] The magnetic moments of the stable nuclei are known only quite roughly. Speck[13] from a spectroscopic determination of the hfs of the 5015 A line obtains

$$\mu_{155}=-0.30\pm0.04 \text{ nm}, \quad \mu_{157}=-0.37\pm0.04 \text{ nm}.$$

These values assume $\mathbf{L}\cdot\mathbf{S}$ coupling and involve questions of configuration mixing. The older values of Murakawa[19] were calculated on the assumption that $I=\frac{7}{2}$ and are therefore not applicable since $I=\frac{3}{2}$. Low[12] calculates $\mu_{155}=-0.24$ nm, $\mu_{157}=-0.32$ nm from the ratio of paramagnetic resonance values of a for $Gd^{155,157}$ to that for Eu^{151}. Assuming identical configuration mixing in Eu^{2+} and Gd^{3+} and the values of $\langle 1/r^3\rangle$ for the two ions calculated by Bleaney, he uses spectroscopic determination of μ_{151} to deduce the above values. If we adopt Speck's value for the moment of Gd^{157}, we obtain $g_{2+}{}^{154}=0.41\pm0.08$.

6. PERTURBATION IN AN IONIC ENVIRONMENT

It has been shown in the preceding sections that aqueous solutions do not provide a suitable medium for the direct determination of the g value since the electronic spin relaxation is both too slow and magnetic field dependent. Sources in ionic crystals are known to yield, in general, strongly perturbed angular correlations. We also found this to be true for Eu^{154} sources and showed that this perturbation could not be decoupled, within the precision of measurement, by a field of $H_{11}=3.3$ kilogauss in agreement with expectation. Perturbed crystalline sources are, of course, unsuitable for direct moment determinations, but it seemed plausible that the same strong interactions might fluctuate sufficiently rapidly in an ionic melt to produce a short electronic relaxation time under all conditions.

Figure 5(d) shows the result obtained with a source of $GdCl_3$ at 980°C, well above its melting point. The best exponential fit to the data, indicated by the solid line in the figure, yields $\tau_2=(24\pm5)\times10^{-9}$ sec or $\tau_2/\tau_N=14\pm3$, about three times the value obtained in aqueous solution. More important than the reduced attenuation, however, is the fact that the value of G_2 was not changed (within the statistical accuracy of about 3%) by a field of $H_\perp=15$ kilogauss. This source was deemed suitable for a direct determination of g. The nature of the small remaining attenuation is not fully understood. It may reflect a time-dependent quadrupole interaction in the liquid. In any case it can be accounted for in the g-value measurement with an uncertainty which is surely smaller than τ_N/τ_2.

[19] K. Murakawa, Phys. Rev. **96**, 1543 (1954).

7. DIRECT MEASUREMENT OF g

The gyromagnetic ratio of the Gd^{154} state was measured by observing the change in coincidence rate of the 1.28- and 0.123-Mev gamma rays emitted at 135° to each other when the direction of a field $H_\perp=15$ kilogauss was reversed. This method has been described repeatedly in the literature.[9] In our experiment the counting rate difference between the two field directions was $2.16\pm0.28\%$. In order to evaluate the results of this measurement, it is necessary to know with precision the angular correlation under the conditions of the experiment, the attenuation $G_2(t)$, the nuclear lifetime τ_N, and the magnitude H_{eff} of the average magnetic field at the nucleus.

The mean life of the 2^+ state has been reported by Sunyar[20] and by Birk *et al.*[21] to be 1.72 ± 0.15 nsec and 1.78 ± 0.15 nsec, respectively. We have carefully remeasured this quantity using delayed coincidences between beta rays from Eu^{154} and conversion electrons from the 0.123-Mev level. The time analysis was calibrated against the velocity of gamma rays in air using annihilation radiation. The experimental part of the decay curve beyond the range of possible contributions from "prompt" coincidences was analyzed by Peierls' method. The time resolution was better than 1 nsec. The result is $\tau_N=(1.70\pm0.05)\times10^{-9}$ sec. The error indicated represents primarily the uncertainty of the time calibration.

The stability, uniformity, and calibration of $H_\perp$ are better than two percent. The paramagnetic correction to the field at the nucleus is presumably due to the field of a penetrating s-wave function and therefore positive. The ionic g value is known to be very close to 2.[12]

The additional field at the nucleus is given by

$$\Delta H=\langle H(0)\rangle_{\text{av}}\frac{\langle J_z\rangle}{J}=\langle H(0)\rangle_{\text{av}}\frac{2\mu_0}{3kT}(J+1)H_\perp,$$

and $\langle H(0)\rangle_{\text{av}}$ is the value used in Sec. 5 to calculate g_N, namely,

$$a\equiv\frac{\mu_N g_N}{J}\langle H(0)\rangle_{\text{av}},\quad \langle H(0)\rangle_{\text{av}}=2.9\times10^5 \text{ gauss},$$

$$J=\tfrac{7}{2},\quad T=1.2\times10^3\ ^\circ\text{K}.$$

Thus

$$\Delta H=0.05H_\perp.$$

The diamagnetic reduction of the field at the nucleus is $\Delta H=-0.0071H_\perp$. Thus, we obtain $H_{\text{eff}}=1.042H_\perp$.

The attenuation of the angular correlation was determined in Sec. 6:

$$G_2(t)=e^{-t/14},\quad t \text{ in nanoseconds}.$$

The proper value of the angular correlation to use in our experiment requires some discussion. From the several curves in Fig. 5 and the integral values of G_2A_2 we obtain an average $A_2=0.227\pm0.006$. The experimentally observed correlation in the actual geometry used differs from this for several reasons. The effect of the solid angle subtended by the counters and of the value of G_2 are readily understood. In addition, there is an appreciable effect of scattering by the pole pieces of the magnet, the vacuum system and the filament. Careful comparison of the value of G_2A_2 obtained in a nearly scatter-free geometry and with the source located between the pole pieces indicates a reduction of A_2 by about 5%. We verified experimentally that the form of the angular correlation remained the same in the presence of the scattering effects, i.e.,

$$W(\theta)=1+A_2,P_2\times(\cos\theta),$$

where we find $A_2'=0.180\pm0.004$. We assume that $A_2'(t)=A_2'(0)e^{-t/\tau_2}$, where $\tau_2=14\tau_N$ (see Sec. 6) and that at any time t it is the instantaneous correlation which rotates with the Larmor frequency ω_L,

$$W(\theta,t,\pm H_\perp)=1+A_2'(t)P_2[\cos(\theta\pm\omega_L t)],$$

$$A_2'(0)=A_2'/G_2=A_2'\left(1+\frac{\tau_N}{\tau_2}\right)=1.07A_2'=0.192.$$

This procedure of using an empirical value of A_2 implies that the effect of a physical rotation of one counter on the coincidence rate is the same as that of a precession of the nuclear spin through the same angle. This is not necessarily true in a situation such as ours in which the scattering material is not distributed with cylindrical symmetry. In particular, an appreciable contribution to the counting rate due to Compton scattering of quanta with initially higher energy than the radiations investigated may cause serious distortions. It is clear from Figs. 3(a) and 3(c) that this cannot occur in our case since the only gamma ray in coincidence with the 1.28-Mev gamma ray is the 0.123-Mev ray and, on the other hand, the most energetic gamma ray in coincidence with the 0.123-Mev ray is the 1.28-Mev ray. Lower energy gamma rays are easily separated from this by pulse-height selection. Elastic scattering of the 0.123-Mev gamma ray is therefore the main cause of the reduction in A_2. In determining the angular correlation with the source located in the magnet, we rotated the large counter, detecting the high-energy gamma ray, leaving the 0.123-Mev gamma detector stationary. Under these circumstances the use of the observed correlation in the manner described is justified.

Integrating over t we obtain, after some simple manipulation,

$$R=\Delta W/\bar{W}=4B_2'\omega_L\tau'/[1+(2\omega_L\tau')^2].$$

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

Here $\tau'=G_2\tau_N$, $\omega_L=g_N H_{\text{eff}}\mu_N/\hbar$, $B_2'=3A_2'/(4+A_2')$,

$$\Delta W=[W(3\pi/4;+H)-W(3\pi/4;-H)],$$

$$\bar{W}=\tfrac{1}{2}[W(3\pi/4;+H)+W(3\pi/4;-H)].$$

Using the experimental value $R=0.0216\pm0.003$, we get $\omega_L=(2.66\pm0.35)\times10^7$ and with $H_{\text{eff}}=15\,600\pm400$ gauss,

$$g_{2^+}{}^{154}=0.36\pm0.06.$$

8. CONCLUSIONS

The gyromagnetic ratio $g=0.36\pm0.06$ obtained from the direct determination and the value $g=0.41\pm0.08$ from the hyperfine structure measurement are rather close to Z/A, but the errors are still too large to allow any far-reaching conclusions. It is gratifying that the agreement between the two determinations is so good. In fact, it may be that one can determine the moments of the stable isotopes by using our directly determined value and proceeding backwards through the calculations in Sec. 5.

The main source of the uncertainty in our result arises from the statistical counting error of the direct moment determination. Measurements in a stronger magnetic field are now planned and should reduce the error somewhat. It appears, however, that the uncertain nature of the residual attenuation, the difficulty of measuring τ_N with great precision and uncertainty concerning the nature of the ionic state during the initial fraction of a nanosecond after beta decay will make it difficult to reduce the error much below ten percent. This is true despite the fact that the conditions of this experiment are probably more thoroughly investigated than for previous measurements of this type. It is obvious that the usual procedure of applying corrections for perturbed angular correlations cannot be employed without detailed study of each case, at least in the case of paramagnetic ions. The incidental observation that at least in liquids, the ground state of the *daughter* ion is reached after beta decay in less than one nanosecond may be of general interest. The technique of obtaining short electronic relaxation times by the use of ionic melts may also be of general applicability. Another incidental result is the fact that the unperturbed angular correlations of the $2-\rightarrow2+\rightarrow0+$ cascades in Gd^{154} and Sm^{152} are nearly identical. Ofer[11] concluded that the Sm^{152} correlation was best fitted with a $\frac{1}{4}\%$ admixture of $M2$ radiation to the $E1$ transition. Our value for Gd^{154} indicates an admixture of the same order of magnitude.

We note that our result for the field-free attenuation in Sec. 3 is not in complete agreement with that of Goldring and Scharenberg[3] who used Coulomb excitation to produce the 2+ state, and found an attenuation corresponding nearly to the free hfs case for Gd^{154}.

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APPENDIX I

The attenuation of angular correlations by a free ionic hyperfine structure has been given explicitly by Alder.[10] We have carried out the numerical calculations for the case of $J=\frac{7}{2}$, $I=2$ appropriate to our case. Since the calculations are somewhat tedious we present the results in Figs. 7(a) and 8(a).

In the case of transverse magnetic decoupling (Sec. 5) the appropriate expansion of $W(\theta)$ is of the form

$$W(\theta)=1+\bar{G}_2B_2\cos2\theta+\bar{G}_4B_4\cos4\theta,$$

rather than in Legendre polynomials since the interaction now has cylindrical symmetry. The relation be-

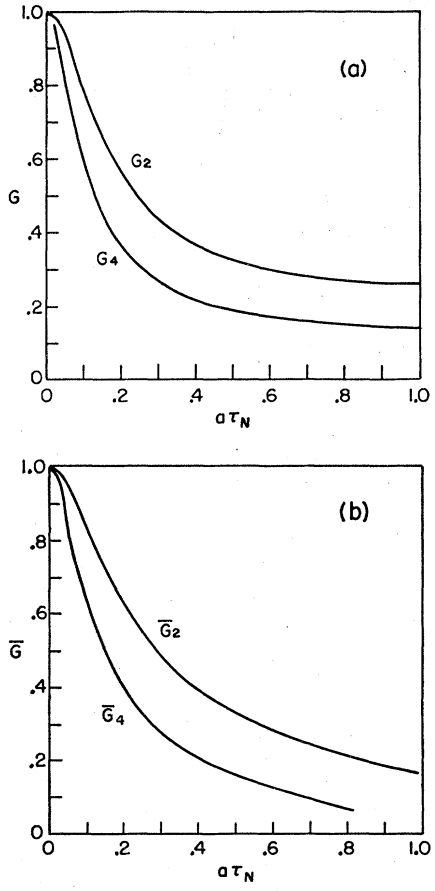

FIG. 7. Effect of magnetic hfs on the time-integrated angular correlation for $J=7/2$, $I=2$. (a): Without magnetic field. (b): Strong transverse magnetic field. The meaning of the symbols is explained in Appendix I.

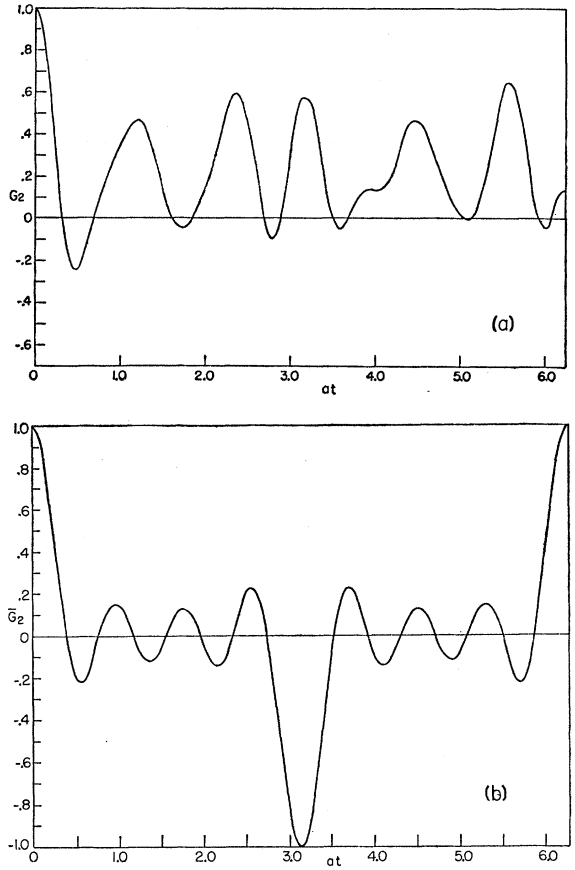

FIG. 8. Time dependence of the attenuation coefficients due to magnetic hfs interaction. (a): Without magnetic field. (b): Strong transverse field.

tween the two expansions is

$$B_2=\frac{48A_2+20A_4}{64+16A_2+9A_4},\quad B_4=\frac{35A_4}{64+16A_2+9A_4},$$

$$\tilde{G}_4=G_4\left(\frac{1+0.25A_2+0.141A_4}{1+0.25G_2A_2+0.141G_4A_4}\right).$$

If $A_4=0$, $B_2=3A_2/(4+A_2)$ and $\tilde{G}_2=G_2(4+A_2)/(4+G_2A_2)$.

$\tilde{G}_2$, $\tilde{G}_4$ are easily calculated for a purely magnetic hfs coupling with $J=\frac{7}{2}$ in a strong transverse field.

$$\tilde{G}_2(t)=\tfrac{1}{4}\sum_{\nu=0}^{3}\cos(2\nu+1)at,$$

$$\tilde{G}_4(t)=\tfrac{1}{4}\sum_{\nu=0}^{3}\cos(4\nu+2)at,$$

and the time integral values are

$$\tilde{G}_2=\tfrac{1}{4}\sum_{\nu=0}^{3}\frac{1}{1+[(2\nu+1)a\tau_N]^2},$$

$$\tilde{G}_4=\tfrac{1}{4}\sum_{\nu=0}^{3}\frac{1}{1+[(4\nu+2)a\tau_N]^2},$$

where the hfs coupling constant a is expressed in radians/sec. These expressions are plotted in Figs. 7(b) and 8(b). It should be kept in mind that the curves (a) and (b) in Figs. 7 and 8 are not strictly comparable because they correspond to different expansions of $W(\theta)$. We note that for the zero-field case, after the first rapid drop $G_2(t)$ oscillates around the "hard-core" value $G_2(\text{min})=0.22$. In the decoupled cases $\tilde{G}_2$ oscillates around zero and there is no "hard-core" limit. For strong hfs coupling (large $a\tau_N$), transverse decoupling will therefore always result in a reduction of the angular correlation. On the other hand, the initial slope of the attenuation curves is steeper in the field-free than in the decoupled case so that for $a\tau_N\lesssim 0.5$ the transverse field will cause an *increase* in the correlation.

The time dependence shown in Fig. 8 will, of course, lose its periodic character if the quadrupole hfs interaction is not negligible. In our analysis in Sec. 5 we have also tacitly assumed that there are no residual perturbations due to the surrounding medium.