

Nuclear Spins and Moments of the 86.5- and 105.3-keV States in Gd¹⁵⁵†

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The recoilless nuclear resonance absorption of the γ rays from the 86.5- and 105.3-keV states in Gd¹⁵⁵ has been studied to determine the spins, the quadrupole moments, and magnetic moments of these states. The following quantities were determined: $I(105.3) = \frac{3}{2}$, $|Q(86.5)/Q(0)| = 0.20 \pm 0.10$, $0.80 < Q(105.3)/Q(0) \lesssim 1.00$, $\mu(86.5) = -0.53 \pm 0.05 \mu_N$, and $\mu(105.3) = 0.13 \pm 0.04 \mu_N$ or $-0.38 \pm 0.06 \mu_N$. The results imply strong bandmixing in the 86.5-keV state and $I(86.5) = \frac{5}{2}$.

INTRODUCTION

THE lower energy states of the nucleus Gd¹⁵⁵ have been the object of numerous experiments of various kinds,¹⁻¹³ and it is well established that the nucleus is strongly deformed in the ground state ($I^\pi = \frac{3}{2}^-$). The lowest states of positive parity (86.5 and 105.3 keV) have not, however, been convincingly assigned as rotational bandheads; neither have the spins of these states been established with certainty. Harmatz *et al.*¹ proposed, on the basis of conversion electron measurements, the Nilsson configurations $|651 \frac{3}{2}\rangle_{I=3/2}$ and $|642 \frac{5}{2}\rangle_{I=5/2}$ for 86.5 and 105.3 keV levels. Unique assignment of the spins cannot be made from the γ -ray transition probabilities⁴ or the branching ratios,² but most authors have preferred the assignment of Harmatz *et al.* The assumption of the spin $\frac{5}{2}$ for the 105.3 keV state was in disagreement, however, with the angular correlation experiment of Subba Rao.⁶ A measurement⁸ of the g factor of the 86.5 keV state

using the spin rotation technique has been interpreted assuming the state to be the configuration $|651 \frac{3}{2}\rangle_{I=3/2}$ with a small admixture of the configuration $|660 \frac{1}{2}\rangle$ caused by Coriolis coupling, but the assignment $|642 \frac{5}{2}\rangle_{I=5/2}$ could not be excluded. Several Mössbauer experiments⁹⁻¹³ on the 86.5 keV transition have recently been reported. The primary data from these experiments cannot be consistently interpreted and the spins $\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$ have all been suggested. Extensive bandmixing involving several Nilsson configurations was very recently proposed¹⁴⁻¹⁶ for the 86.5 and 105.3 keV states.

In the present paper we report on Mössbauer experiments in which the quadrupole and magnetic hyperfine interactions of the 86.5 and 105.3 keV states have been studied. The values derived for the quadrupole and magnetic moments indicate a very strong bandmixing in the 86.5 keV state and provide strong evidence for spins $\frac{5}{2}$ and $\frac{3}{2}$ for the 86.5 and 105.3 keV states. The lowest excited state (60 keV) has also been studied, but due to its short lifetime the hyperfine interactions could not be measured.

EXPERIMENT

The nuclear hyperfine interactions have been studied in Mössbauer absorption experiments. Because the recoil-free fractions are small, source and absorbers have in most cases been kept at a temperature of 4.2°K to obtain as large an effect as possible. The source was Eu¹⁵⁵ in a host matrix of Sm₂O₃. The most accurate results for the quadrupole moments have been obtained from the hyperfine splitting in the source. Absorbers of (Gd_{0.05}Y_{0.95})Al₂ and GdAl₂ were used for these measurements since they provide rather narrow absorption lines and, for practical purposes, approximate unsplit absorbers. The magnetic hyperfine interactions in GdFe₂, used as an absorber, provided the most accurate determination of the magnetic moments.

A constant acceleration velocity drive was used; the apparatus and its performance has been described previously.¹⁷ A Ge(Li) detector with thin windows was

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¹ B. Harmatz, T. H. Handley, and J. W. Mihelich, *Phys. Rev.* **128**, 1186 (1962).

² A. Krusche, D. Bloess, and F. Münnich, *Z. Physik* **192**, 490 (1966).

³ R. E. McAdams and E. N. Hatch, *Nucl. Phys.* **82**, 372 (1966).

⁴ E. N. Hatch and F. Boehm, *Z. Physik* **155**, 609 (1959).

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⁶ R. N. Subba Rao, *Nucl. Phys.* **28**, 503 (1961).

⁷ P. H. Blichert-Toft, E. G. Funk, and J. W. Mihelich, *Nucl. Phys.* **A96**, 190 (1967).

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⁹ R. R. Stevens, Jr., Y. K. Lee, and J. C. Walker, *Phys. Letters* **21**, 401 (1966).

¹⁰ A. E. Balabanov, N. N. Delyagin, and Hussein el Sayes, *Yadern. Fiz.* **3**, 209 (1966) [English transl.: *Soviet J. Nucl. Phys.* **3**, 150 (1966)].

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¹² F. F. Tomblin and P. H. Barret, in International Conference on Hyperfine Interactions Detected by Nuclear Radiation, Asilomar, California, 1967 (unpublished).

¹³ H. Blumberg, B. Persson, and M. Bent, in International Conference on Hyperfine Interactions Detected by Nuclear Radiation, Asilomar, California, 1967 (unpublished).

¹⁴ J. Kormicki, H. Niewodniczanski, Z. Stachura, K. Zuber, and A. Budziak, *Nucl. Phys.* **A102**, 253 (1967).

¹⁵ M. E. Bunker and C. W. Reich, *Phys. Letters* **25B**, 396 (1967).

¹⁶ M. Jaskola, P. O. Tjöm, and B. Elbek (to be published).

¹⁷ D. Agresti, E. Kankeleit, and B. Persson, *Phys. Rev.* **155**, 1342 (1967).

used to detect the γ rays. The resolution of about 3 keV ensured a negligibly small background in the analyzer window when set on the 86.5 and 105.3 keV peaks.

The 10-mCi source was prepared from carrier-free $Eu^{155} Cl_3$ in HCl solution by adding $SmCl_3$ as carrier and precipitating with oxalic acid. The precipitated oxalate was converted into oxide by heating in oxygen at 800–900°C to get the cubic C-type Sm_2O_3 structure. In this oxide the samarium ions are situated in two non-equivalent lattice sites. One type of site has C_2 symmetry and is populated by $\frac{3}{4}$ of the samarium ions; the other type of site has C_{8i} symmetry. Random distribution of the europium impurity over the samarium sites cannot be assumed *a priori* because of dissimilarity of the europium and samarium ions.

The intermetallic compound YAl_2 has cubic symmetry at the Y site and is nonmagnetic. Because of its nearly identical properties, Gd substitutes easily for Y in this compound and with sufficiently small Gd concentration the ternary compound $(GdY)Al_2$ retains the nonmagnetic properties of YAl_2 at 4.2°K. Such a ternary compound, therefore, provides a single absorption line. In the present experiment $(Gd_{0.05}Y_{0.95})Al_2$ [hereafter denoted by $(GdY)Al_2$] has been used. The compound was prepared by heating a pellet of fine powders in the stoichiometric composition in an evacuated quartz tube. The temperature was gradually raised to 1000°C to allow for a few days of diffusion and slow reaction; thereafter the pellet was melted in argon using the levitational melting technique. It was finally annealed in vacuum at 800°C for several days.

The intermetallic compounds $GdFe_2$ and $GdAl_2$ have cubic point symmetry at the Gd site. The internal fields at the Gd nucleus have been measured by NMR¹⁸ to be 430 and -163 kG, respectively, at 4.2°K. The $GdFe_2$ lattice provides the largest known internal field at the Gd nucleus; thus this compound is the best available for the study of the magnetic hyperfine interactions. The internal field in $GdAl_2$ is rather small; therefore this compound may, in some cases, replace a true single line absorber. The $GdFe_2$ was prepared by pressing the stoichiometric amount of high-purity gadolinium metal into a cavity in a pellet of ultrapure iron. By the use of a high-frequency field, the pellet was levitated on a watercooled silver boat and melted in an argon atmosphere. The material was kept liquid for some time to homogenize it. Finally, the material was kept at 810°C in an evacuated quartz tube for 14 days to form a pure phase of $GdFe_2$.¹⁹ The compound $GdAl_2$ was prepared in a similar way, but the final annealing was not necessary.

Upper limits for impurity phases present in the intermetallic compound has been determined to be $<5\%$ from x-ray diffraction patterns. The absorbers for the

¹⁸ R. E. Gegenwarth, J. I. Budnick, S. Skalski, and J. H. Wernick, *Phys. Rev. Letters* **18**, 9 (1967).

¹⁹ M. Mansmann and W. E. Wallace, *J. Chem. Phys.* **40**, 1167 (1964).

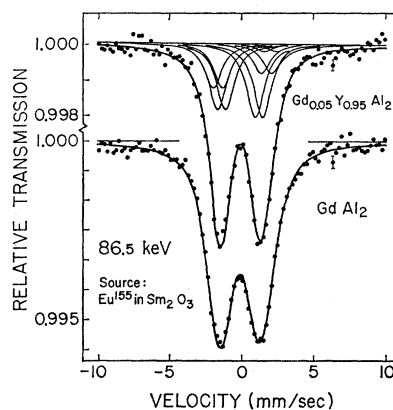


FIG. 1. Velocity spectra for the 86.5 keV transition in Gd^{155} obtained at 4.2°K with a Sm_2O_3 source matrix and absorbers of $Gd_{0.05}Y_{0.95}Al_2$ (8 mg Gd^{155}/cm^2) and $GdAl_2$ (14 mg Gd^{155}/cm^2). The curves of Figs. 1 and 2 result from one of many equally likely fits obtained by fitting the pairs of velocity spectra for the 86.5 and 105.3 keV transitions in a single least-squares procedure. The relative contributions from the two types of lattice sites assumed in this fit were those that gave the smallest linewidth for the components of the 86.5 keV spectrum and also accounted for the difference in the average electric field gradients observed in the 86.5 and 105.3 keV spectra. With $I = \frac{5}{2}$ and the two sites contributing in equal amounts for the 86.5 keV state, and $I = \frac{3}{2}$, $Q(105.3)/Q(0) = 0.95$, and the two sites contributing in the ratio 3:1 for the 105.3 keV state, and $\eta = 0$ the following quantities were determined: $eQ(0)V_{zz} = 1.54 \pm 0.06$, 2.31 ± 0.04 μeV ; $Q(86.5)/Q(0) = 0.25 \pm 0.02$; and the widths of the component Lorentzians in the 86.5 keV spectra 1.46 ± 0.05 for $(GdY)Al_2$, 1.61 ± 0.05 for $GdAl_2$; and in the 105.3 keV spectra 1.04 ± 0.13 for $(GdY)Al_2$, 1.21 ± 0.07 for $GdAl_2$ (widths in mm/sec). The magnetic interaction in the $GdAl_2$ absorbers was taken into account with $H_{int} = -163$ kG and the values of the magnetic moments given in the text. For this fit $\chi^2 = 1.09$ and $\Delta\chi^2 = 0.07$.

Mössbauer experiments were made from finely crushed material dispersed in wax.

ANALYSIS AND RESULTS

Quadrupole Interaction

Velocity spectra for the 86.5 keV transition in Gd^{155} obtained with absorbers of $(GdY)Al_2$ and $GdAl_2$ are shown in Fig. 1. The spectra are symmetric and show two absorption dips separated about 2.8 mm/sec. This structure is attributed to quadrupole interaction in the source for reasons that follow.

Identical spectra were observed with $(GdY)Al_2$ absorbers at about 1.5, 4.2, and 77°K, which indicates that magnetic ordering of the absorber material does not occur in this temperature range. The data obtained at the highest temperature, however, did not allow detailed examination because of poor statistics. Spectra observed with absorbers of $GdAs$ and $GdAl_2$ at 4.2°K are similar to the spectra obtained with the $(GdY)Al_2$ absorber. The poorer resolution of the two absorption dips is probably caused by magnetic interaction in these materials. In fact, the spectra measured with absorbers of $(GdY)Al_2$ and $GdAl_2$ (Fig. 1) can be consistently interpreted only if the internal field is much smaller in $(GdY)Al_2$ than in $GdAl_2$. We therefore conclude that

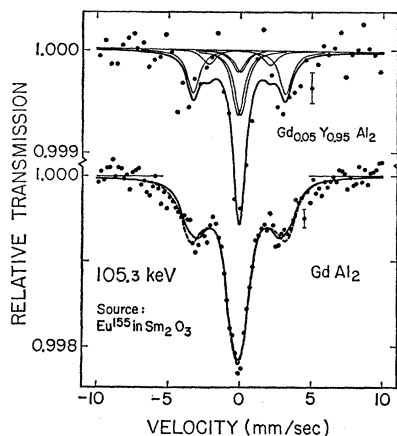


FIG. 2. Velocity spectra for the 105.3 keV transition in Gd^{155} obtained at 4.2°K with a Sm_2O_3 source matrix and absorbers of $Gd_{0.05}Y_{0.95}Al_2$ (16 mg Gd^{155}/cm^2) and $GdAl_2$ (33 mg Gd^{155}/cm^2). The solid curves represent the fit to the experimental points described in the caption to Fig. 1. In this fit the ratio $Q(105.3)/Q(0)$ was constrained to equal 0.95, since only ratios ≤ 1.00 are expected (see text). The dashed curve is the result of a similar fit with the constraint on the ratio of the quadrupole moments relaxed, yielding $Q(105.3)/Q(0) = 1.16 \pm 0.04$.

the spectrum observed with this absorber—its gross structure, at least—is caused by interactions in the source.

Mössbauer spectra obtained with Gd_2O_3 and $(Gd_{0.04}Y_{0.96})_2O_3$ absorbers indicate that the absorption spectra of these materials are very similar to the emission spectrum of the source. This is expected if the hyperfine splittings in these materials are caused by quadrupole interaction. The reason is that the trivalent gadolinium ion has a half-filled $4f$ shell, so the free ion is spherically symmetric ($^8S_{7/2}$). Therefore, the polarization of the Gd ion by the lattice is the sole origin of the electric field gradient at the Gd nucleus. Since the lattice parameters of the investigated sesqui-oxides are nearly equal, the electric field gradients in these compounds should give similar quadrupole splittings. Also, the temperature dependence²⁰ of the quadrupole interaction should be small, as observed, since it is caused only by the lattice expansion. Significant population of electronic configurations other than the ground state are not expected.²¹

Furthermore, the quadrupole splitting of the ground state of Gd^{155} in these oxides can be estimated from the lattice contribution to the splitting of Tm^{169} in Tm_2O_3 .²² With the known quadrupole moments²³ of the states involved, neglecting the small differences of the lattice constants and their temperature dependence, the esti-

mate is 2.1 mm/sec. This value agrees as well as expected with the observed separation of the two absorption dips in Fig. 1. The presence of magnetic interaction of significant magnitude, as has been found for Dy_2O_3 ,²⁴ would have been revealed by an asymmetric spectrum. It should be noted that the Dy_2O_3 is a unique case²⁵ and that magnetic interaction should not be expected generally in the rare-earth sesqui-oxides.

The observed two-dip quadrupole spectrum (cf. Fig. 1) immediately suggests that the quadrupole moment of the 86.5 keV state, $Q(86.5)$, is much smaller than that of the ground state, $Q(0)$. A precise evaluation of the ratio $Q(86.5)/Q(0)$, however, depends on the spin of the 86.5 keV state and on the relative number of γ rays emitted without recoil by the Gd nuclei in the two types of lattice sites in Sm_2O_3 . The experimental data (Fig. 1) have been least-square fitted to superpositions of Lorentzian curves of equal width. We have tried to obtain fits for various combinations of the relevant parameters in order to determine the maximum and minimum values possible for the ratio $Q(86.5)/Q(0)$. Such parameters are the widths of the Lorentzians, the relative contributions of recoil-free γ rays from the two sites, the ground-state splittings and isomeric shifts for the two sites, and the asymmetry of the field gradient. For a large number of fits we cannot give preference for any single one, judging by the quality of the fit and the width of the Lorentzians. Both $I = \frac{3}{2}$ and $I = \frac{5}{2}$ give $1.16 \leq \chi^2 \leq 1.28$ ($\Delta\chi^2 = 0.14$) and the width 1.65 ± 0.1 mm/sec. From the half-life 6.4 nsec² a width ≈ 0.51 mm/sec is expected, which indicates that the nuclear hyperfine interactions have not been accounted for completely. The result for the ratio of the quadrupole moments is $|Q(86.5)/Q(0)| = 0.20 \pm 0.10$. Our experimental data agree essentially with that of Stevens *et al.*⁹ but disagree with the results of the experiments of Balabanov *et al.*¹⁰ and Tomblin *et al.*¹²

Velocity spectra for the 105.3 keV transition in Gd^{155} observed with absorbers of $(GdY)Al_2$ and $GdAl_2$ are shown in Fig. 2. Because of strong photoabsorption, the effect obtained with the $(GdY)Al_2$ absorber is small and the measured spectrum suffers from poor statistics. A larger effect was obtained with the absorber of $GdAl_2$. The magnetic interaction in this material complicates the analysis. However, accurate correction for the magnetic splitting in the absorber can be performed. Also, the magnetic splitting is rather small and its effect on the spectrum, shown in Fig. 2, is to broaden the component lines.

Qualitatively, the appearance of the spectra (cf. Fig. 2) for the quadrupole interaction of the 105.3 keV state is as one would expect for a $\frac{3}{2}^+ \rightarrow \frac{3}{2}^-$ transition, if the quadrupole moment of the excited state is nearly equal to the quadrupole moment of the ground state. The spectra cannot be fitted under the assumption of spin

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²¹ B. G. Wybourne, Phys. Rev. **148**, 317 (1966).

²² R. G. Barnes, R. L. Mössbauer, E. Kankeleit, and J. M. Poindexter, Phys. Rev. **136**, A175 (1964).

²³ *Nuclear Data Sheets*, compiled by K. Way *et al.* (U. S. Government Printing Office, National Academy of Sciences-National Research Council, Washington, D.C., 1965), Appendix 1, compiled by G. H. Fuller and V. W. Cohen.

²⁴ S. Ofer, B. Khurgin, M. Rakavy, and I. Nowik, Phys. Letters **11**, 205 (1964).

²⁵ I. Nowik, Phys. Letters **15**, 219 (1965).

$\frac{5}{2}$ for the excited state. Therefore, we conclude that the spin of the 105.3 keV state is $\frac{3}{2}$.

In analogy with the procedure for the 86.5 keV transition, we have determined the maximum and minimum values of the ratio $Q(105.5)/Q(0)$ that are consistent with the experimental spectra in Fig. 2. Again, many equally likely fits ($1.01 \leq \chi^2 \leq 1.22$, $\Delta\chi^2 = 0.10$) were obtained for various combinations of parameter values. For the ratio of the quadrupole moments, the absolute limits $0.80 < Q(105.3)/Q(0) < 1.20$ were found. Widths of the component Lorentzians in the range 0.64–1.23 mm/sec for GdYAl₂ and in the range 1.16–1.55 mm/sec for GdAl₂ were obtained in these fits, whereas one would expect widths $\geq 2\Gamma = 2.3$ mm/sec.²

The Mössbauer effect in the 60.0 keV transition has been studied at 4.2°K with absorbers of Gd₂O₃ (5 and 12 mg Gd¹⁵⁵/cm²). Widths of about 30 mm/sec were observed in these measurements, consistent with the electronically measured² lifetime ($T_{1/2} = 0.24 \pm 0.06$ nsec, $2\Gamma = 19 \pm 5$ mm/sec). Because of the short lifetime, hyperfine interactions have little effect on the width. In disagreement with these results, a much smaller value for the width (1.4 ± 0.4 mm/sec) has been reported by Balabanov *et al.*¹⁰

Anomalies in the Quadrupole Spectra

The quadrupole hyperfine splitting of the ground state in Gd¹⁵⁵ in a host matrix of Sm₂O₃ can be determined both from the data for the 86.5 keV transition and from that for the 105.3 keV transition. With the assumption that the ratio of the recoil-free fractions for the two types of lattice sites is the same for both transitions, it is found that the splittings derived from the two sets of data differ by about 20%. Imposing the constraint $Q(105.3)/Q(0) \leq 1.00$ (cf. Discussion) increases this discrepancy to 40%. The discrepancy is removed if constraint on the recoil-free fractions is relaxed. Different Debye temperatures for the two sites can then lead to unequal relative contributions from the two sites at the two energies. Reasonably good fits are then obtained with the constraint $Q(105.3)/Q(0) \leq 1$, although the quality of the fit still favors $Q(105.3)/Q(0) > 1$.

The experimental spectra cannot be explained only in terms of superposition of emission spectra from two nonequivalent sites. The following three points need to be considered. (i) The widths (≤ 1.40 mm/sec; 10% thickness broadening is assumed) of the component Lorentzians of the spectra obtained for the 105.3 keV transition are significantly smaller than $2\Gamma = 2.43 \pm 0.06$ mm/sec. The value for the natural width Γ has been calculated from lifetimes measured with electronic methods.^{2,3} It seems unlikely that this value is impaired by any serious error. (ii) The widths (1.50 ± 0.10 mm/sec, assuming 10% thickness broadening) of the component Lorentzians of the spectra obtained for the 86.5 keV transition are considerably larger than expected. Lifetime measurements^{2,3} give $2\Gamma = 0.51$

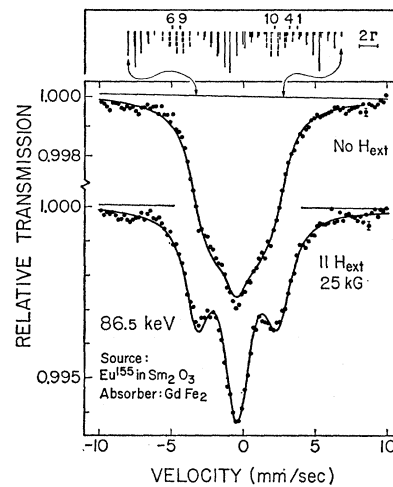


FIG. 3. Velocity spectra for the 86.5 keV transition in Gd¹⁵⁵ obtained at 4.2°K with a Sm₂O₃ source matrix and a GdFe₂ absorber (14 mg Gd¹⁵⁵/cm²) magnetized in the direction of the γ -ray beam (lower diagram) and demagnetized (upper diagram). The curves are synthesized spectra corresponding to a fit to the data under one of the many possible assumptions for the line structure of the source [in the fit shown $eQ(0)V_{zz} = 1.88$ μ eV, $Q(86.5)/Q(0) = 0.21$]. With the NMR value from Ref. 18 for the magnetic splitting of the ground state ($g_{g.s.}^{155}\mu_N H_{int}c/E_\gamma = 0.794$ mm/sec) the following quantities were determined: $\mu(86.5) = 0.520 \pm 0.05$ μ_N , the width of the component Lorentzians = 1.97 ± 0.04 mm/sec, the center shift = -0.41 ± 0.01 mm/sec, and $\chi^2 = 1.73$, where $\Delta\chi^2 = 0.10$. The values of these quantities are insensitive to the particular choice of line structure for the source, provided it fits the source spectrum well. This is demonstrated by the diagram at the top of the figure, where the relative positions of the lines of the emission spectrum and the observed spectrum are shown. The line positions—the scale is arbitrary—and intensities apply for the case of a nonmagnetized absorber. With it magnetized in the direction of the γ -ray beam the dashed lines are not observed. It is evident from a comparison of the separation of the individual lines with 2Γ that only the gross structure of the spectrum can be observed, i.e., five (demagnetized) or three (aligned) absorption dips. Their relative positions and intensities depend on the gross structure of the source spectrum, but only very slightly on its detailed line structure.

mm/sec. It is emphasized that the same source and the same absorber material has been used for the measurements referred to here and in (i). (iii) The average electric field gradient observed with the 86.5 keV transition is smaller than the electric field gradient observed with the 105.3 keV transition.

As possible explanations for these discrepancies one might suggest effects of local disturbances of the surroundings of the decayed europium nucleus. These disturbances must then last a few nanoseconds. A time-dependent Debye-Waller factor caused by local-mode excitation, or large hyperfine fields caused by disturbances of the electron shell, would modify the Lorentzian line shape by reducing the intensity in the wings and narrowing the width.^{26,27} Such effects would explain (i), the narrow width observed for the 105.3

²⁶ S. M. Harris, Phys. Rev. **124**, 1178 (1961); **163**, 280 (1967); J. G. Dash and R. H. Nussbaum, Phys. Rev. Letters **16**, 567 (1966).

²⁷ W. Trifflhäuser and P. P. Craig, Phys. Rev. **162**, 274 (1967).

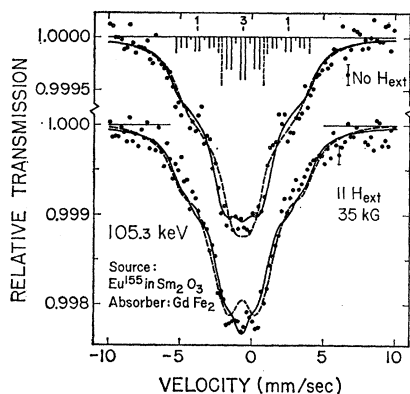


FIG. 4. Velocity spectra for the 105.3 keV transition in Gd^{156} obtained at 4.2°K with a Sm_2O_3 source matrix and a $GdFe_2$ absorber (84 mg Gd^{156}/cm^2) magnetized in the direction of the γ -ray beam (lower diagram) and demagnetized (upper diagram). The curves are synthesized spectra corresponding to a fit to the data under one of the many possible assumptions for the line structure of the source [$eQ(0)\bar{V}_{zz} = 2.22 \mu eV$, $Q(105.3)/Q(0) = 1.00$]. With the NMR value from Ref. 18 for the magnetic splitting of the ground state ($g_{g.s.}^{156}\mu_N H_{int}/E_\gamma = 0.652$ mm/sec) there are two sets of parameters that fit the data. The corresponding calculated spectra are shown as solid and dashed curves, and the following parameters were determined from the fits: $\mu(105.3) = 0.128 \pm 0.009$, $-0.386 \pm 0.016 \mu_N$, the width of the component Lorentzians $= 1.63 \pm 0.13$, 2.12 ± 0.14 mm/sec, the center shift $= -0.63 \pm 0.03$, -0.63 ± 0.03 mm/sec, and $\chi^2 = 1.47$, 1.65 where, $\Delta\chi^2 = 0.10$. The values of these quantities are rather insensitive to the particular line structure chosen to fit the source spectrum. The line structure assumed for the given fit is indicated at the top of the figure; the dashed lines are not observed when the absorber is magnetized parallel to the γ ray-beam.

keV state. Also, the finer details of the spectra in Figs. 2 and 4 would be better fitted with the more triangle-like peak shape.

As another consequence of the suggested time dependence of the surroundings, the observed fraction of γ rays emitted without recoil should be dependent on the elapsed time between the β decay and the formation of the state in question and also on the lifetime of the state. Due to the short lifetime of the 60.0 keV state, no recoil-free emission in this state should be observed when it is fed directly by the β decay. However, this state is populated to 30% by a γ transition from the long-lived 86.5 keV state, so the time spent in the 86.5 keV state allows stabilization of the surroundings. Recoil-free emission in the 60.0 keV state is therefore observed. The observed fraction of γ rays emitted without recoil from the three states investigated are all smaller than expected for a Debye temperature of Sm_2O_3 in the range 200–250°K.²⁸ Also, the energy dependence of the fraction of photon emitted without recoil is quite abnormal. The effect observed for the 60.0 and 105.3 keV transitions are much smaller than expected for these energies from the observed recoil-free fraction for the 86.5 keV transition and a regular energy dependence of the Debye-Waller factor. These observations support the suggestion of a time-dependent surrounding.

²⁸ F. W. Stanek, *Z. Physik* **166**, 6 (1962).

Slow recovery of the electric field gradient at the gadolinium nucleus to the stationary value could be the cause for (ii), the broadening of the 86.5 keV line, as well as for (iii), the differences in the field gradients. A distribution of field gradients would broaden the side peaks of the 105.3 keV spectra, without affecting the center peak. The symmetry of the spectra shows that no long-lived charge states²⁷ but the stationary ones are observed, since, otherwise, the accompanying isomer shifts would have been revealed by asymmetric hyperfine spectra.

Magnetic Interaction

The magnetic hyperfine interactions at 4.2°K in $GdFe_2$ have been studied, using this ferrimagnetic material as absorber. Velocity spectra (Figs. 3 and 4) have been measured with the absorber demagnetized as well as with it magnetized in the direction parallel to the γ -ray beam. In the latter case a superconducting solenoid was used to align the absorber. The magnetic interaction has considerable effect on the spectra observed for the 86.5 keV transition (cf. Figs. 1 and 3) and the shapes of the spectra are much different for the two field configurations. Accurate determination of the magnetic moment of the 86.5 keV state should therefore be possible. The magnetic interactions affects the spectra for the 105.3 keV transition less extensively (cf. Figs. 2 and 4).

The velocity spectra correspond to foldings of a quadrupole emission spectrum and a magnetic absorption spectrum. For the least-squares analysis of the data, the measured spectra were assumed to be superpositions of Lorentzians of equal width. The spins were assumed $\frac{5}{2}$ and $\frac{3}{2}$ for the 86.5 and 105.3 keV states, respectively (cf. Discussion). Since a unique determination of the line structure of the emission spectrum was not possible, various possible line structures were tried. These were the results of numerous fits to the quadrupole spectrum of the source. Fortunately, the values determined for the magnetic moment of the excited state proved to be quite insensitive to the particular assumption of the line structure of the emission spectrum. On the other hand, there was strong correlation among the parameters determining the magnetic hyperfine spectrum. For the determination of the magnetic moments of the excited states we have therefore used the NMR value,¹⁸ 430 kG, for the internal field and the value $-0.254 \pm 0.003 \mu_N$ for the magnetic moment (μ_{155}) of the ground state. The latter value was evaluated from the ground-state magnetic moment²⁹ of Gd^{157} , $\mu_{157} = 0.337 \pm 0.003 \mu_N$, and the ratio $\mu_{155}/\mu_{157} = 0.753 \pm 0.005$, measured with ENDOR technique.³⁰

²⁹ J. P. Hurrell, *Brit. J. Appl. Phys.* **16**, 755 (1965).

³⁰ E. L. Boyd and R. J. Gambino, *Phys. Rev. Letters* **12**, 20 (1964).

TABLE I. Results from Mössbauer experiments in Gd¹⁵⁵.

E (keV)	I	$Q(E)/Q(0)=R$	$\mu(E)^a$ (μ_N)	$ eQ(0)V_{zz} _{av}^b$ (10^{-6} eV)	Width ^c W (mm/sec)	$2\Gamma^d$ (mm/sec)
86.5	$\frac{5}{2}$	$+0.20 \pm 0.10$ -0.20 ± 0.10	-0.53 ± 0.05	1.8 ± 0.1 1.5 ± 0.1	1.65 ± 0.10	0.514 ± 0.007
105.3	$\frac{3}{2}$	$0.80 < R \lesssim 1.00$	0.13 ± 0.04 -0.38 ± 0.06	2.2 ± 0.2	$0.64 < W \pm$ $0.10 < 1.55$	2.43 ± 0.06

^a Evaluated using the NMR value (430 kG) of the internal field in GdFe₂ (Ref. 18).
^b Derived assuming one unique value of the electric field gradient.

^c Not corrected for thickness broadening.
^d Calculated using electronically measured lifetimes (Refs. 2 and 3).

As the result of the experiments, we give the following values for the magnetic moments:

$$\begin{aligned}\mu(86.5) &= -0.53 \pm 0.05 \mu_N, \\ \mu(105.3) &= +0.13 \pm 0.04 \quad \text{or} \quad -0.38 \pm 0.06 \mu_N.\end{aligned}$$

The errors given are conservative estimates, about equal to four times the spread in the values of the individual determinations. The spread was about equal to the standard deviations obtained for the fits. Perfect fits were not obtained; hence the increased values of the errors. The internal fields measured with the NMR technique might differ from those encountered in γ -resonance experiments.³¹ We therefore give the following relations:

$$\begin{aligned}\mu(86.5) &= -0.53 + 0.87 \times 10^{-3} [H_{\text{int}}(\text{kG}) - 430], \\ \mu(105.3) &= 0.13 - 0.73 \times 10^{-3} [H_{\text{int}}(\text{kG}) - 430],\end{aligned}$$

or

$$-0.38 + 1.37 \times 10^{-3} [H_{\text{int}}(\text{kG}) - 430],$$

which may serve for reevaluation of the magnetic moments if it proves that significantly modified internal fields are measured with the NMR technique.

[*Note added in proof.* The application of a magnetic field to GdFe₂, e.g., in alignment of the GdFe₂ absorber with the superconducting coil, induces a magnetic field of significant magnitude at the Gd nucleus. This effect, which will be the subject of a forthcoming paper, has been considered in the determination of the magnetic moments.]

Our results from the study of the magnetic interaction of the 86.5 keV state is in disagreement with the results of Delyagin *et al.*¹¹ On the other hand, the experimental data of Tomblin *et al.*¹² seem to be consistent with ours, since with the assumption of spin $\frac{3}{2}$ for the 86.5 keV state we would deduce a value of the magnetic moment in good agreement with their value.

Isomer Shifts

The velocity spectra measured in the present work are symmetric around a velocity that is only slightly different from zero (< 0.1 mm/sec), with the exception of the spectra obtained with Gd metal and GdFe₂ absorbers. In the latter case center shifts of -0.41 ± 0.02 and -0.63 ± 0.03 mm/sec were found for the 86.5 and

105.3 keV transitions, respectively. A unique interpretation of these centershifts in terms of isomer shifts is not possible without knowledge of the relative intensities of the γ rays emitted without recoil from the two sites in the Sm₂O₃ source matrix.

A summary of the properties of the 86.5 and 105.3 keV states in Gd¹⁵⁵ measured and deduced in the present work is given in Table I.

DISCUSSION

There are the following possibilities for the explanation of the small quadrupole moment of the 86.5 keV state:

(1) The spin of the state is $\frac{1}{2}$ and the quadrupole moment vanishes as tentatively suggested by Stevens *et al.*⁹ This contradicts, however, observation of an anisotropy in the angular-correlation experiments of Hryniewicz *et al.*^{5,8}

(2) The state has much smaller deformation than the ground state. This possibility is excluded, since it implies³² an isomeric shift 10–100 times larger than consistent with our observations.

(3) The state is a spin- $\frac{5}{2}$ rotational state of $K = \frac{3}{2}^+$ pure rotational band. The ratio $Q(86.5)/Q(0) = -5/14$ is then expected, but no $K = \frac{3}{2}^+$ bandhead has been observed.

(4) The state is mixed. With this assumption we can explain the experimental result. The quadrupole moment is then given by

$$Q(86.5) = Q_0 \sum_i c_K^2(i) \times [3K^2(i) - I(I+1)/(I+1)(2I+3)],$$

where $c_K(i)$ are the amplitudes of the various Nilsson configurations with K quantum number $K(i) < I$, and Q_0 is the intrinsic quadrupole moment. The regularity of the rotational band built on the ground state indicates that it is a very pure Nilsson state with $K = \frac{3}{2}$; hence, $Q(0) = \frac{1}{3}Q_0$.

³² This conclusion is based on a comparison of the center shift (-0.4 ± 0.1 mm/sec) measured with Gd-metal absorber and the isomer shift (0.26 ± 0.09 mm/sec) for the $0^+ - 2^+$ in Gd¹⁵⁶. The latter has been measured by D. Yeboah-Amankwah and L. Grodzins, Bull. Am. Phys. Soc. **12**, 580 (1967), with the same source and absorber matrices as used in the present experiment. Thus, the change in deformation is almost as small as deformation changes caused by centrifugal stretching.

³¹ R. L. Mössbauer (private communication).

The following constraints are imposed on the amplitudes c_K by the experimental result. If

$$I = \frac{3}{2}, \quad \text{then} \quad |p_{1/2} - p_{3/2}| = 0.20 \pm 0.10,$$

where

$$p_K = \sum_i c_K^2(i).$$

It is very unlikely, however, that both the 105.3 and the 86.5 keV states can have spin $\frac{3}{2}$, because of the small energy separation. Alternatively, if $I = \frac{5}{2}$, the amplitudes must obey the relation $|4p_{1/2} + p_{3/2} - 5p_{5/2}| = 0.5 \pm 0.3$. Indeed, this is the only acceptable way the present experimental results can be accounted for.

The magnetic moment of the 86.5 keV state determined in the present experiment, $\mu(86.5) = -0.53 \pm 0.05 \mu_N$ is in disagreement with the value $-0.94 \pm 0.07 \mu_N$ obtained from the g factor (-0.376 ± 0.026) measured by Hryniewicz *et al.*⁸ Magnetic moments ≈ -0.31 and $\approx 0.35 \mu_N$ are expected for the pure Nilsson states $|642 \frac{5}{2}\rangle_{I=5/2}$ and $|651 \frac{3}{2}\rangle_{I=5/2}$. These values have been calculated using the wave functions of Rasey³³ with deformation³⁴ $\beta = 0.31$, the effective g factor³⁵ $g_R = 0.32$, and an effective value of the intrinsic g factor of the odd neutron equal to 0.6 times the free-neutron value. The measured magnetic moment deviates from either of the values expected for the pure Nilsson states, and the strong bandmixing implied by the small quadrupole moment is also required for the explanation of the magnetic moment. In fact, both the quadrupole and the magnetic moments are consistent with the wave function

$$\psi_{I=5/2} = 0.47 |642 \frac{5}{2}\rangle_{I=5/2} + 0.88 |651 \frac{3}{2}\rangle_{I=5/2}$$

for the 86.5 keV state, although several other states are expected to contribute to $\psi_{5/2}$.

The spin $\frac{3}{2}$ for the 105.3 keV state has been determined from the appearance of the quadrupole spectrum. This spin value has already been concluded, however erroneously,³⁶ as the result of the γ - γ angular-

correlation experiments of Subba Rao.⁶ The quadrupole moment of the 105.3 keV state cannot be significantly larger than that of the ground state, since both states have spin $\frac{3}{2}$ and the ground state is a very pure Nilsson state. We can therefore reduce the experimental upper limit of the measured ratio of the quadrupole moments, i.e., $0.80 < Q(105.3)/Q(0) \leq 1.00$. The presently determined ratio $Q(105.3)/Q(0)$, which implies $p_{1/2}(I = \frac{3}{2}) < 0.10$, is not sufficiently accurate to allow one to determine whether the 105.3 keV state is a reasonably pure $|651 \frac{3}{2}\rangle_{I=3/2}$ Nilsson state or whether other states are admixed in significant amounts. On the other hand, the experimental values of the magnetic moment differ from the magnetic moment ($-0.15 \mu_N$) expected for a pure $|651 \frac{3}{2}\rangle_{I=3/2}$ state, indicating mixing in the 105.3 keV state also.

Similar conclusions regarding the mixing of the 86.5 and 105.3 keV states have recently been reached by Kormicki *et al.*¹⁴ and Bunker *et al.*¹⁵ on the basis of the level scheme and by Jaskola *et al.*¹⁶ from studies of (d, p) and (d, t) reactions. These authors have suggested contributions from the Nilsson states $|642 \frac{5}{2}\rangle$, $|651 \frac{3}{2}\rangle$, $|660 \frac{1}{2}\rangle$, $|402 \frac{3}{2}\rangle$, and $|400 \frac{1}{2}\rangle$. The assignments proposed for the level at 118 keV in these papers¹⁴⁻¹⁶ are all different, as are the suggested^{14,15} positions of the $|660 \frac{1}{2}\rangle_{I=1/2}$ state. The fit to the level scheme proposed by Bunker *et al.*¹⁵ gives amplitudes of the states mixing in the 86.5 keV level that are inconsistent with the measured nuclear moments. In particular, the ratio $Q(86.5)/Q(0) = 0.83$ is calculated for these amplitudes, while the experimental value is $\pm(0.20 \pm 0.10)$.

With the experimental information available at present, it is not possible to uniquely determine such a complex mixing in the 86.5 keV states and other as yet unassigned states. The spin of the 118 keV level, for example, would be a valuable additional piece of data that should be obtainable from an angular-correlation experiment.

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³³ A. J. Rasey, Phys. Rev. **109**, 949 (1958).

³⁴ B. Elbek, thesis, University of Copenhagen, 1963 (unpublished).

³⁵ F. Boehm, G. Goldring, G. B. Hagemann, G. D. Symons, and A. Tveter, Phys. Letters **22**, 627 (1966).

³⁶ The experiment of Subba Rao was repeated in our laboratory using a Ge(Li) detector for the detection of the 60.0 keV γ rays. We determined the angular-correlation coefficient $A_2 = 0.025 \pm 0.03$. This value disagrees with that of Subba Rao, $A_2 = 0.155 \pm 0.016$. Furthermore, in his work, the incorrect sign of the mixing ratio δ of the 60.0 keV transition has been used. With the correct sign of δ a nearly isotropic angular correlation is expected; the spin of the 105.3 keV state being $\frac{3}{2}$ or $\frac{5}{2}$. A γ - γ angular-correlation experiment is, therefore, not suited for the determination of the spin of the 105.3 keV state.