Hyperfine Interactions in the Ground State and 22-keV State of Sm^{149} in Ferrimagnetic Compounds of Samarium*

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Hyperfine interactions in $SmFe_2$, $SmFeO_3$, Sm metal, and particularly in samarium iron garnet, SmIG, have been studied at various temperatures using the Mössbauer effect in Sm^{149}. Large magnetic hyperfine interactions are observed in SmIG even at room temperature, in contrast with the observed Sm sublattice magnetization. A value of $(1.55\pm0.25)\times10^6$ Oe was found for the magnetic field acting on the Sm nucleus in samarium iron garnet at 20°K. For $SmFe_2$ an upper limit of 10^6 Oe was found for H_{eff} at 77°K. The experimental results support an assignment of a spin $\frac{5}{2}$ for the 22-keV level and yield a value of 1.26 ± 0.04 for the ratio of the magnetic moments of the 22-keV state and the ground state. A small isomeric shift -0.9 ± 0.3 mm/sec was found between Sm metal and Sm_2O_3 absorbers at 300° and 80°K. Upper limits for H_{eff} in $SmFeO_3$ were found to be 4×10^5 Oe 16°K, 1.9×10^5 Oe at 80°K, and 1.5×10^5 Oe at 300°K. The origin of the temperature dependence of the hyperfine fields acting on Sm nuclei in SmIG is discussed, including the effects of the molecular exchange field and the electrostatic crystalline field. Comparison is also made with the theory of magnetization of Sm sublattices. Exchange effects are found to be strong and dominant at high temperatures, whereas at low temperatures, crystalline-field effects are found to predominate. These results are in fair agreement with theoretical predictions if it is assumed that at low temperatures, H_{eff} is determined mainly by the splitting of the quartet Γ^8 ground state (which is the lowest of the two levels into which the $^6H_{5/2}$ ground state is split by a cubic electric field) by the exchange interaction.

I. INTRODUCTION

EVER since the early work of Van Vleck[1] and collaborators[2,3] on the magnetic properties of rare-earth ions, considerable interest has centered on understanding the behavior of Sm^{3+} and Eu^{3+} systems. For these two ions the splitting between the lowest J states ($\sim$1500°K in Sm^{3+}) is not much larger than room temperature, and as a consequence, a number of peculiar properties result.[4–7] In this paper, we describe results of experimental and theoretical investigations of hyperfine interactions in a number of samarium ferrimagnetic compounds, particualrly samarium iron garnet, SmIG.

A strong motivation for our work was the appeal of using the Mössbauer effect as a microscopic probe to study the unusual behavior of the magnetization of SmIG as observed by Pauthenet and others.[8] Since these authors found no observable difference between the magnetization of yttrium iron garnet (YIG) and SmIG, the sublattice contribution due to the Sm ions was considered to be negligible. Such a null result was neither expected nor understood on theoretical grounds. On the other hand, Caspari *et al.*[9] found, using the technique of γ-γ correlation, that the hyperfine field

* The two groups, OSN and BGFS, carried out independent theoretical and experimental studies; preliminary results of one of the groups have been reported. [Bull. Am. Phys. Soc. **9**, 226 (1964); **11**, 606 (1964).] The sources, absorbers, temperature ranges and procedures used by the two groups were very similar. Moreover, the results and conclusions, independently arrived at, coincided sufficiently to make desirable the joint pages which follow. It has been a parallel rather than a collaborative effort. Unless stated otherwise, the experimental results presented here have been obtained by both groups. For obvious reasons no attempt was made to combine independent results.

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acting on Sm nuclei as dilute impurities in europium iron garnet is substantial at room temperature, consistent with theoretical predictions which take into account the admixture of levels due to the exchange interaction, but neglecting crystalline field effects. However, as these authors pointed out,[9] it was not certain whether the measurements represented the hyperfine field due to the Sm^{3+} daughter or the Eu^{3+} mother. Recently, magnetization experiments by Geller *et al.*,[10] using single-crystal spheres, showed that each Sm^{3+} ion contributes a moment of 0.14 μ_B to the net magnetization[11] at 0°K and behaves at $T<298$°K similarly to Pr^{3+} and Nd^{3+} in garnets in that its moment *adds* to that of the resultant of the iron sublattice moments. Such a small moment, which is a good deal smaller than the calculated free ion moment, 0.7 μ_B may be understood to be a result of the combined effects of the crystalline electric fields and the molecular exchange fields.

The present work helps to elucidate some of these effects—particularly since the effects of the exchange field differ for the cases of magnetization and hyperfine field. Unlike the case of the magnetization measurements, which arrive at an estimate of the Sm sublattice magnetization by subtracting the magnetization of YIG from the measured magnetization of SmIG, our hyperfine measurements may be compared directly with theoretical predictions. Exchange effects are found to be strong and dominant at high temperatures, whereas, at low temperatures, crystalline field effects are found to predominate, in fair agreement with results of theoretical calculations based on the simple cubic crystal-field approximation.

In the following section (Sec. II), a simplified theoretical description of the origin and temperature dependence of the hyperfine fields acting on Sm nuclei (particularly in SmIG) is given. Comparison is made with the theory of magnetization of the Sm sublattices. Section III describes the experiments, and Sec. IV presents results for Sm_2O_3, SmIG, Sm metal, $SmFe_2$, and $SmFeO_3$. Finally, Sec. V compares the experimental results with theoretical predictions and presents some conclusions. We infer from the experimental results a spin of $\frac{5}{2}$ for the 22-keV excited state and a ratio of 1.26 ± 0.04 for the magnetic moments of the 22-keV state and the ground state.

II. HYPERFINE FIELDS ACTING ON Sm NUCLEI: SmIG

In free rare-earth ions, the magnetic hyperfine field H_{eff} arises from the orbital and spin angular momenta of the $4f$ electrons and from any contact interaction of s electrons penetrating into the nucleus. The first terms are, within the lowest J manifold, proportional to $\mathbf{J}$, and since contact terms induced by core polarization are small[12] for other than the ^8S state ions, Eu^{2+} and Gd^{3+}, H_{eff} is proportional to the magnetic moment of the level.[13]

In ferrimagnetic compounds of rare earths, H_{eff} at a rare-earth nucleus often has the same temperature dependence as $\mathbf{M}_{\text{RE}}$, the spontaneous magnetization of the rare-earth lattice.[14–17] For these materials, one must consider the role of electrostatic crystalline fields and the exchange fields from the iron sublattices. The correlation between H_{eff} and $\mathbf{M}_{\text{RE}}$ is expected to be valid for the second half of the rare-earth series because the spin-orbit interaction is much larger than the crystalline and exchange field interactions and because any mixing between J states is small. In the first half of the series, the mixing of J states may have a large effect on the values of $\mathbf{M}_{\text{RE}}$ and H_{eff}—as has been shown for EuIG, i.e., the Eu^{3+} ions exhibit paramagnetism[5] and have H_{eff} different from zero,[18] although the electronic ground state is $^7F_0$. Similarly, since the energy of the first excited state of the Sm^{3+} ion, $^6H_{7/2}$, is only 1100 cm^{-1} above the ground state, the mixing of the two states is expected to result in different temperature dependences for the hyperfine field and the sublattice magnetization.

Consider the effect of a molecular exchange field, $H_{\text{exch}}(T)$, on the observed H_{eff} and $\mathbf{M}_{\text{RE}}$ of Sm^{3+} ions, neglecting for the moment crystalline fields. We assume Russell-Saunders coupling, i.e., $\mathbf{L}$ and $\mathbf{S}$ remain good quantum numbers. The spin-orbit interaction splits the otherwise J degenerate free ion states into the usual fine structure multiplets characterized by the quantum numbers $LSJM$. The application of an exchange field now mixes the different J states. Remembering that H_{exch} acts only on the spin $\mathbf{S}$, unlike an external field which acts on $\mathbf{J}$, the weak field (Zeeman region) eigenfunctions and energies are from second-order perturbation theory and using the notation of Ref. 13,

$$\Psi_n=\Psi^0(SLJM)+2\mu_B H_{\text{ex}} \times\left\{\frac{\langle J+1\|\Lambda\|J\rangle[(J+1)^2-M^2]^{1/2}}{E_J{}^\circ-E_{J+1}{}^\circ}\Psi(SLJ+1M) +\frac{\langle J\|\Lambda\|J-1\rangle[J^2-M^2]^{1/2}}{E_J{}^\circ-E_{J-1}{}^\circ}\Psi(SLJ-1M)\right\}, \quad (1)$$

$$E_n=E_J{}^\circ+2\mu_B H_{\text{ex}}[(g_J-1)/(g_J)]M\langle J\|\Lambda\|J\rangle. \quad (2)$$

[9] S. Geller, H. J. Nillians, R. C. Sherwood, J. P. Remeika, and G. P. Espinosa, Phys. Rev. **131**, 1080 (1963).

[10] See, also the measurements reported by C. H. Nowlin [Scientific Report No. 7, Harvard University, Craft Laboratory, 1963 (unpublished)]; J. Perel and M. Schieber, J. Appl. Phys. (Japan) **1**, 243 (1962).

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[18] I. Nowik and S. Ofer, Phys. Rev. **132**, 241 (1963).

Assuming thermal equilibrium, the hyperfine field and magnetization are calculated from a Boltzmann average of the hyperfine operator[13] and μ_z, respectively. We follow the method of operator equivalents,[13] and using

$$\langle\Psi(SLJM)|H_{\mathrm{eff},z}{}^{op}|\Psi(SLJM)\rangle=2\mu_B M\langle r^{-3}\rangle\langle J\|N\|J\rangle,$$

$$\langle\Psi(SLJM)|H_{\mathrm{eff},z}{}^{op}|\Psi(SLJ-1M)\rangle=2\mu_B\langle r^{-3}\rangle[J^2-M^2]^{1/2}\langle J\|N\|J-1\rangle,$$

results in the following expressions for H_{eff} and M_{Sm} at high temperatures:

$$H_{\mathrm{eff}}=[6+8\exp(-E_{7/2}/kT)]^{-1}\mu_B{}^2 2\langle r^{-3}\rangle H_{\mathrm{exch}}\times\left\{-\frac{2440}{63kT}-\frac{3496}{105E_{7/2}}+\exp(-E_{7/2}/kT)\left(-\frac{222\,464}{14\,175kT}-\frac{109\,928}{2025(E_{9/2}-E_{7/2})}+\frac{3496}{105E_{7/2}}\right)\right\} \quad (3)$$

and

$$M_{\mathrm{Sm}}=[6+8\exp(-E_{7/2}/kT)]^{-1}\mu_B{}^2 H_{\mathrm{exch}}\times\left\{-\frac{50}{7kT}+\frac{240}{7E_{7/2}}+\exp(-E_{7/2}/kT)\left(-\frac{2288}{189kT}+\frac{1456}{27(E_{9/2}-E_{7/2})}-\frac{240}{7E_{7/2}}\right)\right\}. \quad (4)$$

$E_{7/2}$ and $E_{9/2}$ denote the energies of the unperturbed $J=\frac{7}{2}$ and $\frac{9}{2}$ states relative to the ground state. For completeness we have included the contributions of these states, although it is only at temperatures well above room temperature that these are appreciable.

From Eq. (4), it is readily seen that M_{Sm} changes sign[4] at a temperature for which $T\approx 5E_{7/2}/24k$, which is independent of H_{exch}. Such a transition point may thus be expected for many of the Sm ferrimagnetic compounds. The measurements of Geller *et al.*[10] suggest that the crossover temperature in SmIG is probably about 300°K, the temperature predicted by Eq. (4).

Unfortunately, no such transition point exists for H_{eff}, as its measurement would provide a striking verification for the simple theory of the effects of molecular exchange fields. At high temperatures, the mixing of J states by the exchange field does have a large effect on the temperature dependence of H_{eff}. However, at low temperatures ($T\approx 0$), the mixing of the $J=\frac{7}{2}$ excited state makes a small contribution to the hyperfine field—about 2% for $\mu_B H_{\mathrm{exch}}(0)/k=25$°K. Using a value[19] of 45×10^{24} cm^{-3} for $\langle r^{-3}\rangle$, the "free-ion" value of H_{eff} at 0°K is 3.3×10^6 Oe.

It is instructive to consider the hyperfine field and magnetization induced by an externally applied magnetic field H_0. To obtain the counterpart expressions to Eqs. (3) and (4), one replaces $2H_{\mathrm{exch}}((g_J-1)/g_J)$ by H_0 in all the diagonal matrix elements which enter the analysis given above. We obtain

$$H_{\mathrm{eff}}=[6+8\exp(-E_{7/2}/kT)]^{-1}\mu_B{}^2 2\langle r^{-3}\rangle H_{\mathrm{ext}}\times\left\{\frac{488}{63kT}-\frac{1748}{105E_{7/2}}+\exp(-E_{7/2}/kT)\left(\frac{525\,824}{14\,175kT}-\frac{54\,964}{2025(E_{9/2}-E_{7/2})}+\frac{1748}{105E_{7/2}}\right)\right\}, \quad (3a)$$

$$M_{\mathrm{Sm}}=[6+8\exp(-E_{7/2}/kT)]^{-1}\mu_B{}^2 H_{\mathrm{ext}}\times\left\{\frac{10}{7kT}+\frac{120}{7E_{7/2}}+\exp(-E_{7/2}/kT)\left(\frac{5408}{189kT}+\frac{728}{27(E_{9/2}-E_{7/2})}-\frac{120}{7E_{7/2}}\right)\right\}. \quad (4a)$$

By contrast with the results for a molecular exchange field, the external field produces no crossover in M_{Sm} but does produce a crossover in H_{eff}. This latter temperature is $T\approx(610E_{7/2}/1311k)\approx 700$°K. Nuclear magnetic resonance techniques are more practical for these observations than are Mössbauer studies.

For a general direction of the magnetic field, there are six magnetically inequivalent sites in SmIG for the rare-earth ions in each unit cell. If the magnetization is in the [111] direction of the unit cell, as is the case for most of the rare-earth iron garnets,[20] the number of inequivalent sites is reduced to two. In dysprosium iron garnet and in europium iron garnet, the two sites could not be distinguished in recoilless absorption measurements,[14,18] whereas, in thulium iron garnet,[16] absorption spectra corresponding to the two sites could be resolved. Pearson[21] has found that at 4°K, the easy magnetization axis for SmIG appears to be along the [110] instead of along the [111] axis, as found for the other garnets. If the magnetization is in the [110] direction, three inequivalent sites exist with relative populations of 4:1:1.

Nowlin's measurements[11] on a single crystal, with the field in the [111] direction showed a drop in the magnetization curve of SmIG below 60°K which could be explained by a shift of the easy magnetization axis from the [111] to the [110] direction below this temperature. (Extrapolating this 60°K data to 0°K, he found a contribution of about $0.13\mu_B$ per Sm^{3+} ion to the total magnetization.)

Including crystalline field effects alters the picture radically, with a resulting decrease in the values of H_{eff} and a change in its temperature dependence. Each rare-earth ion in the rare-earth iron garnet sits in the center of a deformed cube of eight oxygen ions and has

[19] We use an $\langle r^{-3}\rangle$ value extrapolated from the known μ values by taking the shape of the computed Hartree-Fock $\langle r^{-3}\rangle$ values versus atomic number [M. Blume, A. J. Freeman, and R. E. Watson, Phys. Rev. **134**, A320 (1964); and Ref. 12].

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orthorhombic point symmetry. Neglecting the rhombic part of the potential, the effect of a cubic crystalline field on the $J=\frac{5}{2}$ and $\frac{7}{2}$ states of Sm^{3+} has been discussed for some time now.[22,1,2,7] The $J=\frac{5}{2}$ state is split into a doublet (Γ_7) and a quartet (Γ_8); the $J=\frac{7}{2}$ state is split into a Γ_7, a Γ_8, and another doublet Γ_6.

At high temperatures (for which kT is larger than the exchange interaction splittings), the effect of the crystal field can be taken into account[4,2] by multiplying the first term in the bracket of each of the Eqs. (3) and (4) by [i.e., the $(kT)^{-1}$ term]

$$f(T)=(1+2\exp(\Delta E/kT))^{-1}\left[\frac{5}{21}+\frac{26}{21}\exp(\Delta E/kT)+\frac{32kT}{21\Delta E}(1-\exp(\Delta E/kT))\right], \quad (5)$$

where ΔE, the energy separation between Γ_7 and Γ_8, is positive when Γ_8 is the ground state. The crystalline field shifts the transition point of M_S to lower temperatures thus making the difference in the temperature dependence of M_S and H_{eff} more striking.[23] Similarly, the crystal field splitting also affects the Van Vleck term [the second term in the brackets of Eqs. (3) and (4)] but this is neglected in the present crude theoretical description.

When the crystal-field splitting is larger than the exchange interaction, the magnetic interaction can be treated as a perturbation which splits and mixes the two states Γ_8 and Γ_7. The Γ_8 quartet is not isotropic. Its splitting in the presence of a magnetic field depends on the direction of the magnetic field, relative to the cubic axes. The anisotropy of Γ_8 may be responsible for the appearance of magnetically nonequivalent sites in the Sm compounds, even in cases where the crystal field acting on the Sm ion has pure cubic symmetry.[7]

There is experimental evidence for the assumption[20,24,25] that in the case of Sm in SmIG, the crystalline field interactions are larger than the exchange interactions. If we assume that the separation between the ${}^6H_{5/2}$ ground state and the ${}^6H_{7/2}$ state is larger than the crystal field splitting, and that the latter is larger than the exchange splitting, then it is possible to estimate quite easily $H_{\text{eff}}(0)$ for the various sites in the cubic approximation. If these conditions are fullfilled, one is left with a pure Γ_8 ground state (Γ_8 lies below[7] Γ_7 in SmIG) and in the presence of H_{exch} it is split according to[26]

$$W=\pm 2(g_J-1)\mu_B H_{\text{exch}} \times\left\{\frac{65}{36}\pm\frac{1}{9}[270(l^4+m^4+n^4)-74]^{1/2}\right\}^{1/2}, \quad (6)$$

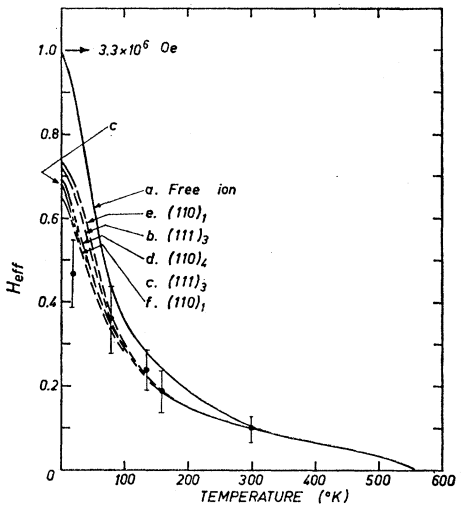

FIG. 1. Theoretical and experimental temperature dependence of H_{eff} acting on the samarium nucleus in SmIG. The theoretical curves are for (a) "free-ion" approximation, (b) and (c), H_{eff} at the two magnetically inequivalent sites that exist if H_{exch} is along the [111] direction of the unit cell, taking into account crystalline field effects. (d), (e), and (f) H_{eff} at the three magnetically inequivalent sites that exist if H_{exch} is along the [110] direction of the unit cell taking into account crystalline field effects (last number in each line is the relative population of the site).

where g_J is the gyromagnetic Landé factor and (l,m,n) are the direction cosines of H_{exch} relative to the *local* axes.

Using Eq. (6), the wave function of the ionic ground state and the value of H_{eff} at 0°K can be calculated for the various sites of the Sm^{3+} ions in SmIG. If H_{exch} is along the [111] direction of the unit cell, two equally populated inequivalent sites exist, and the values of $H_{\text{eff}}(0°\text{K}):H_{\text{eff}}(0°\text{K}, \text{free ion})$ for these two sites are 0.72 and 0.68. If H_{exch} is along the [110] direction, three inequivalent sites exist. For the most populated site $H_{\text{eff}}(0°\text{K})/H(0°\text{K}, \text{free ion})$ is 0.69, and for the two other sites the corresponding ratios are 0.72 and 0.65.

Figure 1 summarizes some of the theoretical results as a function of temperature for various approximations. In these calculations, we use[7,25] $\Delta E=350°\text{K}$ and assume that $H_{\text{exch}}(T)$ is proportional to $M_{\text{Fe}}(T)$, the net magnetization of the iron sublattices and that $\beta H_{\text{exch}}(0°\text{K})/k=22°\text{K}$, which is equal to the average value of $H_{\text{exch}}(0°\text{K})$ for the other rare-earth iron garnets.[27] Curve (a), labeled "free ion," represents the temperature dependence of H_{eff}, neglecting crystal field effects, calculated using Eq. (3) at high temperature and its Brillouin function counterpart at low temperatures. The values of H_{eff} in curves (b), (c), (d), (e), and (f) were calculated taking into account crystal field effects. Curves (b) and (c) represent $H_{\text{eff}}(T)$ for the two magnetically inequivalent sites, assuming that the direc-

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[23] J. A. White, Bull. Am. Phys. Soc. **9**, 464 (1964).
[24] K. A. Wickersheim, Phys. Rev. **122**, 1376 (1961).
[25] R. L. White, Phys. Rev. **115**, 1435 (1959).
[26] B. Bleaney, Proc. Phys. Soc. (London) **73**, 937, 939 (1959).

[27] We have not used the value of 55°K, suggested by White (Ref. 7) on the basis of calorimetric measurements, because this value was deduced assuming that the magnetization is along the [111] direction, which is not correct (Ref. 21).

tion of magnetization is along the [111] direction of the unit cell. Curves (d), (e), and (f) represent $H_{\text{eff}}(T)$ for the three magnetically inequivalent sites, assuming that the magnetization is along the [110] direction.

The values corresponding to temperatures higher than 150°K were calculated from Eqs. (3) and (5), and those corresponding to 0° and 20°K using Eq. (6). The values for temperatures between 20° and 150°K were interpolated from the values of 20°K and above 150°K.

It is clear that the various curves which include crystal-field effects differ significantly from the "free-ion" approximation. We compare these predictions with our experimental data after discussing the experiments. We must bear in mind the many approximations which make the theory crude. Aside from those already mentioned, we assume H_{exch} to be isotropic and given by simple molecular field theory. We have stayed solely within the simple "cubic approximation," thereby neglecting all rhombic contributions despite their apparent importance in the case of[28] YbIG; these rhombic splittings are expected to have important repercussions on the cubic results at low temperatures. The admixture of the $^6H_{7/2}$ state into the ground state by the crystalline fields was neglected. Calculations are currently underway which consider the simultaneous effect of the exchange and crystalline fields on the wavefunctions and eigenvalues of the $\frac{5}{2}$ and $\frac{7}{2}$ states, and which allow for their interaction. Within the remaining approximations the chief uncertainty lies in the definition of the crystalline field parameters.

The effective electric field gradient[13,14] in the free-ion approximation at 0°K can be calculated using the formula

$$q_{\text{eff}} = -e^2\langle 1/r^3\rangle(1-R)\alpha\langle J|3J_z^2-J(J+1)|J\rangle,$$

where R is the Sternheimer shielding factor.[29] Using the same value for $\langle r^{-3}\rangle$ as for the magnetic interaction we obtain $q_{\text{eff}}(0, \text{ for free ion})/h = -650(1-R)$ Mc/sec per barn. For our purposes, we have not been concerned with calculating in detail the effects of $H_{\text{exch}}(T)$ and the crystal field on q_{eff}. This may be done in a manner similar to that given above for H_{eff}.

III. EXPERIMENTAL DETAILS

Jha *et al.*[30] observed the Mössbauer effect in the 22-keV transition of Sm^{149}, using oxide sources and absorbers at room temperature. Recently, the half-life of the 22-keV level was measured electronically by Kistner *et al.*[31] to be $(7.6\pm0.5)\times10^{-9}$ sec.

[28] M. T. Hutchings and W. P. Wolf, Phys. Rev. (to be published).

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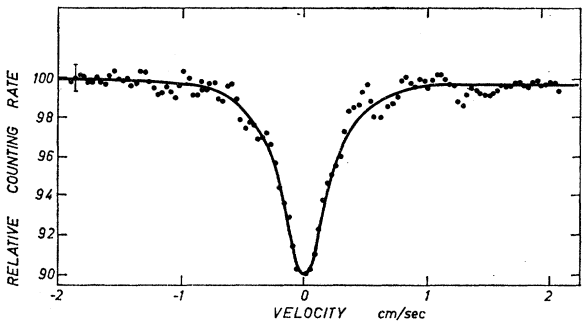

FIG. 2. The absorption by 30 mg/cm² Sm_2O_3 at room temperature of the 22-keV γ-ray emitted from an oxide source of Eu^{149} at room temperature as a function of relative velocity between source and absorber.

In the present experiments, the sources used were Eu^{149} in the form of Eu_2O_3, produced by a (p,n) reaction on enriched $Sm^{149}{}_2O_3$ and by a $(p,2n)$ reaction on enriched $Sm^{150}{}_2O_3$. The Eu^{149} sources were separated from the irradiated Sm by ion exchange. The absorbers used were Sm_2O_3, SmIG, and $SmFeO_3$ containing enriched Sm^{149} of 90% isotopic abundance. Some measurements were also carried out with absorbers of nonenriched Sm metal and $SmFe_2$. The 22-keV radiation was detected either by a 1-mm-thick NaI(Tl) scintillation counter with a 0.001-in. aluminum cover or by a larger argon-methane filled proportional counter.

The absorption as a function of relative velocity between source and absorber was recorded automatically on a multichannel pulse-height analyzer as described in Ref. 18 by one group (OSN), and with a parabolic drive and multiscalar mode operation of the analyzer by the other group (BGFS). In most measurements, the source was at room temperature. The lower temperatures for the absorbers were obtained by placing them in liquid nitorgen, hydrogen, or helium cryostats. The radiation reached the absorbers and detectors through thin Mylar or Be windows. In some measurements source and absorber were kept at 80°, using the cryostat described in Ref. 18. The structure of all the absorber samples used was confirmed by x-ray powder photographs.

IV. EXPERIMENTAL RESULTS

(1) Measurements with Sm_2O_3

The absorption spectrum obtained with the source produced by (p,n) reaction on $Sm^{149}{}_2O_3$ with an enriched $Sm^{149}{}_2O_3$ absorber of about 30 mg/cm² thickness using the scintillation counter is shown in Fig. 2. The maximum effect here is about 10%, the width at half-height (4 ± 0.5) mm/sec. The spectrum obtained with the source produced by the $(p,2n)$ reaction on $Sm^{150}{}_2O_3$ with a thin, enriched $Sm^{149}{}_2O_3$ absorber of about 5 mg/cm² thickness using the proportional counter is shown in Fig. 3. The maximum effect here is about 4%, the width at half-height, (3.3 ± 0.2) mm/sec.

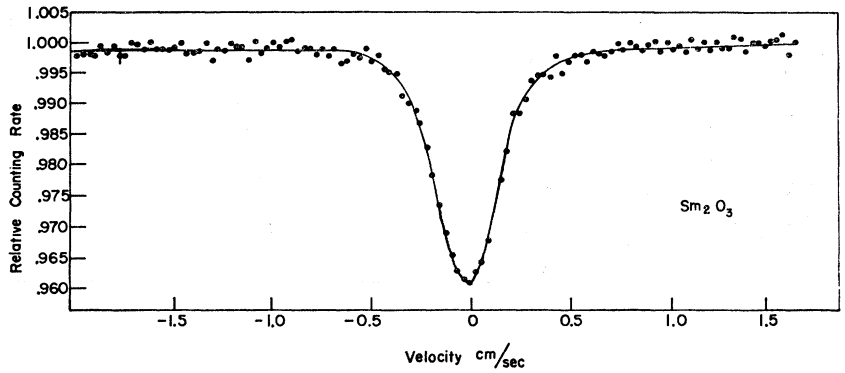

FIG. 3. The absorption by 5 mg/cm² Sm_2O_3 at room temperature of the 22-keV γ ray emitted from an oxide source of Eu^{149} at room temperature as a function of relative velocity between source and absorber.

The value of the natural width of the 22-keV radiation as deduced from the lifetime of the 22-keV level[31] is 0.85 mm/sec. If we assume that the emission line from an oxide source and the absorption line of an oxide absorber have the same width, then the experimental widths at room temperature are about twice the natural linewidth.

In Fig. 4, the absorption spectrum of 30 mg/cm² Sm_2O_3 at 20°K is shown. In this case, too, the source was at room temperature. The maximum change in counting rate was about 16% and the width at half height was about 6 mm/sec, the broadening probably arising mainly from absorber thickness.

From the temperature and absorber thickness dependence of the over-all effect observed with the Sm_2O_3 absorber, a Debye temperature of about 150°K can be deduced for Sm_2O_3 at room temperature.

(2) Measurements with SmIG Absorbers

a. SmIG in Zero External Field

The measurements of the absorption by SmIG were carried out at six different temperatures, most of them at two velocity ranges (Figs. 5–7). At 20° and 16°K, eight resolved absorption lines are seen. The spin of the ground state of Sm^{149} is $\frac{7}{2}$.[32] As the 22-keV transition is of $M1$ character,[33] the possible spin assignments for the 22 keV are $\frac{5}{2}$, $\frac{7}{2}$, or $\frac{9}{2}$. No reasonable fit to the data has been obtained assuming either spin $\frac{7}{2}$ or $\frac{9}{2}$. A good fit has been obtained assuming that the spin of the excited state is $\frac{5}{2}$,and that values of H_{eff} acting on the Sm^{149} nucleus situated at nonequivalent sites do not differ by more than 10%.

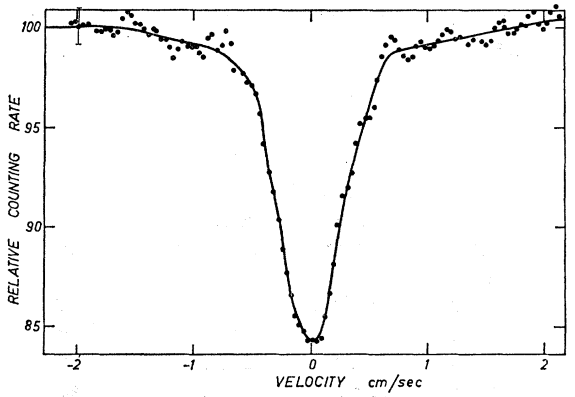

FIG. 4. The absorption by 30 mg/cm² Sm_2O_3 at 20°K of the 22-keV γ ray emitted in the decay of Eu^{149}, as a function of relative velocity between source and absorber.

In the analysis of the results, an axial symmetric quadrupole interaction was assumed in order to account for the asymmetric spectra. Thus, each level was split according to

$$W=-mg\mu_n H_{\text{eff}}+\tfrac{1}{4}eqQ[3m^2-I(I+1)]/[I(2I-1)],$$

where g is the gyromagnetic ratio of the level and Q its quadrupole moment. In addition, a possible shift ΔE between the centroid of the absorption and the emission lines was considered. The results obtained for the splitting parameters at 20°K from the analysis of the experimental peak positions are given in Table I.

The "best-fit" reconstructed spectra, assuming a width of 3.4 mm/sec for each line, are shown in Figs. 6(a) and 8. Each curve assumes a different set of quadrupole interactions, the ratio of $g_1/g_0=1.76$ remaining constant. The agreement is equally good for all three sets of parameters, so that these measurements do not strongly delimit the values of the quadrupole interactions. On the other hand, the agreement is much poorer when either the quadrupole interactions are much outside the range given in the figures[34] or when the ratio of g_1/g_0 is changed appreciably. The discrepancies between experimental and theoretical curves may result from the combination of the following factors: (1) Small deviations from axial symmetry in the quadrupole

[32] A. J. Cooke and H. J. Duffus, Proc. Roy. Soc. (London) **A229**, 407 (1955).

[33] B. Harmatz, T. H. Handley, and J. W. Mihelich, Phys. Rev. **123**, 1758 (1961).

[34] Recent measurements by G. K. Woodgate, F. M. J. Pichanick, and P. G. H. Sandars (private communication, G. K. Woodgate) yield a preliminary value for the quadrupole moment of the ground state of Sm^{149} of +0.061 b, uncorrected for shielding effects. Such a small moment implies the values of the parameters used in curve 3 of Fig. 8. A positive value for Q_0 results in a negative quadrupole interaction in agreement with the data of Fig. 8.

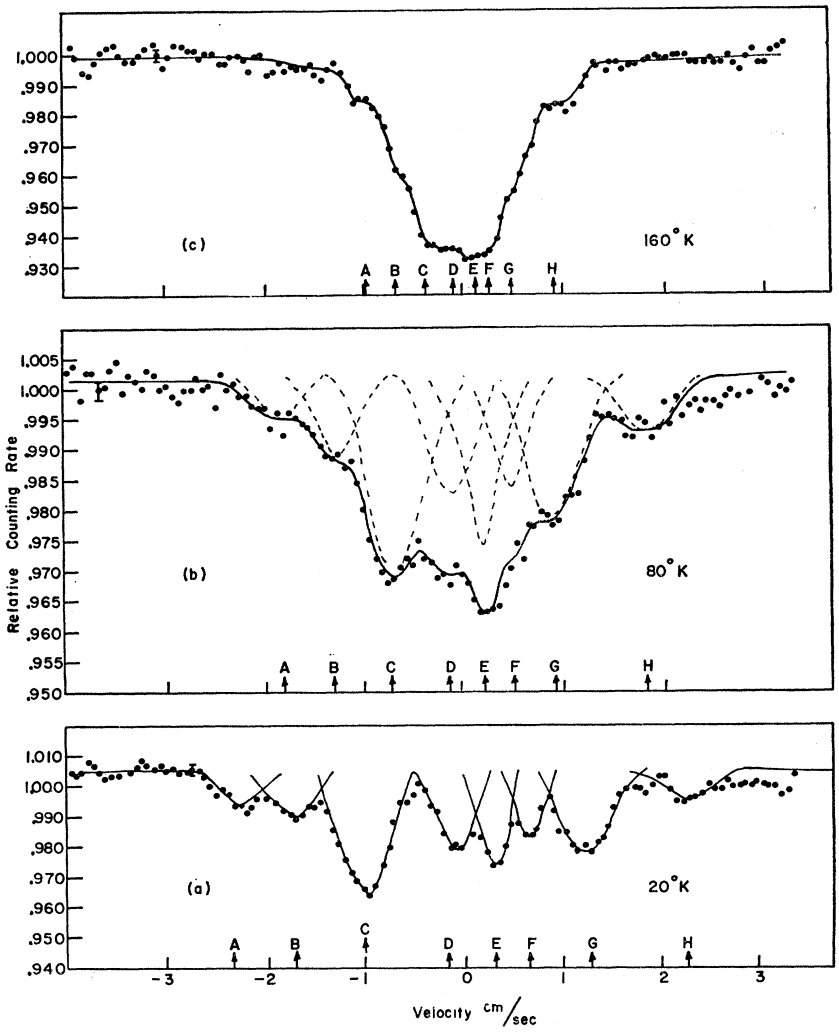

FIG. 5. The absorption by samarium iron garnet of the 22-keV γ ray emitted in the decay of Eu^{149} as a function of relative velocity between source and absorber (range of velocities -3 to $+3$ cm/sec). (a) SmIG at 20°K, (b) SmIG at 80°K, (c) SmIG at 160°K. The position of specific lines are labeled by A, B, C., etc.

interactions. (2) In the reconstruction, the possibility of the existence of different Sm^{3+} sites was neglected. (3) The assumption of a width of 3.4 mm/sec for each absorption line may not be exact.

A wide, though not quite exhaustive, range of parameters have been examined for each spin, covering different quadrupole interactions as well as three nonequivalent sites. As remarked above, we have been unsuccessful in fitting our results with excited state spins of $\frac{7}{2}$ or $\frac{9}{2}$.

It is possible to reconstruct a spectrum which exhibits some similarity to that obtained experimentally at 20°K, by assuming that $g_1/g_0 \approx 1$, that three magnetically inequivalent sites exist with relative populations of 4:1:1, and that the ratios of H_{eff} in the three sites are about 1:1.8:2.4, respectively. We do not prefer this analysis for a number of reasons: (1) The reconstructed spectrum is in poorer agreement with the experimental points than are the spectra of Fig. 8. (2) If it is assumed that the ratios of H_{eff} in the three sites are 1:1.8:2.4, then the values of H_{eff} in the two less populated sites reach the saturation "free-ion" values. This is not probable in the light of the fact that the spontaneous magnetization of the Sm lattices at 0°K is only 20% of the saturation value.[10] (3) The temperature dependence of all the experimental peaks appears to be the same, making it unlikely that the peaks belong to three very different sites. (4) The negative results of an external field run are described briefly below.

The sign of $g_1\mu_n H_{\text{eff}}$ relative to $g_0\mu_n H_{\text{eff}}$ is determined by the experimental results. As seen from Fig. 8, the

TABLE I. Results for the hyperfine interaction and isomeric shift observed in SmIG at 20°K; g_0 corresponds to the ground state of Sm^{149}, g_1 to the 22-keV level.

$\Delta E/h$ (Mc/sec)	$g_0\mu_n H_{\text{eff}}/h$ (Mc/sec)	$g_1\mu_n H_{\text{eff}}/h$ (Mc/sec)
-7 ± 10	240 ± 10	425 ± 20

[35] E. Matthias, W. Schneider, and R. H. Steffen, Arkiv Fysik 24, 97 (1963).

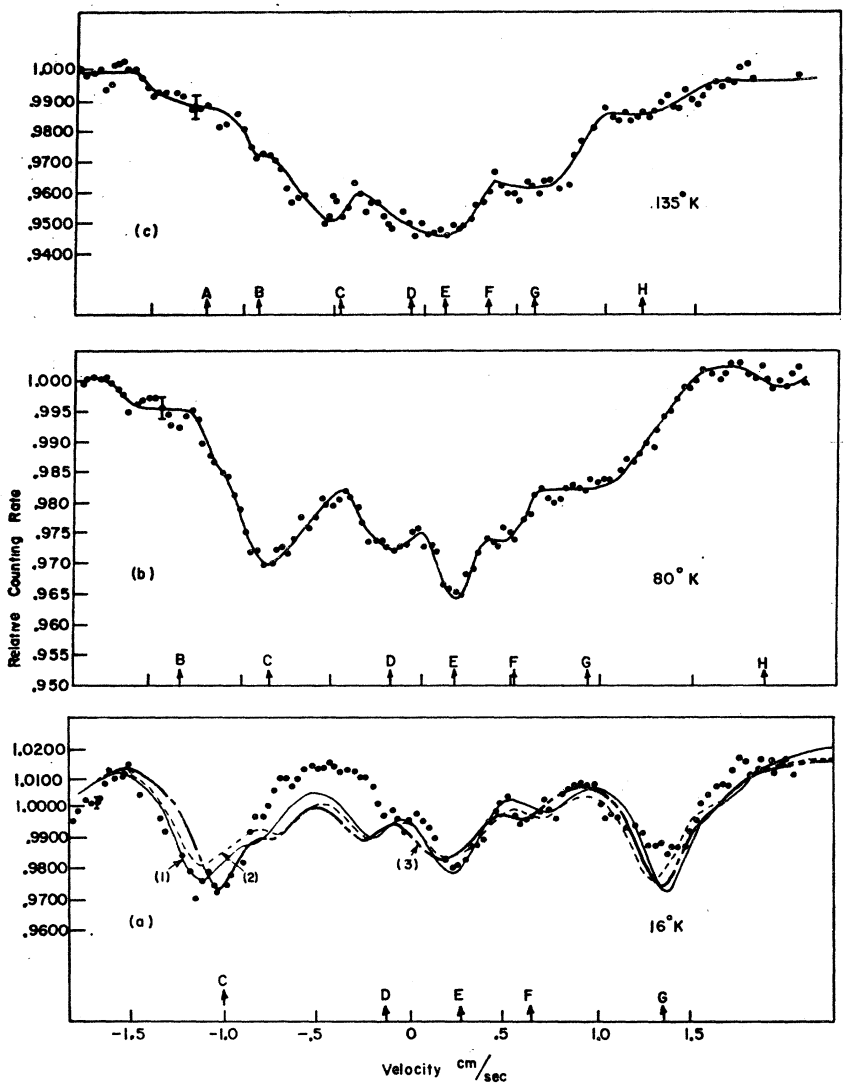

FIG. 6. The absorption by samarium iron garnet of the 22-keV γ ray emitted in the decay of Eu^{149} as a function of relative velocity between source and absorber (range of velocities -1.5 to $+1.5$ cm/sec). (a) SmIG at 16°K, (b) SmIG at 80°K, (c) SmIG at 135°K. The parameters for the theoretical curves drawn in Fig. 6(a) are given in Fig. 8.

sign of the quadrupole interaction in the ground state is determined to be negative by the experimental results in agreement with Woodgate *et al.*[34] The sign of the excited state quadrupole interaction is not uniquely determined from our results; Woodgate's result implies curve 3, Fig. 8, which shows a positive value. Neither the sign of H_{eff} relative to the direction of the magnetization of the Sm sublattice in SmIG nor the sign of g_0 can be derived from Mössbauer measurements without an external field.

We find that $g_1/g_0 = 1.76 \pm 0.05$. The absolute value of the magnetic moment of the ground state of Sm^{149} was measured by Bleaney[36] and found to be (0.68 ± 0.10) nm and by others[37] (-0.64 ± 0.10) nm. Assuming Bleaney's value, we therefore deduce that $\mu_1 = (0.85 \pm 0.15)$ nm and the value of H_{eff} at 16°K is $(1.55 \pm 0.25) \times 10^6$ Oe.

The experimental spectra at higher temperatures could not be reconstructed simply by reducing the parameters obtained at 16°K by a scale factor. They are consistent, however, with the experimental spectra at 16° or 20°K. By taking the eight individual peaks seen at these temperatures and moving them to lower velocities as shown in Fig. 5(b), the solid line in this figure is obtained. In this way, the eight peaks can be followed as a function of temperature so that from the respective splittings of these lines, the temperature dependence of H_{eff} may be deduced. Table II and the experimental points in Fig. 1 show this temperature dependence. The value of $(0.34 \pm 0.10) \times 10^6$ Oe we obtain at 300°K is in good agreement with that determined by Caspari *et al.*[9]

[36] B. Bleaney, Proc. Phys. Soc. (London) **A68**, 937 (1955).
[37] See discussion by I. Lindgren, *Perturbed Angular Correlations*, edited by E. Karlsson, E. Matthias, and K. Siegbann (North-Holland Publishing Company, Amsterdam, 1946).

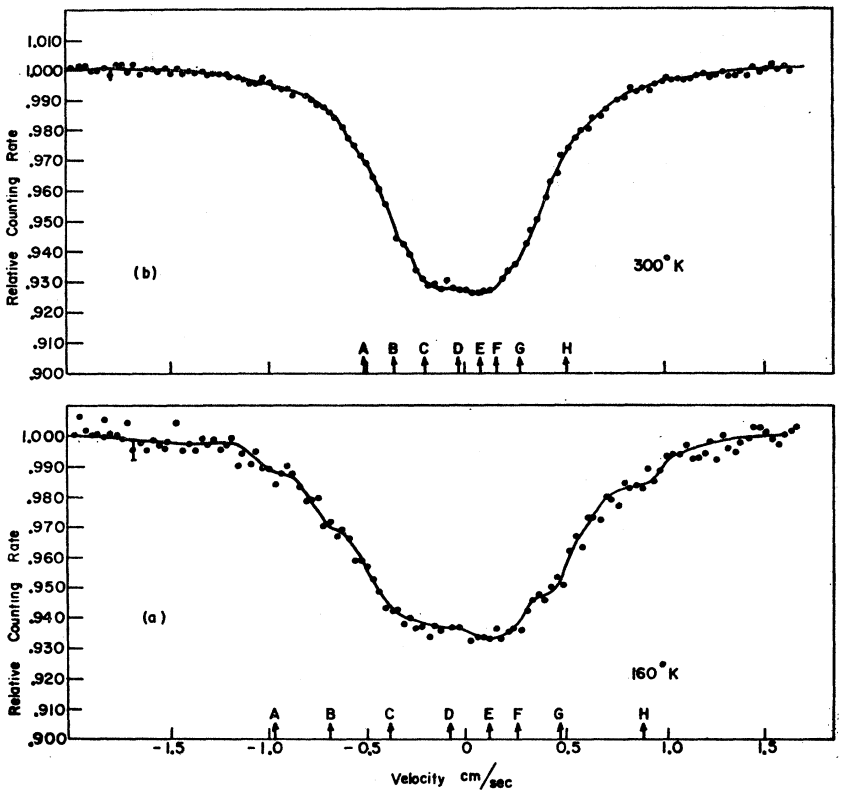

FIG. 7. The absorption by samarium iron garnet of the 22-keV γ ray emitted in the decay of Eu^{149} as a function of relative velocity between source and absorber (range of velocities -1.5 to $+1.5$ cm/sec.) (a) SmIG at 160°K, (b) SmIG at 300°K.

b. SmIG in an External Field

A spectrum (Fig. 9) was obtained at 4.2°K in an external field of 18 kOe provided by a superconducting magnet.[38] Such a magnetic field should result in a substantial alignment of the magnetization[10,11] and hence a reduction in the intensity of the $\Delta m=0$ hyperfine transitions. Little change in the spectrum is expected if the ratio of g_1/g_0 is 1.76 (Fig. 8). Gross changes should be observed if the ratio g_1/g_0 is approximately one (see discussion above), since in this case all the middle peaks (D, E, F, Fig. 5) are $\Delta m=0$ transitions. No reduction in the strengths of these peaks is observed.

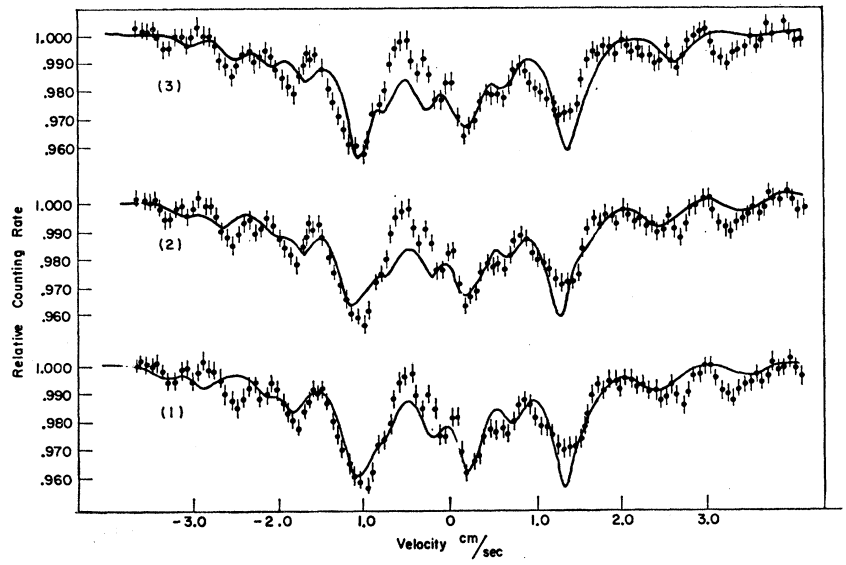

FIG. 8. The absorption by samarium iron garnet at 16°K of the 22-keV γ ray emitted in the decay of Eu^{149} as a function of relative velocity between source and absorber (range of velocities -3 to $+3$ cm/sec.) Solid lines are theoretical reconstructed curves under following assumptions:

(1) $g_0\mu_n H_{\text{eff}}=\pm 1.39$ cm/sec; $eQ_0/4=-0.42$ cm/sec; $g_1\mu_n H_{\text{eff}}=\pm 2.44$ cm/sec: $eQ_1/4=-0.2$ cm/sec.
(2) $g_0\mu_n H_{\text{eff}}=\pm 1.34$ cm/sec; $eQ_0/4=-0.22$ cm/sec; $g_1\mu_n H_{\text{eff}}=\pm 2.35$ cm/sec; $eQ_1=-0.07$ cm/sec.
(3) $g_0\mu_n H_{\text{eff}}=\pm 1.34$ cm/sec; $eQ_0/4=-0.10$ cm/sec; $g_1\mu_n H_{\text{eff}}=\pm 2.35$ cm/sec; $eQ_1/4=+0.10$ cm/sec.

[38] We thank P. Craig of Brookhaven National Laboratory for allowing us to use his equipment to make this measurement.

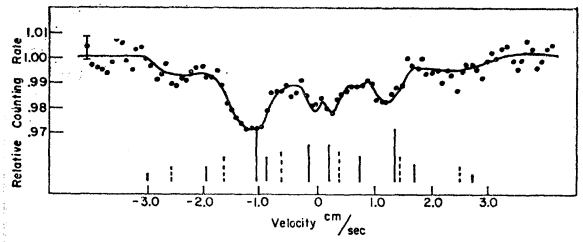

FIG. 9. The absorption by SmIG at 4°K of the 22-keV γ ray emitted by $Eu^{149}{}_2O_3$ as a function of relative velocity between source and absorber with both source and absorber in longitudinal magnetic field of 18 kG. Dashed lines indicate position of $\Delta m=0$ transition lines with parameters of curve (1) in Fig. 8. Full lines indicate positions of $\Delta m=\pm 1$ transitions.

The spectrum is consistent with the reconstructed curve of Fig. 8 with $\Delta m=0$ transitions omitted.

(3) Measurements with Sm Metal, $SmFe_2$, and $SmFeO_3$ Absorbers

a. Sm Metal

Measurements with a Sm metal absorber were carried out mainly for the purpose of measuring the isomeric shift between the absorption of Sm metal and Sm_2O_3. From such a measurement, it is possible to estimate the difference in charge radii of the nuclear excited and ground states.

The absorption spectra of Sm metal obtained at 300° and 80°K are shown in Figs. 10 and 11, respectively. In this case too the source was Eu_2O_3 at room temperature. The asymmetry of the unsplit line, which was obtained at both temperatures, may be attributed to quadrupole splittings in the absorber. Unfortunately, the lines are not entirely resolved and different ratios of quadrupole moments could give this absorption spectrum. One obtains for the isomer shift, $\Delta E = -0.9\pm 0.3$ mm/sec. An isomer shift between Sm metal and Sm_2O_3 could be caused by the "6*s*" conduction electrons of the metal. Using the usual relativistic expression for the isomer shift and Shirley's[39] approximate values for the parameters involved, one gets a very crude estimate of $(R_{exc}-R_{gr}):R_{gr}$ to be approximately -5×10^{-5}, where R_{exc} and R_{gr} are the charge radii of the nuclear excited and ground state, respectively.

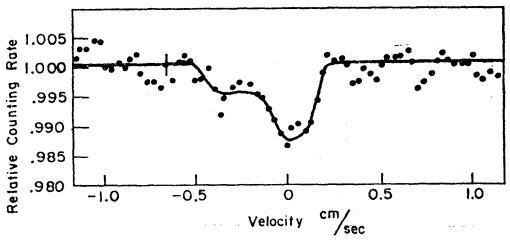

FIG. 10. The absorption by samarium metal at room temperature of the 22-keV γ ray emitted from an oxide source of Eu^{149} as a function of relative velocity between source and absorber.

[39] D. A. Shirley, Rev. Mod. Phys. **36**, 339 (1964).

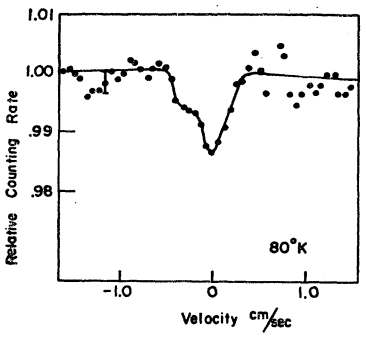

FIG. 11. The absorption by samarium metal at 80°K of the 22-keV γ ray emitted by $Eu^{149}{}_2O_3$ as a function of relative velocity between source and absorber.

b. $SmFe_2$

$SmFe_2$, one of the cubic Laves-phase compounds which have the $MgCu_2$ structure, is ferrimagnetic[40] below 674°K. Because of technical reasons the $SmFe_2$ used in the present work could not be synthesized from samarium enriched in Sm^{149} and therefore only small effects (less than 1%) were obtained in the Mössbauer effect experiments carried out with it. Measurements with a $SmFe_2$ absorber were carried out only at liquid nitrogen temperature. Figure 12 shows the absorption spectrum of the 22-keV γ ray emitted from an oxide source of Eu^{149} at 81°K by a $SmFe_2$ absorber at 81°K. An unsplit line having a width at half-height of (18 ± 1) mm/sec was obtained. No unambiguous isomeric shift ΔE between the emission and absorption line was found; the experimental value is (-1.5 ± 1.5) mm/sec. From the width of the spectrum obtained, it can be concluded that H_{eff} acting on the Sm nuclei in $SmFe_2$ at 81°K is smaller than 10^6 Oe.

c. $SmFeO_3$

$SmFeO_3$ has a pseudoperovskite structure.[41] It is antiferromagnetic below 730°K.

Figures 13 to 15 show the absorption spectra obtained with a $Sm^{149}FeO_3$ absorber at 300, 80, and 16°K,

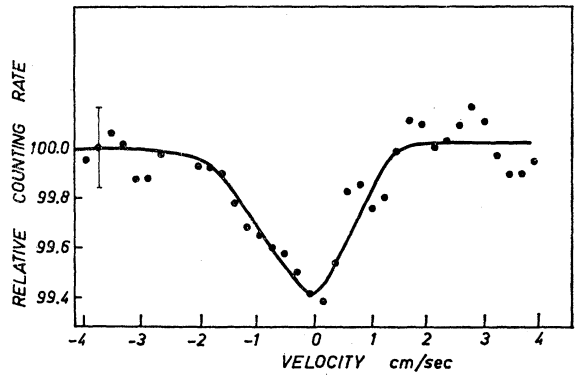

FIG. 12. The absorption by $SmFe_2$ at 81°K of the 22-keV γ ray emitted from an oxide source of Eu^{149} at 81°K as a function of relative velocity between source and absorber.

[40] G. K. Wertheim and J. H. Wernick, Phys. Rev. **125**, 1937 (1962).

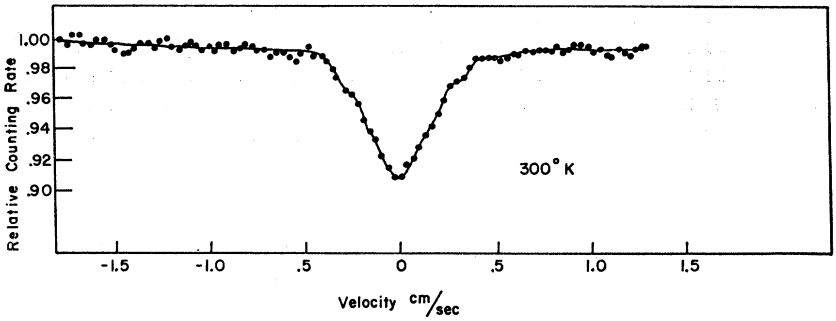

FIG. 13. The absorption by $SmFeO_3$ at 300°K of the 22-keV γ ray emitted from an oxide source of Eu^{149} as a function of relative velocity between source and absorber.

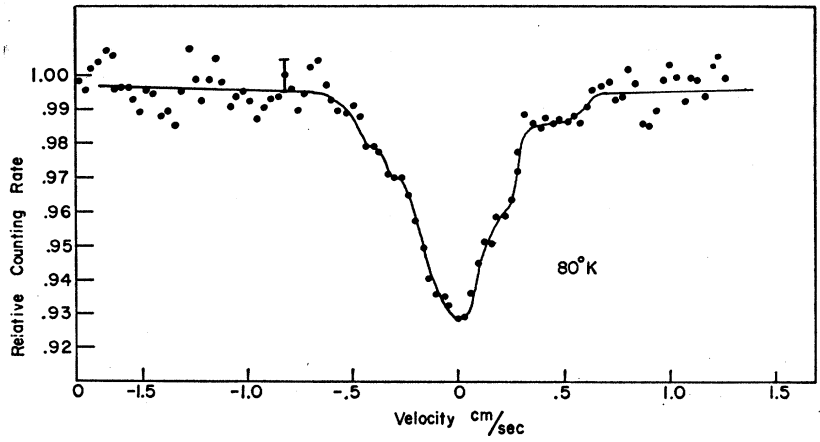

FIG. 14. The absorption by $SmFeO_3$ at 80°K of the 22-keV γ ray emitted from an oxide source of Eu^{149} as a function of relative velocity between source and absorber.

respectively. Unsplit lines with some structure in them were obtained. From the over-all width, upper limits for the effective field acting on $SmFeO_3$ at these temperatures were deduced to be $H_{\text{eff}}(16°\text{K}) \leq 4\times10^5$ Oe, $H_{\text{eff}}(80°\text{K}) \leq 1.9\times10^5$ Oe, and $H_{\text{eff}}(300°\text{K}) \leq 1.5\times10^5$ Oe.

V. SUMMARY AND CONCLUSIONS

(1) SmIG

The present experimental results for the temperature dependence of H_{eff} acting on the Sm^{3+} ions in SmIG, as well as the magnetization measurements[10,11,21] on SmIG, indicate that at low temperatures, the magnetic behavior of the samarium ion is strongly affected by the crystalline electric fields. The experimental results for H_{eff} are consistent with the simplified theoretical predictions if it is assumed that at low temperatures H_{eff} is determined mainly by the splitting of the quartet Γ_8 ground state by the exchange field.

In Fig. 1, our experimental values of $H_{\text{eff}}(T)$ are compared with the various theoretical calculations described in Sec. II. The average theoretical value of H_{eff} at 20°K is about 0.6 times the "free-ion" saturation value. The corresponding experimental value is (0.48±0.05), where the error given includes uncertainties in the present measurements and in Bleaney's measurement of the magnetic hyperfine interaction in the ground state of Sm^{149}. The theoretical value of $H(80°\text{K}):H(20°\text{K})$ is about 0.65, compared with the

TABLE II. Hyperfine fields versus temperature at a Sm nucleus in SmIG.

°K	$H_{\text{eff}}/H_{16°\text{K}}$	$H_{\text{eff}}\times10^6$
16	1	1.55±0.25
20	1±0.01	1.55±0.25
80	0.76±0.02	1.18±0.25
135	0.50±0.03	0.78±0.15
160	0.40±0.03	0.62±0.15
300	0.22±0.05	0.34±0.10

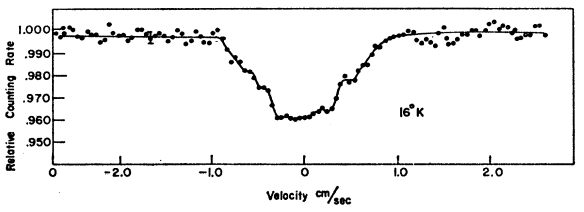

FIG. 15. The absorption by $SmFeO_3$ at 16°K of the 22-keV γ ray emitted from an oxide source of Eu^{149} as a function of relative velocity between source and absorber.

[41] R. C. Sherwood, J. P. Remeika, and H. J. Williams, J. Appl. Phys. 30, 217 (1959); J. B. Goodenough, *Landolt Börnstein II* (Springer-Verlag, Berlin, 1962), Vol. 9.

experimental ratio of 0.76±0.02. Thus, taking into account the crudeness of the theory (discussed in Sec. II) and the uncertainty in the values of some of the parameters, the agreement is (surprisingly) good.

It was found, in the crude cubic approximation that the theoretical values of H_{eff} are only slightly dependent on the direction of the magnetization ([111] or [110]) and on the site in which the nucleus is situated (cf., Fig. 1). If H_{exch} is along the [111] direction of the unit cell, the values of H_{eff} for the two equally populated magnetically inequivalent sites differ by about 6%. If H_{exch} is along the [110] axes, the calculated values of H_{eff} at 16°K for the three magnetically inequivalent sites differ by about 10%. Our experimental data (Fig. 8) indicate that the values of H_{eff} acting on samarium nuclei situated in magnetically inequivalent sites do not differ at 16°K by more than 10% and do not distinguish between the two possibilities. They are therefore consistent with both Pearson's[21] and Nowlin's[11] measurements showing that the direction of easy magnetization in SmIG at low temperatures is along the [110] axes, but they do not exclude the possibility of a [111] orientation for H_{exch}.

The experimental value of the magnetization of the samarium lattice in SmIG at 0°K is about three[11] to five[10] times smaller than the "free-ion" theoretically predicted value, whereas we find $H_{\text{eff}}(0°\text{K}):H_{\text{eff}}(0°\text{K};$ "free-ion")~ 0.5. This difference in behavior of M_{Sm} and H_{eff} is probably a result of the admixture of the ${}^6H_{7/2}$ state into the ground state by the crystalline field and the exchange interactions. Such an admixture has a much larger effect on M_s than on H_{eff} because $\langle J\|L+2S\|J+1\rangle/\langle J\|L+2S\|J\rangle$ is about five times larger (and with negative sign) than the value of $\langle J\|N\|J+1\rangle/\langle J\|N\|J\rangle$.[13] In addition, because of the difference in the experimental techniques used for the determination of M_S and H_{eff}, anisotropy effects may decrease the measured values of M_S, without affecting the measured values of H_{eff}.

(2) $SmFe_2$

The low value of H_{eff} acting on the Sm nucleus in $SmFe_2$ (less than 10^6 Oe at 77°K) is probably due to the fact that in $SmFe_2$ the Γ_7 doublet is the ground state, similar to the situation in[4] $SmAl_2$. In such a case, making the same assumption as was made for SmIG, a value of $\frac{1}{3}$ is obtained for $H_{\text{eff}}(0°\text{K}):H_{\text{eff}}(0°\text{K};$ "free-ion"). This value is consistent with the experimental upper limit obtained at 77°K.

(3) Nuclear Data

The shell model predicts an $f_{7/2}$ state for the ground state of Sm^{149}. This spin assignment has been confirmed experimentally. It therefore seems reasonable to assume that the ground state of Sm^{149} is $f_{7/2}$, and to assign an odd parity to this state. The present experimental results support strongly an assignment of a spin $\frac{5}{2}$ for the 22-keV level of Sm^{149}. Based on measurements of angular correlations of Sm^{149} γ-ray cascades, Harling *et al.*[42] suggest a $\frac{7}{2}$ spin assignment to the 22-keV level. Their objection to a $\frac{5}{2}$ assignment is based on their experimental results for the angular correlation of the 178.4–327.7-keV γ-ray cascade. The angular correlation coefficients which they obtained for this cascade were $A_2=-0.3977\pm0.0398$ and $\bar{A}_4=+0.1220\pm0.0244$ but, as described in Ref. 42 (cf. Fig. 2 in the reference), about 50% of the coincidence counts did not belong to the relevant cascade and the data were appropriately corrected. If the errors in A_2 and A_4 are larger by a factor of about 2 than those estimated by Harling *et al.*, then the results of the angular correlation measurements are not in disagreement with the present $\frac{5}{2}$ spin assignment to the 22-keV level.

Since the 22-keV transition is[33] $M1$, the 22-keV level has the same parity as the ground level of Sm^{149}. Therefore, the only possible single-particle state assignment to the 22-keV level is $f_{5/2}$. However, the magnetic moments of single neutron $f_{7/2}$ and $f_{5/2}$ states should have opposite signs, whereas, the present experimental results indicate that the ratio of the magnetic moments of the 22-keV excited and ground state of Sm^{149} is $+1.26\pm0.04$. Thus, the 22-keV level probably has some collective features. According to the collective model, the small experimental quadrupole moment[36,34] for the ground state of Sm^{149}, results in a deformation factor $\delta<0.05$. For such a small deformation, the theory of Mottelson and Nilsson concerning the intrinsic states of odd-A nuclei, having ellipsoidal equilibrium shapes is not applicable.[43] A possible interpretation of the 22-keV level is that it is not a regular Nilsson level, but a combination of an individual particle state with some collective excitation, such as, for example, a combination of a $2+Sm^{148}$ collective state as a core plus a neutron with spin $\frac{7}{2}$. Assuming this latter oversimplified model, and a g factor for the 2+vibration core of 0.4, coupled to a single particle $\frac{7}{2}$ spin state of magnetic moment -0.68 nm, the ratio of the magnetic moment of the 22-keV state to that of the ground state of Sm^{149} is $+0.9$ compared with the experimental ratio of $+1.25$. The states involved cannot be as simple as the agreement would indicate since the calculated ratio is very insensitive to the g factor of the core, and precise agreement with the experimental ratio necessitates an unrealistically high value for g_R.

ACKNOWLEDGMENTS

We would like to thank J. H. Van Vleck, S. G. Cohen, and J. A. White for discussions and A. Mustachi for help with chemical problems.

[42] O. K. Harling, C. A. Ventrice, and J. J. Pintrice, Phys. Rev. **132**, 807 (1963).

[43] B. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat-Fys. Skrifter **1**, No. 8 (1959).