Hyperfine Interactions in the Ground State and 21.7-keV State of Eu^{151} in Europium Iron Garnet*

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The hyperfine interaction of the ground state and 21.7-keV state of Eu^{151} in europium iron garnet have been measured at 300, 81, and 20°K using the technique of Mössbauer absorption. Values of (335±50), (570±35), and (670±100) kOe were found for effective magnetic fields acting on the Eu nuclei in europium iron garnet at the temperatures of 300, 81, and 20°K, respectively. These values are in agreement with theoretical calculations of the nondiagonal matrix elements associated with the admixture of states produced by the exchange interaction. The value of the quadrupole interaction eQ_0q_{eff} of the ground state of Eu^{151} in europium iron garnet was found to be (−240±100) Mc/sec at 81°K. The results yield an assignment of a spin of $\frac{7}{2}$ for the 21.7-keV level and a value of $\mu=(2.54\pm0.15)$ nm for the magnetic moment of this level.

INTRODUCTION

THE hyperfine Zeeman splitting in the recoil-free absorption spectrum of the 21.7-keV γ rays from Eu^{151m} ($\tau=1.34\pm0.1\times10^{-8}$ sec[1]) in Eu^{151} situated in europium iron garnet (EuIG) have been observed at 20, 81, and 300°K, using the technique of recoil-free absorption. This is possible since the sublattice of the rare earth ions in EuIG displays a spontaneous magnetization below the Curie temperature (about 550°C), which is correlated with the existence of an effective magnetic field at the Eu nuclei.[2] The spontaneous magnetization of the Eu sublattice is temperature-dependent and this implies that there will be a corresponding dependence of the effective fields acting on the Eu nuclei on temperature.

The ionic ground state of Eu^{3+} is $^7F_0$, which is diamagnetic. Wolf and Van Vleck[3] have pointed out, however, that the presence of nondiagonal matrix elements connected with the admixture of the excited $^7F_1$ state into the $^7F_0$ ground state produced by the exchange interaction accounts essentially for the behavior of the spontaneous magnetization of EuIG below its Curie temperature. Using this approach, Wolf and Van Vleck succeeded in calculating the temperature dependence of the spontaneous magnetization of EuIG below its Curie temperature. Their results are in good agreement with the experimental data.[4] In a recent paper, Gilat and Nowik[5] using the same approach have calculated the corresponding nondiagonal matrix elements determining the hyperfine interaction on the Eu nucleus in EuIG, and have in this way estimated, as a function of temperature, the magnitude of the effective magnetic field (H_{eff}) and the part of the effective electric field gradient (q_{eff}) associated with the orientation of the orbital wave functions produced by the exchange interaction through the spin-orbit coupling. Their calculated values of H_{eff} and of eq_{eff}/h at 0°K were 7×10^5 Oe and −550 Mc/sec per b, respectively. One of the main purposes of the present work was to measure H_{eff} and q_{eff} acting on Eu^{3+} ion in EuIG as a function of temperature and to compare the results to the theoretical predictions. A very good agreement between the experimental and theoretical values of H_{eff} was found.

The spin of the ground state of Eu^{151} is $\frac{5}{2}$. The value of its magnetic moment is[6] (3.419±0.004) nm and of its quadrupole moment is (0.95±0.10) b.[7] The 21.7-keV transition is $M1$. From the point of view of nuclear information, concerning the 21.7-keV level, the results presented in this article yield an assignment of a spin of $\frac{7}{2}$ for this state, and a value of $\mu=(2.54\pm0.15)$ nm for the magnetic moment for this state. The value of the magnetic moment is interpreted in terms of two possible models.

Shirley *et al.*[8] already observed the Mössbauer effect in the 21.7-keV transition of Eu^{151}, using oxide sources and absorbers. The absorption patterns obtained by Shirley *et al.* were single lines of approximately natural widths.

EXPERIMENTAL DETAILS

The source used was Gd^{151} in the form of Gd_2O_3. It was produced by a (p,n) reaction on enriched Eu^{151} oxide. The bulk of europium was removed by a Na amalgam reduction[9] and the activity then precipitated with NH_4OH using 5 mg Nd_2O_3 carrier. The precipitate was then ignited at 800°C. The absorber used was 40 mg/cm² EuIG containing enriched Eu^{151} of 91% isotopic abundance. The 21.7-keV radiation was detected by a 1-mm thick NaI(Tl) scintillation counter with a

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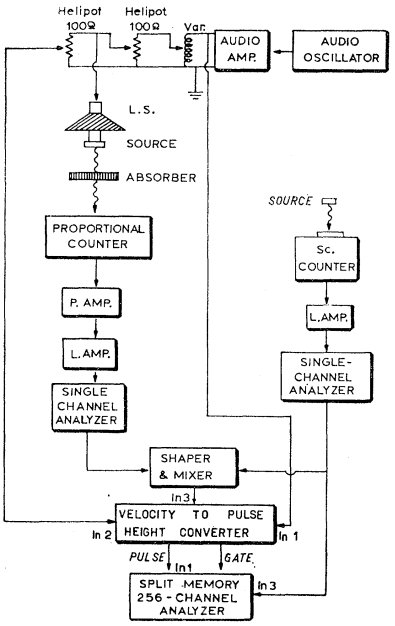

FIG. 1. Block diagram of the electronics used for the automatic measurement of the recoilless absorption as a function of relative velocity between source and absorber.

0.001-in.-aluminum cover. The absorption as a function of relative velocity between source and absorber was recorded automatically on a multichannel pulse-height analyzer as already described.[10] A scale of velocities was established by using as a frequent calibration the absorption spectrum of α-Fe_2O_3 which has been previously investigated.[11] It turned out that the spectra obtained with the EuIG extended to velocities of about 6 cm/sec. On the other hand, the α-Fe_2O_3 spectrum extends to about 1 cm/sec. In order that the calibration should cover the required range on the pulse-height analyzer the ac voltage applied to the loudspeaker coil during the calibration run was a suitable fraction of the voltage used in the EuIG measurement. The ratio which determines the conversion factor between the two scales of voltage was measured accurately using a helipot potentiometer. The procedure assumes linearity of the loudspeaker which was confirmed separately in the relevant range. A block diagram of the electronics is shown in Fig. 1.

The measurements at 81°K were carried out using the cryostat shown in Fig. 2. The cryostat is suitable for work at a continuous range of temperatures between 81°K and room temperature. In the present experiment the heating coils were not used and the measurements were made with both the source and absorber at 81°K. Very thin rubber was used to close the hole between the Lucite rod which held the source and the cryostat in order to prevent freezing of moisture in the cryostat which would impede the motion.

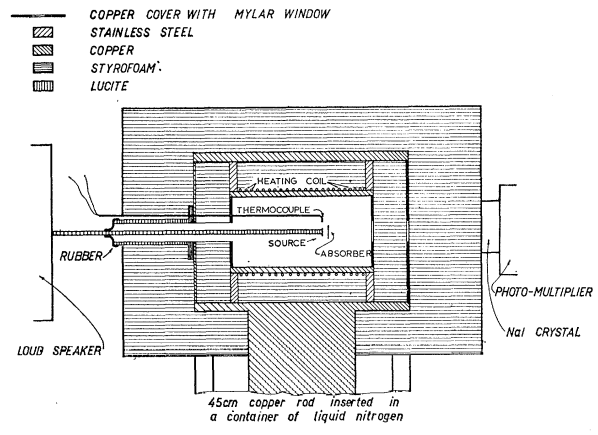

FIG. 2. Experimental arrangement for carrying out recoil-free absorption measurement with source and absorber at liquid-nitrogen temperature.

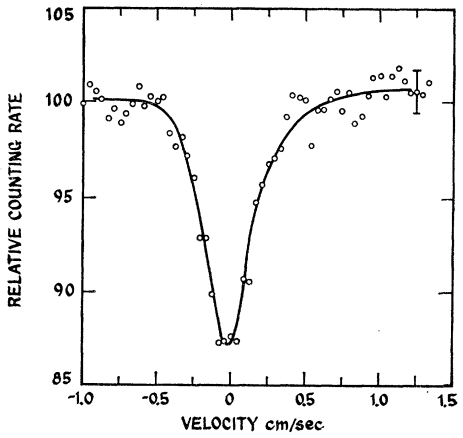

FIG. 3. The absorption by Eu_2O_3 at 81°K of the 21.7-keV γ ray emitted from an oxide source of Gd^{151} at 81°K, as a function of relative velocity between source and absorber.

For the measurements at 20°K, the EuIG absorber was placed in a liquid-hydrogen cryostat. The source was at room temperature. The radiation reached the absorber and detector through thin Mylar windows.

EXPERIMENTAL RESULTS

In order to measure the width of the recoilless emission line, measurements with Eu_2O_3 absorbers were carried out. Our results are in full agreement with those obtained by Shirley *et al.*[8] The spectrum obtained with oxide source and absorber at 81°K is shown in Fig. 3.

The measurements of the absorption by EuIG at 81°K were carried out in two velocity ranges (Figs. 4 and 5). Six resolved absorption lines are seen. The spin of the ground state of Eu^{151} is $\frac{5}{2}$. As the 21.7-keV transition is of $M1$ character, the possible spin assignments for the 21.7-keV level are $\frac{3}{2}$, $\frac{5}{2}$, or $\frac{7}{2}$. The Zeeman effect splits the ground and 21.7-keV level into magnetic sub-

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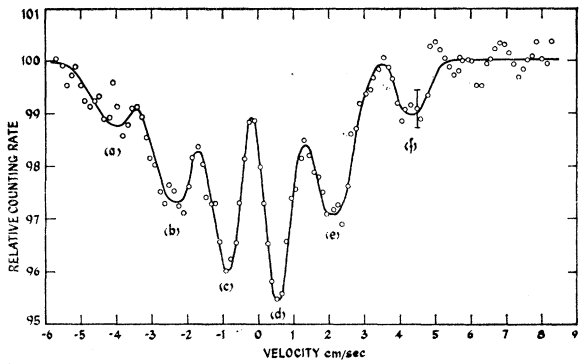

FIG. 4. The absorption by europium iron garnet at 81°K of the 21.7-keV γ ray emitted in the decay of Gd^{151}, as a function of relative velocity between source and absorber. (Range of velocities -6 to $+8$ cm/sec.)

levels. The selection rules for a pure $M1$ transition permit 12, 16, or 18 transitions between the sublevels, corresponding to values of $\frac{3}{2}$, $\frac{5}{2}$, or $\frac{7}{2}$ for the spin of the 21.7-keV level, respectively. The fact that only six resolved absorption lines are obtained shows that the ratio of the gyromagnetic ratios of the 21.7-keV state and ground state is such that many of the possible absorption lines overlap. A preliminary analysis of the experimental results showed that the general shape of the absorption spectrum is consistent with either one of the following three assumptions: (a) the spin of the 21.7-keV level is $\frac{3}{2}$ and the ratio of the g factors of the excited state and ground state, respectively, is about 1.5; (b) the spin is $\frac{7}{2}$ and the ratio of g factors is about $\frac{2}{3}$; (c) the spin is $\frac{7}{2}$ and the ratio of g factors is about 0.5. In order to decide which of the three assumptions fits the experimental results, the ratio of the areas corresponding to peaks "c" and "d" to the areas corresponding to peaks "b" and "e" was determined from Figs. 4 and 5. The ratio was found to be 1.05 ± 0.20. The theoretical ratio corresponding to assumption (a), (b), and (c) are 1.6, 2.3, and 1, respectively. Thus, the present experimental results indicate that the spin of the 21.7-keV level is $\frac{7}{2}$ and that the ratio of the g factors of the 21.7-keV level and ground level is about 0.5. Following this interpretation, the transitions between the sublevels are classified in Table I according to their positions in the experimental absorption spectra. The calculated relative transition probabilities are also given in this Table. The absorption lines corresponding to the $+\frac{5}{2}\rightarrow+\frac{3}{2}$ and the $-\frac{5}{2}\rightarrow-\frac{3}{2}$ transitions were not detected experimentally because of their very low relative intensity.

In the final analysis of the results, a possible axially symmetric quadrupole interaction was taken into account and it was assumed that each level was split according to the following expression[2]:

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

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 lines and the emission line was taken into account. The results obtained for the splitting parameters at 81°K from the analysis of the experimental peak positions are given in Table II. The sense of H_{eff} was chosen to be positive and as g_0 is positive[6] the sign of $g_0\mu_n H_{\text{eff}}$ is positive. The relative signs of the other hyperfine interactions are determined by the experimental results. Our experimental

TABLE I. Identification of observed transitions between nuclear Zeeman levels.

Peak observed	Transition between Zeeman substates	Relative intensity
"a"	$+\frac{5}{2}\rightarrow+\frac{5}{2}$	0.28
	$+\frac{3}{2}\rightarrow+\frac{1}{2}$	0.14
"b"	$+\frac{5}{2}\rightarrow+\frac{7}{2}$	1.0
	$+\frac{3}{2}\rightarrow+\frac{3}{2}$	0.48
	$+\frac{1}{2}\rightarrow-\frac{1}{2}$	0.28
"c"	$+\frac{3}{2}\rightarrow+\frac{5}{2}$	0.71
	$+\frac{1}{2}\rightarrow+\frac{1}{2}$	0.58
	$-\frac{1}{2}\rightarrow-\frac{3}{2}$	0.48
"d"	$+\frac{1}{2}\rightarrow+\frac{3}{2}$	0.48
	$-\frac{1}{2}\rightarrow-\frac{1}{2}$	0.58
	$-\frac{3}{2}\rightarrow-\frac{5}{2}$	0.71
"e"	$-\frac{5}{2}\rightarrow-\frac{7}{2}$	1.0
	$-\frac{3}{2}\rightarrow-\frac{3}{2}$	0.48
	$-\frac{1}{2}\rightarrow+\frac{1}{2}$	0.28
"f"	$-\frac{3}{2}\rightarrow-\frac{1}{2}$	0.14
	$-\frac{5}{2}\rightarrow-\frac{5}{2}$	0.28
	$-\frac{5}{2}\rightarrow+\frac{3}{2}$	0.04
	$-\frac{5}{2}\rightarrow-\frac{3}{2}$	0.04

TABLE II. Results for the hyperfine interaction and chemical shift observed in EuIG at 81°K. g_0 and Q_0 correspond to the ground state of Eu^{151}. g_1 and Q_1 correspond to the 21.7-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)	eQ_0q_{eff}/h (Mc/sec)	eQ_1q_{eff}/h (Mc/sec)
-17 ± 25	595 ± 35	315 ± 35	-240 ± 100	-320 ± 100

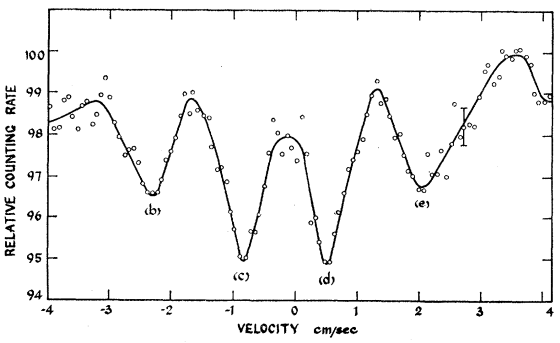

FIG. 5. The absorption by europium iron garnet at 81°K of the 21.7-keV γ ray emitted in the decay of Gd^{151}, as a function of relative velocity between source and absorber. (Range of velocities -4 to $+4$ cm/sec.)

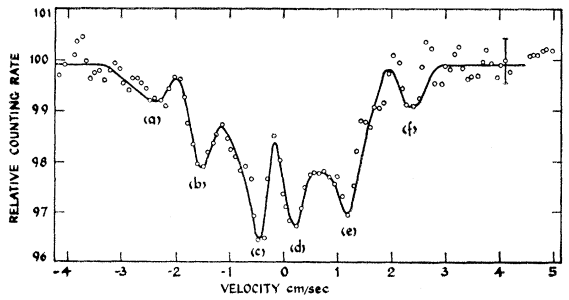

FIG. 6. The absorption by europium iron garnet at 300°K of the 21.7-keV γ ray emitted in the decay of Gd^{151}, as a function of relative velocity between source and absorber.

results do not determine the sign of H_{eff}, relative to the direction of the magnetization of the Eu sublattice in EuIG.

From these results we see that $g_1/g_0=0.53\pm0.03$ and $Q_1/Q_0=1.3\pm0.5$. The magnetic moment of the ground state was measured by Pichanick *et al.* and found to be[6] (3.419±0.004)nm and, therefore, our results show that the magnetic moment of the 21.7-keV level is (2.54±0.15)nm. The present results show that the value of H_{eff} at 81°K is $(570\pm35)\times10^3$ Oe.

The recoilless resonance absorption spectrum by EuIG at 300°K is shown in Fig. 6. The peaks "*a*," "*b*," "*c*," "*d*," "*e*," and "*f*" can be identified in this spectrum too, although they are not very well resolved. By comparing this spectrum to that obtained at 81°K, a value of 0.59±0.08 was obtained for $H_{\text{eff}}(300°\text{K})/H_{\text{eff}}(81°\text{K})$. Thus, the value of H_{eff} at 300°K is $(335\pm50)\times10^3$ Oe.

The absorption spectrum obtained at 20°K is shown in Fig. 7. The measurement was carried out in this case for a relatively narrow range of velocities with the intention of measuring accurately the separation between the peaks "*c*" and "*d*." By comparing the splitting between peaks "*c*" and "*d*" at 20°K to the corresponding splitting at 81°K, a value of (1.18±0.15) was obtained for $H_{\text{eff}}(20°\text{K})/H_{\text{eff}}(81°\text{K})$. Thus, the value of H_{eff} at 20°K was found to be $(670\pm100)\times10^3$ Oe.

Although a $\frac{3}{2}$ spin assignment to the 21.7-keV level seems very unprobable, an analysis of the results was also made assuming such an assignment. Then, a value of ~3 nm was obtained for the magnetic moment of the 21.7-keV level and the values derived for H_{eff} were about 10% lower than those given before.

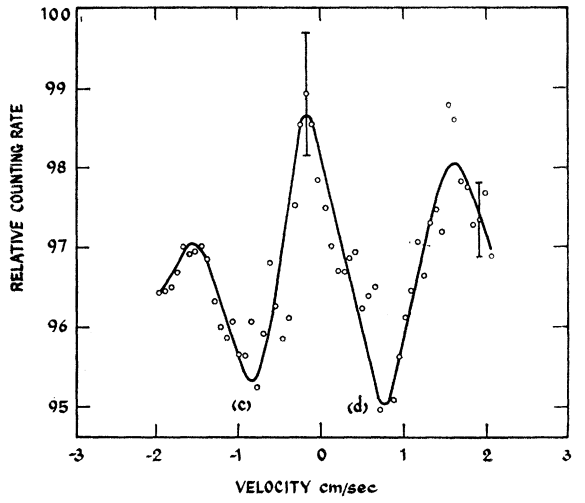

FIG. 7. The absorption by europium iron garnet at 20°K of the 21.7-keV γ ray emitted in the decay of Gd^{151}, as a function of relative velocity between source and absorber.

DISCUSSION

In Table III, a summary of the experimental values of H_{eff} and eQ_0q_{eff}, together with their theoretical values according to Ref. 5, is given. Figure 8 also displays the results for H_{eff} as a function of temperature. From the table and the figure it can be seen that the magnitude of H_{eff} and its temperature dependence are in good agreement with the theoretical predictions. The absolute values are also very close to the theoretical results. On the other hand, one must take into account the following qualifications with respect to the theoretical calculations. (a) Possible effects of core polarization were neglected (the contribution to H_{eff} due to core polarization was estimated roughly by Freeman and Watson to be -9×10^4S, where S is the spin.[12] This contribution is negative and therefore diminishes the total effective magnetic field). (b) The values of the exchange field

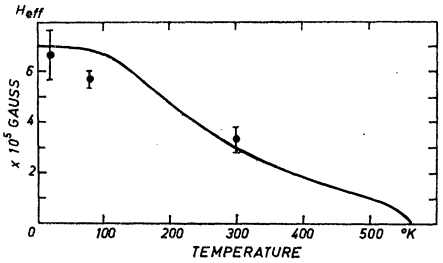

FIG. 8. Solid line represents the theoretical temperature dependence of H_{eff} acting on the Eu nuclei in EuIG. Points represent the experimental values of H_{eff} obtained in the present work.

TABLE III. Summary of experimental and theoretical values of H_{eff} and eQ_0q_{eff}.

Temperature (°K)	H_{eff}(exp) (kOe)	H_{eff} (theory) (kOe)	eQ_0q_{eff}(exp) (Mc/sec)	eQ_0q_{eff} (theory) (Mc/sec)
300	335±50	320		
81	570±35	665	−240±100	−480
20	670±100	700		

[12] A. J. Freeman and R. E. Watson, Phys. Rev. **127**, 2058 (1962).

used in these calculations are obtained in a very approximate way. (c) There is still considerable doubt concerning the true value of $\langle 1/r^3 \rangle$ to be used in the theoretical formula for the hyperfine interaction.[13]

There are large errors in the experimental value of eQ_0q_{eff}. It is interesting, however, to note, that at least the negative sign of the observed quadrupole interaction is in agreement with the theoretical estimate. As pointed out in the Introduction, the calculated quadrupole interaction assumes that the only contribution to the gradient arises from the presence of the exchange field. In Ref. 5 reasons are given to justify the assumption that other contributions are relatively small. Good agreement between experiment and theory for the value of eQ_0q_{eff} at 81°K is obtained if we assume a value of about $R=0.4$ for the Sternheimer shielding correction factor, relevant to the $4f$ electrons.

The properties of the ground and first excited states of Eu^{151} are, in many respects, very similar to those of the neighboring lower Z, odd-proton even-neutron isotopes. For example, in $_{59}Pr^{141}$ the spin of the ground state is $\frac{5}{2}$ and of the first excited state $\frac{7}{2}$. In $_{57}La^{139}$ and in $_{55}Cs^{133}$, the spin of the ground state is $\frac{7}{2}$ and of the first excited state is $\frac{5}{2}$. The value of the magnetic moment of the ground state of Eu^{151} (3.4 nm) is not too far from that of Pr^{141} (4.9 nm) and the value found in the present work for the magnetic moment of the 21.7-keV level of Eu^{151} (2.54 nm) is very close to those of the $\frac{7}{2}$ ground states of La^{139}(2.76 nm), Cs^{133} [(2.56 nm) and Cs^{135} (2.71 nm [14])]. In these cases mentioned, the $\frac{5}{2}$ spin state are identified as $d_{5/2}$ shell-model states and the $\frac{7}{2}$ spin states as $g_{7/2}$ states with positive parities. One possibility, therefore, is to interpret analogously the ground state of Eu^{151} as a $d_{5/2}$ level and the 21.7-keV level as a $g_{7/2}$ level.

On the other hand, the quadrupole moment of the ground state of Eu^{151} is about 1 b.[7] This moment corresponds to a deformation factor δ of about 0.15.[15] According to the unified model, the ground state of Eu^{151} is probably a [532, $\frac{5}{2}$], negative parity Nilsson state.[15] For such a state the calculated magnetic moment for this ground state is a function of its deformation and for $\delta=0.15$ one obtains a value of about 3.1 nm, which is not too far from the experimental value. But, there does not seem to be any individual Nilsson state of negative parity which is suitable for the 21.7-keV level. The [523, $\frac{7}{2}-$] state is too greatly separated rofm the [532, $\frac{5}{2}-$] state, and in addition the magnetic moment for the latter state corresponding to $\delta\sim0.15$, is not in agreement with the experimental value of the magnetic moment of the 21.7-keV level. Likewise, there is no pure rotational state which could occur at an energy so low as 21.7 keV. With a deformation of $\delta\sim0.15$, the lowest rotational state should be several hundred keV above the ground state.

A possible conclusion is that Nilsson's collective model does not describe in a complete and satisfactory way the properties of nuclei with small deformations and the spins and parities of the ground and 21.7-keV levels of Eu^{151} are $\frac{5}{2}+$ and $\frac{7}{2}+$, respectively.

Another possibility which has been suggested is that the 21.7-keV level is not a regular Nilsson level and not a regular collective state, but a combination of an individual particle state with some collective excitation, for example, a combination of a 2+ Sm^{150} rotational state as a core plus a proton with spin $\frac{5}{2}$.[16] We have calculated the magnetic moment of the $\frac{7}{2}$ state, assuming this model, in which we couple vectorially the magnetic moment associated with a 2+ vibration state with g factor 0.4, to a single particle $\frac{5}{2}$ spin state of magnetic moment 3.4 nm (as in the ground state of Eu^{151}) and we obtain a value of 2.9 nm close indeed to the measured value of 2.54 nm.

An abstract of an article by Barrett and Shirley describing Mössbauer resonance experiments performed on europium metal was very recently published.[17] The value given there for the magnetic moment of the 21.7-keV level of Eu^{151} is in excellent agreement with the present results. The value of H_{eff} in the metal at 4°K was found to be (264±8) kOe, about 2.5 times smaller than the field acting on Eu in EuIG.

ACKNOWLEDGMENTS

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