PHYSICAL REVIEW VOLUME 143, NUMBER 1 MARCH 1966

Gamma-Gamma Angular-Correlation Study of Tm^{169} in Lutetium Iron Garnet*†

S. Koicki,‡ A. Koicki,‡ G. T. Wood,§ and M. E. Caspari
Department of Physics and Laboratory for Research on the Structure of Matter,
University of Pennsylvania, Philadelphia, Pennsylvania

(Received 13 September 1965)

The attenuation and rotation of the integral angular correlation of the $\frac{7}{2}$(177-keV)$\frac{7}{2}$(131-keV)$\frac{3}{2}$ γ-γcascade in Tm^{169} following electron capture of Yb^{169} was measured in dilute solutions of Yb^{169} in polycrystalline samples of lutetium iron garnet at room temperature and at liquid-nitrogen temperature with and without applied magnetic field. The same measurements were also performed on the $\frac{7}{2}$(198-keV)$\frac{5}{2}$(110-keV)$\frac{3}{2}$ γ-γ cascade at liquid-nitrogen temperature. From these experiments the magnetic hyperfine field acting at the nuclei of the Tm^{3+} ions, the electronic relaxation time at the Tm^{3+} ions, and the ratio of the g factors of the $\frac{7}{2}+$ and $\frac{5}{2}+$ spin states in Tm^{169} could be deduced. At room temperature the time-averaged magnetic hyperfine field in the direction of the magnetizing field $\langle H_{\mathrm{int}}{}^z\rangle$ was 206 kOe and agreed within 10% with that calculated from molecular-field theory using the free-ion model with an exchange field of 196 kOe and neglecting inner-core exchange-polarization effects. The value of $\langle H_{\mathrm{int}}{}^z\rangle$ at liquid-nitrogen temperature averaged over all sites was 523 kOe. Neglecting quadrupole effects, the electronic relaxation time of the Tm^{3+} electrons was estimated to be 3.8×10^{-13} sec at room temperature and 1.8×10^{-12} sec at liquid-nitrogen temperature, consistent with an inverse dependence of the relaxation time on the absolute temperature. The ratio of the g factors was determined to be $g_{7/2}/g_{5/2}=1.28\pm0.12$.

I. INTRODUCTION

Much interesting information has been obtained from measurements of the magnetic hyperfine field acting at the nuclei of magnetic ions in magnetic materials. In the ferrimagnetic compounds of the garnet type ($5Fe_2O_3$, $3M_2O_3$,where M is a triply ionized rare-earth ion) the nuclei of the rare-earth ions are subjected to effective internal magnetic fields ranging from 10^5 to 10^7 Oe. These fields produce Zeeman splitting of the nuclear gamma transitions and can in certain cases be detected in Mössbauer experiments. Some time ago Caspari, Frankel, and Wood[1] (hereafter designated I) investigated the application of gamma-gamma angular-correlation experiments to the measurement of magnetic hyperfine fields and nuclear magnetic moments of short-lived excited states in nuclei of magnetic atoms or ions in ferro- or ferrimagnetic materials. Such experiments also yield information regarding the electronic relaxation time at the magnetic atom or ion and the direction of the magnetic hyperfine field at the nucleus. The method was applied to the measurement of the direction and magnitude of the magnetic hyperfine field and electronic relaxation at the Sm^{3+} ions in europium iron garnet as a function of temperature.

In this paper we present a similar study on dilute solutions of Tm^{3+} in lutetium iron garnet (LuIG). The experiments were carried out on the Tm^{3+} ions incorporated as an impurity in LuIG rather than in TmIG itself in order to facilitate the alignment of the magnetic moment of the sample in an applied magnetic field which is necessary to obtain the full rotation of the correlation. The measurements were performed in Tm^{169} following the 32-day electron capture decay of Yb^{169}. Less than 0.1 at.% of radioactive Yb^{169} was incorporated in polycrystalline samples of LuIG. A partial decay scheme[2] showing the relevant transitions is shown in Fig. 1. Two different correlations having different intermediate states, namely the 177–131-keV and the 198–110-keV gamma-gamma cascades, were measured. The intermediate states have energies of 139 and 118 keV, spins $\frac{7}{2}+$ and $\frac{5}{2}+$, and mean lifetimes of 4.6×10^{-10} sec [3] and 9×10^{-11} sec,[4] respectively. Both angular correlations have been previously measured in liquid sources and large positive anisotropies of 40 to 50% have been reported.[5] Measurements of the g factors of the $\frac{7}{2}$ and $\frac{5}{2}$ spin states in liquid sources with applied magnetic fields have been carried out by Bowman *et al.*,[6] and Ristinen and Sunyar.[7]

The initial state of both cascades is the metastable 660-nsec 316-keV level. It can therefore be stated with a very high degree of probability that the electronic configuration of the Tm^{169} ion following the electron capture of Yb^{169} has reached the ground state of the Tm^{3+} ion before the emission of the first gamma-ray transition in the cascade. The elimination of "after

---

* This work was supported by the U. S. Army Research Office (Durham) and the National Science Foundation.

† A summary of the results reported in this paper has been published in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (Institute of Physics and the Physical Society, London, 1965), p. 666.

‡ Present address: Boris Kidric Institute, Belgrade, Yugoslavia.

§ Present address: Argonne National Laboratory, Argonne, Illinois.

[1] M. E. Caspari, S. Frankel, and G. T. Wood, Phys. Rev. **127**, 1519 (1962).

[2] *Nuclear Decay Sheets*, compiled by K. Way *et al.* (National Academy of Science–National Research Council, Washington 25, D. C.).

[3] T. Sundström, P. Sparrman, J. O. Lindstrom, and J. Lindskog, Arkiv Fysik **26**, 377 (1964).

[4] A. E. Blaugrund, Y. Dar, and G. Goldring, Phys. Rev. **120**, 1328 (1960).

[5] S. Koički, J. Simič, and A. Kukoc, Nucl. Phys. **10**, 412 (1959).

[6] J. D. Bowman, J. deBoer, and F. Boehm, Nucl. Phys. **61**, 632 (1965).

[7] R. A. Ristinen and A. W. Sunyar (private communication). We are indebted to Dr. A. W. Sunyar for his permission to use the results of their measurements prior to publication.

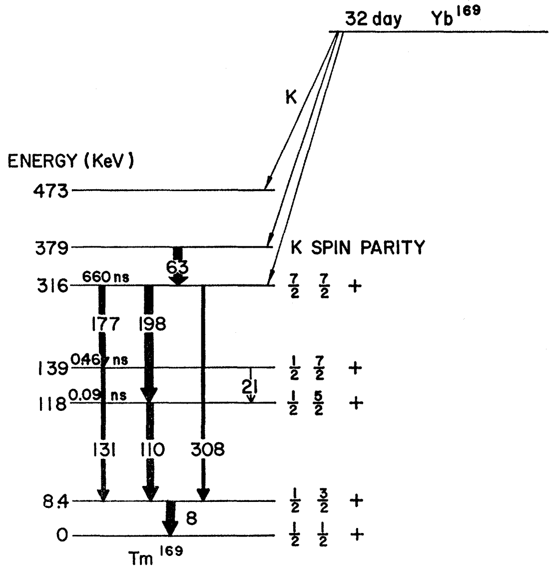

FIG. 1. The decay scheme of 32-day Yb^{169} electron capture to Tm^{169}.

effects" in this experiment also implies that the nuclear magnetic moments of both the intermediate states are subjected to the same internal magnetic field. In addition to information regarding the magnetic hyperfine field and the electronic relaxation at the Tm^{3+} ions in LuIG, the experiment can therefore be expected to yield a reliable value of the ratio of the g factors of the two short-lived intermediate states. Since the value of the internal field is greater than the applied fields readily available, our experiment can be expected to give a somewhat more accurate value of this ratio than that obtained from similar experiments in nonmagnetic materials.

Mössbauer experiments have been performed at liquid-nitrogen termperature and below in TmIG using the 8-keV gamma-ray transition in Tm^{169}.[8] The experiments were carried out without applied field and the magnetic hyperfine field acting at the nuclei of the Tm^{3+} ions was found to be significantly different for the two inequivalent Tm sites. No direct measurements of the electronic relaxation time at the Tm^{3+} ion has so far been performed.

II. SOURCE PREPARATION

Radioactive Yb^{169} of very high specific activity (about 10 mc/μg) was prepared by neutron capture in enriched (approximately 30%) samples of Yb^{168} and was used in the preparation of the garnets.

Radioactive ytterbium chloride was mixed with milligram quantities of lutetium chloride in solution. The chlorides were transformed to powdered oxides and mixed with Fe_2O_3 in stoichiometric proportions. Finally the mixed powder was pressed into cylindrically shaped cylinders (less than 1.5 mm in diam and 0.5 to 4 mm high) and sintered.

The structure of the samples was checked by x-ray analysis. Several samples prepared under somewhat different conditions of sintering time and temperature were used, but all gave the same results.

III. EXPERIMENTAL TECHNIQUES

In order to separate the two γ-γ cascades of interest it is necessary to use detectors of good resolution. Two 3-in. diam by 3-in. NaI(Tl) scintillation crystals (in integral mount) having approximately 7.4% energy resolution for Cs^{137} were used. Because of the resolution problem, the stability of the gain was also important and electronic pulse-height stabilizers were therefore employed on each channel. Coincidences were detected by a conventional transistorized fast-slow system.

The magnetic fields used to align the magnetization of the samples were derived from permanent magnets and ranged up to 18 kOe in a gap length of 7 mm. The photomultipliers were well shielded by a concentric set of Mumetal shields and residual magnetic effects were eliminated by electronic pulse-height stabilization.

The angular-correlation coincidence measurements were performed in an automated setup at several angles. The magnetic field was applied perpendicular to the counter plane, alternately in opposite directions. The field up and down measurements were fitted simultaneously by the nonlinear method of least-squares to the six parameters A, B, b_2, b_4, δ, and R in the formulas

$$\begin{aligned} W_{\mathrm{up}}(\theta) &= A[1+b_2\cos2(\theta+\Delta\theta_2+\delta) \\ &\quad +b_4\cos4(\theta+\Delta\theta_4+\delta)], \qquad (1) \\ W_{\mathrm{down}}(\theta) &= B[1+b_2\cos2(\theta-\Delta\theta_2+\delta) \\ &\quad +b_4\cos4(\theta-\Delta\theta_4+\delta)], \end{aligned}$$

where

$$\tan2\Delta\theta_2=2R,$$
$$\tan4\Delta\theta_4=4R \quad [\text{see Ref. } 9].$$

The inclusion of the phase angle δ compensates for any systematic errors in the measurement of the angle between the counters θ.

IV. EXPERIMENTAL RESULTS

1. Attenuation of the 177–131-keV Angular Correlation at Room Temperature without Magnetic Field

The attenuation of the angular correlation of the 177–131-keV cascade in Tm^{169} in LuIG was measured without external magentic field at room temperature. The experiments were carried out in extremely small samples in order to reduce scattering inside the source. A typical set of measurements of the coincidence rate as a function of the angle between the counters is shown

---

[8] R. L. Cohen, Phys. Letters 5, 177 (1963).

[9] In correlations in which there is a time-dependent wipe-out $e^{-\lambda t}$, this equation assumes that $\lambda_2=\lambda_4$. This approximation is justified since in our case, $b_4 \ll b_2$.

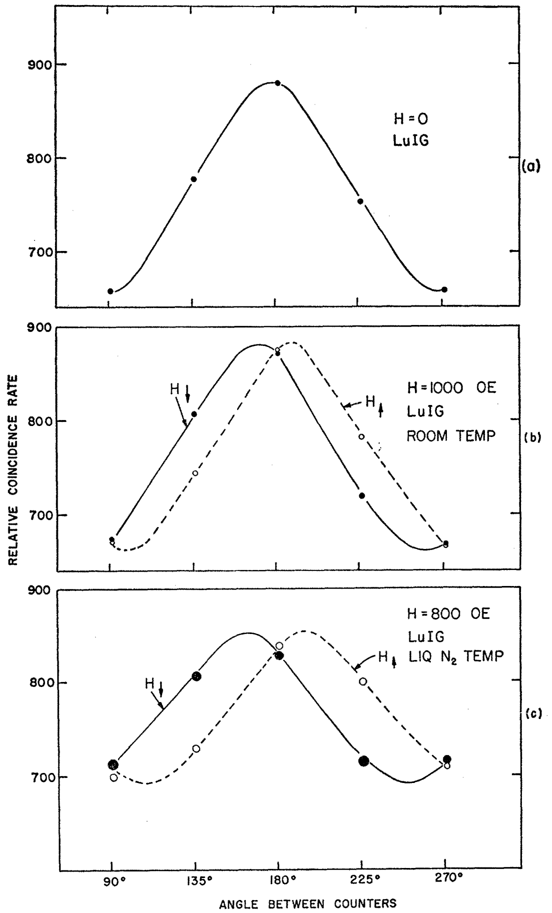

FIG. 2. Representative angular correlations of the 177–131-keV gamma-gamma cascade in Tm^{169} in polycrystalline samples of lutetium iron garnet (LuIG) for (a) room temperature without applied field, (b) room temperature with applied field, and (c) liquid-nitrogen temperature with applied field (uncorrected).

in Fig. 2(a). The angular-correlation function for this case can be written in the form $W(\theta)=1+G_2A_2P_2(\cos\theta)$, where A_2 is the unperturbed angular-correlation coefficient and G_2 is the attenuation factor. A detailed fitting of the data displayed in Fig. 2(a) to Eqs. (1) shows that the P_4 term can be neglected in agreement with other measurements.[5] The coefficient A_2 can be extracted from the electron-internal-conversion data of Kelman *et al.*[10] In this way one obtains $A_2=0.282$. To check this value of A_2 the angular correlation has been measured with the source in HCl solution. Assuming $A_2=0.282$ the attenuation factor in the liquid was 0.985 ± 0.013. Thus the value of A_2 obtained from internal-conversion data is compatible with the angular-correlation results in liquid sources if one assumes practically no attenuation in the liquid source. It was found necessary to use very small volumes of liquid source in a thin-walled glass container to reduce scattering sufficiently to achieve this small attenuation.

The measured value of G_2A_2 in the garnet without applied field was found to be 0.253 ± 0.003. Using the value of A_2 quoted above we obtain $G_2=0.90\pm0.01$.

2. Attenuation and Rotation of the 177–131-keV Angular Correlation at Room Temperature with Applied Field

The 177–131-keV gamma-gamma angular correlation in Tm^{169} in LuIG was measured at room temperature with an applied magnetic field perpendicular to the plane of the counter in order to align the magnetization of the sample. In this case one obtains a rotation of the correlation as well as an attenuation [Fig. 2(b)]. The sense of the rotation changes sign as one changes the orientation of the applied field.

It is known that the electronic relaxation time at the Tm^{3+} ions in TmIG and LuIG is very short as is the case at the other rare-earth ions in garnets (except Gd^{3+}). The analysis of the experimental results was therefore carried out in the same way as that of previous similar experiments on Sm^{152} in EuIG.[1] Under these circumstances, it was shown in I that the rotation of the angular correlation is determined by the Larmor angular-precession frequency ω of the nuclear magnetic moment of the intermediate state in the time-averaged magnetic hyperfine field $\langle H_{\text{int}}{}^z\rangle$ along the direction of the external magnetic field perpendicular to the counter plane. Hence, $\omega=g_N\mu_{nm}\langle H_{\text{int}}{}^z\rangle/\hbar$, where g_N is the g factor of the intermediate nuclear state and μ_{nm} is the nuclear magneton. Based on the assumptions that the electronic relaxation time τ_s at the Tm^{3+} ions is small compared with τ_N, the lifetime of the intermediate nuclear state, that $\langle H_{\text{int}}{}^z\rangle\ll\langle H_{\text{int}}{}^2\rangle^{1/2}$ and that $\omega_s\tau_s\ll1$, where ω_s is the hyperfine interaction energy divided by $\hbar$, one obtains the following expression for the time differential and integral angular correlations:

$$w(\theta,t)=1+A_2e^{-\lambda t}P_2[\cos(\theta+\omega t)],$$
$$W(\theta)=1+G_2'A_2P_2[\cos(\theta+\Delta\theta)].$$

In these equations λ is a coefficient which determines the time-dependent attenuation of the correlation. The attenuation factor G_2' can be written approximately (see I) as $G_2'=\alpha^{-1}(1+4R^2)^{-1/2}$, where $\alpha=1+\lambda\tau_N$ and $R=\omega\tau_N/\alpha$. The angle of rotation of the correlation $\Delta\theta=\frac{1}{2}\tan^{-1}2R$. The P_4 terms have again been neglected as in Sec. IV.1.

The time-dependent part of the attenuation factor α^{-1} can therefore be evaluated from the measured values of $\Delta\theta$ and G_2'. In Eq. (16) of I, it was shown that the attenuation of the correlation of the unrotated pattern obtained without applied field G_2 can also be expressed in terms of α and R. It is therefore in principle possible to determine α from either the measured values of G_2 or G_2'. We have chosen to use the value of G_2 in the analysis of our data since the experiments without mag-

---

[10] V. M. Kelman, R. Ya. Metskhvarishvili, B. K. Preobrazhenskii, V. A. Romanov, and V. V. Tuchkevich, Zh. Eksperim. i Teor. Fiz. 37, 639 (1959) [English transl.: Soviet Phys.—JETP 10, 456 (1960)].

netic field were carried out with smaller samples, thus reducing scattering in the source, and since such experiments are not subject to errors arising from scattering from the pole pieces of the magnets.

In Fig. 3 we have plotted the quantity $R=\omega\tau_N/\alpha$ as a function of the applied external magnetic field H_{ext}. It can be seen that R decreases with increasing H_{ext} which can be attributed to a decrease of the time-averaged magnetic hyperfine field. This effect can be explained as follows: Above the compensation temperature in rare-earth iron garnets the magnetic exchange field acting at the rare-earth ions, which is responsible for the alignment of the magnetic moment of the rare earth ions, is directed opposite to the applied field which tends to align the net magnetic moment of the iron sublattices. The alignment of the magnetic moment of the $4f$ electrons by the exchange field is therefore reduced by the direct action of the applied magnetic field on the $4f$ electrons. While the applied field is of course very much smaller than the exchange field (~200 kOe) it must be remembered that the applied field acts on both the orbital and spin magnetic moments while the exchange field acts only on the spin. The effect of the applied field relative to that of the exchange field is enhanced by the factor $g_J/(2g_J-1)$, where g_J is the Lande g factor, which can be large for the heavier rare-earth ions. For Tm^{3+}, the value of $g_J/(2g_J-1)$ is 3.5. This is the same effect which also leads to the marked magnetic field dependence of the magnetization, i.e., the large susceptibility of the garnets.[11] This magnetic field dependence of the magnetization in TmIG has recently been investigated by us in some detail[12] and has been used to obtain a value of the magnetic exchange field acting at the Tm^{3+} ions at room temperature of 1.96×10^5 Oe.

3. The Magnetic Hyperfine Field at Room Temperature

The internal magnetic field acting at the Tm^{169} nuclei for zero applied field can be determined from the extrapolated value of $R=0.160\pm0.012$ at $H_{\mathrm{ext}}=0$. The factor α is calculated from the attenuation factor G_2 obtained without applied magnetic field using[1]

$$G_2=(5\alpha)^{-1}[1+2/(1+R^2)+2/(1+4R^2)].$$

With $G_2=0.90\pm0.01$ one obtains $\alpha^{-1}=0.945\pm0.01$. In carrying out this calculation we have assumed that the value of R is the same for all the Tm^{3+} sites. Although this is known to be not true at liquid-nitrogen temperature,[8] the effect of the crystalline field on the magnetic hyperfine field at room temperature is usually not very large. The neglect of the crystalline field in this case will be checked later for internal consistency by comparing our measured value of the magnetic hyperfine field with the free-ion value.

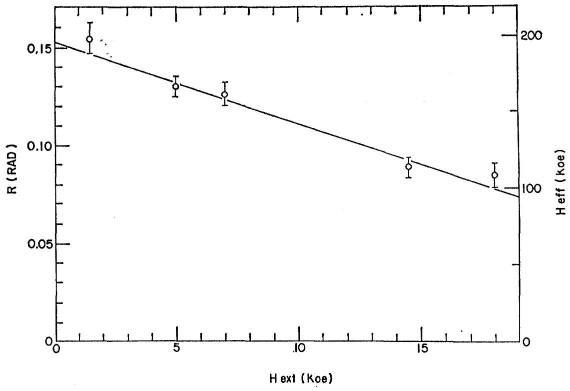

FIG. 3. Dependence of the effective magnetic field acting on the Tm^{3+} nuclei in LuIG at room temperature as function of an applied magnetic field.

Using[3] $\tau_N=(0.46\pm0.02)$ nsec we obtain, at zero applied field $\omega=R\alpha/\tau_N=(0.37\pm0.03)\times10^9$/sec. The g factor of the $\frac{7}{2}+$ state was deduced from an average value of the measurements of Bowman *et al.*[6] and Ristinen and Sunyar[7] using a paramagnetic enhancement factor of 5.08 and a mean lifetime of 4.62 nsec. The value of the g factor is then 0.37 ± 0.04. Hence, one obtains for the time-averaged value of the magnetic hyperfine field at room temperature, extrapolated to zero external field,

$$\langle H_{\mathrm{int}}{}^z\rangle_{H\mathrm{ext}=0}=\hbar\omega\mu_{nm}{}^{-1}g_N{}^{-1}=206\pm30\ \mathrm{kOe}.$$

On the right-hand side of Fig. 3 we have expressed the ordinate in terms of H_{eff}, the total field acting at the Tm nuclei, using the conversion procedure explained above and assuming that α is independent of H_{ext}. It should be noted that H_{eff} is the sum of the time-averaged magnetic hyperfine field $\langle H_{\mathrm{int}}{}^z\rangle$ and the external field H_{ext}.

In complete analogy with the dependence of the magnetization on the external field,[12] the time-averaged internal magnetic field acting at the rare-earth nuclei can be written

$$\langle H_{\mathrm{int}}{}^z\rangle=(\beta-1)[2(g_J-1)/g_JH_{\mathrm{exch}}-H_{\mathrm{ext}}],$$

where H_{exch} is the exchange field acting at the Tm^{3+} ion. Since above the compensation temperature in rare-earth iron garnets $\langle H_{\mathrm{int}}{}^z\rangle$ and H_{ext} act in opposite directions (Fig. 4) it can be seen that the H_{eff} versus H_{ext} curve is a straight line with slope $-\beta$. The slope β was experimentally determined from a least-squares fit of Fig. 3 to be 5.4 ± 0.8. In nonmagnetic substances $H_{\mathrm{exch}}=0$ and the absolute value of the parameter β defined by the relation $H_{\mathrm{eff}}=\beta H_{\mathrm{ext}}$ is the so-called paramagnetic enhancement factor commonly employed by research workers in the field of perturbed angular cor-

---

[11] R. Pauthenet, Ann. Phys. (Paris) 3, 424 (1958).

[12] M. E. Caspari, A. Koicki, S. Koicki, and G. T. Wood, Phys. Letters **11**, 195 (1964).

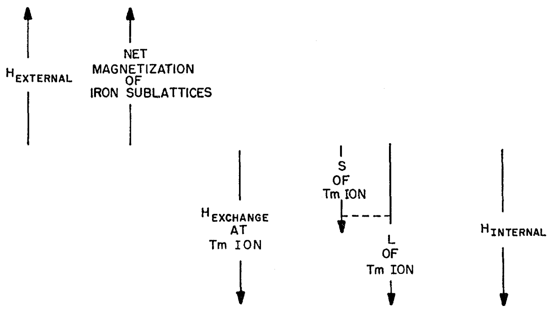

FIG. 4. The direction of the magnetic hyperfine field acting at the nuclei of the Tm^{3+} ions in LuIG relative to that of the external applied field.

relations to describe the magnetic field acting at the nuclei of paramagnetic ions.[13]

The value of the internal field extrapolated to zero applied field $\langle H_{\mathrm{int}}{}^{z}\rangle_{H_{\mathrm{ext}}=0}$, and β may be compared with theoretical predictions from molecular-field theory neglecting crystalline-field effects (free-ion approximation). Using $\langle 1/r^3\rangle = 11.47$ atomic units,[14] one obtains $\beta = 5.08$, and using our value[12] for the magnetic exchange field $H_{\mathrm{exch}} = 196$ kOe obtained from room-temperature susceptibility measurements on TmIG we calculate $\langle H_{\mathrm{int}}{}^{z}\rangle_{H_{\mathrm{ext}}=0} = -228$ kOe.

Although the measured time average of the magnetic hyperfine field at zero external field is in agreement within experimental error with this value calculated from molecular-field theory using the free-ion model, the experimental results suggest a slight reduction of $\langle H_{\mathrm{int}}{}^{z}\rangle_{H_{\mathrm{ext}}=0}$ from the free-ion value by the crystalline field. The experimental value of β is also in agreement with the free-ion value within experimental error.

The value of the magnetic exchange field determined from the intercept of the H_{eff} versus H_{ext} line with the abscissa is $H_{\mathrm{exch}} = (157 \pm 16)$ kOe, which can be compared with the value of (196 ± 14) kOe obtained from susceptibility measurements.[12] It is possible that the smaller value of the exchange field obtained from the angular-correlation measurements is caused by a systematic error due to the scattering of gamma rays from the pole pieces of the permanent magnet used to produce the external field. This scattering may result in a smaller value of the rotation of the angular correlation at the higher magnetic fields because the field was increased by placing the pole pieces closer to the source.

The sense of rotation of the angular correlation may be predicted directly from Fig. 4. It can be seen that, above the compensation temperature, if the effect of the exchange field acting at the rare-earth ions is larger than that of the applied field, the sense of the rotation will be negative. By this we mean that it is opposite in sign to that produced by the Larmor precession of a positive nuclear-magnetic moment in the applied field. The predicted sense of the rotation is based on the assumption that the g factor of the intermediate state is positive and that the magnetic hyperfine field arises mainly from the direct effect of the $4f$ electrons, the indirect effect due to the exchange polarization of the inner-core electrons being insufficient to change the sense of the magnetic hyperfine field from that produced by the direct effect. The sense of the rotation thus predicted is in agreement with our observations.

4. Attenuation and Rotation of the 177–131-keV γ-γ Angular Correlation at Liquid-Nitrogen Temperature with Applied Field

The 177–131-keV gamma-gamma angular correlation in Tm^{169} in LuIG was measured with an applied magnetic field perpendicular to the plane of the counters at liquid-nitrogen temperature. A typical result is shown in Fig. 2(c). From the amplitude of the angular correlation, and again using $A_2 = 0.282$, we obtain for the attenuation factor $G_2' = 0.646 \pm 0.019$. This value was corrected for the effects of internal gamma-ray scattering in the source, scattering from the walls of the Dewar used to cool the sample, and scattering from the pole pieces of the magnet used to produce the applied field. The pole-piece scattering correction was made by measuring the ratio of the amplitudes of the 198–110-keV γ-γ angular correlation with and without applied magnetic field in the same sample. It is assumed that, apart from external scattering effects, the amplitude of this correlation is nearly unattenuated due to the short lifetime (9×10^{-11} sec) of its intermediate state. The effect of scattering from the Dewar and of internal source scattering was found by comparing the amplitude of the 177–131-keV angular correlation from the sample as measured inside the Dewar without external field at room temperature to that obtained from a very small sized garnet without Dewar. The Dewar system was constructed in such a way that the amount of liquid nitrogen between the source and the counters was quite small and its effect on the gamma-ray scattering could be neglected. From the rotation of the 177–131-keV γ-γ angular correlation at liquid-nitrogen temperature, we found $R = 0.337 \pm 0.011$ after extrapolating to zero external field (see Sec. IV.3).

5. The Magnetic Hyperfine Field at Liquid-Nitrogen Temperature

The analysis of the results at liquid-nitrogen temperature is more difficult than that at room temperature because the influence of the crystalline field on the magnetic hyperfine field becomes very pronounced at these low temperatures. From Mössbauer measurements on TmIG at liquid-nitrogen temperature without applied field (magnetization in the [111] direction) it has been

---

[13] For this and other information regarding the technique of perturbed angular correlations the reader is referred to *Perturbed Angular Correlations*, edited by E. Karlsson, E. Matthias, and K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1964).

[14] I. Lindgren, in Ref. 13, p. 326.

shown that the ratio of the two internal fields acting at the nuclei of the Tm^{3+} ions at the two inequivalent sites is about 2 to 1.[8] In a single crystal in which the magnetization and hence the exchange field acting at the rare-earth ions is forced to lie in an arbitrary orientation relative to the crystal axes by an external field there are 6 inequivalent Tm^{3+} sites and one must expect a different hyperfine field at each site which varies with the direction of the magnetization relative to the crystal axes. In the present case, where we are dealing with experiments on powdered samples with an applied magnetic field, the direction of magnetization relative to the crystal axes is different for every crystallite in the sample and hence there will be a wide distribution of different magnetic hyperfine fields.

In the Appendix we have analyzed the effect of this distribution of hyperfine fields at various sites on the measured integral correlation. The following assumptions were made:

(1) The experimental results are to be analyzed using the formulas applicable to short electronic relaxation times at the Tm^{3+} ions as in Sec. IV.2.

(2) The value of the coefficient λ(or α) is the same at every site.

(3) The direction of the magnetic hyperfine field $\mathbf{H}_{\mathrm{int}}$ at every site is perpendicular to the counter plane. We have therefore neglected the effect of anisotropic exchange interactions.

The results of the analysis show that if $2\bar{R}<1$, which is the case in our measurements both at room and liquid-nitrogen temperature, the angle of rotation $\Delta\theta$ and the attenuation factor G_2 are

$$\Delta\theta=\tfrac{1}{2}\tan^{-1}2\bar{R}C_1,$$
$$G_2=(1+4\bar{R}^2)^{-1/2}\alpha^{-1}C_2. \qquad (2)$$

Here $\bar{R}=\bar{\omega}\tau_N/\alpha=g_N\mu_{Nm}\bar{H}_{\mathrm{eff}}\tau_N/(\hbar\alpha)$, where $\bar{H}_{\mathrm{eff}}$ is the average value of the effective field averaged over all Tm sites. C_1 and C_2 are correction factors given by

$$C_1=1+4(\langle R^2\rangle_{\mathrm{Tm}}-\langle R^3\rangle_{\mathrm{Tm}}/\bar{R})+\cdots,$$
$$C_2=1+4(\bar{R}^2-\langle R^2\rangle_{\mathrm{Tm}})+\cdots,$$

where $\langle\ \rangle_{\mathrm{Tm}}$ (like the bar) means an average over the Tm sites. It is shown in the Appendix that in the present case the correction factors C_1 and C_2 cannot be expected to differ from unity by more than 10%. Hence, if C_1 and C_2 can be neglected, the angular-correlation experiment will determine the true effective magnetic field perpendicular to the counter plane acting at the nuclei of the Tm^{3+} ions averaged over all the sites.

From the experimental determination of $\bar{R}$ we obtain $(1+4\bar{R}^2)^{-1/2}=0.83\pm0.01$ and using Eq. (2), $\alpha=1.28\pm0.04$, or $\lambda\tau_N=0.28\pm0.04$. Hence $\omega=(0.94\pm0.06)/$nsec and the time-averaged value of the internal magnetic field acting at the nuclei of the Tm^{3+} ions at liquid-nitrogen temperature averaged over all the sites is $\langle\bar{H}_{\mathrm{int}}{}^z\rangle=523\pm75$ kOe, where we have used the same value of τ_N and g_N as above. The ratio of $\langle\bar{H}_{\mathrm{int}}{}^z\rangle$ at room temperature to $\langle\bar{H}_{\mathrm{int}}{}^z\rangle$ at liquid-nitrogen temperature is 0.44 ± 0.03.

6. The Attenuation of the 177–131-keV γ-γ Angular Correlation at Liquid-Nitrogen Temperature without Applied Field

We have also measured the attenuation of the 177–131-keV γ-γ angular correlation at liquid-nitrogen temperature without an applied magnetic field. We find from the amplitude of this correlation, again using $A_2=0.282$, an attenuation factor $G_2=0.670\pm0.014$. If the magnetic hyperfine field is not the same at every site because there are two or more inequivalent sites depending on the direction of the magnetization then the value of G_2, using Eq. (16) in I, can again be expressed in terms of the value of R averaged over all sites. If we assume that $2\bar{R}<1$ and that α is the same at every site then it is shown in the Appendix that

$$G_2=(5\alpha)^{-1}[1+2/(1+\bar{R}^2)+2/(1+4\bar{R}^2)]C_3, \qquad (3)$$

where $C_3\sim1+2(\bar{R}^2-\langle R^2\rangle_{\mathrm{Tm}})+\cdots$. In our experiments the correction factor C_3 can be expected to differ from unity by not more than $2\tfrac{1}{2}\%$ (see Appendix).

If one takes the value of $\bar{R}$ to be equal to that obtained in the previous section from measurements of the correlation with an applied magnetic field, then the value of α from the attenuation measurements without applied field using Eq. (3) and neglecting C_3 becomes 1.25 ± 0.03. This is the same, within experimental error, as that obtained from the rotation and attenuation experiments with an applied field. This result is somewhat surprising since in the experiments carried out without an applied field the magnetization has the same direction in every crystallite of the sample relative to the crystal axes while in the experiments with an applied field this direction is different for every crystallite in the sample.

7. Electronic Relaxation Time at Room and Liquid-Nitrogen Temperature

It is possible to make a rough estimate of the electronic correlation time of the Tm^{3+} ion in LuIG from our data. At room temperature $\alpha=1+\lambda\tau_N=1.059\pm0.01$ (Sec. IV.3) and hence $\lambda=0.128\pm0.025$/nsec. The factor λ represents essentially the transition rate among the magnetic sublevels of the intermediate nuclear state which is induced by fluctuating hyperfine interactions set up by electronic fluctuations at the Tm^{3+} ion. If one assumes that transitions are caused by fluctuating magnetic hyperfine fields alone one can estimate roughly the electronic correlation time τ_s using Eq. (10) in I. This formula is actually general for any spin I of the intermediate state (although applied only to the case

$I=2$ in I). We have

$$\tau_s=\lambda\hbar^2/[2\langle H_{\text{int}}^2\rangle g_N^2\mu_{nm}^2], \qquad (4)$$

where $\langle H_{\text{int}}^2\rangle$ is the expectation value of the square of the magnetic hyperfine field operator.[15] This can be straightforwardly evaluated using $\langle 1/r^3\rangle=11.47$ (atomic units).[14] We find $\langle H_{\text{int}}^2\rangle=5.28\times10^{13}$ (Oe)^2 and hence $\tau_s=(3.8\pm1.0)\times10^{-13}$ sec at room temperature.

The value of the relaxation time at liquid-nitrogen temperature can be obtained from the value $\lambda\tau_N=0.248\pm0.041$ (Sec. IV.5). The ratio of the value of the electronic relaxation time at room temperature to that at liquid-nitrogen temperature is $(\tau_s)_{\text{RT}}/(\tau_s)_{\text{LN}}=(\lambda_{\text{RT}}\tau_N)/(\lambda_{\text{LN}}\tau_N)=0.21\pm0.05$. The ratio of the relaxation times is therefore compatible with an inverse dependence of the relaxation time on the absolute temperature since $T_{\text{LN}}/T_{\text{RT}}=77/300=0.257$. The value of the electronic relaxation time of the Tm^{3+} ions at liquid-nitrogen temperature is therefore estimated to be $(\tau_s)_{\text{LN}}=(1.8\pm0.4)\times10^{-12}$ sec.

The errors in the relaxation time indicated above refer to the random statistical errors in the experiment. The relaxation times are calculated from the parameter λ with the assumption that the correlation function of the hyperfine field exhibits an exponential time decay. Under these circumstances the time constant of the decay is the electronic relaxation time. In order to calculate this relaxation time the strength of the hyperfine interaction must be known. In this paper we have neglected the contribution of time-dependent quadrupole interactions. This assumption introduces certain systematic errors into the absolute value of the relaxation time evaluated from our experiment. It is expected, however, that the absolute values of the relaxation times quoted in this paper are correct at least to within a factor of 2 and that the temperature dependence of the parameter λ follows that of the relaxation times τ_s.

8. The Angular Correlation of the 198–110-keV γ-γ Cascade at Liquid-Nitrogen Temperature and the Ratio of the *g* Factors of the $\frac{7}{2}$ and $\frac{5}{2}$ Spin States

The angular correlation of the 198–110-keV γ-γ cascade in Tm^{169} in LuIG was measured at liquid-nitrogen temperature with an applied field perpendicular to the plane of the counters. The lifetime τ_N of the intermediate nuclear state has been determined to be $(9.0\pm0.4)\times10^{-11}$ sec.[4] From the rotation of the angular correlation we obtain a value of $R=0.064\pm0.004$. Because the effect of gamma-ray scattering in the source and Dewar was not evaluated for this case, the attenuation factor G_2 could not be correctly determined for this cascade. However, measurements without external applied filed yielded uncorrected values of $G_2A_2=0.271\pm0.006$ at room temperature and $G_2A_2=0.279\pm0.007$ at liquid-nitrogen temperature. Thus, on the basis of this nontemperature dependence of G_2 one can conclude that the attenuation factor G_2 is nearly unity.

Since the electronic relaxation time is more than an order of magnitude smaller than the lifetime of the intermediate state even for the fast cascade, the results are again evaluated in terms of the fast electronic relaxation procedure used throughout this paper. Straightforward considerations show that if the static interactions of the nuclear magnetic moment in the full hyperfine field were to contribute significantly to the rotation one would also expect the attenuation factor to differ significantly from unity in contradiction to our evidence.

The ratio of the *g* factors of the $\frac{5}{2}$ and $\frac{7}{2}$ spin states can be obtained from the values of $R=\omega\tau_N/\alpha$, for the two cascades. Since $\langle H_{\text{int}}^2\rangle$ is the same for both cascades, we have

$$g_{5/2}/g_{7/2}=(R_{5/2}/R_{7/2})(\tau_{7/2}/\tau_{5/2})(\alpha_{5/2}/\alpha_{7/2})=0.756\alpha_{5/2}. \qquad (5)$$

Since $\alpha_{5/2}$ is close to unity $g_{5/2}/g_{7/2}\approx0.756$. A better approximation is obtained by using this approximate *g*-factor ratio to calculate $\alpha_{5/2}$ from $\alpha_{7/2}$ using Eq. (3) in Sec. IV.6. Since the values of τ_s and $\langle H_{\text{int}}^2\rangle$ are the same for both cascades we may write at liquid-nitrogen temperature $\alpha_{5/2}-1=(\alpha_{7/2}-1)(g_{5/2}^2/g_{7/2}^2)(\tau_{5/2}/\tau_{7/2})=0.032$.

Hence $\alpha_{5/2}=1.032$ which is compatible with our experimental results. With this correction for $\alpha_{5/2}$ we obtain from Eq. (5) $g_{7/2}/g_{5/2}=1.28\pm0.12$. From this ratio we obtain a value for the *g* factor of the $\frac{5}{2}$ spin state of 0.29 ± 0.05.

V. DISCUSSION AND CONCLUSION

The value of the time-averaged magnetic hyperfine field acting at the nuclei of the Tm^{3+} ions incorporated as dilute impurities in polycrystalline samples of LuIG at room temperature was determined to be 206 ± 30 kOe. This value agrees to within 10% with that calculated from molecular-field theory neglecting crystalline-field effects and indirect contributions to the magnetic hyperfine field due to the exchange polarization of the inner-core *s* electrons by the 4*f*-electron spins. This result is in agreement with the well-known fact that the magnetic properties of rare-earth ions in rare-earth iron garnets are not greatly influenced by crystalline-field effects at room temperature.

At liquid-nitrogen temperature the time-averaged magnetic hyperfine field acting at the nuclei of the Tm^{3+} ions can be expected to be different for ions at the various inequivalent sites due to crystalline-field effects. The measured value of the internal field at liquid-nitrogen temperature, averaged over all sites, was (523 ± 75) kOe. As expected, this value differs considerably from that calculated on the basis of the free-ion model.

Cohen[8] has determined the internal field acting at the

---

[15] A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. (London) **A205**, 135 (1951).

nuclei of the Tm^{3+} ions at liquid-nitrogen temperature in TmIG from Mössbauer experiments and has obtained the values of 1240 and 570 kOe for the fields acting at the two inequivalent sites, respectively. It can be seen that the lower value is compatible with our experimental results but that the arithmetical mean of the two fields is not. The two experiments may however not be comparable, since the angular-correlation results were deduced from experiments in polycrystalline samples in which the magnetization was aligned by an external field while the Mössbauer experiments were carried out without external field so that the magnetization of each crystallite would lie in the [111] direction. The average value of the magnetic hyperfine field may therefore be different in the two experiments (see Sec. IV.5). However, the attenuations of the angular correlation measured with and without external field are consistent if one assumes that the hyperfine interaction constant and the electronic relaxation time τ_s(i.e., α) are the same at every site and that the average internal field is the same in the two cases.

The following possible explanations of this observation can be suggested:

1. The value of the internal field does not vary greatly among the various inequivalent sites in contradiction to the Mössbauer experiments.
2. It has been reported[16] that the easy axis of magnetization in LuIG at liquid-nitrogen temperature lies in the [100] direction instead of the [111] direction as in TmIG. Since our experiments were carried out in dilute solutions of Tm^{3+} in LuIG and the Mössbauer experiments were made in TmIG, the average values of the internal fields may not be the same even if both experiments are carried out without external fields. It is then possible that the average values of the hyperfine field acting at the nuclei of the Tm^{3+} ions in LuIG are accidentally the same in experiments with and without applied fields.
3. The value of α differs significantly among inequivalent Tm^{3+} sites and the agreement between the attenuation factors as evaluated in Sec. IV.5 may be purely accidental.

In order to resolve the questions raised above, more complete theoretical and experimental results of the crystalline-field effects in TmIG must become available. Further clarification of this problem may come from a Mössbauer study of the magnetic hyperfine field at Tm nuclei in LuIG with and without applied field.

The ratio of the average internal fields at room and liquid-nitrogen temperature was 0.44±0.03. This value is in agreement with the corresponding ratio of the magnetic moments of the Tm^{3+} ions in TmIG of 0.41 which was obtained by Perel and Schieber[17] from magnetization measurements in which the magnetization was also aligned by an applied field. This agreement is to be expected since in Tm^{3+} the expectation values of both the hyperfine field and the magnetic moment can be determined from matrix elements within the ground-state multiplet alone.[18]

In a recent paper[12] we have interpreted the increase of the magnetization as a function of an applied field in TmIG as the result of a competition between the action of the exchange field and the applied field on the alignment of the magnetization of the $4f$ electrons at the rare-earth ions. This hypothesis is strikingly borne out by the observed *decrease* of the magnetic hyperfine field as a function of the external field in an angular-correlation experiment. Just as with this decrease in the hyperfine field, the large susceptibility of TmIG has its origin in the decrease of the Tm^{3+} ion sublattice magtization with applied field and should not be attributed to difficulties in achieving saturation in the samples.

The electronic relaxation time at the Tm^{3+} ions was estimated from our experiments to be 4×10^{-13} sec at room temperature and 2×10^{-12} sec at liquid-nitrogen temperature. These values are consistent with an inverse dependence of the relaxation time on the absolute temperature, in agreement with measurements on the temperature dependence of ferrimagnetic resonance linewidths of Tm substituted YIG by Miyadai.[19,20] The absolute value of the relaxation time at the Tm^{3+} ions is about an order of magnitude smaller than the corresponding value at the Sm^{3+} ions in EuIG.[1] At present the experimental results on the ferrimagnetic resonance linewidth of Tm-doped garnets as well as our knowledge of the fine structure of the Tm^{3+} ion in garnets is not sufficient to determine whether the small value of the relaxation time is compatible with the presently accepted "slow"-relaxation theory[21] of the linewidth.

The ratio of the g factors of the $\frac{7}{2}+$ and $\frac{5}{2}+$ spin states in Tm^{169} was measured to be 1.28±0.12. This ratio is in agreement, within experimental error, with the values obtained by Bowman *et al.*[6] and by Ristinen and Sunyar[7] who have used applied magnetic fields in liquid sources. Their values are 1.43±0.35 and 1.19 ±0.19, respectively, where in order to make this comparison we have adjusted their values by taking $\tau_{7/2}=0.46$ nsec and $\beta=5.08$, corresponding to our analysis. Bowman *et al.*[6] have calculated the magnetic moment of the $\frac{5}{2}+$ and $\frac{7}{2}+$ spin states from the experimentally known magnetic moments $\mu_{1/2}$ and $\mu_{3/2}$, the lifetime $\tau_{5/2}$, and the reduced transition rate $B(E2, \frac{1}{2}\rightarrow\frac{5}{2})$ by applying the formulas of the rotational

---

[16] Ernest R. Czerlinsky and Ferdinand Euler, *Rare Earth Research*, edited by Eugene V. Kleiber (The Macmillan Company, New York, 1961), p. 113.

[17] J. Perel and M. Schieber, Japan. J. Appl. Phys. **1**, 243 (1962).

[18] S. Ofer, E. Segal, I. Nowik, E. R. Bauminger, L. Grodzins, A. J. Freeman, and M. Schieber, Phys. Rev. **137**, A627 (1965).

[19] T. Miyadai, J. Phys. Soc. Japan **17**, 1899 (1962).

[20] P. E. Seiden, Phys. Rev. **133**, A728 (1964).

[21] J. H. van Vleck, in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (Institute of Physics and the Physical Society, London, 1965), p. 401.

model to the $K=\frac{1}{2}$ ground band in Tm^{169}. Their values agree within experimental error with the present measurements of the g-factor ratio.

APPENDIX

It is desired to obtain an expression for the integral correlation if the internal magnetic field is different at different nuclear sites. We assume that the correlation is under the influence of time-dependent perturbations which are described by the factor λ. The value of λ is assumed to be the same at every site. The internal field at every site is assumed to point in the direction perpendicular to the counter plane. Under these circumstances the angular-correlation function is

$$W(\theta)\cong 1+(B_2/\alpha)\frac{1}{N}\sum_{i=1}^{N}\frac{\cos 2(\theta+\Delta\theta_i)}{(1+4R_i^2)^{1/2}},$$

where the summation is over the N nuclear sites contributing to the angular correlation. $\Delta\theta_i$ is defined by

$$\Delta\theta_i=\tfrac{1}{2}\tan^{-1}2R_i,$$

where $R_i=2\omega_i\tau_N/(1+\lambda\tau_N)$ and B_2 is the theoretical angular-correlation coefficient.

The equation above may be expressed in the form

$$W(\theta)=1+G_2B_2\cos 2(\theta+\Delta\theta),$$

where $\Delta\theta$ is the rotation of the angular-correlation pattern and G_2 is the attenuation coefficient. Provided all $2R_i<1$ we may obtain the following expansion relating $\Delta\theta$ and G_2 to the R_i:

$$\tan 2\Delta\theta=2\bar{R}C_1,$$
$$G_2=(1+4\bar{R}^2)^{-1/2}\alpha^{-1}C_2,$$

where

$$C_1\cong 1+4(\langle R^2\rangle_{\text{Tm}}-\langle R^3\rangle_{\text{Tm}}/\bar{R})+\cdots,$$
$$C_2\cong 1+4(\bar{R}^2-\langle R^2\rangle_{\text{Tm}})+\cdots,$$

where the nth moment of R is related to the corresponding moment of $\langle H_{\text{int}}{}^z\rangle$ through

$$\langle R^n\rangle_{\text{Tm}}\equiv\frac{1}{N}\sum_{i=1}^{N}R_i{}^n=(g_N\mu_{nm}\tau_N/\hbar)^n\langle\langle H_{\text{int}}{}^z\rangle^n\rangle_{\text{Tm}}$$

and

$$\langle R\rangle_{\text{Tm}}=\bar{R}.$$

In order to estimate the importance of the correction factors C_1 and C_2 in our experiment we consider the action of two fields at the nuclei situated at two inequivalent equally populated sites. The ratio of the two fields is taken to be 2:1 to correspond to the actual observations of the two fields in Mössbauer experiments[8] at liquid-nitrogen temperature without applied field. According to the Mössbauer results, this should apply to angular-correlation experiments in a single crystal of TmIG in which the [111] direction points perpendicular to the angular-correlation plane. It is expected that the deviation of the fields at the various sites in the actual experiments discussed in this paper would be no larger than this. In this case if $2R_1=R_2$, $\bar{R}=\frac{3}{2}R_1$, $\langle R^2\rangle_{\text{av}}=(10/9)\bar{R}^2$, and $\langle R^3\rangle_{\text{av}}=\frac{4}{3}\bar{R}^3$. Consequently, $C_1=1+(8/9)\bar{R}^2$ and $C_2=1+(4/9)\bar{R}^2$. If we take $\bar{R}=0.33$ corresponding to our experimental results under the assumption that $C_1=1$, we obtain for the corrected values $C_1=1.10$ and $C_2=1.05$. Hence, the neglect of C_1 and C_2 in Eq. (2) [Sec. IV.5] should not affect the results by more than 10%.

The attenuation of the angular correlation in experiments without applied field in which the magnetic hyperfine field is different at various nuclear sites can be analyzed in a similar manner.

In this case

$$G_2=(5\alpha)^{-1}\left[1+\frac{1}{N}\sum_{i=1}^{N}\left(\frac{2}{1+R_i^2}\right)+\frac{1}{N}\sum_{i=1}^{N}\frac{2}{1+4R_i^2}\right],$$

which expands to

$$G_2=(5\alpha)^{-1}\left(1+\frac{2}{1+\bar{R}^2}+\frac{2}{1+4\bar{R}^2}\right)C_3,$$

where $C_3=1+2(\bar{R}^2-\langle R^2\rangle_{\text{Tm}})+\cdots$.

The relations between $\bar{R}$, $\langle R^2\rangle_{\text{Tm}}$, and $\langle R^3\rangle_{\text{Tm}}$ noted above apply directly to polycrystalline samples without applied field. In this case we obtain $C_3\cong 1.025$ for $\bar{R}=0.33$.