and

$$\chi=1+\Gamma/|\omega|\,.$$

In the same way, we obtain

$$\tilde{G}_\omega(k,\alpha)\tilde{G}_{-\omega}(-k,-\beta)=\delta_{\alpha\beta}S_\omega(k,\alpha)$$

and

$$\frac{1}{|\lambda|}=T_c\sum_\omega\sum_{kk'}\Big[S_\omega(k,k')-n_I(2l+1)\frac{U}{1+UT_c\sum_{\omega j}S_\omega(i,j)}\times T_c\sum_{\omega'}S_{\omega'}(k,i)S_\omega(i,k')\Big].$$

Now, we have

$$\sum_k^D\sum_{k'}^D S_\omega(k,k')\sim\frac{2g(0)}{|\omega|}\tan^{-1}\frac{\omega_D}{|\omega|}-(2l+1)n_I\times\frac{\Gamma}{|\omega|[E^2+(|\omega|+\Gamma)^2]}+O(I^2)n\,,$$

$$\sum_k^D S_\omega(k,d)\sim\frac{1}{|\omega|[E^2+(|\omega|+\Gamma)^2]}+O(n_I)\,,$$

$$\sum_j S_\omega(i,j)\sim\frac{1}{E^2+(|\omega|+\Gamma)^2}+O(n_I)\,.$$

In that case, the relaxation time $\tau_R(0)$ is given by

$$\frac{1}{2\tau_R(0)}=\frac{n_I(2l+1)}{\pi g(0)}\frac{\Gamma^2}{E^2+\Gamma^2}\,.$$

We can write, exactly as in the nondegenerate case,

$$\frac{\Delta T_c}{T_{c0}}\simeq-\alpha\frac{\tau_d}{\tau_R(0)}[1+\alpha g_d(0)U_{\rm eff}]\,.$$

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Sublattice Magnetization in Yttrium and Lutetium Iron Garnets*

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The temperature dependence of the a and d sublattice magnetization in yttrium iron garnet (YIG) and lutetium iron garnet (LuIG) has been observed by means of the NMR of Fe^{57} between 4 and 373°K, with special emphasis on the temperature range where the spin-wave theory is expected to hold. An analysis of these results in terms of the spin reversals due to acoustical and optical spin-wave modes has been carried out. From a comparison of theory and experiment in YIG, the value of the dispersion parameter $D=30.0\pm0.6$ cm^{-1} is obtained. A reasonable estimate of the separate exchange parameters is $J_{ad}=22.5\pm1$ cm^{-1}, $J_{dd}=2.0\pm0.5$ cm^{-1}, and $J_{aa}=0.5\pm0.5$ cm^{-1}, subject to the constraint that their linear combination gives $D=30$ cm^{-1}. The agreement between experiment and theory is improved when a small transferred hyperfine interaction from the d sites to the a sites (and vice versa) is postulated. For LuIG one obtains a value $D=27.3$ cm^{-1}. The values of D and of the exchange parameters J_{ij} for YIG and LuIG are compared with those from other experiments. In YIG, the magnetic field dependence of the sublattice magnetizations has been observed up to 10 kG at 63 and 77°K, and there is good agreement with the results from spin-wave theory. It is believed that this is the first such investigation in a ferrimagnet. In the course of these experiments, the gyromagnetic ratio of Fe^{57} was determined to be $\gamma/2\pi=137.4\pm0.2$ Hz/G. In LuIG a broad spectrum of resonances between about 40 and 76 MHz was observed at 4.2°K. This is attributed to the resonance of Lu^{175} and Lu^{176} in a transferred hyperfine field (~100 kG) broadened by quadrupole interaction. In Y^{89} in YIG, this transferred field seems to be smaller than ~10 kG, from NMR and specific-heat evidence.

I. INTRODUCTION

IN recent years there have been several accurate investigations of the sublattice and total magnetizations in ordered magnetic materials at low temperatures.[1–3] The purpose was to test the spin-wave theory and to derive the respective exchange parameters in the material under investigation. In this paper

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we shall present a detailed study of the sublattice magnetization in yttrium iron garnet in (YIG) and lutetium iron garnet (LuIG) by means of the NMR of Fe^{57} and the interpretation in terms of a spin-wave analysis.

There are several reasons that make YIG and LuIG particularly interesting choices for such an investigation. First, these compounds are considered ideal examples of ferrimagnetic insulators with small anisotropy, and YIG in particular has been the subject of much research.[4] Although the unit cell is rather complicated, an analysis of the spin-wave modes, both acoustical and optical, has been carried out.[5] Therefore it is possible by means of certain types of measurements to compare theory and experiment. The information to be expected from sublattice magnetization is then the following: (1) an accurate determination of the spin-wave dispersion parameter D, which can be compared with values obtained from other experiments, such as specific heat[6-8] and microwave instability.[9] This seemed particularly important because of inconsistencies between the results from these methods; (2) a moderately accurate determination of the separate exchange parameters J_{aa}, J_{dd}, and J_{ad} can be obtained.

The sharpness of the Fe^{57} NMR in the domains in YIG makes it possible to detect small frequency changes with temperature (approximately ± 1 kHz out of 76 MHz on the a sites). Therefore a detailed analysis of the changes in the sublattice magnetization with temperature can be made.

As will be mentioned in this paper, the comparison of theory and experiments in YIG shows some evidence of hyperfine field transfer effects from the Fe ions on the d sublattice to those of the a sublattice and vice versa (Sec. VIA). Furthermore, in the course of magnetization measurements in applied fields, a determination of the gyromagnetic ratio of Fe^{57} gives a value in good agreement with recent ENDOR experiments (Sec. VIC). The work to be reported here has been presented elsewhere in more preliminary fashion.[10]

II. PREVIOUS WORK ON THE MAGNETIZATION OF YIG

The technique of NMR is a very convenient way to measure sublattice magnetizations. The first such measurements in YIG were carried out by Robert,[11] who, however, was not concerned with a detailed study of the magnetization, but showed that his results were consistent with the total magnetization data of Pauthenet.[4] The more recent work of Boyd, Smart, and Moruzzi[12] aimed at an analysis of the sublattice magnetization of both YIG and GdIG in terms of a Weiss molecular-field model at high temperatures. The work of Buyle-Bodin and LeDang Khoi[13] consisted of a study of YIG, LuIG, and several other garnets and a comparison of their respective NMR frequencies of Fe^{57}. Litster and Benedek[14] presented (simultaneously with the present authors[10]) the first detailed investigation of the sublattice magnetization in the spin-wave region, their aim being also to determine the dispersion constant D. In their analysis, however, they did not allow for the dipolar interaction term in the spin-wave spectrum,[15] which somewhat influences the determination of the parameter D. As will be shown in Sec. VI, there is also some difference in the experimental data below 100°K obtained by the two groups of workers.

Precise measurements of the total magnetization in YIG were carried out by Solt[16] below 55°K. The best available total magnetization data above this temperature are those of Geller *et al.*[17] and of Anderson.[18] These data will be compared in Sec. VIB, with results derived from NMR data.

There has been relatively little research done on LuIG, which may be due to the greater difficulty in preparing pure samples. Nevertheless, some NMR data at certain temperatures exist,[13] as well as good magnetization data.[17]

III. THE EXCHANGE PARAMETERS IN YIG AND LuIG

There have been several attempts in estimating the exchange parameters J_{aa}, J_{ad}, and J_{dd} from magnetic measurements. Anderson's Weiss-molecular-field (WMF) analysis[18,19] probably overestimates the intrasublattice exchange. Harris[5] analyzed the magnetization data of Solt using the spin-wave theory. To fit

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[19] (a) E. E. Anderson, in *Proceedings of the International Conference on Mangetism, Nottingham, 1964* (Institute of Physics and The Physical Society, London, 1965), p. 660. It is known that the WMF model overestimates the intrasublattice exchange parameters. (See, for instance, J. S. Smart, in Ref. 3, Vol. I, p. 63.) (b) However, a WMF analysis of these parameters from magnetization measurements of $(YGa_xFe_{1-x})G$ by B. Lüthi [Phys. Rev. **148**, 519 (1966)] indicates that J_{aa} and J_{dd} are small.

the data, he used the value of $D=27$ cm^{-1} from specific heats. Had he used the larger value of D from microwave instabilities, he would have found a smaller ratio of J_{aa}/J_{ad} and J_{dd}/J_{ad} than he did. Wojtowicz[20] has determined J_{dd} and J_{ad} from a high-temperature expansion analysis of Aleonard's[21] susceptibility data. He concluded that $J_{dd}\simeq 0.1J_{ad}$. From other evidence he estimated $J_{aa}\approx 0.1J_{dd}$. Unfortunately, Aleonard's data differ by about 10–15% from the more recent data of Anderson[18] in the overlapping temperature region. Therefore, there is still some doubt about the correct values of the exchange parameters, and this, in part, motivated the present authors into making a fresh attempt to determine them.

IV. THEORETICAL SURVEY

We give the relevant results of the spin-wave theory, considering at first only the acoustical modes and later the optical modes. Also, we discuss the problem of deducing the sublattice magnetization from NMR data.

A. Acoustical Mode

Recently, Harris[22] has derived the dispersion relation of the acoustical mode in YIG and LuIG using the method of Holstein and Primakoff.[15] Including the effects of the terms of order k^2 and k^4 in the dispersion curve, the average $S_i(T)$ of the spin S_i for the ith sublattice is then calculated to be

$$1-\frac{S_i(T)}{S_i}=\xi_i\delta+\tfrac{2}{5}\zeta(\tfrac{3}{2})\left(\frac{k_BT}{16\pi D}\right)^{3/2}\times\left[f_M-\frac{\zeta(\frac{5}{2})}{\zeta(\frac{3}{2})}\frac{k_BT}{D^2}(6\xi_iAD+3F+15E)\right]. \quad (1)$$

Here the coefficients E and F are those given in a previous paper,[5] and

$$D=(40J_{aa}-25J_{ad}+15J_{dd})/16\,,$$
$$A=(-48J_{aa}+25J_{ad}-12J_{dd})/64J_{ad}\,, \quad (2)$$

$\xi_a=1$, $\xi_d=\frac{2}{3}$, and δ is the zero-point deviation. The $\zeta(n)$ are the Riemann functions. The effect of the magnetic field and dipolar interactions are represented by the quantity

$$f_M=1-\frac{\sqrt{\pi}}{2\zeta(\frac{3}{2})}\left[\left(\frac{9g\beta H_{\rm eff}}{k_BT}\right)^{1/2}+(H_{\rm eff}+4\pi M)\times\left(\frac{g\beta}{4Mk_BT}\right)^{1/2}\sin^{-1}\left(\frac{4\pi M}{H_{\rm eff}+4\pi M}\right)^{1/2}\right], \quad (3)$$

which multiplies the term in $T^{3/2}$. In this derivation, one neglects higher-order terms in $g\beta H/k_BT$ and $4\pi g\beta M/k_BT$. The field $H_{\rm eff}$ is defined by

$$H_{\rm eff}=H_0+H_A+H_D\,, \quad (4)$$

where H_0 is the externally applied field, $H_A=\frac{4}{3}(K/M)$ is the anisotropy field, and H_D is the demagnetizing field. One has[23] for a spherical sample:

$$H_D=-H_0 \quad \text{for} \quad H_0\lesssim\tfrac{4}{3}\pi M\,,$$
$$H_D=-\tfrac{4}{3}\pi M \quad \text{for} \quad H_0\gtrsim\tfrac{4}{3}\pi M\,. \quad (5)$$

Inclusion of terms of order $\mathbf{k}^6$ in the dispersion relation leads to an additional term $\varphi_iT^{7/2}$ where φ_i is an extremely lengthy sum of various combinations of the exchange integrals for which Harris[22] has tabulated numerical values.

A more simple dispersion relation than that of Holstein and Primakoff is obtained if the angular dependence of the dipolar "energy gap" is averaged over all directions. This corresponds to a Lorentz field-type approximation.[24] Thus one obtains a dispersion relation of the form

$$\hbar\omega=\lambda_0+Da^2k^2+\sum_{l,m}\alpha_{lm}k_l^2k_m^2+\sum_{l,m,n}\alpha_{lmn}k_l^2k_m^2k_n^2. \quad (6)$$

Walker[25] has derived the partition function for a ferromagnet with such a dispersion law. The corresponding magnetization can then be written in the form

$$1-\frac{S(T)}{S}=AT^{3/2}\frac{F(\frac{3}{2},\chi)}{\zeta(\frac{3}{2})}+BT^{5/2}\frac{F(\frac{5}{2},\chi)}{\zeta(\frac{5}{2})}+CT^{7/2}\frac{F(\frac{7}{2},\chi)}{\zeta(\frac{7}{2})}, \quad (7)$$

where $\chi=(g\beta H_{\rm eff}+H_L)/k_BT$ and where $F(\sigma,\chi)$ are the Bose-Einstein integral functions discussed and evaluated by Robinson.[26] For small values of χ, Eq. (5) becomes

$$1-\frac{S(T)}{S}=AT^{3/2}(1-1.354\chi^{1/2}+0.56\chi)+BT^{5/2}(1-1.98\chi)+CT^{7/2}(1-1.17\chi). \quad (8)$$

It should be noted that there is no zero-point deviation for a ferromagnet. If we now assume that Eq. (7) is

[20] P. Wojtowicz, Ref. 19(a), p. 11.
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[23] This assumption is verified experimentally for YIG at 4.2°K, where the spontaneous magnetization is very close to its maximum value. When the field at a Fe^{57} nucleus in a domain is observed as a function of external applied field, it stays constant until H_0 has reached a value of approximately $\frac{4}{3}\pi M$. Then it changes linearly with H_0.
[24] S. M. Charap and E. L. Boyd [Phys. Rev. **133**, A811 (1964)] have discussed in detail various approximations that can be made for calculating the effect of dipolar interaction in a ferromagnet. See also Ref. 2.
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[26] J. E. Robinson, Phys. Rev. **83**, 678 (1951).

valid for sublattices of a ferrimagnet, account must be taken of the zero-point deviation in the same way as in Eq. (1). Then the terms A, B, and C can be identified with the coefficients for the terms in $T^{3/2}$, $T^{5/2}$, and $T^{7/2}$ calculated by Harris.[22]

For small χ, the Holstein-Primakoff[15] treatment gives, of course, the correct answer for the magnetization. However Eq. (8) is useful because it includes the effect of terms linear in χ, which appear at higher values for χ.

In the experimental part, Sec. VI, both approaches will be compared with magnetization data at constant temperature as a function of applied field.

B. The Optical Modes

The energies of the optical modes have been calculated analytically as a function of the exchange parameters by Douglass[27] at the center and the edge of the Brillouin zone. Harris[5] has made numerical calculations of the dispersion curves for $\mathbf{k}$ lying along the [111] directions. For a not unlikely ratio of the parameters, namely, $J_{aa}/J_{dd}\equiv X=0$, $J_{dd}/J_{ad}\equiv Y=0.2$ he finds that only the optical mode $E=-10J_{ad}$ (for $\mathbf{k}=0$) is strongly k-dependent. Since this mode lies lowest, a careful numerical estimate of its effect on the magnetization has to be made. The dispersion relation for this mode is calculated to be

$$\hbar\omega=-10J_{ad}+\tfrac{5}{16}(12J_{aa}+2J_{dd}-5J_{ad})a^2k^2 + \text{terms in } k^4\cdots. \quad (9)$$

The spin-wave amplitudes of the optical modes on the different sublattices can be calculated from the secular equations of Dreyfus[28] (Table II of his reference). In Table I we give the energies of the modes for $\mathbf{k}=0$ and their respective amplitudes in the a and d sublattices and the total magnetization. For the total magnetization, the results are consistent with those of Douglass.[27] We note that

(1) In each sublattice, the spin-wave modes decrease the magnetization. However, since the magnetization on the a sublattice is opposite to the total one, a spin reversal on the a site increases the total magnetization.

(2) The spin-wave reversals for modes 2–14 are roughly $\mathbf{k}$-independent. The reversals for the modes 15–20 on the sublattices might be more $\mathbf{k}$-dependent than those of the other modes and this matter needs further study. This makes the calculation of the change in magnetization more uncertain for the sublattices than for the total magnetization especially because modes 18–20 have a low energy and give a significant contribution to $\{1-[S_i(T)/S_i]\}$.

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TABLE I. The energies and spin reversals at $\mathbf{k}=0$ of the acoustical and optical modes in YIG and LuIG. Here $u=17J_{ad}^2-20J_{ad}\times(J_{dd}+2J_{aa})+4(J_{dd}+2J_{aa})^2$; $2\epsilon=\{-1+\rho(\rho^2-200)^{-1/2}\}$; $\rho=25-20X-10Y$; $n_{a\mu}$, $n_{d\mu}$=spin reversal per mode μ per unit cell on the a or on the d sublattice; and n_μ=total spin reversal per unit cell. Mode No. 1 is the acoustic mode. The others are optical modes.

Mode No. μ	Energy E_μ	Spin reversal per mode $n_{a\mu}$	$n_{d\mu}$	n_μ
1	0	-2	3	1
2	$-10J_{ad}$	-3	2	-1
3,4,5	$40J_{aa}-30J_{ad}$	-1	0	-1
6	$80J_{aa}-30J_{ad}$	-1	0	-1
7,8	$10J_{dd}-20J_{ad}$	0	1	1
9,10,11	$20J_{dd}-20J_{ad}$	0	1	1
12,13	$30J_{dd}-20J_{ad}$	0	1	1
14	$40J_{dd}-20J_{ad}$	0	1	1
15,16,17	$5u^{1/2}-5J_{ad}$ $+20J_{aa}-10J_{dd}$	$-(1+\epsilon)$	ϵ	-1
18,19,20	$5u^{1/2}+5J_{ad}$ $-20J_{aa}+10J_{dd}$	$-\epsilon$	$(1+\epsilon)$	1

The change in sublattice magnetization due to optical modes is now calculated as shown for instance in Eq. (24) of Ref. 5. We have then, in summary for the sublattices, in terms of the noninteracting acoustical and optical spin-wave modes:

$$\frac{S_i(1-\xi_i\delta)-S_i(T)}{S_i}=\alpha f_M T^{3/2}+\beta_i T^{5/2}+\varphi_i T^{7/2} + \sum \text{optical-mode reversals}. \quad (10)$$

Arguments presented in the Appendix show that up to about 80°K the contribution from the effect of spin-wave interactions is less than 1% of the total change. A representative example for the relative contributions of the spin reversal is given in Table II for the parameter $D=30$ cm^{-1}, $J_{aa}/J_{dd}=0$, $J_{dd}/J_{ad}=0.1$. In view of uncertainties in the calculation of the optical modes, the comparison of experiment with theory will not be made beyond about 70°K.

C. The Sublattice Magnetization as Obtained from NMR Data

One commonly assumes the hfs field $H_i(T)$ at the nucleus i (after subtracting the Lorentz and dipolar fields) to be proportional to the time average $S_i(T)$ of the ionic spin S_i. What therefore can be measured is the ratio

$$\frac{\Delta\nu_i}{\nu_i(0)}=\frac{\nu_i(0)-\nu_i(T)}{\nu_i(0)}=1-\frac{S_i(T)}{S_i(0)} =\frac{S_i(1-\xi_i\delta)-S_i(T)}{S_i(1-\xi_i\delta)}, \quad (11)$$

where

$$\nu_i(T)=(\gamma/2\pi)H_i(T). \quad (12)$$

We define $1-S_i(T)/S_i(0)\equiv 1-\sigma_i$, the reduced magnetization change of sublattice i. Equation (11) is

TABLE II. Calculated magnetization change from acoustic and optical spin-wave modes for YIG or LuIG with $D=30.0$ cm^{-1}, $J_{aa}/J_{ad}=X=0$; $Y=J_{dd}/J_{ad}=0.1$, $\delta=0$. The energy for mode 2 was taken according to Eq. (9) and those for the modes 2–17 have been taken at $\mathbf{k}=0$; those of the modes 18–20 have been taken as the average between center and corner of the Brillouin Zone.

	$T=50$°K			$T=80$°K		
	$10^5\times(1-\sigma_a)$	$10^5\times(1-\sigma_d)$	$10^5\times(1-\sigma)$	$10^5\times(1-\sigma_a)$	$10^5\times(1-\sigma_d)$	$10^5\times(1-\sigma)$
	Acoustic mode					
$T^{3/2}f_M$	346	346	346	709	709	709
$T^{5/2}$	11	51	133	35	169	431
$T^{7/2}$	−2	9	30	−12	44	153
	Optical modes					
#2	2	1	−1	23	10	−15
#18–20	0	4	12	14	86	230
Total optical	2	6	15	38	140	344
			Total			
	356	412	524	770	1060	1640

different from the theoretical expression Eq. (10) by the factor $(1-\xi_i\delta)^{-1}$ and this gives the possibility of determining δ by combining the experimental data for both sublattices. Unfortunately, the linewidth of the Fe^{57} resonance in YIG (and therefore the scatter in the data) is too large to allow a meaningful[29] determination of δ, expected to be of the order of a few percent for antiferromagnets,[25] but smaller in ferrimagnets.

The total reduced magnetization for YIG and LuIG is

$$1-\sigma=3(1-\sigma_d)(1-\tfrac{2}{3}\delta)-2(1-\sigma_a)(1-\delta). \quad (13)$$

Let us suppose now that hyperfine fields $H_{ij}(T)$ at the nucleus i from interaction transfers from other ions j may not be negligible. What is most likely is a hyperfine field transfer on a d-site Fe^{3+} ion from the 4 nearest a-site ions and vice versa on the a sites from the surrounding 6 nearest d sites. Then one will have

$$\begin{aligned} H_i(T)&=H_{ii}(T)+H_{ji}(T),\\ \nu_a(T)&=A_{aa}|S_a(T)|+A_{da}|S_d(T)|,\\ \nu_d(T)&=A_{ad}|S_a(T)|+A_{dd}|S_d(T)|. \end{aligned} \quad (14)$$

In terms of a reduced sublattice magnetizations, one has then the relation

$$1-\sigma_a=\frac{\Delta\nu_a(T)}{\nu_a(0)}-\frac{B_{da}}{(1-B_{da}-B_{ad})}\left(\frac{\Delta\nu_d}{\nu_d(0)}-\frac{\Delta\nu_a}{\nu_a(0)}\right),$$

$$1-\sigma_d=\frac{\Delta\nu_d(T)}{\nu_d(T)}+\frac{B_{ad}}{(1-B_{ad}-B_{da})}\left(\frac{\Delta\nu_d}{\nu_d(0)}-\frac{\Delta\nu_a}{\nu_a(0)}\right),$$

where (15)

$$B_{da}=\left(1+\frac{A_{aa}}{A_{da}}\left|\frac{S_a(0)}{S_d(0)}\right|\right)^{-1},\quad B_{ad}=\left(1+\frac{A_{dd}}{A_{ad}}\left|\frac{S_d(0)}{S_a(0)}\right|\right)^{-1}.$$

[29] From our experience with YIG, it is estimated that if the Fe^{57} resonance frequency could be read to ±50 Hz, then δ could be determined to better than ±0.01.

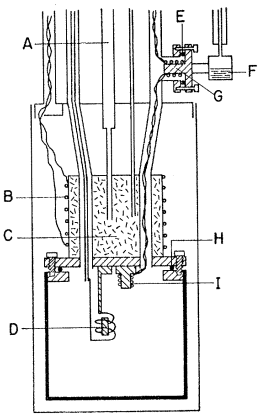

FIG. 1. Schematic drawing of the cryostat for the temperature region between 4 and 250°K. A, helium transfer tube; B, heater; C, charcoal; D, garnet sample and coil; E, H, indium O rings; F, N_2 vapor pressure pot; G, thermocouple reference junction; I, thermocouple measuring junction. Not shown on the figure is the aluminum radiation shield surrounding the sample chamber.

For B_{da} and $B_{ad}=0$ (no hyperfine field transfer) Eq. (15) reduces, of course, to Eq. (11). Because of the relative number of nearest neighbors, one would expect that $|B_{da}|>|B_{ad}|$. One might expect them to be of the same sign.

V. EXPERIMENTAL

A. The Cryostat

The chamber containing the sample was attached to a copper cylinder filled with adsorption charcoal. Figure 1 shows a schematic drawing of the relevant part of the cryostat. The sample chamber and charcoal pot were suspended in a vacuum space surrounded by a liquid-nitrogen bath. The rf coil containing the garnet was a copper ribbon adapted to the size of the different samples and thermally grounded to the charcoal pot. Thermal contact between sample and the surroundings was achieved by the exchange gas in the can and by vacuum grease between the sample and the coil. An aluminum radiation shield surrounded the sample chamber.

Temperatures between 4 and 77°K were reached and maintained by boiling off the liquid helium and creating a pressure in the charcoal pot limited to about 3 atm. When the desired temperature was reached, the heater was shut off and the temperature kept constant by bleeding a small amount of He gas through a fine needle valve and a sensitive flowmeter. When equilibrium was maintained, the heat leak into the system was then exactly compensated by the energy of desorption of the escaping helium gas. The temperature could be kept constant within the sensitivity of the thermocouple thermometer. Usually the temperature was kept constant for up to 20 min to allow equilibrium to be reached between sample and thermometer. Between 77 and 230°K, the temperature of the charcoal pot was regulated by supplying heat electrically, and therefore compensating for the heat loss through conduction and radiation. Measurements at higher temperatures could be carried out by plunging the sample container in a water bath between 273 and 373°K. In this temperature range, a mercury thermometer, which could be read to $\frac{1}{10}$°, was used.

B. The Thermometer

The temperature was measured with a gold-cobalt versus normal silver thermocouple.[30] One junction was brought into thermal contact with the charcoal pot, the other junction was in thermal contact through a copper post with the main liquid-N_2 bath and with a liquid-N_2 vapor-pressure thermometer, so that its temperature could be continuously monitored. Any change of thermoelectric emf of this junction due to a fluctuation in temperature of the main N_2 bath could be then corrected for. The sensitivity of the thermocouple combined with that of the potentiometer made it possible to measure changes as small as 0.05°K over most of the temperature range.

The thermometer was calibrated by substituting the rf coil by a vapor-pressure bulb and a helium-gas thermometer rigidly attached to the charcoal pot and the thermocouple junction. Simultaneous measurements of the gas pressure at fixed volume and of the saturated vapor pressure of pure He^4, Ne, and N_2 were carried out. These data, together with the ice point thus calibrated the gas thermometer, which in turn established the temperature scale of the thermocouple up to 273°K. The calibration was estimated to be about 0.1°K, an accuracy entirely sufficient for the measurements of the magnetization. The reproducibility of the thermocouple was checked at every NMR experiment by measuring the voltage with the junctions, respectively, at 77 and 4.2°K.

C. The rf Spectrometer

A phase-coherent NMR spectrometer was used to detect the Fe^{57} resonance. A General Radio Type 1001-A Standard Signal Generator supplied the rf signal, the frequency of which was measured by a Hewlett-Packard electronic counter. The spectrometer was connected to the sample coil in the cryostat through a resonant coaxial transmission line. To detect the signal, the superheterodyne technique was used. The signal was converted to about 18 MHz, the center frequency of the if amplifier. The recovery time of the receiver was of the order of 5 μsec. By applying the appropriate harmonic from the signal generator to the receiver, phase sensitive detection could be employed. With this method of "zero beating" the harmonic with the nuclear signal, the limiting factor in determining the resonant frequency was the width of the echo. A beat frequency of a few tenths of a cycle could be detected and thus for an echo width of about 40 μsec, a resolution of a few kHz was obtained. The reproducibility of the peak frequency was of the order of 1–2 kHz under the most favorable circumstances (a' sites in YIG).

[30] Sigmund Cohn Corp., Mount Vernon, New York. Detailed measurements of the thermoelectric power of this thermocouple have been described by R. L. Powell, M. D. Bunch, and R. J. Corruccini, Cryogenics **1**, 1 (1961).

D. The Samples

The garnet samples had a natural abundance of Fe^{57}. Two ceramic samples of YIG of great purity were used for measurements in zero field. They were both prepared by Dr. J. P. Remeika at Bell Telephone Laboratories. Sample YIG I (prepared in 1960) had relaxation times short enough to allow an easy measurement of the resonance frequency. Sample YIG II (prepared in 1965) of still purer material had a long relaxation time T_1 of the order of seconds, which made measurements more tedious and less accurate. Sample YIG I was a sphere about 1 cm in diam and sample II was a cylinder 2 cm in diam and 5 cm long. They were also used for specific-heat measurements.[8,31] The LuIG sample was of sintered material also.

VI. RESULTS AND DISCUSSION

A. YIG in Zero Applied Field

Under the conditions of strong rf irradiation, the nuclear signal from the domains was observed.[10] The detection of two lines on the a sites with comparable intensities is evidence that the nuclear signal comes from domains magnetized in an easy, [111], direction, and most of the splitting is due to the inequivalence of the a sites because of dipolar fields. The rest of the splitting may be due to anisotropy of the hyperfine field.[32] From Boutron and Roberts's calculation[32] one can deduce the size of the dipolar fields $H_{\rm dip}$ for the two sites. If one also takes account of the Lorentz field H_L and the field $H_{\rm eff}$, the frequency due to the hyperfine field alone in YIG, $\nu_i(T)$ is then

$$(2\pi/\gamma)\nu_i(T)=(2\pi/\gamma)\nu_i(\text{obs.})+H_{\rm dip}+H_L+H_0+H_D. \quad (16a)$$

In particular,

$$(2\pi/\gamma)\nu_{a'}(T)=(2\pi/\gamma)\nu_{a'}(\text{obs.})+4088\sigma_d-818\sigma-|H_0+H_D|\ (\text{in gauss}) \quad (16b)$$

for a site (a') with symmetry axis along the direction of magnetization;

$$(2\pi/\gamma)\nu_{a''}(T)=(2\pi/\gamma)\nu_{a''}(\text{obs.})-1363\sigma_d-818\sigma-|H_0+H_D|\ (\text{in gauss}) \quad (16c)$$

for a site (a'') with symmetry axis along any of the three other ⟨111⟩ directions; and

$$(2\pi/\gamma)\nu_d(T)=(2\pi/\gamma)\nu_d(\text{obs.})+818\sigma+|H_0+H_D|\ (\text{in gauss}) \quad (16d)$$

for d sites. For the gyromagnetic ratio $\gamma/2\pi$ we will adopt the value 137.4 Hz/G. (See Sec. VI C.) The theoretically expected intensity ratio (a'' sites: a'

[31] D. G. Onn, J. P. Remeika, H. Meyer, and J. Henderson, J. Appl. Phys., **38**, 1023 (1967).

[32] F. Boutron and C. Robert, Compt. Rend. **253**, 433 (1961). See also the detailed Report by F. Hartmann-Boutron, Rapport C.E.A.R. 2336, Centre d'Etudes Nucléaires de Saclay (unpublished).

sites) is 3:1 in the domains.[32] The difference $\nu_{a''}(T)-\nu_{a'}(T)$ which may indicate the extent of the hfs anisotropy is about 250 kHz until 150°K, and above this temperature tends to increase slightly.

A computer fitting of the data to the form

$$1-\sigma_i=\alpha_i f_M T^{3/2}+\beta_i T^{5/2} \tag{17}$$

was made for the data below 60°K (a sites) and below 40°K (d sites). The factor f_M was calculated from values of the anisotropy field[33] $H_A=170$ G and the magnetization measurements of Geller *et al.*[17] Representative values of f_M are 0.901 and 0.939 at 20 and 50°K, respectively. The values of α_i were the same within experimental error for the three sublattices, as expected from theory when δ is negligible. The values are

	$10^5\alpha$	$10^8\beta$
a' sites	1.04±0.02	1.3±0.2
a'' sites	1.01±0.03	2.0±0.5
d sites	1.04±0.03	3.7±2.0

The error limits are twice the standard deviation and do not include the possible systematic deviation due to neglect of the zero-point deviation. The value of $\alpha\times10^5=1.04\pm0.02$ corresponds to a value of $D=30.0\pm0.6$ cm^{-1}. This is good agreement with measurements on microwave instability[9] which at 0°K gave 31 cm^{-1} and 29 cm^{-1}. Litster and Benedek,[14] who did not take into account dipolar effects in their analysis, obtained $D=31.5$ cm^{-1}. Had they considered these effects, their result would be $D\cong30.7$ cm^{-1}. The values of D obtained from specific-heat results[6-9] are all systematically lower.

A very sensitive way to compare experimental data and theoretical curves is to plot $\Delta\nu_i/[\nu_i(0)T^{3/2}f_M]$ versus T/f_M. This reduced plot will also allow the comparison of experiments carried out in applied fields. It shows, of course, an increasing scatter as the temperature tends to zero, because then the ratio $\Delta\nu_i/[\nu_i(0)T^{3/2}f_M]$ becomes undetermined; but this plot gives more detailed information than the usual $\nu_i(T)$-versus-$T^{3/2}$ graph. The results on the sublattices in YIG are presented in Fig. 2 and 3, together with the data of Litster and Benedek. Figure 3 shows the data of the a' sites, where $\nu_{a'}(T)$ could be measured most accurately. The solid lines represent the ratio $(1-\sigma_i)/T^{3/2}f_M$ calculated from Eq. (10) for $D=30.0$ cm^{-1} and several combinations (X,Y), as labeled for each curve.

It should be noted first that the predictions of the spin-wave theory are in good qualitative agreement with experiment, namely, the $T^{3/2}$ coefficients are the same for both sublattices and the $T^{5/2}$ term and optical-mode reversals are larger for the d sublattice. The deviation for the combination $(X=0,\ Y=0.1)$ is only about 5% at 80° for the a sites. However a more quantitative fit for a given (X,Y) combination cannot be made for both sublattices at the same time.

A trend is noticeable: For a certain (X,Y) set, the theoretical curves tend to lie higher in relation to the experimental points for the d sites than for the a sites, as seen for instance for the curves (0,0.2).

This discrepancy can be removed to some extent by the assumption of hyperfine interaction transfer effects: In deducing the temperature dependence of $1-\sigma_a$ from experimental curves of $\nu_a(T)$, we have initially taken $B_{da}=B_{ad}=0$ in which case, Eq. (15) reduces to Eq. (11). Now bearing in mind that $\Delta\nu_d/\nu_d(0)\geqslant\Delta\nu_a/\nu_a(0)$ it can be seen that by making B_{da} and B_{ad} positive, the experiment can be made to fit the theory better at least up to temperatures of about 70°K. A criterion that the magnitude of the hyperfine field transfer is correct is that the total magnetization calculated from NMR data agrees with that determined from direct magnetization measurements.

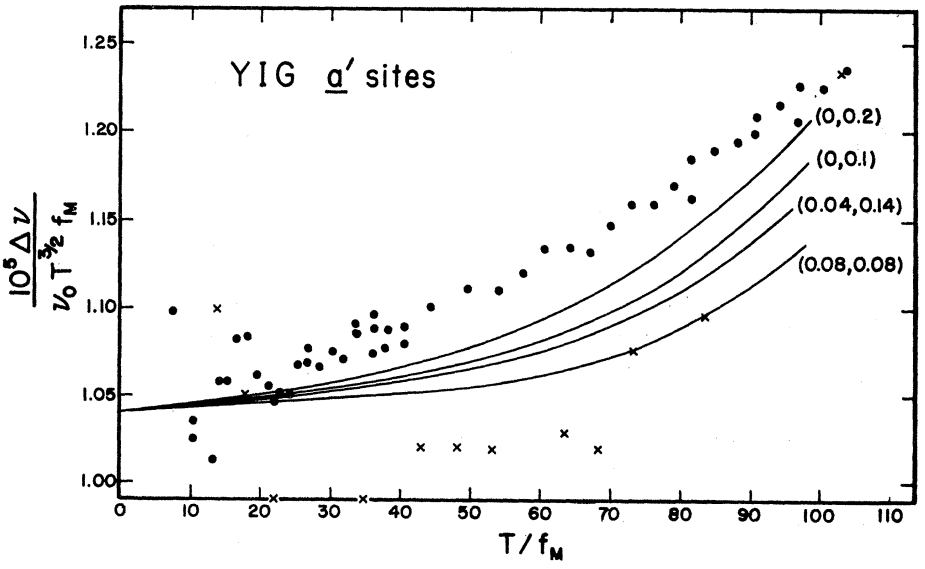

FIG. 2. The ratio $\Delta\nu_a/[\nu_a(0)T^{3/2}f_M]$ versus T/f_M for the a sites in YIG; closed circles, present results; crosses, Litster and Benedek (Ref. 14). The solid lines are the ratios $(1-\sigma_a)/T^{3/2}f_M$ calculated from spin-wave theory [Eq. (10)] for different combinations $(X=J_{aa}/J_{ad},\ Y=J_{dd}/J_{ad})$, and for $\alpha=\frac{2}{5}\zeta(\frac{3}{2})(k_B/16\pi D)^{-3/2}=1.04\times10^{-5}$.

[33] G. P. Rodrigue, H. Meyer, and R. V. Jones, J. Appl. Phys. **31**, 376S (1960).

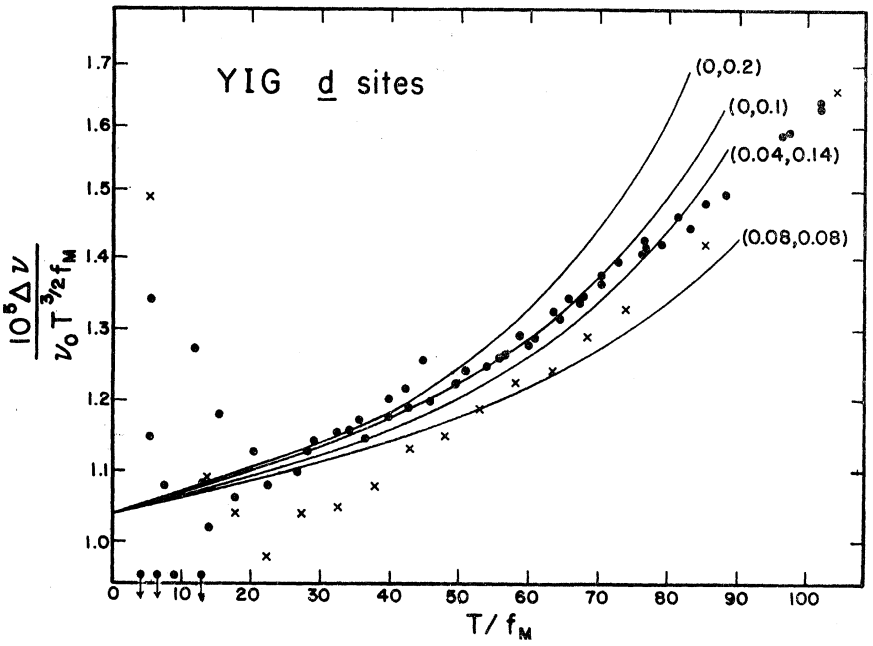

FIG. 3. The ratio
$\Delta\nu_d/[(\nu_d(0)T^{3/2}f_M]$
versus T/f_M for the d sites in YIG. Closed circles, present results; crosses, Litster and Benedek (Ref. 14). The solid lines are the ratios $(1-\sigma_d)/T^{3/2}f_M$ calculated from spin-wave theory as in Fig. 2.

The conclusions from various fitting attempts that took into account the experimental error in α and the possibility of a value of δ up to 0.02 and also that assumed B_{da} and B_{ad} to have the *same* sign, are as follows: (1) B_{da} can be of the order of $+0.05$, and $B_{ad}<B_{da}$. This means that the fields H_{ii} and H_{ji} at the nucleus add up, which is in qualitative agreement with calculations on hyperfine field transfer in several ordered Mn^{2+} and Fe^{3+} compounds made by Huang, Orbach, and Simanek,[34] and by Owen and Taylor.[35] The authors thank Dr. R. Orbach for communicating the calculations on Fe^{3+} compounds. (2) The parameter J_{aa} is smaller than J_{dd} but not necessarily negligible, and $J_{dd}/J_{ad} \approx 0.1$. The constraint is always that D, which is a linear combination of the J_{ij}, is 30.0 cm^{-1}. A reasonable set of values is then $J_{aa}=0.5\pm0.5$ cm^{-1}, $J_{dd}=2.0\pm0.5$ cm^{-1}, $J_{ad}=22.5\pm1$ cm^{-1} and $B_{da}=0.05\pm0.05$, $B_{ad}=0.02\pm0.02$. Again it must be pointed out that above 70°K, the fit between experiment and theory becomes worse because of the steep rise of the calculated curve. This may be due to the neglect of the **k** dependence of the energy and reversals of the optical modes. The value of J_{ad} is in good agreement with that deduced for (Eu,Ga)IG from infrared absorption,[36] 23.5 cm^{-1}.

TABLE III. Experimental magnetization reversals in YIG.

T(°K)	$10^4(1-\sigma_a)$	$10^4(1-\sigma_d)$	$10^4(1-\sigma)$
0	0	0	0
10	3.0±0.2	3.1±0.3	3.3±1.2
20	8.6	8.9	9.6
30	16.4	17.4	19.5
40	25.8	28.2	33.0
50	37.0	41.6	51.0
60	49.8	57.9	74.6
70	64.2	77.3	103.9
80	80.8	99.6	137.5
90	99.0	127.1	183.2
100	120.1	161.0	242.3
110	142.7	199.8	313.2
120	167.5	242.8	392.9
140	225.5	343.7	579.6
160	294.3	468.7	816.5
180	378.8	603.9	1054
200	475.5	748.8	1295
220	584.5±1.0	908.5±1.5	1556 ± 6
240[a]	708	1084	1836
260[a]	844	1273	2129
280	993	1475	2436
300	1158 ±2	1687 ±3	2745 ±12
320	1342	1916	3065
340	1539	2158	3398
360	1745	2403	3718
380[b]	1975	2673	4069

[a] Interpolated.
[b] Extrapolated.

The total magnetization has been calculated from Eq.

[34] N. L. Huang, R. Orbach, and E. Simanek, Phys. Rev. Letters **17**, 134 (1966); R. Orbach (private communication). The authors thank Dr. R. Orbach for communicating the calculations on Fe^{3+} compounds.
[35] J. Owen and D. R. Taylor, Phys. Rev. Letters **16**, 1164 (1966).
[36] P. L. Richards and J. P. Remeika, J. Appl. Phys. **37**, 1310 (1966).
[37] J. D. Litster (private communication).

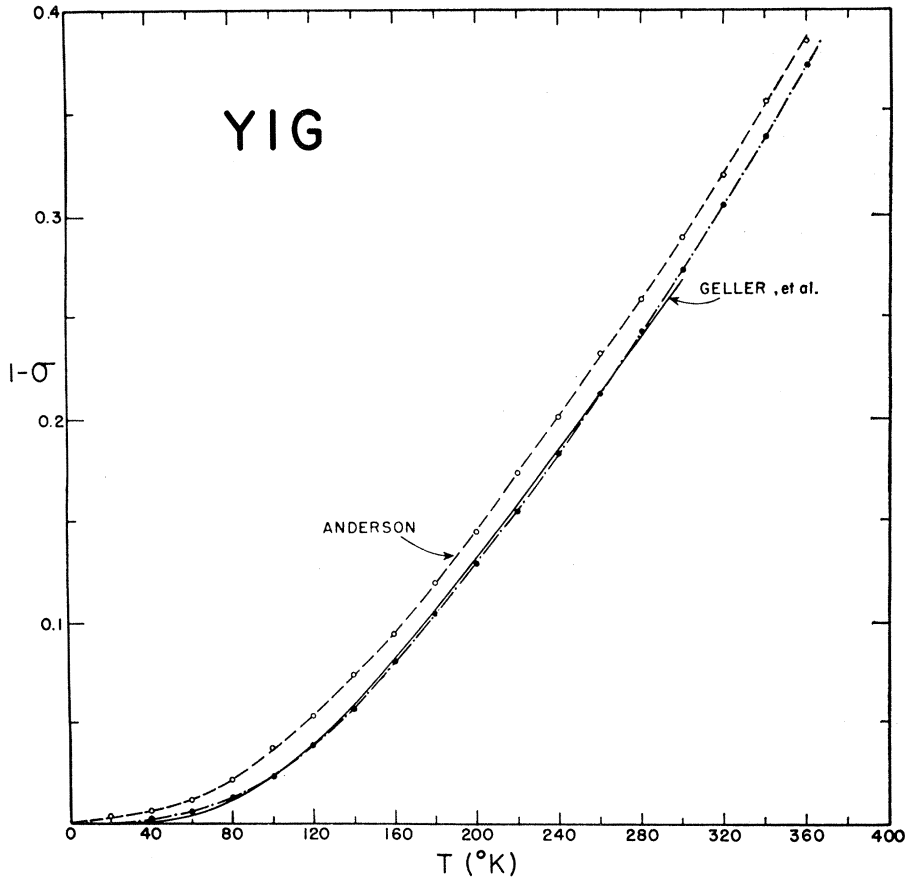

FIG. 4. Change of the reduced total magnetization $(1-\sigma)$ in YIG as a function of T. Closed circles, present smoothed results, for $B_{da}=B_{ad}=0$. Litster and Benedek's data agree with the present data within the experimental scatter above 100°K (Refs. 17 and 18).

(13) for $B_{da}=B_{ad}=0$ and compared in Fig. 4 with the data of Geller *et al.*[17] and of Anderson.[18] The agreement is very good with the data of Geller *et al.*, except for some small systematic deviations. For $B_{da}=0.05$ and $B_{da}=0.02$, the result up to temperatures of 380°K only changes by a few percent, as can be seen from Eqs. (13) and (15), and excellent agreement is obtained with Geller up to ~250°K.

The change in total magnetization in YIG has also been measured very precisely by Solt[16] from the spacing of the magnetostatic modes of a spherical crystal in a microwave cavity. This experiment was carried out in a

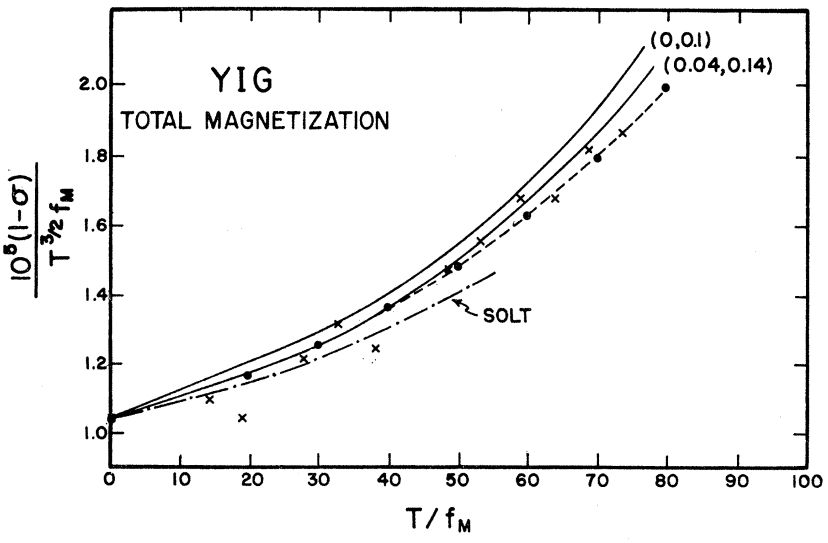

FIG. 5. The reduced total magnetization in the spin-wave region, plotted as $(1-\sigma)/T^{3/2}f_M$. Closed circles, present smoothed results, for $B_{da}=B_{ad}=0$; crosses, Litster and Benedek (Ref. 14); dot-dashed line, Solt (Ref. 16). The solid lines are calculated from spin-wave theory as explained in Fig. 2.

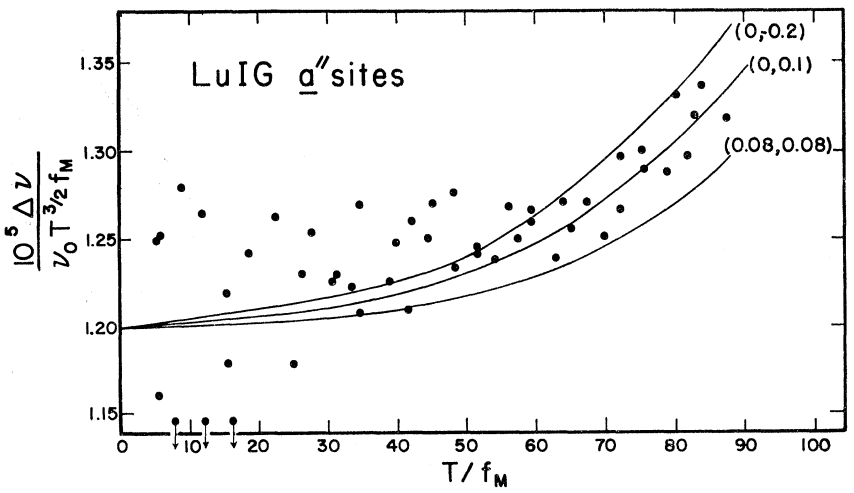

FIG. 6. The ratio $\Delta\nu_a/[\nu_a(0)T^{3/2}f_M]$ versus T/f_M for the a'' sites in LuIG. The solid lines are the ratios $(1-\sigma_a)/T^{3/2}f_M$ calculated from spin-wave theory for the value $\alpha=1.20\times10^{-5}$.

field that was on the average about 4000 G and his data are presented on a reduced plot in Fig. 5. Here a factor f_M, corresponding to the total applied field, has been used. This plot of Solt's data gives also an intercept of $\alpha=1.04\times10^{-5}$ corresponding to $D=30$ cm^{-1}. There is good agreement with our smoothed data, obtained from Eq. (13). Using $B_{da}=0.05$ and $B_{ad}=0.02$ would raise our experimental curve by about 1.5% at 70°K.

It is interesting, but perhaps not very meaningful, that the total magnetization derived from Litster and Benedek's[14] results agree with ours, although the sublattice magnetization data are systematically different below 100°K.

On the other sample YIG II, data were only taken up to temperatures of 40°K. For this sample, an identical analysis gave the value of $D=30.0\pm1.0$ cm^{-1} for both sublattices.

B. LuIG in Zero Applied Field

The results for LuIG were less satisfactory than for YIG because of the broadness of the lines and the scatter was correspondingly larger than for YIG. Two lines were again found for the a sites, the distance between which was 1100 kHz. The line from the a' sites became rather broad above about 80°K, and no frequency measurements were made on it beyond this temperature. The frequencies due to the hyperfine field alone are given by an equation similar to Eq. (16), the numerical factors being slightly different because of the different molar volume of LuIG. It is possible that in LuIG, the resonance was observed in the domain walls because the line was broader and the relative intensities of the a' and a'' lines changed with temperature.

TABLE IV. Experimental magnetization reversals in LuIG.

T(°K)	$10^4(1-\sigma_a)$	$10^4(1-\sigma_d)$	$10^4(1-\sigma)$
10	3.1±0.7	3.2±1.0	3.4±2.0
20	9.9	10.2	10.8
30	18.6	19.6	21.6
40	29.2	31.4	35.9
50	41.5	46.0	55.0
60	55.4	63.4	79.3
70	71.0	85.5	114.5
80	89.9	112.0	156.2
90	110.5	144.6	213
100	131.9	181.0	280
110	156.4	223.5	357
120	183.0	270	443
140	245.0	375	640
160	325.0	502	877
180	415	658	1140
200	517±5	824±7	1444±20

The value of D was found from a plot of the data to be 27.3±1.0 cm^{-1} for both sites. This is in good agreement with the results from specific-heat data. A reduced plot $(\Delta\nu/\nu)/(T^{3/2}f_M)$ for the a'' sites, is shown in Fig. 6. No conclusions could be reached about the possibility of hyperfine field transfer. It was estimated that $J_{dd}/J_{ad}\lesssim0.2$. Possibly J_{aa} is smaller than J_{dd}. The total magnetization is plotted in Fig. 7 and compared with the data of Geller *et al.* Table IV presents the magnetization reversals in LuIG.

As mentioned elsewhere,[10] there is at 4.2°K a strong spectrum of broad resonances between about 40 and 70 MHz that can be attributed to the NMR of Lu^{175} and Lu^{176}. Such a resonance was predicted from specific-heat measurements[38] and it is broadened by the large quadrupole moment of both Lu isotopes (respectively, $I=\frac{7}{2}$ and 7). Therefore the magnetic field at the nucleus of Lu^{3+} could not be studied as a function of temperature. It was noted though, that the pattern of the spectrum changes with temperature. The field at the nucleus of Lu^{3+} is an interesting effect of induced nuclear hyperfine field through charge transfer from the Fe ions. Such an effect in iron garnets has also been investigated by Mössbauer effect in Sn embedded in

[38] D. G. Onn, R. Gonano, and H. Meyer, in *Proceedings of the Ninth International Conference on Low Temperature Physics, Columbus, Ohio, 1964*, edited by J. G. Daunt, D. V. Edwards, F. J. Milford, and M. Yaqub (Plenum Press, Inc., New York, 1965), p. 897.

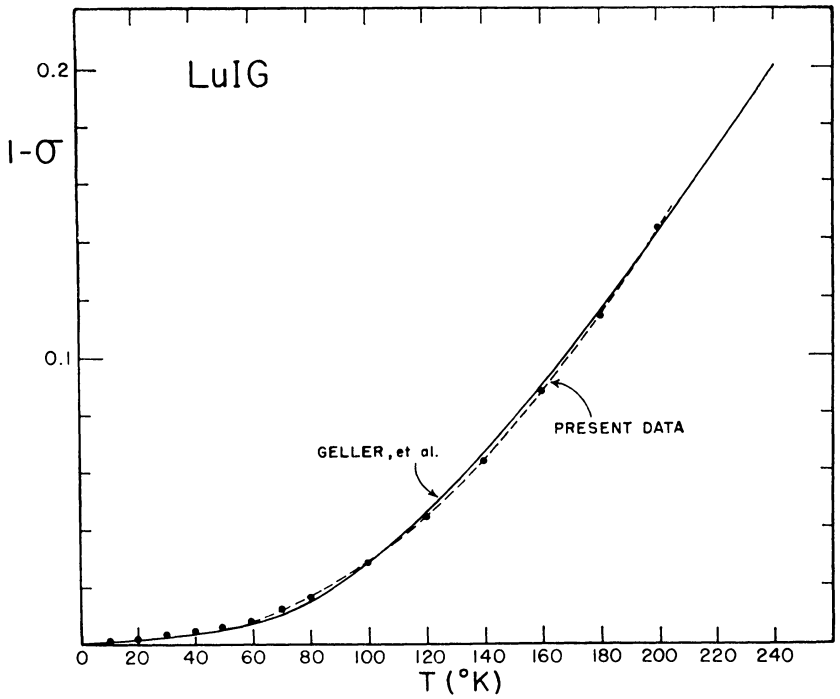

FIG. 7. The total reduced magnetization of LuIG plotted as $1-\sigma$ versus T. —●— present smoothed results for $B_{da}=B_{ad}=0$. Solid line, Geller *et al.* (Ref. 17). Had a value of $B_{da}=0.05$, $B_{ad}=0.02$ been taken, the present data would be in even better agreement with those of Geller *et al.*

YIG.[39] In this case, it was possible to study the field at the nucleus as a function of temperature. Because of the existence of a nuclear field in Lu^{3+}, it was assumed that there should also be a charge-transfer effect for Y^{89} in YIG. A search over a frequency range from 2–80 MHz was therefore made at 4.2°K, but no resonance belonging to this nucleus could be detected. Combining evidence from NMR and specific-heat data,[31] the field at the nucleus seems to be smaller than about 10 kG.

C. YIG in an Applied Magnetic Field

We have investigated the effect of a magnetic field on the sublattice magnetization of YIG at 63.1 and 77.6°K. The purpose was to show that the field dependence is as predicted from spin-wave theory, Eqs. (3) and (8). As far as we know, the only similar work has been done on ferromagnets. The most detailed work is that of Argyle, Charap, and Pugh,[2] who showed that the magnetization-versus-temperature curve for two ferromagnets, iron and nickel, depends on the applied field in the way predicted by spin-wave theory.[40]

Although the change of magnetization with field is small, it can be conveniently measured by Fe^{57} nuclear magnetic resonance. Since the quantity of interest is the hyperfine field $H_i(T)$ [proportional to $S_i(T)$], the effect of the applied field[41] and other "external" fields has to be taken into account, as has been done in Eq. (15). Because of the small change of the magnetization, the gyromagnetic ratio $\gamma/2\pi$ must be known to a high accuracy.

For this determination, a very pure 5-mm-diameter single-crystal sphere[42] of YIG was aligned inside an rf coil with an easy [111] direction parallel to the applied field. The NMR frequency for both a and d sites was measured at 4.2°K as a function of field with an accuracy of about 1–2 kHz. The magnetic field was measured by proton resonance. The highest applied magnetic field was about 10 kG. At still higher fields the ^{57}Fe signal became too weak because of the decrease in the enhancement factor.

Since the sample is almost completely magnetically saturated at 4°K, the variation of frequency with field is almost exactly $\pm\gamma H/2\pi$. The frequency increases for the a sites and decreases for the d sites by this amount. The very small increase in the magnetization with field which would tend to increase frequencies on both sites by the same amount was eliminated by averaging the absolute values of the slopes of the two curves of frequency versus field. The result[43] was $\gamma/2\pi=137.4\pm0.2$ Hz/G, in excellent agreement with the most recent value of 137.6 ± 0.1 Hz/G obtained by Locher and Geschwind[44] from ENDOR experiments on Fe^{57} in MgO.

[39] V. I. Goldanski, V. A. Turkhanov, M. N. Devisheva, V. F. Belov, Phys. Letters **15**, 317 (1965).

[40] Some other work reported in less detail is that of N. V. Zavaritsky and V. A. Tsarijw, in *Proceedings of the Eighth International Conference on Low-Temperature Physics, London, 1962*, edited by R. O. Davies (Butterworths Scientific Publications Ltd., London, 1963), p. 260.

[41] In the discussion of the field dependence of $H_i(T)$ we can neglect the possible effect of charge transfer.

[42] Airtron Co., 200 East Hanover Ave., Morris Plains, New Jersey.

[43] R. Gonano, E. Hunt and H. Meyer, Bull. Am. Phys. Soc. **11**, 221 (1966). In a similar earlier experiment, C. Robert [Compt. Rend. **152**, 1442 (1961)] obtained the value $\gamma/2\pi=139.2\pm1$ Hz/Oe.

[44] P. R. Locher and S. Geschwind, Phys. Rev. **139**, A991 (1965).

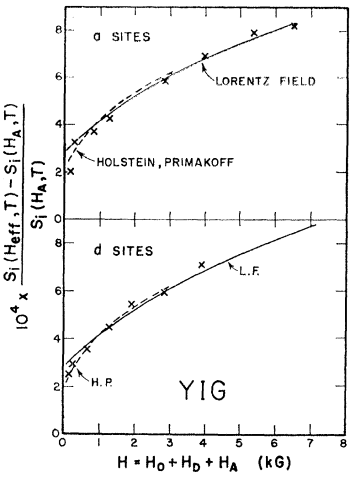

FIG. 8. Relative change of sublattice magnetization $[S_i(H_{eff},T)-S_i(H_A,T)]/[S_i(H_A,T)]$ as a function of applied field. Crosses, present data. The solid lines represent the results from spin-wave theory, respectively Eqs. (3) and (8).

The change of sublattice magnetization upon application of a magnetic field was observed on the same YIG sphere at 63°K (triple point of N_2) which is a very convenient temperature, and still in the range where optical modes are barely excited.

The effective change in magnetization was obtained in plotting

$$\frac{S_i(H_{\rm eff},T)-S_i(H_A,T)}{S_i(H_A,T)} = \frac{\nu_i(H_{\rm eff},T)_{\rm obs}\pm\gamma/2\pi(H_0+H_D)-\nu_i(H_A,T)_{\rm obs}}{\nu_i(H_A,T)_{\rm obs}} \quad (18)$$

versus $H_{\rm eff}$, where $H_{\rm eff}=H_A$ when the external field is zero. The + and − before $\gamma/2\pi$ apply, respectively, to the d and a sublattices. In Fig. 8 the experimental curves are shown and compared with theoretical expectations. As one can see, the predictions of Eqs. (3) and (8) differ at low fields and agree at intermediate fields. The experiment is not conclusive enough to distinguish between the theories at low fields, but agrees well with them in the region of intermediate fields.

It should be noted here that in accordance with the expectations of spin-wave theory, the magnetization of the a sublattice, though antiparallel to the applied field, increases with this field. Also, the susceptibility is approximately proportional to $H^{1/2}$ and not constant (as is assumed for higher temperatures).

The same experiment was also performed at 77°K. Although the signal-to-noise ratio of the Fe^{57} signal was not as good as at lower temperatures, the results were consistent with those at 63°K, and in agreement with theory.

VII. CONCLUSION

We have shown that the NMR of Fe^{57} is a precise tool for measuring accurately the small changes of the sublattice magnetization of iron garnets at low temperatures. The data demonstrated that the sublattice magnetization in these ferrimagnets followed the predictions from the spin-wave theory. In order to improve the agreement between theory and experiment, a small field enhancement at the Fe^{57} nucleus (particularly on the a sites) brought about by the presence of the neighboring magnetic ions has been invoked. It is gratifying that this assumed hyperfine transfer effect has the same sign and order of magnitude as that calculated for Mn^{2+} and Fe^{3+} compounds.[34] The remaining discrepancy between experiment and spin-wave theory (a few percent in the spin reversal) could conceivably be attributed to the approximations made in the spin-wave treatment or possibly a change of D with temperature.

The quantity obtained with the most precision from the spin-wave analysis is the parameter $D=30.0$ cm^{-1} in YIG and 27.3 cm^{-1} in LuIG. The exchange constants J_{ij} are less well defined, but the conclusion is that for YIG at least, $J_{aa}<J_{dd}$ and $J_{dd}\approx 0.1J_{ad}$. We therefore agree with Wojtowicz's analysis from the susceptibility data, except that we find that J_{aa} is possibly not negligible in comparison with J_{dd}.

There is good agreement in the value of D for YIG obtained from NMR, total magnetization and microwave instability. Surprisingly, there is a discrepancy with D obtained from specific-heat experiments[8,31] on very pure samples. For LuIG, there is no such discrepancy, although the sample investigated is probably less perfect.

Experiments in a magnetic field up to 10 kG have also shown that the field variation of the sublattice magnetization is in agreement with that predicted from spin-wave theory. As a byproduct of this study, we have determined the gyromagnetic ratio $\gamma/2\pi$ for Fe^{57} with an accuracy comparable with that of ENDOR experiments. This was made possible by the very sharp lines observed in YIG.

Lu^{175} and Lu^{176} in LuIG show the effects of charge transfer and the field at the nucleus is of the order of 100 kOe, which corresponds to the resonance band observed between about 40 and 70 MHz. For Y^{89} in YIG, this induced field is probably less than 10 kOe.

Extensive data of the sublattice magnetization have also been taken for GdIG [10] and TmIG as a function of temperature. The resonance of Fe^{57} in DyIG and ErIG has been observed at 4.2°K. The sublattice magnetization in EuIG is presently being measured. These results and their analysis and comparison with other data will be presented in another paper.

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APPENDIX

The Effect of Magnon Exchange Interaction

We attempt here a crude order-of-magnitude calculation of the effect of the interaction between spin waves. This estimate is based on the paper by Keffer and Loudon[45] and the discussions by Kittel[46] and Walker.[25] One takes into account the reduction of the exchange field as the quantity $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ changes with temperatures. We assume that this change can be represented by an effective decrease in the exchange integrals J_{ij} and that the relative change is the same for J_{aa}, J_{dd}, and J_{ad}. We consider (a) *Changes due to acoustical spin waves.* This case has been discussed in detail before.[45] For long-wavelength spin waves, the motion of the neighboring spins is highly correlated and the exchange field acting on its neighbors varies with the mean angle between the spins. The result is an effective change in J given by

$$\left[\frac{J(T=0)-J(T)}{J(0)}\right]_{\text{acous}}=\left[\frac{\Delta J}{J}\right]_{\text{acous}}=\frac{\text{magnon energy}}{\text{total magnetic exchange energy}}\simeq\frac{0.0113D(k_BT/D)^{5/2}}{|300J_{ad}-150J_{dd}-200J_{aa}|}\cdots\simeq 0.02\% \text{ at } 80^\circ\text{K}. \quad \text{(A1)}$$

[45] F. Keffer and R. Loudon, J. Appl. Phys. **32**, 25 (1961).
[46] C. Kittel, *Low-Temperature Physics* (Gordon and Breach Science Publishers, Inc., New York, 1962), p. 469.

(b) *Changes due to optical modes.* These modes involve out-of-phase spin motions so their effect on J is greater than that of long acoustical modes. We assume a molecular field approximation to be reasonable. Since we consider the effects of uncorrelated optical-mode motions only, the magnetization reversal in question is the optical-mode reversal. We obtain

$$\left[\frac{\Delta J}{J}\right]_{\text{opt}}=(1-\sigma)_{\text{opt}}\simeq 0.3\% \text{ at } 80^\circ\text{K}\cdots. \quad \text{(A2)}$$

We then assume that the changes of J are additive;

$$\frac{\Delta J}{J}=\left[\frac{\Delta J}{J}\right]_{\text{acous}}+\left[\frac{\Delta \tau}{J}\right]_{\text{opt}}\simeq 0.32\% \text{ at } 80^\circ\text{K}. \quad \text{(A3)}$$

The change in J will modify the power series in the acoustical mode [Eqs. (1) and (8)]. For instance, the change of the term in $T^{3/2}$ will be given by $-\frac{3}{2}\Delta J/J = -\frac{3}{2}\Delta D/D$. Then $(1-\sigma)_{\text{acous}}$ changes by 0.6% at 80°K.

In the optical modes, the energy will be changed by the amount

$$\Delta E_\mu/E_\mu=\Delta J/J. \quad \text{(A4)}$$

The change in spin reversal N_μ will be

$$\Delta N_\mu/N_\mu\approx\Delta E_\mu/k_BT. \quad \text{(A5)}$$

For YIG, the optical modes that count most, have an energy of the order of 250°K. Therefore, using (A4) and (A5), we have

$$\frac{\Delta N_\mu}{N_\mu}=\frac{\Delta(1-\sigma)_{\text{opt}}}{(1-\sigma)_{\text{opt}}}\lesssim 1\% \text{ at } 80^\circ\text{K}. \quad \text{(A6)}$$

The total spin reversal is the sum from the acoustical and optical branch and the relative change due to the interacting spin waves is found to be less than about 1% at 80°K. It can therefore be considered negligible in the calculations outlined in Sec. IV B.