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Sublattice Magnetization in Several Rare-Earth and Yttrium Iron Garnets by Means of ^{57}Fe NMR

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Abstract

We have observed the NMR of ^{57}Fe on the *a* and *d* sublattices of YIG, GdIG, and LuIG between 4° and 200°K. Using the spin-echo technique the frequencies could be determined to within a few kHz. For LuIG the determination of the frequency was less accurate due to the presence of the lutetium resonances which overlay the iron resonances. Assuming the frequency is proportional to the sublattice magnetization, the data were fitted to the equation derived from spin-wave theory. For YIG and LuIG inclusion of the $T^{7/2}$ term was necessary to represent the data above 40°K. Using the theory of noninteracting spin waves we have derived expressions for the sublattice magnetizations in terms of the exchange integrals J_{aa}, J_{ad}, J_{dd}, and J'_{ad}, where J'_{ad} describes interactions between next-nearest-neighboring pairs of spins on the *a* and *d* sublattices. Our experimental results indicate that J_{aa} and J_{dd} are not negligible. The $T^{3/2}$ coefficients were found to be about 15% larger for LuIG than for YIG. Our value of the exchange stiffness constant of YIG is 10% larger than those determined calorimetrically and hence agrees with values determined by microwave instability measurements. For LuIG our value of this constant agrees with the specific heat value.

Disciplines

Physics

We may regard[15] the Gd ions as a paramagnetic sublattice in an exchange field caused by the iron sublattices and neighboring Gd ions. The reduced magnetization (σ_c) of the Gd sublattice will simply be given by a Brillouin function of the total exchange field:

$$\sigma_c = B_{7/2}(X_c),$$

$$X_c = Ng^2\beta^2 S_c(2S_a\lambda_{ca}\sigma_a + 3S_d\lambda_{cd}\sigma_d + 3S_c\lambda_{cc}\sigma_c)/8kT,$$

where $S_a = S_d = \frac{5}{2}$, $S_c = \frac{7}{2}$, k is Boltzmann's constant, and N is the number of magnetic ions per unit volume. In an ordinary molecular field calculation one must solve self-consistently three such expression for the sublattice magnetizations.[3] Here, we require the iron magnetizations (σ_a, σ_d) to assume the temperature dependence we have measured and solve self-consistently for σ_c. The solid line in Fig. 3 was obtained with $\lambda_{ca} = -795$, $\lambda_{cd} = -4175$, $\lambda_{cc} = -313$ G²·g·erg⁻¹. Since this calculation fits the data quite well, we regard these coefficients as reliable. The exchange constants calculated[16] from them are $J_{ca} = -0.22$ cm^{-1}, $J_{cd} = -3.46$ cm^{-1}, and $J_{cc} = -0.13$ cm^{-1}.

In conclusion, we have measured the temperature dependence at constant volume of the sublattice magnetizations in YIG. Analyzing our data below 25°K, we have found the dispersion coefficient D of the acoustic spin-wave mode to be 32 cm^{-1}. In addition, our data above 25°K provide accurate values for the sublattice magnetization up to $0.85T_c$. We have also made precise measurements of both iron sublattice magnetizations at constant pressure (corrections to constant volume will be significant only above 300°K) in GdIG and have used them to determine the Gd magnetization up to $0.9T_c$. These data should prove valuable for spin-wave theories which include the effect of optical modes and renormalization.

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Sublattice Magnetization in Several Rare-Earth and Yttrium Iron Garnets by Means of ^{57}Fe NMR*

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We have observed the NMR of ^{57}Fe on the a and d sublattices of YIG, GdIG, and LuIG between 4° and 200°K. Using the spin-echo technique the frequencies could be determined to within a few kHz. For LuIG the determination of the frequency was less accurate due to the presence of the lutetium resonances which overlay the iron resonances. Assuming the frequency is proportional to the sublattice magnetization, the data were fitted to the equation derived from spin-wave theory. For YIG and LuIG inclusion of the $T^{7/2}$ term was necessary to represent the data above 40°K. Using the theory of noninteracting spin waves we have derived expressions for the sublattice magnetizations in terms of the exchange integrals J_{aa}, J_{ad}, J_{dd}, and J'_{ad}, where J'_{ad} describes interactions between next-nearest-neighboring pairs of spins on the a and d sublattices. Our experimental results indicate that J_{aa} and J_{dd} are not negligible. The $T^{\frac{3}{2}}$ coefficients were found to be about 15% larger for LuIG than for YIG. Our value of the exchange stiffness constant of YIG is 10% larger than those determined calorimetrically and hence agrees with values determined by microwave instability measurements. For LuIG our value of this constant agrees with the specific heat value.

THE use of NMR techniques to measure the temperature dependence of the magnetization has enabled several authors[1] to test the validity of spin-wave theory. Previous NMR measurements[2] on yttrium iron garnet (YIG) and GdIG between 4° and 300°K were interpreted using the Weiss molecular field model, but were not sufficiently sensitive for the study of spin-wave effects. Accordingly, we have performed extensive high-resolution temperature and frequency measurements over a range of temperature including the spin-wave region.

These experiments were carried out on samples described previously.[3] The temperature was measured

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by a gold–cobalt vs normal silver thermocouple which was calibrated against the known vapor pressures of liquid He, Ne, and N_2. A gas thermometer was used for calibration in the intermediate ranges and above 77°K. The accuracy was estimated to be better than 0.1°K above 8°K. Using the spin-echo technique the center of the resonance lines could be determined to within a few kHz.

Spin-wave theory[4–6] predicts the a- and d-sublattice magnetization of YIG and LuIG at low temperatures to be

$$M(T)/M(0)=1-\alpha T^{3/2}(1-cT^{-\frac{1}{2}})-\beta T^{5/2}-\gamma T^{7/2}+\cdots, \quad (1)$$

with $\alpha_a=\alpha_d=\alpha=2.94\times10^{-3}(k/D)^{3/2}$, where k is the Boltzmann constant and

$$\beta_a=-\alpha k(6AD+3F+15E)/8D^2, \quad (2a)$$

$$\beta_d=-\alpha k(4AD+3F+15E)/8D^2. \quad (2b)$$

The term $cT^{-\frac{1}{2}}$, arising from the dipolar interaction, has been calculated by Holstein and Primakoff[4];

$$c=0.53(4\pi g\mu_B M/k)^{\frac{1}{2}}.$$

Expressions for E and F assuming only nearest-neighbor interactions between sublattices were given previously.[5] For a nonzero next-nearest neighbor exchange integral, J'_{ad}, between the a and d sublattices,

$$D=(40J_{aa}-25J_{ad}-65J'_{ad}+15J_{dd})/16, \quad (3a)$$

$$A=(-48J_{aa}+25J_{ad}+65J'_{ad}-12J_{dd})/64(J_{ad}+J'_{ad}), \quad (3b)$$

and E and F will be given elsewhere.[6]

In all samples we observed two narrow resonances in the vicinity of the a-site frequency and only one for the d-site. The width of the echo was 20 to 40 μsec—much greater than one gets for domain wall excitations. We believe we are observing the nuclear signal from domains which are magnetized along the easy (111) directions. Boutron and Robert[7] have shown that for this case there are two inequivalent a sites due to the dipolar field and all the d sites are equivalent. The calculated splitting in YIG at low temperatures is 0.76 MHz to be compared with our measured value of 1.00 or 0.92 MHz which can be deduced from Robert's experimental work. This discrepancy of about 0.2 MHz may be due to the anisotropy of the hyperfine field.[7] Further support for this interpretation is obtained from a study at 4.2°K using a single-crystal YIG sphere. With the (111) axis parallel to a saturating magnetic field, we observed the same a-site resonances split by 1.0 MHz and a single d-site line as before.

TABLE I. Experimental frequencies and spin-wave coefficients.

	YIG		LuIG	
	a	d	a	d
$f(0)$ (kHz)	76 058	64 980	75 802	64 560
	75 057		74 900	
$\alpha(\times10^{-5}$ °$K^{-3/2})$	1.02±0.01		1.18±0.02	
$\beta(\times10^{-8}$ °$K^{-5/2})$	0.5	1.5	0.4	0.7
$\gamma(\times10^{-10}$ °$K^{-7/2})$	1.5	4.5	1.4	6.4
D (cm^{-1})	30.3±0.2		27.5±0.4	
A	0.17		0.04	

In LuIG the same lines are observed but are considerably broader. This is due in part to the presence of the ^{175}Lu and ^{176}Lu resonances which were observed from about 40 to 75 MHz.

In GdIG the observed lines were as sharp as for YIG. Extrapolated to 0°K the a-site resonances are 76.252 and 73.990 MHz, and thus are split by 2.26 MHz. The d-site frequency is 66.254 MHz. Assuming the magnetization lies along a (111) direction, and using the values of the lattice sums calculated by Boutron and Robert,[7] we calculated the dipolar splitting to be 1.78 MHz for the a site and zero for the d site. Thus it would appear that the anisotropy of the hyperfine field at the a site is greater for GdIG than for YIG.

By taking account of the dipolar field we find the temperature dependence of the a-sublattice magnetization to be given equally well by either a-site resonance. We have assumed the hyperfine coupling constant to be independent of temperature and volume and have made no corrections for transfer hyperfine interactions. With the coefficients given in Table I, Eq. (1) represents the data of YIG and LuIG to 180°K for the a sites and 130°K for the d sites. No estimate of the errors in β, γ, and A has been made and the values given are only tentative pending computer analysis. At higher temperatures and for GdIG, Eq. (1) does not represent adequately the experimental data, which will be presented more fully elsewhere.[8]

For YIG our value of D agrees well with the microwave instability measurements but is about 10% higher than calorimetric results (see Table V in Ref. 3). For LuIG our result agrees with that deduced from specific heat data. Our values for A for YIG and LuIG are smaller than the value obtained from Eq. (3b) assuming only J_{ad} to be nonzero, indicating that J_{aa} and J_{dd} are not negligible. Furthermore, if J_{aa} and J_{dd} were negligible, the optical mode frequencies would be so low as to cause an appearance of exponential terms in Eq. (1) well within our temperature region.

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