IEEE TRANSACTIONS ON MAGNETICS, VOL. MAG-19, NO. 5, SEPTEMBER 1983

BRILLOUIN LIGHT SCATTERING DETERMINATION OF THE SPIN-WAVE STIFFNESS PARAMETER IN LITHIUM-ZINC FERRITE

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ABSTRACT

The spin-wave stiffness parameter for Zn substituted Li-ferrite has been measured by the technique of Brillouin light scattering. A range of compositions from pure Li-ferrite through Li-Zn ferrite with 0.65 Zn at./formula unit was studied. The magnon frequency vs. field determinations from light scattering and separate magnetization data were used to determine the spin-wave stiffness parameter, D. The data give a D-value of 10.1×10^{-9} Oe$\cdot$cm^2 for pure lithium ferrite, approximately twice as large as for yttrium iron garnet. A rapid decrease occurs with Zn substitution and D is essentially zero for about 0.4 Zn at./formula unit. These are the first reported results of Brillouin scattering from a polycrystalline ferrite. A molecular field calculation of the spin-wave stiffness yields a much slower decrease in D with zinc than is found experimentally.

INTRODUCTION

Zinc substituted Li-ferrite, because of its high magnetization and good high frequency properties, is an important microwave and millimeter wave material [1]. Zinc substitution occurs for A-sublattice Fe^{3+} ions in the spinel structure, with a site distribution given by:

$$(Zn_xFe_{1-x})_A[Li_{0.5-x/2}Fe_{1.5+x/2}]_BO_4 \ . \tag{1}$$

Zinc substitution is known to cause an increase in the magnetization for low levels of substitution, up to about x = 0.3-0.4, followed by a decrease in magnetization at higher levels of substitution [2]. This is explained by magnetic dilution of the A-sublattice with zinc, as evident from (1). This initially causes the net magnetization to increase with x due to the larger B-sublattice moment. As x continues to increase, however, the A-sublattice becomes so magnetically dilute as to reduce the antiferromagnetic exchange to the point where the B-site Fe^{3+} ion moments no longer remain parallel. As a consequence of the antiferromagnetic B-B exchange interaction, the B-site moments cant to some extent. This canting reduces the net B-sublattice magnetization. Canting becomes quite significant above x = 0.3-0.4 and leads to a decrease in the net magnetization with zinc [3].

With recent developments in Brillouin light scattering techniques [4], it is now possible to observe spin-wave excitations directly.

EXPERIMENT

Brillouin light scattering was done on highly polished spheres about 2 mm in diameter. The material composition varied from pure Li-ferrite to Li-Zn ferrite with 0.65 Zn at./formula unit. The samples with 0.55 and 0.65 zinc were single crystals. All other samples were polycrystals prepared by hot pressing [1].

The measurements were carried out using a multipass tandem Fabry-Perot interferometer similar to that described by Sandercock [4]. The present system has a measured contrast of better than 10^{12}. The 4880 Å line of an argon laser was used in the backscattering configuration with a laser power of about 30 mW and incident light perpendicular to the static magnetic field. A polarization analyzer oriented perpendicular to the incident light polarization was positioned after the collecting lens to reduce the amount of strong spectrally reflected light.

Two spectra are shown in Fig. 1, where the channel content is shown as a function of frequency for x = 0.1 (a) and 0.55 (b) with an applied field,

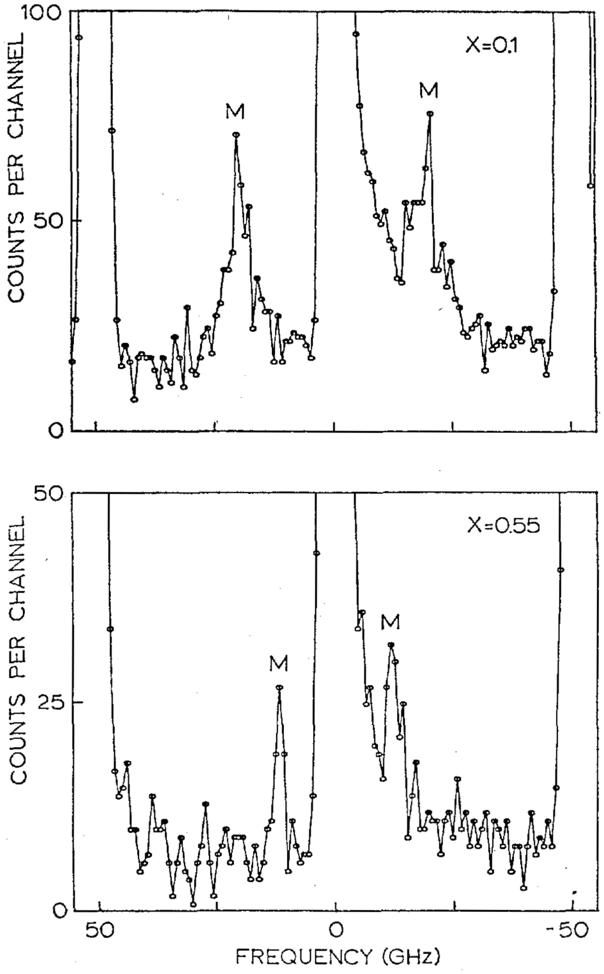

Fig. 1. Brillouin light scattering spectra for two Li-Zn ferrite samples: (a) a polycrystalline sphere with x = 0.1, (b) a single crystal sphere with x = 0.55. The free spectral range was 50 GHz and the accumulated time per channel was 20 seconds for (a) and 10 seconds for (b). The external applied field was 4 kOe.

Manuscript received February 25, 1983.

This work was supported in part by the United States Army Research Office and the National Science Foundation, Grant DMR-8013727.

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H_{ext}, of 4 kOe. The inward shift of the magnon peaks, labeled M, from (a) to (b) provides a clear indication of the effect of zinc on spin-wave frequency.

Based on sets of spectra vs. applied field for the various compositions, data on magnon frequency vs. field were obtained for all samples. Some typical results are shown in Fig. 2 for 0.4 Zn at./formula unit. These data were analyzed using the linearized spin-wave dispersion relation [6],

$$f_k/g = H_{ext} + Dk^2 + 4\pi M_o/6 \ . \qquad (2)$$

In (2), f_k is the magnon frequency, g is the gyromagnetic ratio, D is the spin-wave stiffness parameter, k is the magnon wavenumber, and $4\pi M_o$ is the saturation induction. Note that (2) is specifically for spherical samples magnetized to saturation and for a spin-wave wavevector perpendicular to the applied field.

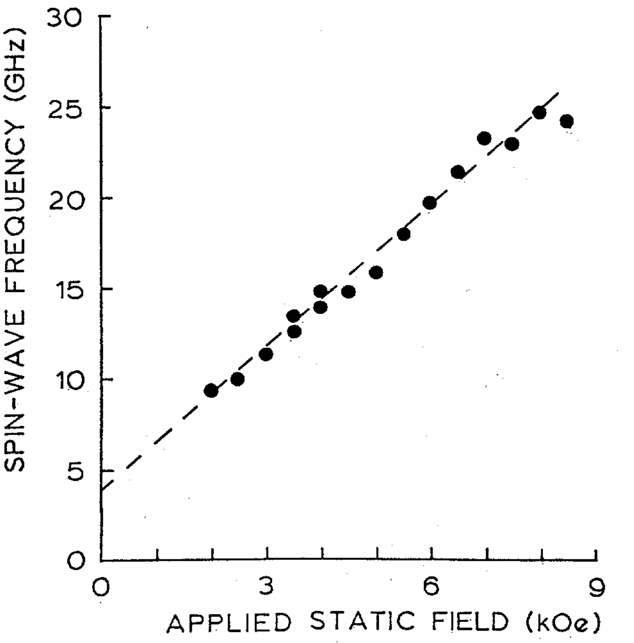

Fig. 2. Data on spin-wave frequency vs. applied field for x = 0.4. The dashed line is the linear least square fit to the data.

Values of $4\pi M_o$ were obtained by vibrating sample magnetometry. This was done at 150°C to account for laser heating in the light scattering experiment. The gyromagnetic ratio was determined for each sphere by ferromagnetic resonance at 9.4 GHz. These values are listed in Table I. The magnon wavenumber in backscattering is given by

$$k = 2(2\pi/\lambda)n \ , \qquad (3)$$

where λ is the laser wavelength (4880 Å) and n is the refractive index. The refractive index was taken to be 2.46, the same as for yttrium iron garnet (YIG) [7].

The analysis consisted of a linear least squares fit to the frequency vs. field data as indicated by the dashed line in Fig. 2. The frequency at 5.5 kOe from the linear fit was then used in (2) along with the known values of k and $4\pi M_o$ to obtain D.

Table I. Gyromagnetic Ratio

Zn at./formula unit	Gyromagnetic Ratio
0.0	2.74 GHz/kOe
0.1	2.78
0.2	2.82
0.4	2.93
0.5	2.80
0.55	2.83
0.6	2.86
0.65	2.82

RESULTS

The fitted spin-wave stiffness parameter determinations as a function of zinc content are listed in Table II and shown in Fig. 3. The spin-wave stiffness for pure Li-ferrite is 11.6 × 10^{-9} Oe·cm^2, approximately twice the 5.4 × 10^{-9} Oe·cm^2 value reported for YIG [5]. This is not unreasonable, since the Curie temperature T_c for lithium ferrite is a factor of 1.64 larger than that of YIG [3]. Figure 3 shows that the stiffness parameter decreases very rapidly with zinc and is essentially zero at x = 0.4. As mentioned in the introduction, this is about the same amount of zinc at which strong canting effects commence. The rapid drop in D is in contrast with the much more gradual decrease in T_c with zinc [3]. At x = 0.6, T_c is still 60% of the value for pure Li-ferrite.

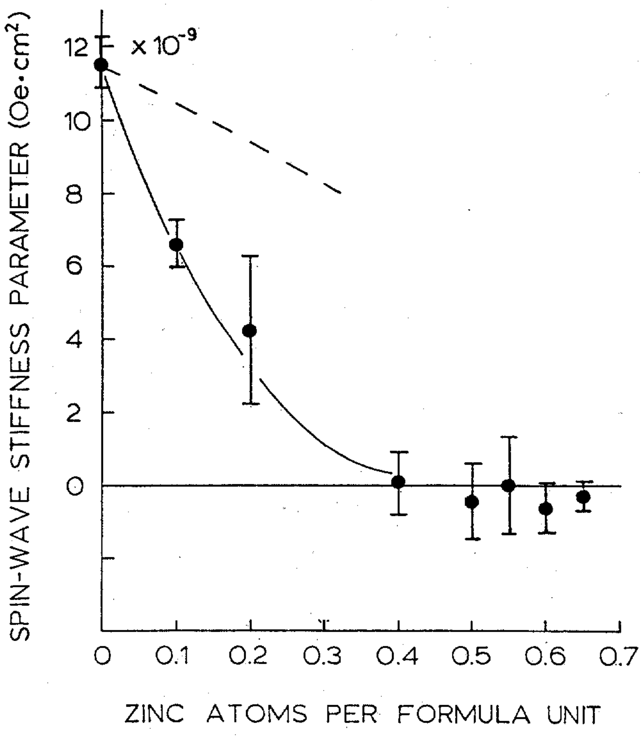

Fig. 3. Experimental spin-wave stiffness parameter vs. zinc content in Li-Zn ferrite. The dashed line shows the results of the spin-wave analysis described in the text.

Table II. Spin-Wave Stiffness Parameter

Zn at./formula unit	Stiffness Parameter
0.0	11.6×10^{-9} Oe·cm^2
0.1	6.6
0.2	4.2
0.4	0.0
0.5	-0.5
0.55	0.0
0.6	-0.7
0.65	-0.3

In a simple spin-wave analysis, based on nearest-neighbor exchange and the two-sublattice model for spinel ferrites, the exchange energy of a magnon with wavenumber k satisfies

$$F_k \propto k^2 \left[\frac{-q_A n_{AA} |J_{AA}|}{16} - \frac{q_B n_{BB} |J_{BB}|}{24} + \frac{11(q_A n_{AB} + q_B n_{BA}) |J_{AB}|}{192} \right] , \qquad (4)$$

where q_A and q_B denote the number of Fe^{3+} ions per formula unit on A and B sites, respectively. The number of nearest neighbor A-ions to an A-ion is denoted by n_{AA}, the number of nearest neighbor B-ions to an A-ion by n_{AB} and similarly for n_{BB} and n_{BA}. Expressions for the q parameters and average n parameters are:

$$q_A = 1-x \qquad (5a)$$

$$q_B = 1.5 + x/2 \qquad (5b)$$

$$\langle n_{AA} \rangle = 4(1-x) \qquad (5c)$$

$$\langle n_{AB} \rangle = 6(1.5+x/2) \qquad (5d)$$

$$\langle n_{BB} \rangle = 3(1.5+x/2) \qquad (5e)$$

$$\langle n_{BA} \rangle = 6(1-x) \qquad (5f)$$

The exchange coefficients of Dionne [8] were then used with the above expressions to calculate the variation in exchange stiffness D as a function of zinc content. Dionne used a molecular field model for collinear spins (no canting) to fit magnetization and T_C data and thereby determine the molecular field parameters λ_{AA}, λ_{AB}, and λ_{BB} vs. zinc content up to x = 0.3. The conversion between molecular field parameters and exchange integrals is given by:

$$|J_{AA}| = \frac{(g\mu_B)^2 q_A |\lambda_{AA}|}{2n_{AA}} , \qquad (6a)$$

$$|J_{BB}| = \frac{(g\mu_B)^2 q_B |\lambda_{BB}|}{2n_{BB}} , \qquad (6b)$$

$$|J_{AB}| = \frac{(g\mu_B)^2 q_A q_B |\lambda_{AB}|}{[q_A n_{AB} + q_B n_{BA}]} . \qquad (6c)$$

These expressions were used to calculate the x-dependence of D. The results of this calculation are shown in Fig. 3 as the dashed line. The proportionality factor in (4) was adjusted to fit the theory to the data at x = 0. It is clear that the predicted decrease in exchange is much weaker than found experimentally.

The reasons for the failure of the model to explain the rapid drop in D with x are not completely clear. The present results indicate that a molecular field approach with averaged concentrations and nearest neighbor parameters provide an inadequate basis for the analysis of spin dynamics in magnetic oxides with random substitutions.

ACKNOWLEDGEMENTS

The authors are indebted to Dr. W. Wettling for his assistance in setting up the light scattering system, Mr. R. G. West, Trans. Tech., Inc., for providing the polycrystalline samples, and Dr. P. Feldman for providing the single crystal samples.

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