BRILLOUIN LIGHT SCATTERING STUDY OF THE SPIN WAVE STIFFNESS PARAMETER IN Sc-SUBSTITUTED LUTETIUM-YTTRIUM IRON GARNET

J. G. Booth, G. Srinivasan, C. E. Patton, and P. de Gasperis

Abstract - The spin wave exchange stiffness coefficient has been measured for a series of Sc substituted lutetium-yttrium iron garnet (YIG) liquid phase epitaxy thin films. Results were obtained for a range of scandium substitutions, $0 < y < 0.83$, based on the formula

$$(Y_{3-x}Lu_x)[Fe_{2-y}Sc_y]Fe_3O_{12}.$$

The exchange determinations were based on magnetization data and spin wave dispersion measurements by Brillouin light scattering. Scandium substitution results in a rapid decrease in the exchange stiffness from the value for pure YIG; the stiffness decreases to zero at approximately $y = 1$. The observed variation of the exchange stiffness as a function of y is in general agreement with theory and with previous microwave data.

Introduction

Scandium substitution occurs for Fe^{3+} ions on the a-sublattice in yttrium iron garnet (YIG). The site distribution for the series investigated here can be written as

$$(Y_{3-x}Lu_x)^c[Fe_{2-y}Sc_y]^a(Fe_3)^d O_{12},$$

where the superscripts denote the pertinent sublattices for the YIG structure. The Lu substitutions are for film substrate lattice matching. An increase in magnetization occurs for low levels of Sc substitution due to the reduction in the net a-site moment. A magnetization maximum occurs for $y \sim 0.7$ [1,2,3]. For larger scandium levels, the magnetization decreases, due to random canting of the d-sublattice iron moments [3]. This canting occurs because the substitution dilution of the a-sublattice also weakens the exchange coupling between the a- and d-sublattices and the a-d interaction is no longer sufficient to maintain the parallelism of the d-sublattice moments.

The purpose of this work was to examine the effect of scandium substitution on the spin wave exchange stiffness parameter D_{ex}. Recent work on the Li-Zn spinel ferrite system revealed a rapid drop in D_{ex} with zinc [4]. Similar effects were anticipated for YIG with Sc substitution. The work was carried out for a range of Sc substitutions, $0 < y < 0.83$. The D_{ex} determinations were based on magnetization data and Brillouin light scattering (BLS) measurements of thermal magnon frequencies. All data were obtained at room temperature.

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J. G. Booth is with the Department of Pure and Applied Physics, University of Salford, Salford M5 4WT, UK. This work was performed while Professor Booth was a Visiting Professor at Colorado State University.

G. Srinivasan and C. E. Patton are with the Department of Physics, Colorado State University, Fort Collins, Colorado 80523.

P. de Gasperis is with the Instituto di Elettronica dello Stato Solidi - CNR, Rome 00156, Italy.

It is important to realize that scandium substitutions result in a continuous decrease in the Curie temperature T_c, also due to the weakening of the average a-d exchange interaction. The Curie temperatures were all above room temperature for the samples measured here. One additional sample with $y = 1.1$ had a T_c - value below room temperature; no BLS magnon line or ferromagnetic resonance could be observed for this material. T_c falls below room temperature at about $y = 0.9$ [2].

Experiment

The single crystal garnet films were prepared by liquid phase epitaxy using horizontal dipping at about 100 rpm. The substrate material was (111) oriented gadolinium gallium garnet; the substrates were 1 inch diameter, about 500 μm thick, and polished on both surfaces. The film thicknesses were in the 3 - 10 μm range. Ferromagnetic resonance (FMR) measurements were used to determine the gyromagnetic ratio, the effective saturation induction $4\pi M_s^{eff}$, and (in combination with literature data on magnetization) the effective anisotropy fields for the films. Compositions were estimated from x-ray lattice parameter determinations.

Brillouin light scattering was carried out by back reflection of argon ion laser light from the surface of the samples which were in the form of flat plates approximately 25 mm^2 in area. The incident light wavelength λ was 4880 Å. A multipass tandem Fabry-Perot interferometer with a contrast of 10^{12}, similar to that described by Sandercock [5], was used to determine the magnon frequencies as a function of field. The incident light was perpendicular to the static applied field and a polarization analyzer oriented perpendicular to the incident light polarization was positioned after the collecting lens to reduce the intensity of the specularly reflected light. Typical spectra are shown in Fig. 1, where the channel content of a multichannel analyzer is shown as a function of frequency for $y = 0$ [Fig. 1(a)] and $y = 0.54$ [Fig. 1(b)]; the in-plane magnetic field was 3 kOe in both cases. The inward shift of the magnon peaks (labelled M) from Fig. 1(a) to Fig. 1(b) indicates the reduction in spin wave frequency (and hence the spin wave stiffness) caused by scandium substitution. Because of the low Curie temperatures for the larger y-values, it was found important to avoid heating effects by using the lowest possible laser powers for the BLS measurements. The incident laser power on the samples was typically below 10 mW.

Data on magnon frequency versus static applied field H were obtained for all the samples except for $y = 1.1$. These data yielded linear plots which were fitted to the spin-wave dispersion equation,

$$f_\kappa = \gamma\,[(H + D_{ex}\kappa^2)(H + D_{ex}\kappa^2 + 4\pi M_s)]^{1/2} \qquad (1)$$

where f_κ is the magnon frequency, γ is the gyromagnetic ratio, κ the magnon wave number, and $4\pi M_s$ is the saturation induction. Equation (1) is specifically for a spin wave propagation perpendicular to the static field. In the present geometry, this direction is also along the film normal.

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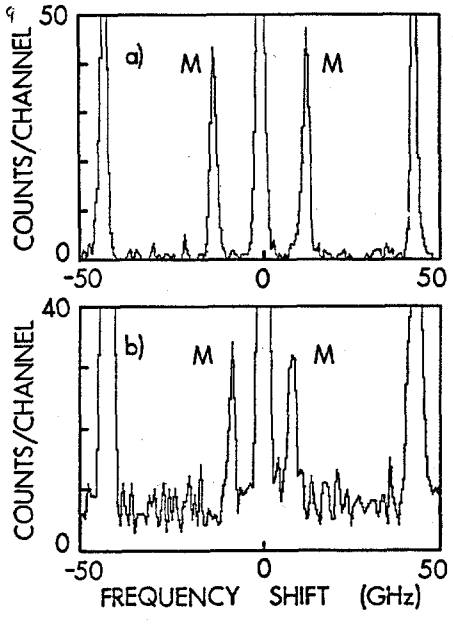

Fig. 1. Brillouin light scattering (BLS) spectra for scandium substituted Lu-YIG samples with (a) 0 and (b) 0.54 Sc atoms per formula unit, both for an applied field of 3 kG. The peaks labelled M are due to thermal magnons propagating perpendicular to the applied field.

The magnon κ-value for back scattering is given by

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

where λ is the light wavelength and n is the refractive index. For the evaluation of κ, an n-value of 2.46 for the pure YIG at 4880 Å was used [6]. From (2), the wave number κ is 6.34×10^5 cm^{-1}. Nonmagnetic substitutions are expected to result in some decrease in the refractive index [7], but the effect on D_{ex} is estimated to be small, ranging from a few percent at y = 0.2 (where D_{ex} is still large) to about 10 % at y = 0.8 (where D_{ex} is already quite small).

Results

The BLS values of D_{ex} as a function of the level of Sc substitution are shown in Fig. 2 by the open squares. The open circle data points correspond to earlier results obtained by Krahn et al. [8] on a similar series of samples by microwave resonance techniques. The BLS data point at y = 0 is for a pure YIG film provided by Westinghouse. Although the BLS values for the Lu-Sc substituted films tend to be somewhat smaller than the microwave results, the agreement is reasonably satisfactory except for the point at y = 0.2. Additional samples in the y = 0.1 - 0.4 range are presently in preparation for additional measurements.

In any case, the rapid drop in D_{ex} with Sc substitution is in marked contrast with the gradual decrease in the Curie temperature T_c. Literature data on T_c versus y [2] are shown in the insert in Fig. 2. The experimental D_{ex} values do extrapolate to zero at about the same y - value for which T_c falls below room temperature (y = 0.9), in contrast with the situation for Li-Zn ferrite [4].

The solid and dotted lines in the main part of the figure show the results from calculations of D_{ex} versus y at 0 K and room temperature, respectively. The details of the calculations are given in the next section.

Theoretical Analysis

Keffer [9] has given the exchange energy of the acoustic spin-wave mode for pure YIG at 0 K as

$$\epsilon^{+} = (5/16)\kappa^2 a_o^2 \{8J_{aa} + 3J_{dd} - 5J_{ad}\}, \qquad (3)$$

where J_{aa}, J_{dd}, and J_{ad} are the exchange integrals for interactions between the sites indicated and a_o is the lattice parameter. Taking into account the random substitution of scandium ions for Fe^{3+} a-site ions, one may rewrite (3) as

$$\epsilon^{+} = (5/16)\kappa^2 a_o^2 \{8(1-y/2)^2 J_{aa} + 3J_{dd} - 5(1-y/2)J_{ad}\}/(1+y). \qquad (4)$$

The spin wave stiffness factor (in energy units) for a composition y at 0 k, D(y,0), is simply the κ^2 coefficient in (4). Using exchange constants which satisfy both the magnetization and susceptibility data for YIG [10], namely, J_{aa} = -6.5 K, J_{ad} = -30.4 K, and J_{dd} = -12.0 K, one obtains numerical values of D_{ex} which are somewhat lower than observed experimentally. The relative change in D(y,0) with scandium content, however, is consistent with the data in Fig. 2, except for the point at y = 0.2. The variation in D(y,0) with y, adjusted to fit the experimental D_{ex} - value for pure YIG (y = 0), is shown by the solid line in Fig. 2. The good correlation between the measured values of D_{ex} and the 0 K theory demonstrates that the basic fall off is due to a compensation point like effect in exchange as in (3), and not simply due to a decrease in T_c.

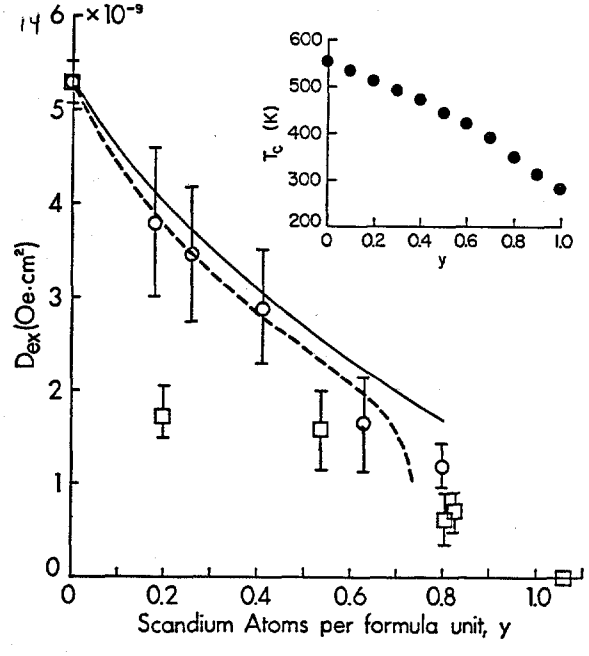

Fig. 2. Brillouin light scattering data on the spin wave stiffness factor D_{ex} in scandium substituted Lu-YIG. The open squares represent the light scattering data and the circles represent the results of microwave measurements from ref. 8. The solid and dotted lines show the theoretical variations in D_{ex} with y at 0 K and 300 K, respectively.

The rapid decrease in T_c with scandium does make it important to consider the effect of temperature on the exchange stiffness. The prescription of Nakamura and Bloch [11] has been used to evaluate the ratio $D(y,T)/D(y,0)$ as a function of temperature. One first writes this ratio as

$$D(y,T)/D(y,0) = 1 - [\{kT/4\pi D(y,T)\}^{3/2} f(S_a,S_d)\epsilon(y,T)]/\Sigma. \quad (5)$$

In (5), the f-function is given by

$$f(S_a,S_d) = 2(S_a^2 + S_d^2)/(S_a - S_d)^2, \quad (6)$$

where S_a and S_d denote the net a- and d-site spin sums for the primitive magnetic cell. The primitive magnetic cell has a volume which is half that of the unit cell. These sums are given by $S_a = (5/2)[4(2-y)]$ and $S_d = (5/2)\cdot 12$.

The energy $\epsilon(y,T)$ is related to the energy of the optical spin wave mode, $\bar{\epsilon}_{y,T}$, according to

$$\epsilon(y,T) = \bar{\epsilon}_{y,T} \exp[-\bar{\epsilon}_{y,T}/kT]. \quad (7)$$

This optical mode energy is assumed to have a temperature dependence which scales with the temperature dependence of the magnetization M_s according to

$$\bar{\epsilon}_{y,T} = \bar{\epsilon}_{y,0} [M_s(T)/M_s(0)], \quad (8)$$

where $\bar{\epsilon}_{y,0}$, the optical mode energy at 0 K, is given by

$$\bar{\epsilon}_{y,0} = -2(n_a S_a - n_d S_d) z_{ad} J_{ad}/ n_d = -10(1 + y)J_{ad}. \quad (9)$$

Finally, Σ is an exchange sum over the sites of the primitive magnetic cell which is given by

$$\Sigma = 2 \sum_{i,j} J_{ij} z_{ij} n_i s_i s_j, \quad (10)$$

in general, and by

$$\Sigma = (200/a_o^3)[2(2-y)^2 J_{aa} + 6J_{dd} - 3(2-y)J_{ad}] \quad (11)$$

for the present situation.

In the above, z_{ij} is the number of j-site nearest neighbors to an i-site ion and n_i (n_j) represents the number of i-site (j-site) magnetic ions in the primitive magnetic cell.

Values of $D(y,T)$ were obtained by first writing (5) in the form

$$[D(y,T)]^{5/2} = D(y,0)\cdot[D(y,T)]^{3/2} - D(y,0)\cdot X, \quad (12)$$

where X is simply a numerical constant for any particular y-value and temperature T. Using values of $D(y,0)$ from (4), one obtains $D(y,T)$ by solving (12) for $[D(y,T)]^{1/2}$ and discarding the extraneous roots. Such calculated values of $D(y,T)$ versus T are shown by the dotted line in Fig. 2, with appropriate scaling to fit the data for pure YIG [as for $D(y,0)$]. Taking the temperature into account in the theory results in a slightly smaller value of D_{ex} for modest values of y. The drop in D_{ex} becomes much more abrupt at large values of y and there appears to be a cutoff in the vicinity of y = 0.75, somewhat below the y - value at which T_c falls below room temperature (y = 0.9). It appears, therefore, that the actual zero in D_{ex} at room temperature is due to the combination of exchange compensation and thermal effects.

Conclusion

The results of the present investigation are as follows: (1) Scandium substitution in Lu-YIG leads to a rapid decrease in the exchange stiffness, qualitatively similar to the previously reported result for Zn substituted Li-ferrite. (2) The exchange stiffness drops to near zero at about 0.9 Sc atoms per formula unit. (3) The observed drop in D_{ex} is consistent with theory and can be loosely viewed as due to a compensation point effect for exchange. (4) The actual zero at y = 0.9 is due to a combination of exchange compensation and thermal effects.

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