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SPIN WAVE STIFFNESS PARAMETERS IN LITHIUM-ZINC FERRITES

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The spin wave exchange stiffness has been calculated for a series of zinc substituted lithium ferrites and compared with measured values obtained by Brillouin light scattering. The theory yields values of 2.9×10^{-9} to 0.9×10^{-9} Oe-cm^2, relative to the observed values of 11.6×10^{-9} to 0 Oe-cm^2, for 0 to 0.4 Zn at./ formula unit, respectively.

INTRODUCTION

Zinc substitution in lithium ferrite occurs for tetrahedral site iron ions in the spinel structure with a site distribution given by

$$(Zn_xFe_{1-x})^A[\,Li_{0.5\,-\,x/2}\,Fe_{1.5\,+\,x/2}]^B\;O_4,$$

where A and B denote the tetrahedral and octahedral sites, respectively. For low levels of zinc substitution, the magnetization increases due to the reduction in the A-site moment. The magnetization reaches a maximum at $x = 0.4$. For larger zinc substitutions, a decrease in magnetization occurs due to canting of the B-site iron moments.[1] The introduction of zinc in the A-sublattice leads to a weakening of the exchange coupling between the iron moments on the A- and B-sites. Consequently, the B-sublattice moments become canted since the A-B interaction is not strong enough to maintain ferromagnetic ordering among the octahedral site spins.

The spin wave stiffness parameter D_{ex} for a range of zinc substitution x from 0 to 0.65 in lithium ferrite has been previously measured at temperatures in the region of 450 K by the Brillouin light scattering (BLS) technique.[2] The BLS data gave a D_{ex} of 11.6×10^{-9} Oe-cm^2 for pure lithium ferrite. A rapid decrease in D_{ex} was observed with Zn substitution and the stiffness parameter was essentially zero for $x = 0.4$. Recent data on scandium substituted lutetium yttrium iron garnet (YIG) show a similar drop in D_{ex} with scandium substitution.[3]

A simple molecular field calculation of the spin wave stiffness in Li-Zn ferrite yielded a much less marked fall off in D_{ex} than was found experimentally.[2] The objective of this work was to extend the theoretical analysis to take temperature effects into account and to make further comparisons with experiment. The theoretical falloff in D_{ex} is in qualitative agreement with but still somewhat weaker than indicated by the data. The situation is similar to that for the Sc-YIG.[3]

THEORY

The ground state of a ferrimagnet is very complex, but the low lying excitations may be adequately described in terms of spin waves and a simple trial ground state.[4] Based on a simple two-sublattice Neel model, the Hamiltonian is given by

$$H_{ex} = -\sum_{i,j} 2\,J_{ij}\,\underline{S}_i\cdot\underline{S}_j\,, \qquad (1)$$

where $\underline{S}_i$ and $\underline{S}_j$ are two nearest neighbor spins located on sublattices i and j respectively and the J_{ij} denote particular exchange constants. At small wave vector $\underline{\kappa}$ the energy for the acoustic spin wave branch is given by [4]

$$\epsilon^+ = \sum_{i,j}[J_{ij}(\underline{S}_i\cdot\underline{S}_j)(\underline{\kappa}\cdot\underline{r}_{ij})^2]/|\textstyle\sum_i \underline{S}_i|, \qquad (2)$$

where the summation is over the primitive magnetic cell only and $\underline{r}_{ij}$ is the usual displacement vector. The spin wave stiffness

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parameter is given by the κ^2 coefficient in Eq.(2). For pure lithium ferrite, the expression for the stiffness parameter at 0 K, D(0,0), can be written as[5]

$$D(0,0) = 5a^2[8J_{AA} + 9J_{BB} - 33J_{AB}]/64 , \quad (3)$$

where a is the lattice spacing and the J's are the exchange constants between nearest neighbor A- and B-ions. Taking into account the substitution of the zinc ions on to the A sites and averaging over the entire randomly substituted crystal, one obtains a stiffness parameter of the form,

$$D(x,0) = 5a^2[f(J_{ij})]/[64(3x + 1)], \quad (4)$$

where D(x,0) is the spin-wave stiffness parameter for a composition x at 0 K. The function $f(J_{ij})$ in Eq.(4) is given by

$$f(J_{ij}) = [8J_{AA}(1 - x)^2 + J_{BB}(3 + x)^2 - 11J_{AB}(1 - x)(3 +x)]. \quad (5)$$

Values of D(x,0) vs. x have been obtained from Eqs.(4) and (5), based on values of the exchange constants from Srivastava et al.[6], $J_{AA} = -20$ K, $J_{AB} = -29$ K, and $J_{BB} = -8$ K. The numerical values of D(x,0) are smaller than the measured D_{ex} values from Ref. 2 by about 75%. Figure 1 shows a comparison of the theoretical x- dependence of D(x,0) and experimental values with all results normalized to values at x = 0. It is clear that the relative drop in stiffness parameter with zinc substitution is much more rapid than expected from the 0 K theory.

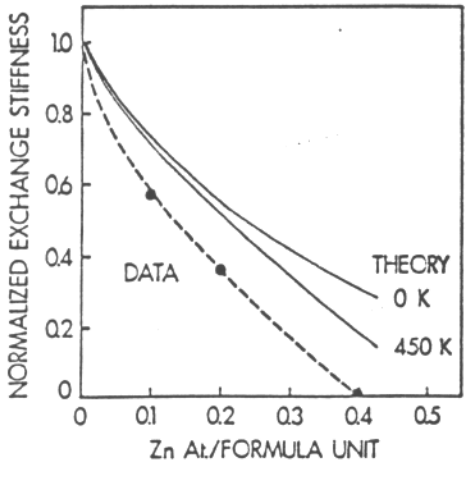

Fig.1 Variation of spin wave stiffness parameter with zinc substitution in lithium zinc ferrites. The solid lines represent the theoretical results at 0 and 450 K, normalized to the values for pure lithium ferrite. The closed circles represent the normalized experimental data measured at 450 K by Brillouin light scattering (Ref. 2).

The temperature variation of D, following the treatment by Nakamura and Bloch,[7] can be expressed as

$$D(x,T)/D(x,0) = 1 - [(kT/4\pi D(x,T))]^{3/2} f(S)\ \epsilon(x,T)]/\ \Sigma. \quad (6)$$

The function f(S) in Eq.(6) is given by

$$f(S) = 2(S_A^2 + S_B^2)/(S_A - S_B)^2, \quad (7)$$

where S_A and S_B represent the net average A- and B-spin sums for the primitive magnetic cell. The primitive magnetic cell consists of four formula units and the spin sums are given by $S_A = 4(5/2)(1-x)$ and $S_B = 4(5/2)(1.5+x/2)$.

The term $\epsilon(x,T)$ is related to the energy of the optical spin wave mode, $\epsilon^-(x,T)$, according to

$$\epsilon(x,T) = \epsilon^-(x,T)\ \exp[-\epsilon^-(x,T)/kT]. \quad (8)$$

The optical mode energy is directly proportional to the magnetization M_s and the temperature dependence of that energy is given by

$$\epsilon^-(x,T) = \epsilon^-(x,0)\ [M_s(x,T)/M_s(x,0)], \quad (9)$$

where $\epsilon^-(x,0)$, the optical mode energy at 0 K, is given by

$$\epsilon^-(x,0) = -2(n_A S_A - n_A S_B) z_{AB} J_{AB}/n_B = 15(3x + 1)J_{AB}. \quad (10)$$

Data on magnetization vs. temperature for the determination of $\epsilon(x,T)$ were obtained from Ref. 8.

Finally, the exchange sum over the sites of the primitive magnetic cell, denoted by Σ in Eq.(6), is given by

$$\Sigma = 2 \sum_{i,j} J_{ij}\ z_{ij}\ n_i\ S_i\ S_j.$$

Here, the n_i (n_j) denote the number of magnetic ions on sublattice i (j) of the primitive unit cell, and z_{ij} is the number of j-site nearest neighbors of an i-site ion. For zinc substituted lithium ferrite, the exchange sum Σ, in terms of average values for the $n_{i,j}$, $S_{i,j}$, and z_{ij} parameters, is given by

$$\Sigma = (25/8a^3)\ [16J_{AA}(1-x)^2 + 3J_{BB}(3+x)^2 - 24J_{AB}\ (1-x)(3+x)]. \quad (11)$$

In order to obtain the explicit variation of D with T for various x-values, Eq.(6) was first written in the form

$$D(x,T)^{5/2} - D(x,0).D(x,T)^{3/2} + C = 0 , \qquad (12)$$

where C is a constant. Using values of D(x,0) from Eq.(4), D(x,T) was obtained by solving Eq.(12) for $[D(x,T)]^{1/2}$. The theoretical variation of D(x,T)/D(0,0) with temperature T is shown in Fig.2 for x = 0, 0.1, 0.2, and 0.4. The roots become complex at the points where the full lines terminate. The normalized theoretical stiffness parameters at 450 K are compared with the reduced experimental data in Fig. 1. One obtains a fall-off in D with x which is slightly more rapid than for T = 0 K, but which still is somewhat less than is found experimentally.

DISCUSSION

The above results show that a molecular field analysis, based on fixed values of the J_{ij} exchange integrals, average n-, S, and z-parameters and with thermal effects taken into account, can account for the essential feature of the spin-wave stiffness found experimentally, namely, the rapid falloff in D with zinc. There are, however, two key problems: (i) D-values which are much smaller than observed experimentally and (ii) a falloff in D with increase in zinc substitution which is somewhat weaker than shown by the data.

Concerning (i), better agreement between theory and experiment could be achieved by increasing the J_{ij} parameters. Such an increase however, would not be consistent with the available data[8] on the temperature dependence of the magnetization. This problem remains unresolved.

For (ii), it is clear from Fig. 1 that the inclusion of the temperature effects in the theory do lead to an additional decrease in D, but this is not sufficient to produce agreement with the data. A further reduction in the theoretical D-values is possible, due to the expected reduction in the J-parameters with zinc substitution. Assuming the empirical dependence of the J_{ij} on the zinc substitution level used by Dionne to fit magnetization vs. temperature data,[8] a decrease in D ranging from 2% for x = 0.1 to about 10% for x = 0.4 is estimated. Such a decrease is insufficient to remedy the discrepancy between theory and the data in Fig. 1. Canting effects may also be important for larger levels of zinc substitution. Dionne's fits do not include canting effects.

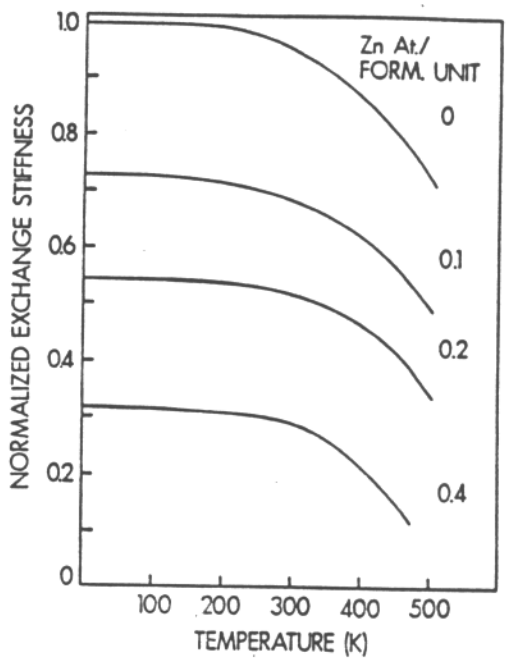

Fig.2 Calculated variation of the theoretical spin wave stiffness parameter with temperature for 0, 0.1, 0.2, and 0.4 Zn at./formula unit. The curves are all normalized to the theoretical value at 0 Zn at./formula unit and 0 K.

It is possible, of course, that the experimental values are too small. A reduction in index of refraction with zinc would lead to such an error. There are no data available to estimate the effect for Li-Zn ferrite, but estimates for yttrium iron garnet with Lu-Sc substitutions indicate that the errors in D are less than a few percent.[3] This effect is too small to account for the discrepancy in Fig. 1.

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