Magnetic properties of lithium zinc ferrite

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The magnetization of Li-Zn ferrite, $Li_{0.5-x/2}Zn_xFe_{2.5-x/2}O_4$, with $x = 0$ to $x = 1.0$ Zn at./f.u. has been examined over the temperature range 4–950 K and for fields up to 90 kOe. Magnetization M and high-field incremental susceptibility χ_{inc} were compared with a calculation based on a localized canting model. A qualitative fit was found for M versus x for an exchange ratio $J_{AB}/J_{BB} = 3$–4, but the experimental and theoretical χ_{inc} versus x results were completely different. On the other hand, a good correlation was found between χ_{inc} and the probability of a B-site Fe having only one A-site Fe neighbor. The data also reveal low-temperature maxima in M in the 30–80 K range for $x = 0.4$ up to 1.0; the effect is most pronounced for $x = 0.7$. Weak mictomagnetic behavior is found for $x = 0.5$ and 0.6. Samples with $x = 0.9$ and 1.0 show no spontaneous moment down to 4 K, but exhibit initial susceptibility maxima at about 15 K with extrapolated Curie temperatures of 10 K and 4 K, respectively. For $x = 0.8$, a Curie temperature of 238 K is obtained. These are considerably lower than literature values.

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INTRODUCTION

Zinc substitutions in lithium ferrite are known to cause an increase in the magnetization for low levels of substitution (below x = 0.3–0.4 Zn at./F.U.) followed by a decrease in magnetization for larger amounts (1). This is generally explained by a magnetic dilution of the A-sublattice with zinc which initially causes the net magnetization to increase with x, due to the dominance of the B-sublattice. As x continues to increase, however, the A-sublattice becomes so magnetically dilute as to reduce the A-B exchange to the point where the B-spins no longer remain parallel but cant to some extent due to the antiferromagnetic B-B exchange (2,3). Canting has, in fact, been observed in NiZn and CoZn ferrite (4,5,6) and Mössbauer evidence for a non-colinear spin structure in high-x LiZn ferrite has recently been found (7).

The objectives of this work were to examine the magnetization M and the high field incremental susceptibility χ_{inc} over a wide range of field and temperature, and compare the results with quantitative canting models. Agreement is qualitative, at best, for M and completely lacking for χ_{inc}. The high field susceptibility appears to be more related to local environment considerations. Furthermore, magnetization temperature maxima and field history effects reminiscent of mictomagnetism are found for x = 0.4 to 1.0 Zn at./F.U.

RESULTS

Magnetic moment data are shown in Fig. 1, plotted as μ_B/atom vs. x, assuming 2.5 μ_B/atom at x = 0. Data were obtained at 4 K and 20 kOe. The straight line indicates the theoretical result expected in the absence of canting, based on a site distribution of

$$(Zn_x^{2+}Fe_{1-x}^{3+})_A\ [Li_{0.5-x/2}^{1+}Fe_{1.5+x/2}^{3+}]_B\ O_4^{2-},$$

and 5 μ_B per Fe^{3+} ion. The solid curves were obtained from a split B'-B" sublattice localized canting scheme similar to that of Ref. 8 with two significant differences. First, a free energy formulation was used to calculate the local B'B" molecular field. It was found that Ref. 8 overestimates that field by a factor of two. Second, an iterative approach was used to obtain self-consistent local canting angles for B-sublattice spins and the average canting angle. Fits for various ratios of the inter- to intrasublattice exchange parameters J_{AB}/J_{BB} are indicated in the figure. It is seen that a qualitative fit is obtained for a J_{AB}/J_{BB} ratio in the range 3-4. In view of the shortcomings of the split-sublattice models, the fit is not unreasonable.

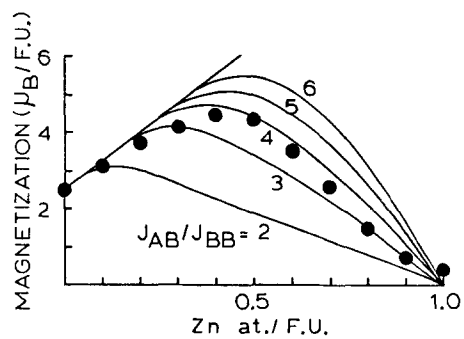

Fig. 1. Magnetization vs. Zn content at 20 kOe and 4 K for Li-Zn ferrite. Theoretical curves are for a self-consistent localized canting model described in text for various ratios of the inter-to-intrasublattice exchange parameters.

A second aspect of canting concerns the high field incremental susceptibility. For a colinear spin structure (e.g., no canting), one would expect the low temperature high field incremental susceptibility to be quite small. For a non-colinear structure, however, the field would serve to straighten up the canted moments to some extent and result in a somewhat larger incremental susceptibility. Experimental results up to x = 0.8 are shown in Fig. 2 for measurements at 4 K from about 60 to 80 kOe. Below 60 kOe, M vs. field exhibits curvature. Above 60 kOe, M vs. field is essentially linear. It is seen that χ_{inc} is essentially negligible below $x \approx 0.4$ and increases rapidly for $x > 0.4$. A theoretical curve based on the model described above for $J_{AB}/J_{BB} = 4$ is also shown. It is clear that the data and the theory are not even functionally similar.

It is not clear why the results are so different for M vs. x and χ_{inc} vs. x, in terms of the localized

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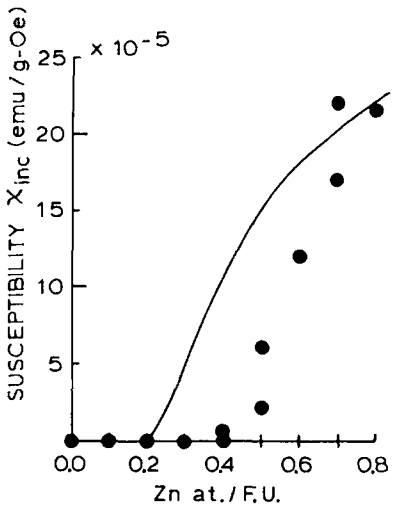

Fig. 2. High field incremental susceptibility vs. Zn content at 4 K for Li-Zn ferrite, from 60 to 80 kOe. The theoretical curve was obtained from the canting model described in the text with J_{AB} = 8 K and J_{BB} = 2 K.

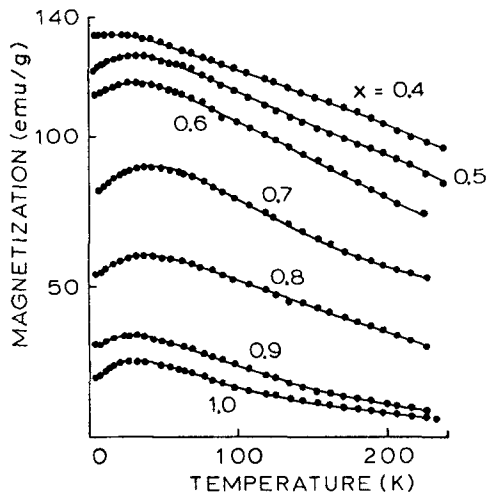

Fig. 4. Magnetization vs. temperature for Li-Zn ferrite with 0.4 to 1.0 Zn At./F.U. at 40 kOe, upon warming from 4 K after cooling from room temperature in zero field.

canting model. One other comparison, however, provides a possible clue to the origin of χ_{inc}. In Fig. 3, χ_{inc} is plotted vs. x, normalized to the value at x = 0.7. The solid lines are simply the probabilities $P_m(x)$ for a B-site Fe ion to have m A-site Fe nearest neighbors. It is seen that χ_{inc} tracks a curve between m = 0 and m = 1 reasonably well. The implication is that B-site iron spins with less than two A-site iron neighbors are the moments responsible for the high field χ_{inc}. This does not necessarily imply that B-site iron with m $\geq$ 2 are colinear. Rather, these moments are simply realigned at fields less than 60 kOe.

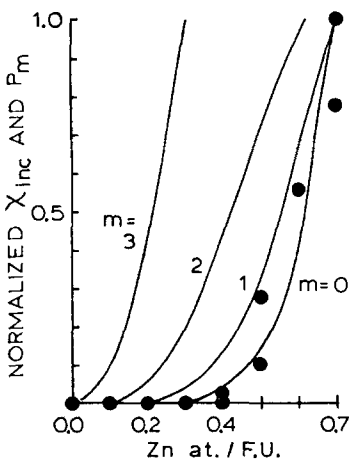

Fig. 3. High field incremental susceptibility vs. Zn content as in Fig. 2, but normalized to the value at 0.7 Zn at./F.U. The solid curves represent the probability P_m of a B-site Fe surrounded by m A-site Fe neighbors, with m = 0, 1, 2, and 3.

It is clear that large Zn substitutions lead to a noncolinear spin arrangement in Li-Zn ferrite. There is further evidence that the magnetic order for large x also involves mictomagnetic tendencies as well as canting. Figure 4 shows data on magnetization vs. temperature, measured upon warming from 4 K in a field of 40 kOe after cooling in zero field. The data show that M vs. T peaks at 30-40 K for x $\gtrsim$ 0.4, with the biggest effect for x $\sim$ 0.7. Mictomagnetic-like field cooling effects were found for x = 0.5 and 0.6, as shown in Fig. 5. Cooling the x = 0.5 sample to 4 K in zero field yields the magnetization maximum indicated upon warming in 40 kOe. The same measurement made after cooling to 4 K in 40 kOe yielded the upper curve. This result is quite reminiscent of mictomagnetism. Higher cooling or measuring fields, up to 80 kOe, yield results not appreciably different from those shown.

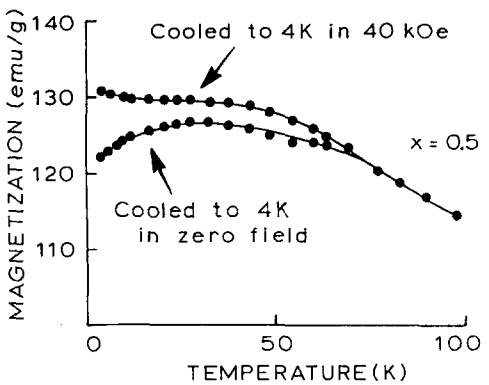

Fig. 5. Magnetization vs. temperature for Li-Zn ferrite with 0.5 Zn at./F.U. at 40 kOe, upon warming from 4 K after cooling from about 250 K in 40 kOe (upper data set) or zero field (lower data set).

Values of the Curie temperature T_c obtained in the course of this work are tabulated in Table I. For x = 0.0 to 0.8, values are based on 8 kOe M vs. T data extrapolated to M = 0. For x = 0.9 and 1.0, M vs. field curves were linear and exhibited no remanence down to 4 K. In these cases, T_c was obatined from extrapolated $1/\chi$ vs. T data. Both samples show minima in $1/\chi$ vs. T at 10-20 K, characteristic of antiferromagnetic (or mictomagnetic) order. It is interesting that the extrapolated T_c values are still positive. The values for 0.8-1.0 are substantially lower than literature values (9). Normalized T_c values are plotted vs. Zn content in Fig. 6. The decrease is linear from x = 0 to x = 0.7, and then drops sharply to the low values of Table I for x = 0.9 and 1.0. The linear decrease in T_c is weaker than one would expect from simple A-sublattice dilution (the dotted line).

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Table I. Curie Temperatures of Li-Zn Ferrite vs. Zn Content.

Zn at./F.U.	T_c(K)
0.0	907
0.1	844
0.2	784
0.3	720
0.4	667
0.5	600
0.6	538
0.7	479
0.8	238
0.9	10
1.0	4

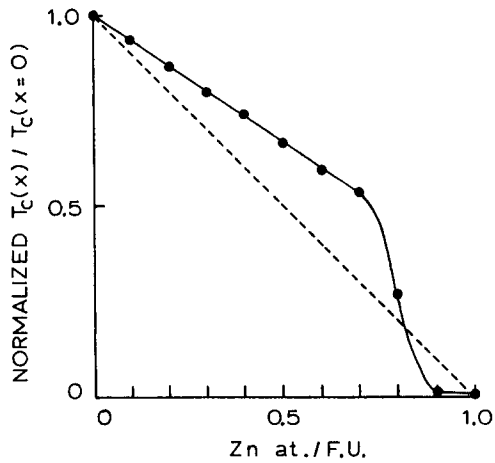

Fig. 6. Normalized Curie temperature vs. Zn content.

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