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A NEW MODEL FOR THE CALCULATION OF THE MAGNETIC PROPERTIES OF DOUBLY SUBSTITUTED FERRITES †

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A model has been developed for the self-consistent calculation of the saturation magnetization and incremental susceptibility of ferrites with nonmagnetic substitutions. The key new feature is an average over all possible local environments which occur at a given substitution level. The model is applied to lithium ferrite with Zn and Ti substitutions. Good agreement with previous data is found. This is only true for $x < 0.6$.

The magnetic structure of lithium ferrite is that of two opposing A and B magnetic sublattices. The magnetic ions are all coupled antiferromagnetically. The A–B coupling generally dominates. There are more B sites than A sites so the net magnetization is simply the difference between the B sublattice magnetization and the A sublattice magnetization. It is well known that Zn substitution in lithium ferrite initially leads to an increase in magnetization. At higher doping levels, a decrease in magnetization occurs. The reason for this is that low doping concentrations lead to a decrease in the number of spins occupying the A sublattice, and this leads to an increased net magnetization in the direction of the B sublattice. As the Zn doping is increased, the B spins are no longer held rigidly in place by the antiferromagnetic coupling to the few remaining A spins, and the antiferromagnetic interaction between B spins leads to canting on the B sublattice. This in turn leads to a decrease in the net magnetization.

There have been many attempts to explain this type of behavior in various substituted spinels and garnets. One of the earliest [1] proposed that the B sublattice splits into two further sublattices B′ and B″, each of which is canted from the direction of net magnetization. It has become apparent, however, that *local* effects control the canting [2]. Recent calculations [3–5] have considered the different possible local environments and then weighted the results of the local picture according to the statistical probability of occurrence for those different environments.

In the present approach, a single spin which is nominally part of the B′ sublattice is considered to interact with nearest neighbor (n.n.) spins on *all three* sublattices. These n.n. spins are taken to be oriented as indicated in fig. 1. The B′ spin under consideration has an energy

$$E = -J_{AB} n_A \cos\theta + J_{BB} n_{B'} \cos(\theta - \theta_{av}) + J_{BB} n_{B''} \cos(\theta + \theta_{av}), \quad (1)$$

where J_{AB} is the A–B n.n. exchange energy, J_{BB} is the B–B n.n. exchange energy, n_A is the number of n.n. A spins for a B spin, $n_{B'}$ is the number of n.n. B′ spins for a B′ spin, and $n_{B''}$ is the number of n.n. B″ spins for a B′ spin.

Energy minimization, $\partial E/\partial\theta = 0$, leads to a condition on the local canting angle θ, in terms of the average canting angle θ_{av},

$$\cot\theta = \frac{n_A J_{AB} - (n_B + n_{B''}) J_{BB} \cos\theta_{av}}{(n_{B''} - n_{B'}) J_{BB} \sin\theta_{av}}. \quad (2)$$

Various approximations can now be used to determine the quantities n_A, $(n_{B'} + n_{B''})$ and $(n_{B''} - n_{B'})$. In the present model, n_A and $(n_{B'} + n_{B''})$ are treated exactly. We consider all of the allowed configurations for different n.n. A sites and B sites occupied by magnetic ions.

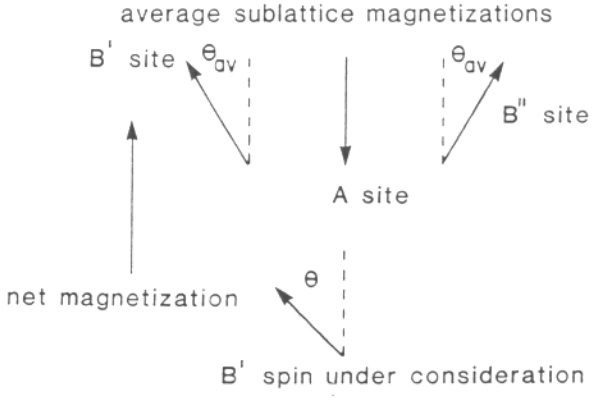

Fig. 1. Model spin configuration. The spin under consideration makes an angle θ with the net magnetization axis. The surrounding spins are directed in accordance with the *average* canting angle θ_{av}.

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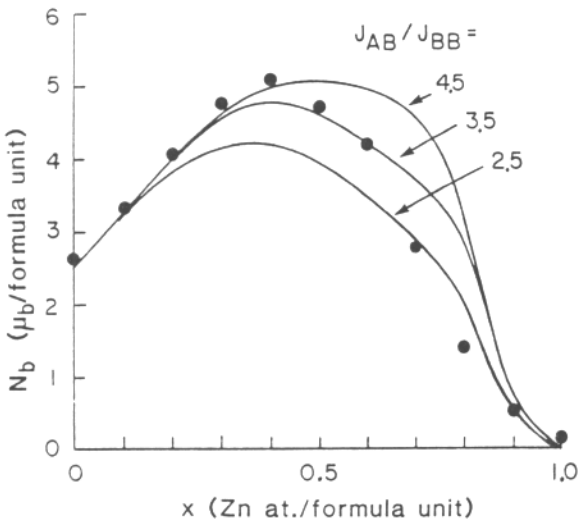

Fig. 2. Moment at 0 K, N_b, in μ_B per formula unit, vs. x, the level of zinc substitution in atoms per formula unit, for LiZn ferrite. The solid lines correspond to the theory and the solid points show data from ref. [5].

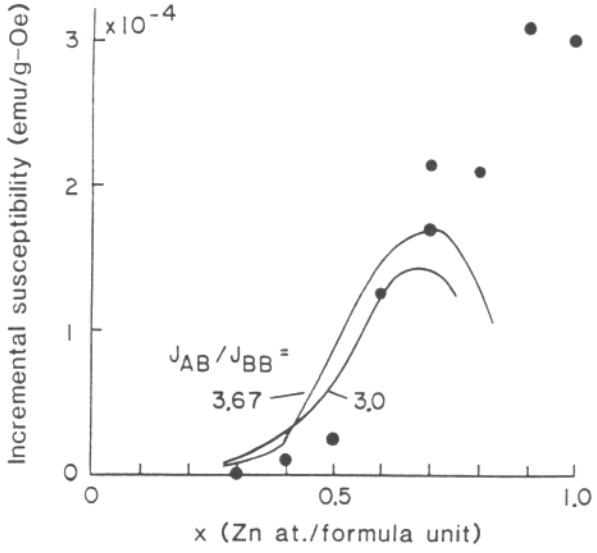

Fig. 3. Incremental high field susceptibility vs. x, the level of zinc substitution in atoms per formula unit, for LiZn ferrite. The solid lines correspond to the theory and the solid points show data from ref. [5].

calculate the local angle in each case, and then perform a weighted average over the local angles to get the average angle θ_{av}. The quantity $(n_{B''} - n_{B'})$ is set equal to unity. That is, for the B′ spin under consideration, there will be on the average one more n.n. B″ spin than B′ spin.

The numerical calculation starts with an initial guess at θ_{av}. One then calculates the different local θ-values appropriate to the different possible environments. These θ-values are then statistically averaged and a new average θ_{av}-value is calculated. This process is repeated until the results are self-consistent.

Now turn to the results of these calculations. Fig. 2 presents a plot of the net magnetic moment per formula unit, N_b, in units of μ_b (the Bohr magneton), versus x, the level of Zn substitution per formula unit for LiZn ferrite,

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

The solid points show the data of ref. [5]. The theoretical results match the data for a J_{AB}/J_{BB} ratio of about 3.5 and a substitution level x below 0.6. This ratio is consistent with values of the exchange constants reported by Srivastava et al. [6]. Above $x = 0.6$, no single exchange ratio fits the data. There is experimental evidence that the exchange constants change significantly for large levels of substituent [7].

One can easily extend the calculations to include the effect of an applied field and obtain the high field incremental susceptibility. The corresponding theoretical results and previous data [5] for LiZn ferrite are shown in fig. 3. Again, for $x \leq 0.6$, the agreement is satisfactory. The best fit curves again correspond to exchange parameters which are close to those reported by Srivastava et al. In the region $x \geq 0.7$, the theoretical and experimental results do not agree at all. The theoretical susceptibility turns down at these higher x-values while the data continue to show an increase. This discrepancy is related to the use of fixed values for the exchange parameters in the theory. In reality, however, the exchange constants seem to be decreasing. This results in spins which are less strongly bound, so that the magnetic field can still cause some significant shift in the direction of the spins.

We conclude with some results for double substitutions in lithium ferrite. B sublattice substitutions should reduce the average B–B exchange and thereby inhibit canting. This could lead to an increase in net magnetization. Calculations based on the present model show that for the system $(Zn_xFe_{1-x})_A(Li_{0.5-x/2+t/2}Fe_{1.5-x/2+3t/2}Ti_t)_BO_4$ [7], the canting is indeed inhibited to some degree by the addition of Ti. The effect is still insufficient, however, the overcome the decrease in the number of spins on the B sublattice with increasing t. The net result is a decrease in magnetization, except for very high levels of zinc.

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