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Magnetomechanical Ratio and Related Magnetic Properties of Nickel-Zinc Ferrites

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Magnetomechanical ratios (g') have been measured by the Einstein–de Haas method for furnace-cooled samples of the ferrites $Zn_x Ni_{1-x} Fe_2O_4$ ($x = 0$, 0.1, 0.15, 0.2, 0.35, 0.5, 0.63, 0.73) at (300 ± 9) °K. The g' values vary in an irregular way with increasing Zn^{+2} concentration from (1.849 ± 0.002) at $x = 0$ to (1.974 ± 0.006) at $x = 0.73$. Using the experimentally determined ionic distributions and Yafet-Kittel angles reported from neutron diffraction work on similar samples, we have calculated the expected g' values at 0 °K. For our calculations, the g' for Ni^{+2} on a B site was obtained using averaged reported saturation magnetization values. Plots of the experimental and calculated g' values compare quite favorably and substantiate the three-sublattice molecular-field interpretation. The g' values have also been used to determine the effective electronic orbital and spin contributions to the magnetization. The percent orbital contribution varies from 8.2 to 1.4%. The expression $1 = 1.016(g)^{-1} + (g')^{-1}$ is found to relate the Einstein–de Haas g' values with recently reported ferromagnetic-resonance g values to within about 0.5% accuracy.

I. INTRODUCTION

In this paper we report g' values for the ferrites $Zn_x Ni_{1-x} Fe_2O_4$ measured by the Einstein–de Haas method,[1] all at (300 ± 9) °K. The g' values of these ferrites have been determined to be independent of temperature for the range of temperature reported (± 9 °K) and are taken to be temperature independent at all temperatures far below the Néel temperatures.

The measured g' values are generally interpreted in terms of the individual constituent ions and hence are related to the individual ionic g' values, the ionic distribution between simple sublattices, the angles between sublattices, and, of course, ultimately to the saturation magnetization M_T. Here we have used values of M_T,[2–11] ionic distributions, and Yafet-Kittel angles[12] reported in the literature to calculate values of g' as a function of zinc concentration. The shapes of the measured and computed curves with increasing Zn^{+2} concentration are quite similar.

In a later section the measured g' values are used to determine the total effective electronic orbital and spin contributions to M_T. The orbital contribution, of course, is always found to be a small fraction of the total M_T.

Improved ferromagnetic-resonance measurements of g have been recently reported for these ferrites.[13] These are discussed in Sec. VII and compared with the g' values.

II. STRUCTURE

The $Zn_xNi_{1-x}Fe_2O_4$ ferrites are known to have a spinel structure.[14] Furthermore, it is generally accepted that the Zn^{+2} ions occupy tetrahedral locations (A sites) and the Ni^{+2} octahedral locations (B sites). Recent neutron diffraction data confirm this structure and ionic distribution.[12]

Using a three-sublattice model, Satya Murthy *et al.*[12] have found a Yafet-Kittel (Y K) type of magnetic ordering to occur at temperatures below those for Néel ordering. No evidence for a spinel spin configuration was found since no satellite reflections were observed.

The Y K angles α_{YK} between magnetizations of B-site Fe^{3+} ions have been determined both as a function of the temperature (T) and Zn^{+2} concentration (x). At 0 °K, α_{YK} increases with x, approaching $\frac{1}{2}\pi$ at $x = 1$ ($ZnFe_2O_4$). Satya Murthy *et al.* have also given the temperature dependence of α_{YK} for various x.

The temperature ranges of the two types of magnetic ordering (Néel and YK) are shown in Fig. 1 as a function of zinc concentration.[12] Systems in the lightly shaded area possess nonzero value of α_{YK} between Fe^{+3} on a B site. At a fixed concentration the YK temperature is defined as the temperature at which the B-site sublattice angle α_{YK} vanishes and Néel ordering appears. To determine α_{YK} for a sample of given Zn^{+2} concentration in the YK region we have linearly interpolated from Satya Murthy's data of α_{YK} at 0 °K.

III. FERRITE SAMPLE PREPARATION

The ferrite samples, after sintering in the presence of oxygen, were furnace cooled and ground to cylinders of 0.561 in. diam $\times \frac{3}{4}$ in. length. Several of these short samples were then stacked together to form a cylinder about $8\frac{1}{4}$ in. long on which the g' measurements were made.

All the ferrite concentrations given are known to

within about 1%. The ferrite samples used in the g' measurements were later tested and found to have low conductivity.[15] We believe this indicates essentially no Fe^{+2} present, except for the $NiFe_2O_4$ sample in which the maximum Fe^{+2} concentration was estimated to be some small fraction of 1%.

IV. MEASUREMENTS OF THE MAGNETOMECHANICAL RATIO g'

Values of g' for this ferrite series were measured using the Einstein–de Haas equipment at the Kettering Magnetics Laboratory, Oakland University, Rochester, Mich. Details of the experiment are given in earlier papers,[16,17] and sources of error have been recently discussed.[18] The measurements are summarized in Table I and plotted in Fig. 2.

V. CALCULATION OF g' FROM IONIC g' VALUES, IONIC DISTRIBUTIONS, AND SUBLATTICE ANGLES α_{YK}

The quantity g' is related to M_S, the electronic spin magnetization, and M_T by[19]

$$g' = 2M_T/(2M_T - M_S). \tag{1}$$

g' for the ferrite samples can be quite easily calculated as a function of the individual ionic contribution and hence of α_{YK} from Eq. (1).

It can readily be shown by summation of individual ionic moments over all sublattices that the saturation moment per molecule at 0 °K can be written[20]

$$M_T = \left[\sum_{j=\text{all sublattices}} \sum_{i=\text{all ionic species}} x_{ij}\left(\frac{g'_{ij}}{g'_{ij}-1}\right) S_{ij}(\cos\alpha_{YK})_{ij}\right]_A - \left[\sum_{j=\text{all sublattices}} \sum_{i=\text{all ionic species}} x_{ij}\left(\frac{g'_{ij}}{g'_{ij}-1}\right) S_{ij}(\cos\alpha_{YK})_{ij}\right]_B , \tag{2}$$

where x_{ij} is the number of ions per molecule of the ith magnetic species in sublattice j, g'_{ij} is the value of the magnetomechanical ratio for the ith ion in sublattice j, $(\cos\alpha_{YK})_{ij}$ is the cosine of the YK angle α_{YK} for the ith magnetic ion in sublattice j, and S_{ij} is the total spin quantum number of the ith ion in sublattice j.

In our calculations we use values of x_{ij} and $(\alpha_{YK})_{ij}$ taken from the neutron diffraction work of Satya Murthy *et al.*[12]

In a similar manner we can also write M_S at 0 °K as

$$M_S = 2\left[\sum_{j=\text{all sublattices}} \sum_{i=\text{all ionic species}} x_{ij}\, S_{ij}(\cos\alpha_{YK})_{ij}\right]_A - 2\left[\sum_{j=\text{all sublattices}} \sum_{i=\text{all ionic species}} x_{ij}\, S_{ij}(\cos\alpha_{YK})_{ij}\right]_B , \tag{3}$$

where the symbols have the same meaning as in Eq. (2). On the A site, the j summation reduces to one term (one sublattice) and on a B site the j summation reduces to two terms.

Using (2) and (3) in (1) gives g',

$$g' = \left\{\left[\sum_{j=\text{all sublattices}} \sum_{i=\text{all ionic species}} x_{ij}\left(\frac{g'_{ij}}{g'_{ij}-1}\right) S_{ij}(\cos\alpha_{YK})_{ij}\right]_A - \left[\sum_{j=\text{all sublattices}} \sum_{i=\text{all ionic species}} x_{ij}\left(\frac{g'_{ij}}{g'_{ij}-1}\right) S_{ij}(\cos\alpha_{YK})_{ij}\right]_B\right\} \times \left\{\left[\sum_{j=\text{all sublattices}} \sum_{i=\text{all ionic species}} x_{ij}\left(\frac{1}{g'_{ij}-1}\right) S_{ij}(\cos\alpha_{YK})_{ij}\right]_A \times \left[\sum_{j=\text{all sublattices}} \sum_{i=\text{all ionic species}} x_{ij}\left(\frac{1}{g'_{ij}-1}\right) S_{ij}(\cos\alpha_{YK})_{ij}\right]_B\right\}^{-1} . \tag{4}$$

In the Ni-Zn ferrites, g' for Fe^{+3} is thought to be the same for both A and B sites and to equal the pure spin value[20] of 2.0023. Zn^{+2} is nonmagnetic and does not contribute. For Ni^{+2} on a B site, g'

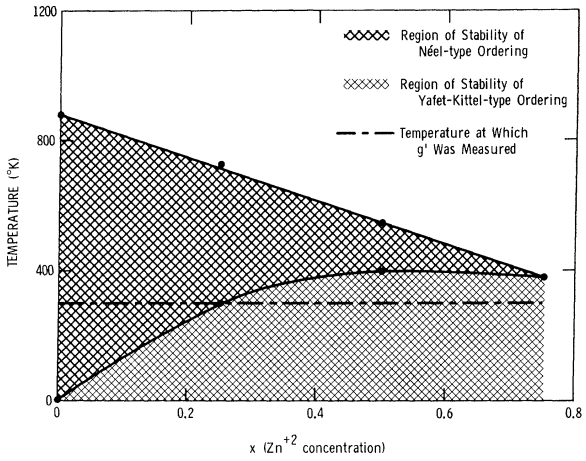

FIG. 1. Magnetic order in $Zn_xNi_{1-x}Fe_2O_4$ systems as determined from neutron diffraction measurements (Ref. 12).

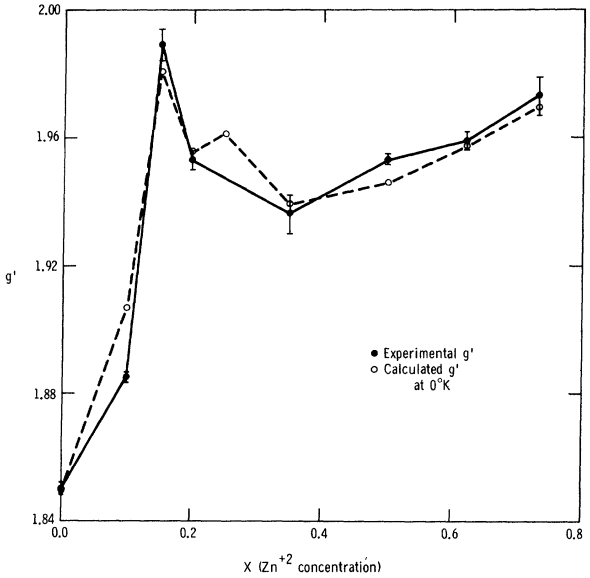

FIG. 2. Experimental [(300 ± 9) °K] and calculated (0 °K) magnetomechanical ratios, g' as a function of Zn^{+2} concentration for $Zn_xNi_{1-x}Fe_2O_4$ ferrites. Mean deviation of calculated g': ±0.013. Mean deviations of experimental g' are given by error bars.

has been obtained using the average of reported values of M_T [Eq. (2)].[2-11] The calculated values of g' appear in Fig. 2.

VI. EFFECTIVE ELECTRONIC ORBITAL AND SPIN MAGNETIZATION

We have used the expressions[19]

$$M_O/M_T = (2-g')/g' \text{ and } M_S/M_T = 2[(g'-1)/g']$$

to derive the orbital and spin contributions to the magnetization from the measured g' values. Our values of the fraction M_O/M_T of orbital magnetization differ from those determined from earlier ferromagnetic resonance g factors by as much as two orders of magnitude. This naturally results because of the relatively smaller errors in g' values as compared with earlier g-factor work. The calculated values are shown in Table II.

VII. COMPARISON OF g' and g

Recently measured values of g for these ferrites together with our g' values allow us to test the generalized Kittel–Van Vleck relationship.[21,22] Measured values of g by Makram[13] are shown as dots in Fig. 3 and interpolated g values used for comparison are indicated by the □'s.

Table III lists interpolated g values, the sum of the reciprocal g and g' values, and ρ, recently defined by Blume, Geschwind, and Yafet.[21] The quantity ρ is the ratio of the expectation value of the real spin to the fictitious spin. In terms of the generalized Kittel–Van Vleck relationship[21,22] we have

$$g' = g(g-\rho)^{-1}.$$

The mean value of ρ for these ferrites is 1.016, where the mean standard deviation is less than 0.005, except for the value at $x = 0.15$ where the interpolation of g was quite uncertain because of its rapidly changing value.

TABLE I. Summary of g' measurements for the ferrite series $Zn_xNi_{1-x}Fe_2O_4$ at (300 ± 9) °K.

x	g'	probable error
0	1.849	±0.002
0.10	1.885	±0.005
0.15	1.989	±0.003
0.20	1.953	±0.006
0.35	1.936	±0.002
0.50	1.953	±0.003
0.63	1.959	±0.003
0.73	1.9(7)	anomalous at room temperature

TABLE II. Electronic orbital and spin contributions to the magnetization of $Zn_xNi_{1-x}Fe_2O_4$ ferrites.

x	M_O/M_T	M_S/M_T
0	0.082±0.001	0.918±0.001
0.1	0.061±0.003	0.939±0.003
0.15	0.006±0.002	0.994±0.002
0.20	0.024±0.003	0.976±0.003
0.35	0.033±0.001	0.967±0.001
0.5	0.025±0.002	0.975±0.002
0.63	0.021±0.002	0.979±0.002
0.73	0.014±0.003	0.986±0.003

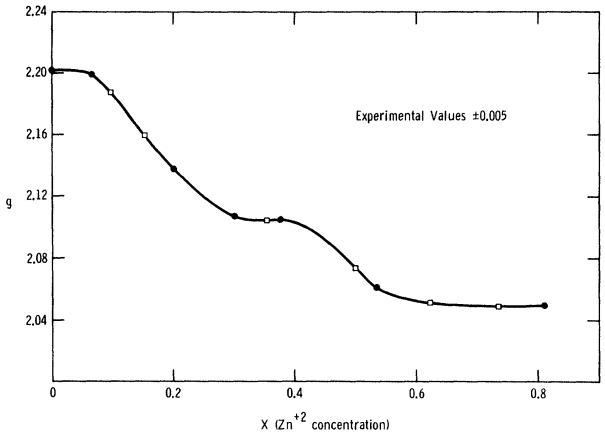

FIG. 3. Ferromagnetic-resonance g factors for $Zn_xNi_{1-x}Fe_2O_4$ as a function of x as reported by Makram (Ref. 13). Dots represent experimental points and □'s refer to "interpolated" values.

VIII. CONCLUSIONS

Measured g' values for Ni-Zn ferrites agree reasonably well with the values predicted from the sum of ionic magnetic moments at 0 °K. Our results seem to indicate that these ferrites can be characterized using the three-sublattice model and YK angles determined from neutron diffraction work. While agreement is not within experimental error at all Zn^{+2} concentrations, the experimental and calculated curves do possess the same general behavior as a function of increasing Zn^{+2} concentration.

We have also calculated g' for $x = 0.25$ at 0 °K using Satya Murthy's α_{YK} and reported M_T. A value of $g' = 1.962$ was obtained (see Fig. 2).

At the two Zn^{+2} concentrations for which both g' and α_{YK} were available in the YK region, the calculated and measured g' values agree within experimental errors.

Considering disagreement among early ferromagnetic resonance experiments and uncertainties in the interpolation of the more recent g values, the correlation between g and g' appears to be unexpectedly good. Although the value of ρ appears to be constant throughout this ferrite series, the limited number of g measurements makes this constancy uncertain.

TABLE III. Comparison of g and g' and validity of the generalized Kittel–Van Vleck relationship (Refs. 21 and 22) for $Zn_xNi_{1-x}Fe_2O_4$ ferrites.

x	g	$1/g' + 1/g$	ρ
0	2.200 ±0.005	0.995 ±0.002	1.01
0.1	2.188[a] > ±0.005	0.988 > ±0.003	1.03
0.15	2.160[a] > ±0.005	0.966 > ±0.001	1.07
0.20	2.118[a] > ±0.005	0.984 > ±0.003	1.03
0.35	2.106[a] > ±0.005	0.991 > ±0.002	1.02
0.50	2.074[a] > ±0.005	0.994 > ±0.002	1.01
0.63	2.052[a] > ±0.005	0.998 > ±0.002	1.00
0.73	2.050[a] > ±0.005	0.995 > ±0.003	1.01
		= 1.016 ± mean deviation < 0.005	
		ρ_{av} (except at $x = 0.15$)	

[a]Interpolated values from Makram (Ref. 13).

[1]G. G. Scott, J. Phys. Soc. Japan Suppl. B-1 17, 372 (1962).
[2]Gmelins Handbuch der Anorganischen Chemie, 8th ed., System 59, Part D, Suppl. p. 371 (unpublished).
[3]E. W. Gorter, Nature 165, 800 (1950).
[4]E. W. Gorter, Phillips Res. Rep. 9, 295 (1954).
[5]J. J. Went and E. W. Gorter, Philips Tech. Rev. 13, 181 (1952).
[6]L. Néel, Ann. Phys. (Paris) 3, 137 (1948).
[7]L. Néel, Compt. Rend. 230, 375 (1950).
[8]C. Guillaud, J. Phys. Radium 12, 258 (1951).
[9]C. Guillaud and M. Roux, Compt. Rend. 229, 1133 (1949).
[10]C. Guillaud and H. Creavaux, Compt. Rend. 230, 1256 (1950); 230 1458 (1950).
[11]C. Guillaud and M. Sage, Compt. Rend. 232, 944 (1951).
[12]N. S. Satya Murthy, M. G. Natera, S. I. Youssef, R. J. Begum, and C. M. Srivastava, Phys. Rev. 181, 969 (1969).
[13]H. Makram, Czech. J. Phys. 17, 387 (1967).
[14]D. H. Martin, *Magnetism in Solids* (MIT Press, Cambridge, Mass., 1967), p. 241.
[15]The authors are indebted to Dr. R. E. Michel of this laboratory for these tests.
[16]G. G. Scott, Rev. Mod. Phys. 34, 102 (1962).
[17]G. G. Scott, Phys. Rev. 148, 525 (1966).
[18]G. G. Scott and H. W. Sturner, Phys. Rev. 184, 490 (1969).
[19]R. A. Reck and D. L. Fry, Phys. Rev. 184, 492 (1969).
[20]E. W. Gorter, Nature 173, 123 (1954).
[21]M. Blume, S. Geschwind, and Y. Yafet, Phys. Rev. 181, 478 (1969).
[22]M. E. Foglio, Phys. Rev. Letters 23, 577 (1969).