On the variation of a.c. susceptibility with temperature for some Cu–Li ferrites

A B NAIK, S R SAWANT, S A PATIL and J I POWAR

Department of Physics, Shivaji University, Kolhapur 416 004, India

MS received 16 September 1988; revised 5 November 1988

Abstract. The a.c. susceptibility vs. temperature of the system $(\text{Li}_{0.5}\text{Fe}_{0.5})_{1-x}\text{Cu}_x\text{Fe}_2\text{O}_4$, for $0 \leq x \leq 1.0$ has been studied. While pure CuFe_2O_4 ($x=1$) shows a dramatic peak in susceptibility just before its Curie temperature (T_c), the same is suppressed with the addition of a small amount of Li to the system. The variation of lattice parameter with Li content has also been investigated. Samples up to $x=0.4$ show tetragonal structure while those with $x>0.4$ are cubic. This change of structure can be explained on the basis of reduction of dsp^2 bonding of Cu ions on B-sites of the spinel lattice.

Keywords. Susceptibility; Curie temperature; lattice parameter; Cu–Li ferrites.

1. Introduction

Lithium ferrite and Li-substituted ferrites, despite their enormous potential for application as microwave devices and memory cores, have not been developed. For application as cores for memory, the squareness ratio and coercive force of ferrites are of importance. From susceptibility–temperature studies of magnetic materials information on the domain state of particles in a bulk sample, such as multidomain (MD), single-domain (SD) and supermagnetic (SP) and the magnetic ordering can be obtained.

The effect of long range ordering in Li-substituted ferrites has been studied by Maddavoma (1974) and Maisin *et al* (1981). Recently, Naik *et al* (1988) carried out some magnetization studies on Cu–Li ferrites. In this paper we report the results of the thermal variation of a.c. susceptibility ($\chi_{a.c.} - T$) and X-ray studies on Cu–Li ferrites.

2. Experimental

Polycrystalline samples having the general formula $(\text{Li}_{0.5}\text{Fe}_{0.5})_{1-x}\text{Cu}_x\text{Fe}_2\text{O}_4$, with $x=0, 0.2, 0.4, 0.6, 0.8$ and 1.0 were prepared by standard ceramic methods. High purity oxides of Fe_2O_3 and CuO and lithium carbonate were mixed in stoichiometric proportions and presintered at 700°C for 4 hr. This was followed by grinding in acetone medium, drying, sintering at 950°C for 20 hr and furnace-cooling at the rate of 40°C/hr . The sieved powders ($38\ \mu$) were pressed into pellets of 1 cm diameter and were sintered at 1000°C for 10 hr. The X-ray diffraction patterns obtained using FeK_α radiation confirmed single phase formation in each of the samples. The $\chi_{a.c.} - T$ measurements were made on polycrystalline samples using a double-coil method operating at 317 Hz and a peak field of 0.5 Oe (Radhakrishnamurty *et al* 1978).

3. Results and discussion

Figure 1 shows the $(\chi/\chi_{RT}) - T$ curves for four of the samples ($x=1.0, 0.8, 0.6$ and

Figure 1. Variation of χ/χ_{RT} with temperature T (K) for samples 1 to 4.

0.4). The lattice parameter and other data for all the samples is given in table 1. The lattice constants shown in table 1 are in good agreement with those reported in literature for Li ferrite (Gorter 1954) and Cu ferrite (Patil 1983). The Li-Cu ferrites up to $x=0.4$ show tetragonal structure and those with $x>0.4$ show cubic structure. Tetragonal distortion decreases with decreasing Cu content at the B-site of the spinel lattice. This is due to the reduction of dsp^2 bonding of Cu ions at the B-site (Goodenough and Loeb 1955). The reduction of covalent bonding is due to more and more of the Li ions replacing Cu at B-sites.

From the curves shown in figure 1, the following points can be noted.

- (i) The $\chi_{RT}-T$ curve of pure CuFe_2O_4 (sample 1) shows a dramatic peak just before T_c . The χ for this sample at the peak is 9.5 times that at the room temperature. Addition of a small quantity of Li to the Cu ferrite suppresses this behaviour as exemplified by the curves of samples 2, 3 and 4 ($x=0.8, 0.6$ and 0.4).
- (ii) Addition of Li increases the Curie temperature (see also table 1).
- (iii) The single sharp decrease in $(\chi/\chi_{RT})-T$ curve for each of the sample signifies one T_c and this implies also a single phase material.

A polycrystalline material may consist of particles of different sizes, depending upon the thermal history of the sample and it is possible to distinguish if these also are of different domain states through susceptibility and hysteresis studies (Radhakrishnamurty *et al* 1982; Joshi and Kulkarni 1986). The appearance of a

Table 1. Curie temperatures obtained from susceptibility measurements for the $(\text{Li}_{0.5}\text{Fe}_{0.5})_{1-x}\text{Cu}_x\text{Fe}_2\text{O}_4$ ferrite system.

Sample		Crystal structure	Lattice constant		Curie temp. from χ/χ_{RT}	Curie temp. observed	Blocking temp T_b
No.	x		a	c	T_c (K)	(K)	(K)
1	1	Tetragonal	8.252	8.731	752	780	742
2	0.8	Tetragonal	8.354	9.816	793	815	773
3	0.6	Tetragonal	8.339	8.813	814	833	804
4	0.4	Cubic	8.341	—	773	885	762
5	0.2	Cubic	8.330	—	—	915	—
6	0	Cubic	8.328	—	—	933	—

sharp cusp in the $\chi - T$ curve is one of the characteristics of a spin-glass system (Connella and Mydson 1972). Also, a peak in the $\chi - T$ curve of a sample can be attributed to a transition of the particles from SD to SP state (Baldha *et al* 1986). In the case of some Fe-Ti ferrites, it has been suggested by Nagarajan and Murty (1981) that there could be regions behaving like SP particles at room temperature and the same could behave like SD at lower temperatures. The specific temperature at which the SD and SP transition for a particle of volume v takes place is known as the blocking temperature, T_b , and the relation governing this is given by

$$vJ_s H_c = 2kT_b,$$

where J_s is the saturation intensity, H_c is the coercive force of the material and k is the Boltzmann constant. For SD particles, H_c is large whereas it tends to zero for SP particles (Bean 1955). Thus χ , which is inversely proportional to H_c , is large for SP of the same material and hence there is a peak in χ at T_b .

From the above concepts, the $\chi_{a.c.} - T$ curves shown in figure 1 imply that there is a pronounced tendency to form SD regions in the case of sample 1 (CuFe_2O_4), which seems to be inhibited by an addition of Li as can be inferred from the behaviour of the other samples. However, the presence of some SD regions even in samples 2 to 4 is indicated by the broad and minor χ -peaks shown by them as well. The predominant T_b for each of these samples, deduced from their respective $\chi_{a.c.} - T$ curves, is given in the last column of table 1. Satyanarayana and Raman Murthy (1985) have observed a χ -peak for cobalt ferrites at 260°C and they termed it an isotropic peak. This type of χ -peak is shown by any sample containing multidomain particles and with an isotropic point. Also, samples with predominantly MD particles show almost constant susceptibility with increasing temperature (flat $\chi - T$ curve) till T_c (Radhakrishnamurty *et al* 1982). Thus it may be concluded that in pure Cu-ferrite, particles with single-domain state are easily formed, which tendency is inhibited by the addition of Li cations to the system.

References

- Baldha G J, Upadhyay R V and Kulkarni R G 1986 *Mater. Res. Bull.* **21** 1051
 Bean C P 1955 *J. Appl. Phys.* **26** 1981
 Connella V and Mydson J A 1972 *Phys. Rev.* **B6** 4220
 Goodenough J B and Loeb A L 1955 *Phys. Rev.* **98** 391
 Gorter E W 1954 *Philips Res. Rep.* **9** 295

- Joshi H H and Kulkarni R G 1986 *J. Mater. Sci.* **21** 2138
- Maddivoma E 1974 *Phys. Status Solidi* **24** K55
- Maisin A M, Feder M and Mites D 1981 *Rev. Roum. Phys.* **26** 69
- Nagarajan R and Murty C R K 1981 *Bull. Mater. Sci.* **3** 217
- Naik A B, Patil S A and Powar J I 1988 *J. Mater. Sci. Lett.* (in press)
- Patil S A 1983 *Indian J. Pure Appl. Phys.* **21** 182
- Radhakrishnamurty C and Nanadikar N G 1979 *Pramana-J. Phys.* **13** 413
- Radhakrishnamurty C, Likhite S D and Sahasrabudhe P W 1978 *Proc. Indian Acad. Sci. (Earth Planet. Sci.)* **87** 245
- Radhakrishnamurty C, Likhite S D, Deutsch E R and Murthy G S 1982 *Phys. Earth Planet. Inter.* **30** 281
- Satyanarayana S and Raman Murthy S 1985 *J. Mater. Sci. Lett.* **4** 606