

Susceptibility and magnetization studies of Gd^{3+} substituted Mg–Cd ferrites

JL BHOSALE, SN KULKARNI, RB SASMILE and BK CHOUGULE*

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

MS received 23 May 1995; revised 22 August 1995

Abstract. Studies on lattice parameters, magnetization and ac susceptibility measurements for the ferrite system $Cd_xMg_{1-x}Gd_yFe_{2-y}O_4$ (with $x = 0.2, 0.3, 0.4$ and $y = 0.1, 0.2, 0.3, 0.4$) are presented. The ac susceptibility was measured from room temperature to 800 K at a constant magnetic field of 7 oersted. X_{ac} vs T variations show that all the samples contain predominantly multidomain (MD) particles. Magnetization measurements of the system indicate that as the Cd^{2+} content increases magnetization increases while the addition of Gd^{3+} reduces the magnetization. The observations further indicate the existence of Y–K type of magnetic ordering in the system.

Keywords. ac susceptibility; magnetization; Mg–Cd–Gd ferrites.

1. Introduction

The zinc and cadmium containing ferrites are a class of very important magnetic materials because of their wide variety of applications in microwaves. Magnetization studies on many of the Zn and Cd substituted ferrites like Ni–Zn, Fe–Zn, Cu–Zn, Cu–Cd and Ni–Cd (Leung *et al* 1973; Srivastava *et al* 1976; Kulkarni and Patil 1982; Kulkarni and Panicker 1984; Panicker *et al* 1984) show canted spin arrangement on the octahedral site. The canting of the spins gives rise to Yafet–Kittel (Y–K) angles. Many workers have reported the ac susceptibility studies for mixed ferrites and concluded that three types of ferromagnetic particles viz. multidomain (MD), single domain (SD) and superparamagnetic (SP) exists within the material (Kulkarni and Baldha 1985; Baldha *et al* 1986). Not much information on the magnetic behaviour of trivalent rare earth ion substituted Mg–Cd ferrites is available in the literature. The present communication reports studies on compositional variation of lattice parameter, magnetization and temperature variation of susceptibility in Gd^{3+} substituted Mg–Cd ferrites. The variation of low field ac susceptibility with temperature was studied especially to determine the nature of magnetic ordering in the system.

2. Experimental

Polycrystalline ferrites having the general formula $Cd_xMg_{1-x}Gd_yFe_{2-y}O_4$ ($x = 0.2, 0.3, 0.4$ and $y = 0.1, 0.2, 0.3, 0.4$) were prepared by the standard ceramic method using AR grade oxides as starting materials. The oxides were mixed in stoichiometric proportions and fired at 1050°C for 24 h and cooled slowly to room temperature.

The ac susceptibility measurements, on powdered samples, were made in temperature range from 300 K to 800 K using a double coil set up operating at a frequency of 263 Hz and in the r.m.s. field of 7 oersted described by Murthy *et al* (1984). The saturation

magnetization of each sample was measured by using the high field hysteresis loop tracer (Radhakrishnamurthy *et al* 1965).

3. Results and discussion

The X-ray powder diffraction patterns of some representative samples are shown in figure 1. The variation of lattice constant with cadmium content for the samples is shown in figure 2. The lattice constant increases as Cd content increases. This is due to larger ionic radius of Cd^{2+} (0.99 Å) as compared to the smaller Mg^{2+} (0.67 Å) ions. The lattice constant does not vary much with Gd^{3+} (1.02 Å) content (table 1). The diffractograms of the Gd containing samples show the presence of a additional small peak on the lower angle side of (311) plane. The ASTM data confirms that this reflection is due

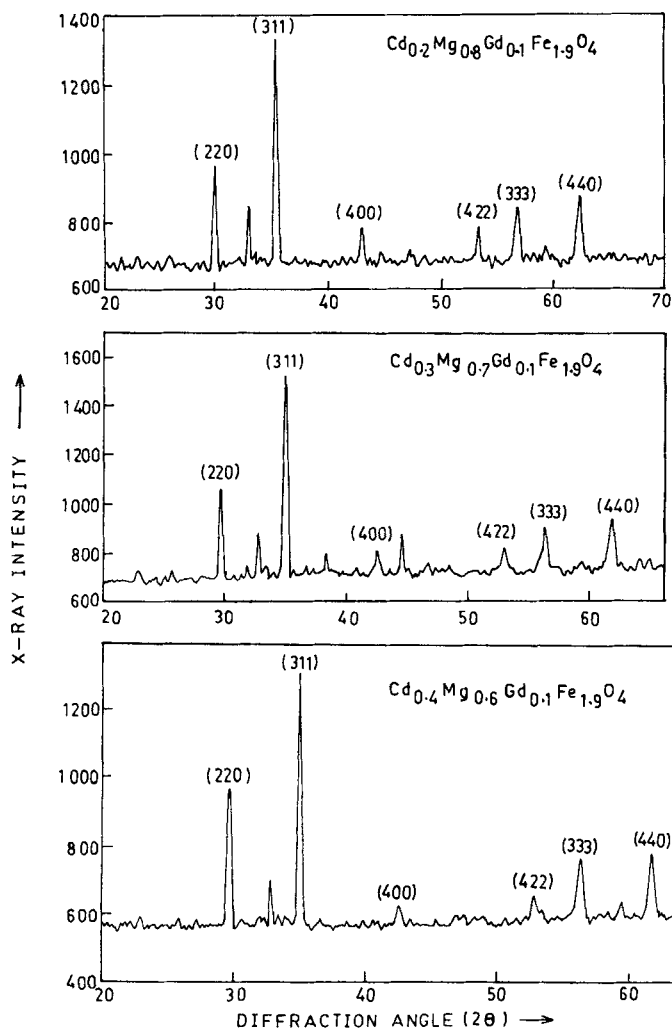


Figure 1. X-ray diffraction patterns for $\text{Cd}_x\text{Mg}_{1-x}\text{Gd}_y\text{Fe}_{2-y}\text{O}_4$ ferrites.

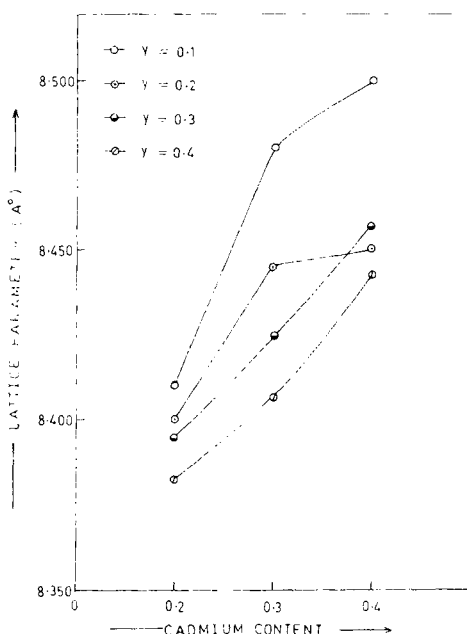


Figure 2. Variation of lattice parameters with content of cadmium for $\text{Cd}_x\text{Mg}_{1-x}\text{Gd}_y\text{Fe}_{2-y}\text{O}_4$ ferrites.

Table 1. The X-ray density (d_x), saturation magnetization (σ_s) and Curie temperature (T_c) for $\text{Cd}_x\text{Mg}_{1-x}\text{Gd}_y\text{Fe}_{2-y}\text{O}_4$ system.

Composition x	y	d_x (gcm^{-3})	σ_s (emu g^{-1}) 300 K	Curie temp. (°K) (susceptibility)	M_r/M_s
0.2	0.1	5.08	34	615	0.523
0.3		5.15	36	582	0.478
0.4		5.30	40	548	0.518
0.2	0.2	5.32	27	613	0.52
0.3		5.44	30	605	0.5
0.4		5.62	31	585	0.533
0.2	0.3	5.56	19	600	0.464
0.3		5.70	23	603	0.472
0.4		5.83	26	598	0.458
0.2	0.4	5.82	15	650	0.5
0.3		5.96	18	575	0.428
0.4		6.08	19	582	0.428

to GdFeO_3 perovskite phase formed at high temperatures due to the higher reactivity of Gd and Fe (Kolekar *et al* 1994). A similar phase has also been reported by Tsagaroyannis *et al* (1992) in gadolinium–iron ferrites.

The data on X-ray and physical densities are given in table 1. The X-ray density increases with x and y , which suggests that the increase in mass overtakes the increase in volume of the unit cell with Cd and Gd content.

The magneton numbers [n_B] calculated from the hysteresis data obtained at liquid nitrogen temperature, by using the standard relation (Smit 1971) are given below.

$$n_B = \frac{\text{Molecular weight} \times \text{saturation magnetization}}{5585}, \quad (1)$$

Table 2. Saturation magnetization per formula unit in Bohr magneton at 78 K and α_{YK} for $Cd_xMg_{1-x}Gd_yFe_{2-y}O_4$ ferrites.

Composition x	y	Bohr magneton n_B	Yafet-Kittel angle (α_{YK}), °
0.2		1.99	0°
0.3	0.1	2.41	0°
0.4		2.75	0°
0.2		1.55	0°
0.3	0.2	2.05	0°
0.4		2.32	0°
0.2		1.36	0°
0.3	0.3	1.55	17°36'
0.4		1.96	23°15'
0.2		0.90	19°30'
0.3	0.4	1.26	26°5'
0.4		1.33	38°35'

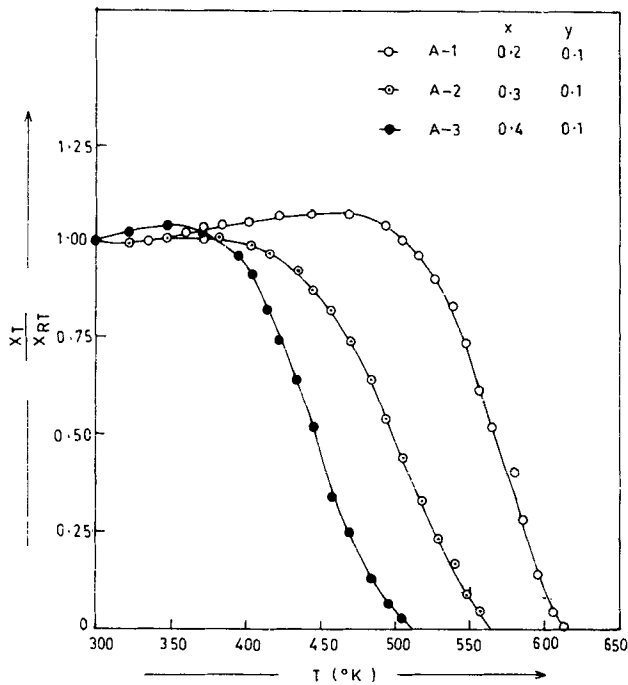


Figure 3. Variation of normalized ac susceptibility as a function of temperature for $Cd_xMg_{1-x}Gd_yFe_{2-y}O_4$ ferrites.

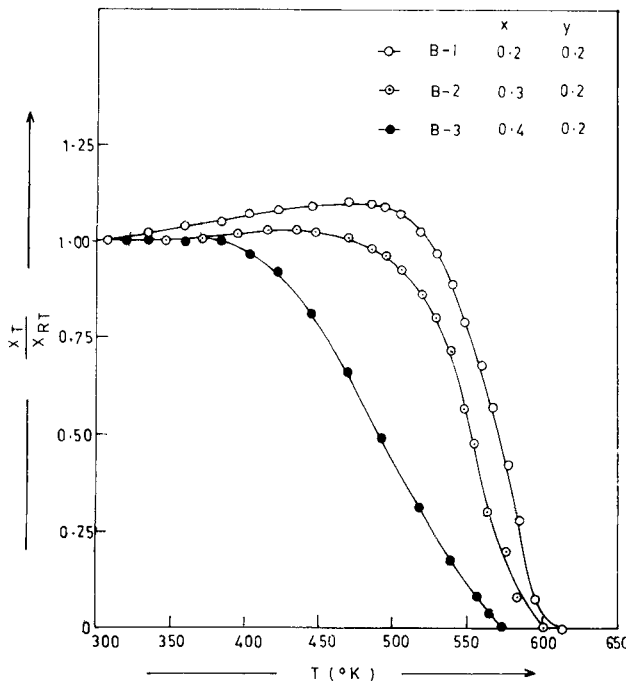
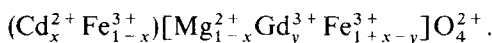


Figure 4. Variation of normalized ac susceptibility as a function of temperature for $Cd_xMg_{1-x}Gd_yFe_{2-y}O_4$ ferrites.

and the Y–K angles calculated using the relation

$$n_B = (5 + X) \cos \alpha_{YK} - 5(1 - X), \tag{2}$$

for each sample are given in table 2. It can be observed that the n_B value increases with the Cd^{2+} content while it decreases with Gd^{3+} content. The Cd^{2+} ions successively reduce the Fe^{3+} ions on the A site and since Gd^{3+} ions have a preference for B site, they proportionately reduce the Fe^{3+} ions on the B site. Hence the cation distribution for the present ferrites can be represented as



The magnetic moments of Cd^{2+} , Mg^{2+} and Gd^{3+} are zero, zero and 7.9 MB respectively. The net magnetic moment is due to the Fe^{3+} ions situated on A and B sites and the Gd^{3+} ions on the B sites. Since the addition of Gd^{3+} replaces Fe^{3+} ions on B site, the decrease in magnetization of the sublattice is due to the antiferromagnetic canting arrangement of the moments of these ions on the B site.

The observed variation of saturation magnetization with Cd content can be explained on the basis of cation distribution and Neel’s two sub lattice model. According to this model, of the three kinds of exchange interactions viz. A–A, B–B and A–B, A–B interaction is more effective and stronger than the other two. The substitution of Cd^{2+} on the A site successively reduces the Fe^{3+} ions on A site and equal number of Fe^{3+} ions are transferred to the B site. This slowly builds up the strength of A–B interaction and, hence, the magnetization of the sample increases with Cd^{2+} content.

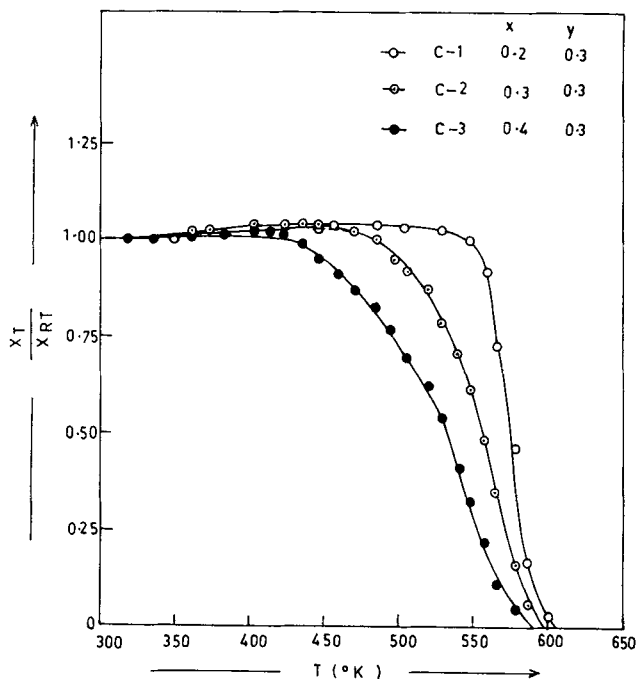


Figure 5. Variation of normalized ac susceptibility as a function of temperature for $\text{Cd}_x\text{Mg}_{1-x}\text{Gd}_y\text{Fe}_{2-y}\text{O}_4$ ferrites.

It can be seen from table 2 that Y-K angles, calculated from magnetization data at liquid nitrogen temperature, go on increasing with the addition of both Cd^{2+} and Gd^{3+} beyond $y > 0.2$. However, they do not exist for lower values of Gd.

It has been a general observation that Y-K angles do not exist for smaller concentrations of cadmium and zinc in case of many Cd and Zn containing pure binary ferrites (Leung *et al* 1973; Kulkarni and Panicker 1984; Panicker *et al* 1984). The fact has been confirmed both from hysteresis and Mössbauer studies. Upadhyay *et al* (1985) have also studied the $\text{Cd}_x\text{Mg}_{1-x}\text{Fe}_2\text{O}_4$ ferrite system and observed that Y-K angles exist beyond $x = 0.3$. Our results on Y-K angles suggest that there is canting of the moments on the B site when Gd content exceeds 0.2.

The plots of X_T/X_{RT} against temperature are shown in figures 3-6 which exhibit normal ferrimagnetic behaviour. The Curie temperatures determined from the plots (where $X_{ac} = 0$) are listed in table 1. It can be seen that there is a decrease in Curie temperature with the addition of both cadmium and gadolinium. This is attributed to the decreasing A-B interaction resulting from replacement of Fe^{3+} by Cd^{2+} on A site and Gd^{3+} on the B site. It can further be observed that the susceptibility remains almost constant with temperature which is characteristic of MD particles.

Bean (1955) suggested that the values of H_c and M_r/M_s are larger for SD particles and almost zero for SP particles. It has been shown further that X_{ac} does not vary much with temperature and drops off sharply near T_c (Kulkarni and Upadhyay 1986) for MD samples.

Taking into consideration the above, the observations which can be noted for our samples are (i) the temperature invariance of X_{ac} suggests that these compositions

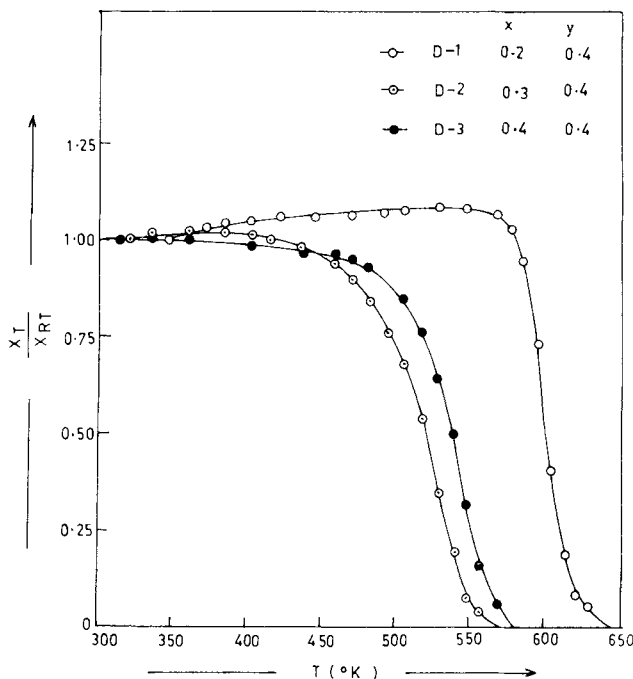


Figure 6. Variation of normalized ac susceptibility as a function of temperature for $\text{Cd}_x\text{Mg}_{1-x}\text{Gd}_y\text{Fe}_{2-y}\text{O}_4$ ferrites.

contain MD particles in predominance, (ii) the X_{ac} does not exhibit peaking behaviour and drops slowly to zero near T_c , (iii) the Curie temperature, where $X_{ac} = 0$, decreases with the addition of both Cd^{2+} and Gd^{3+} , (iv) double T_c behaviour is absent suggesting that there are no impurity phases present within the material and (v) the samples have low values of M_r/M_s .

Acknowledgements

The authors thank Prof. R N Patil for encouragement. One of the authors (JLB) wishes to thank UGC, New Delhi for the award of a Teacher Fellowship.

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** 1381
- Kolekar C B, Kamble P N and Vaingankar A S 1994 *J. Magn. & Magn. Mater.* **138** 211
- Kulkarni R G and Patil V U 1982 *J. Mater. Sci.* **17** 843
- Kulkarni R G and Panicker V G 1984 *J. Mater. Sci.* **19** 890
- Kulkarni R G and Baldha G J 1985 *Solid State Commun.* **53** 11
- Kulkarni R G and Upadhyay R V 1986 *Mater. Lett.* **4** 168
- Leung L K, Evans B J and Morrish A H 1973 *Phys. Rev.* **B8** 29
- Murthy C R K, Likhite S D and Sahasrabudhe P W 1984 *J. Phys. E: Sci. Instrum.* **18** 401
- Panicker V G, Upadhyay R V, Rao S N and Kulkarni R G 1984 *J. Mater. Sci. Lett.* **3** 385

- Radhakrishnamurthy C, Likhite S D and Sahasrabudhe P W 1965 *Rev. Sci. Instrum.* **36** 1558
- Smit J 1971 *Magnetic properties of materials* (McGraw Hill Book Co.) p. 89
- Srivastava C M, Shringi S N and Srivastava R G 1976 *Phys. Rev.* **B14** 2032
- Tsagaroyannis J, Haralambous K J, Loizos Z and Spyrellis N 1992 *Mater. Lett.* **14** 214
- Upadhyay R V, Rao S N and Kulkarni R G 1985 *Mater. Lett.* **3** 273