

Magnetic ordering in $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ system

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Abstract. Magnetization measurements of the ferrite system $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ were undertaken primarily to study variation of the saturation magnetization with manganese concentration and to gain information about the type of magnetic ordering. The observations indicate the existence of Yafet–Kittel (YK) type of magnetic ordering in this system which is well supported by results of Mössbauer effect measurements. A molecular field analysis of the YK spin ordering using the three-sublattice model is shown to explain the experimental data satisfactorily.

Keywords. Ferrites; magnetic ordering; collinear; non-collinear.

1. Introduction

This possibility of changing continuously the concentration of non-magnetic ions in the different sublattices makes the ferrites very interesting materials to study the appearance of various magnetic structures (Bhargava and Zeeman 1980; Brand *et al* 1985; Muralidharan *et al* 1985; Dormann *et al* 1987; Chakravarthy *et al* 1988; Dormann and Nogues 1990; Jotania *et al* 1992). Among these, systems with spinel structure seems to be particularly attractive, as they allow a variety of magnetic disorder and frustration to be introduced. This is due to the fact that, in spinel intra-sublattice interactions are weaker than the inter-sublattice interactions, as a result there are unsatisfied bonds in the ferrimagnetic phase. Because of these unsatisfied bonds increasing magnetic dilution accentuates the competition between the various exchange interactions resulting in a variety of magnetic structures (Bhargava and Zeeman 1980; Brand *et al* 1985; Muralidharan *et al* 1985; Dormann *et al* 1987; Chakravarthy *et al* 1988; Dormann and Nogues 1990; Jotania *et al* 1992).

A large number of investigators have studied Ti-substituted spinels in order to understand their magnetic properties. Brand *et al* (1985) found in the case of $\text{Mg}_{1+x}\text{Ti}_x\text{Fe}_{2-2x}\text{O}_4$ that on increasing dilution x , the collinear ferrimagnetic phase breaks down before reaching the ferrimagnetic percolation threshold and for higher concentration of Ti, system approaches toward spin-glass. Dormann *et al* (1987) have also studied the magnetization measurements of Ti^{4+} substituted lithium ferrite using different techniques and suggested the possibility of canted spin arrangements to explain the magnetization measurements.

In order to understand the influence of magnetic ion Mn^{4+} in place of non-magnetic ion Ti^{4+} , we have modified the magnetic phase of $\text{Mg}_{1+x}\text{Ti}_x\text{Fe}_{2-2x}\text{O}_4$ by replacing Ti^{4+} by Mn^{4+} , so that intra-sublattice interaction (J_{BB}) will become predominant over the inter-sublattice interaction (J_{AB}), which may lead to a frustration in the spin sub systems. In our recent communication (Jani *et al* 1997), we have reported magnetic properties on the mixed spinel $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$. The present paper reports the results of X-ray, magnetization and Mössbauer effect measurements on the $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ system. The variation of the saturation magnetization per formula unit in Bohr magneton number (n_{B}) with Mn-concentration in this system is similar in nature to that in the Ni–Zn (Satyamurthy *et al* 1969) and Cu–Zn (Kulkarni and Patil 1982) ferrites and the existence of Y–K angles on the B-site, observed in the latter systems is strongly suspected. A molecular field analysis of the Y–K spin ordering using a three-sublattice model is shown to explain the experimental data satisfactorily.

2. Experimental

Nine samples of Mn-substituted $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ system were prepared by the usual double sintering ceramic method for $0.1 \leq x \leq 0.9$ in steps of 0.1. The starting materials were analytical reagent grade oxides, Fe_2O_3 , MgO and MnO_2 . These oxides were mixed in proper proportions and presintered at 990°C for 24 h. In the final sintering process the material was held at 1050°C for 12 h and slowly cooled to room temperature (2°C/min). The X-ray diffractograms were obtained using FeK_α radiation on a Philips X-ray diffractometer model PW-1820. The diffraction patterns show the sharp lines corresponding to a single phase spinel for all the samples.

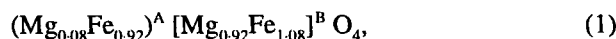
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The magnetization measurements of each sample at 300 K were carried out using high-field hysteresis loop technique (Radhakrishnamurthy *et al* 1971). The Mössbauer spectra were obtained at 12 K and 300 K in transmission geometry with source of 10 mCi $^{57}\text{Co}(\text{Pd})$ and a constant acceleration transducer interface to PC-based 1024 multi channel analyser.

3. Results and discussion

All the compositions of the system $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ exhibit single phase cubic spinel structure. No reflections other than those belonging to a spinel structure were observed in the patterns (figure 1). The values of lattice constant 'a' (Å) determined from X-ray data with an accuracy of ± 0.002 Å for $x=0.1$ to 0.9 are shown in figure 2 as a function of x . It is clear from figure 2 that the 'a' parameter initially increases up to $x=0.4$ and thereafter it levels off for further increase in $x \geq 0.5$. The observed nonlinear behaviour in 'a' with x may be attributed to the simultaneous replacement of Fe^{3+} (0.64 Å), Mg^{2+} (0.60 Å) and Mn^{4+} (0.60 Å). The cation distribution of MgFe_2O_4 ($x=0$) has been reported

to be



where A is the tetrahedral site and B the octahedral site. In order to determine the cation distribution, XRD intensities were calculated using the formula suggested by Buerger (1960):

$$I_{\text{hkl}} = |F_{\text{hkl}}|^2 PL_p, \quad (2)$$

where notations have their usual meanings. The distribution of divalent, trivalent and tetravalent cations amongst octahedral and tetrahedral sites in the $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ samples are determined from the ratios of intensities of X-ray diffraction lines: I_{220}/I_{440} , I_{220}/I_{400} and I_{400}/I_{422} . In table 1, the results of X-ray intensity calculations for typical samples $x=0.2, 0.6, 0.8$ and 0.9, are listed along with experimental intensity ratios. It is evident from table 1 that tetravalent Mn-ions occupy B-site replacing B-site iron and the added Mg^{2+} ions substitute for A-site iron without any change with x in the initial degree of inversion i.e. (1).

In deriving the cation distribution of $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ our X-ray intensity calculations (table 1) and relative Fe^{3+} intensities of Mössbauer spectra (table 2), for A and B site ions were taken into account. Accepting the cation distribution of MgFe_2O_4 (1), an approximate cation distribution for the $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ can be written as

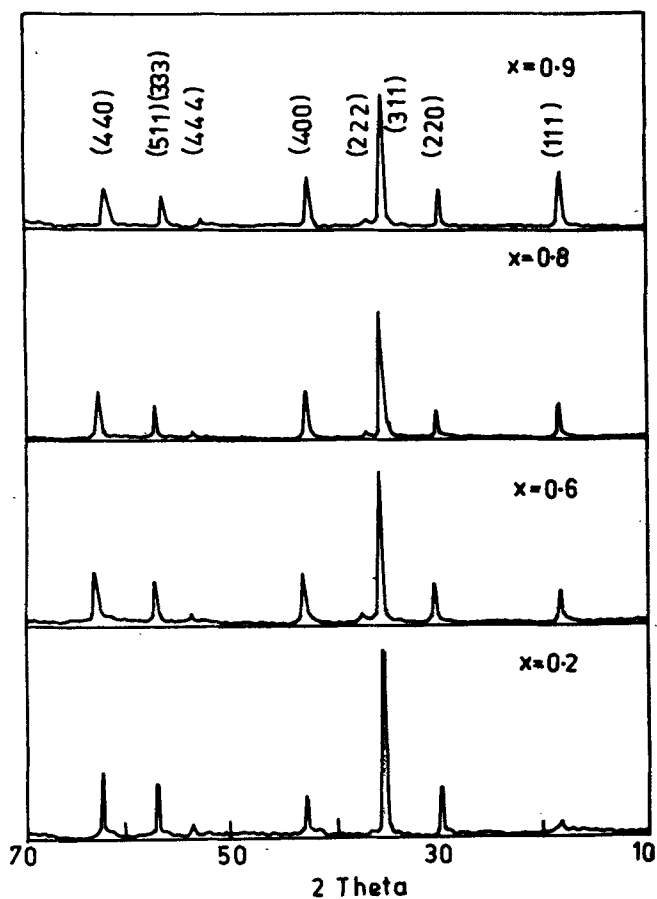


Figure 1. Typical X-ray diffractograms of the system $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ for $x=0.2, 0.6, 0.8$ and 0.9.

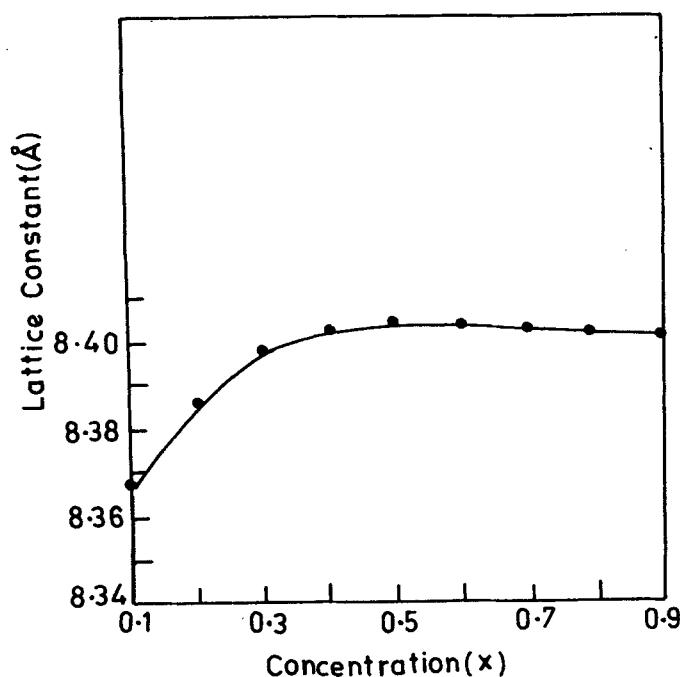


Figure 2. Variation of lattice parameter a (Å) with Mn concentration x .

Table 1. Comparison of X-ray intensities ratio for $Mg_{1+x}Mn_xFe_{2-2x}O_4$ in estimating cation distribution.

x	Lattice constant a (Å)	A-site cation	B-site cation	I_{220}/I_{440}		I_{220}/I_{400}		I_{400}/I_{422}	
				Obs.	Cal.	Obs.	Cal.	Obs.	Cal.
0.2	8.3853	$Mg_{0.28}Fe_{0.72}$	$Mg_{0.92}Mn_{0.20}Fe_{0.88}$	0.82	0.82	1.33	1.35	2.49	2.14
0.6	8.4320	$Mg_{0.66}Fe_{0.34}$	$Mg_{0.94}Mn_{0.60}Fe_{0.46}$	0.78	0.53	0.76	0.66	6.10	4.46
0.8	8.3984	$Mg_{0.82}Fe_{0.18}$	$Mg_{0.98}Mn_{0.80}Fe_{0.22}$	0.74	0.33	0.66	0.43	6.80	6.30
0.9	8.3965	$Mg_{0.91}Fe_{0.09}$	$Mg_{0.99}Mn_{0.90}Fe_{0.11}$	0.81	0.29	0.74	0.35	6.23	7.87

* a (Å) error ± 0.002 Å.**Table 2.** Mössbauer parameters of $Mg_{1+x}Mn_xFe_{2-2x}O_4$ obtained at 12 K. Average sublattice magnetization, $\mu(x)$ in μ_B .

x	Isomer shifts (mm/s)*		Hyperfine fields (KOE)		Mössbauer intensity ratio $I_B(Fe^{3+})/I_A(Fe^{3+})$	$\mu(x) = n_B^M(x)$ in μ_B
	IS_A	IS_B	H_A	H_B		
0.2	0.37 ± 0.05	0.37 ± 0.05	506 ± 3	523 ± 3	1.22	1.39
0.6	0.32 ± 0.05	0.32 ± 0.05	489 ± 3	510 ± 3	1.35	2.50
0.8	0.32 ± 0.05	0.32 ± 0.05	469 ± 3	495 ± 3	1.22	2.98
0.9	0.32 ± 0.05	0.32 ± 0.05	448 ± 3	480 ± 3	1.22	3.15

* IS w.r.t. α -Fe.

$$(Mg_{0.08+x}Fe_{0.92-x})^A [Mg_{0.92}Mn_xFe_{1.08-x}]^B O_4 \quad (3)$$

Equation (3) expresses that the added Mg^{2+} ions substituted for A-site Fe^{3+} and the Mn^{4+} ions for B-site Fe^{3+} without any change in the initial degree of inversion except $x = 0.6$. The experimental integrated intensity ratios of the A and B sites, ^{57}Fe spectra and those calculated on the basis of cation distribution obtained from X-ray intensity data given in table 1, are in very good agreement with each other (table 2) for $0.2 \leq x \leq 0.9$.

The saturation magnetization (σ_s) and the magnetization number (n_B) at 300 K obtained from magnetization data for $x = 0.1-0.9$ are summarized in table 3. From field dependence of magnetization and observed magnetic moments at 300 K (table 3), it is clear that the samples with $x = 0.0-0.6$ show ferrimagnetic behaviour in agreement with Mössbauer data at 300 K (figure 3) which decreases with increasing x values and samples with $x \geq 0.7$ display paramagnetic behaviour. Figure 4 shows the variation of n_B with x for $x = 0.0-0.9$ at 300 K, n_B increases with x up to $x \leq 0.3$ and thereafter it decreases.

According to Neel's (1950), two sublattice models of ferrimagnetism, the magnetic moment per formula unit in μ_B , n_B^N , is expressed as

$$n_B^N(x) = M_B(x) - M_A(x), \quad (4)$$

where M_B and M_A are B and A sublattice magnetic moments in μ_B . The $n_B^N(\mu_B)$ values for $x = 0.0-0.9$ were calculated using the ionic magnetic moments of Fe^{3+} , Mn^{4+} and Mg^{2+} with respective values $5\mu_B$, $3\mu_B$ and

Table 3. Saturation magnetization σ_s (emu/g) and saturation magnetization per formula unit in Bohr magnetons, n_B at 300 K. The values of the exchange constants in K used in calculating α_{YK} are given as footnote.

x	Magnetization (300 K)		Yafet-Kittel angles	
	σ_s (emu/gm)	n_B (μ_B)	α_{YK} (exp.)	α_{YK} (cal.)
0.0	23.2	0.80	0°	0°
0.1	29.2	1.03	0°	0°
0.2	43.2	1.50	0°	0°
0.3	45.3	1.55	14°36'	19°40'
0.4	39.5	1.32	31°27'	30°05'
0.5	38.4	1.26	40°09'	39°22'
0.6	28.5	0.92	53°07'	48°03'
0.7	22.2	0.70	63°11'	57°44'
0.8	11.4	0.36	83°39'	71°06'
0.9	—	—	—	—

 $J_a = -14.0; J_b = -13.0; J_d = -10.0; J_e = -4.5$ and $J_c = -7.5$.

$0\mu_B$ in (3) and the results are shown in figure 4 as a solid line. The calculated n_B^N values for $x = 0.0-0.3$ at 300 K are in good agreement with the experimentally found n_B values confirming the collinear spin ordering, while for $x = 0.4-0.9$ they clearly differ from the observed values (figure 4) indicating that significant canting exists on B-sites, suggesting magnetic structure to be non-collinear. Thus the change of spin ordering from collinear to non-collinear displays a strong influence on the variation of the saturation magnetic moment per molecule

as observed by magnetization (figure 4) with chemical composition.

In order to confirm the presence of a canted spin (non-collinear) structure indirectly, we have recorded Mössbauer spectra at 12 K for typical samples $x=0.2$, 0.6, 0.8 and 0.9 as shown in figure 5 in addition to Mössbauer spectra taken at 300 K (figure 3). The spectra exhibit the coexistence of magnetic as well as very small paramagnetic component. The full lines in figure 5 correspond to the least squares fitted spectra using two magnetic sextets and a central doublet. The magnetic components, which are well resolved at these temperatures, have been assigned to magnetically ordered Fe^{3+} ions at the tetrahedral (A) and octahedral (B) sites. The

hyperfine fields along with isomer shifts obtained from Mössbauer data analysis are listed in table 2 as a function of x for both A and B sites. Both hyperfine fields (H_A and H_B) decrease with increasing x demonstrating reduction in ferrimagnetic behaviour and magnetic coupling J_{AB} , with increasing x values in agreement with magnetization results (table 3). Isomer shifts $IS(A)$ and $IS(B)$ show very little change with Mn-content. The quadrupole shift is absent for all Mn-contents.

An indirect check for the presence of a canted spin (non-collinear) structure follows from the apparent proportionality between $|H_n|$ and the average sublattice magnetization.

It can be shown that

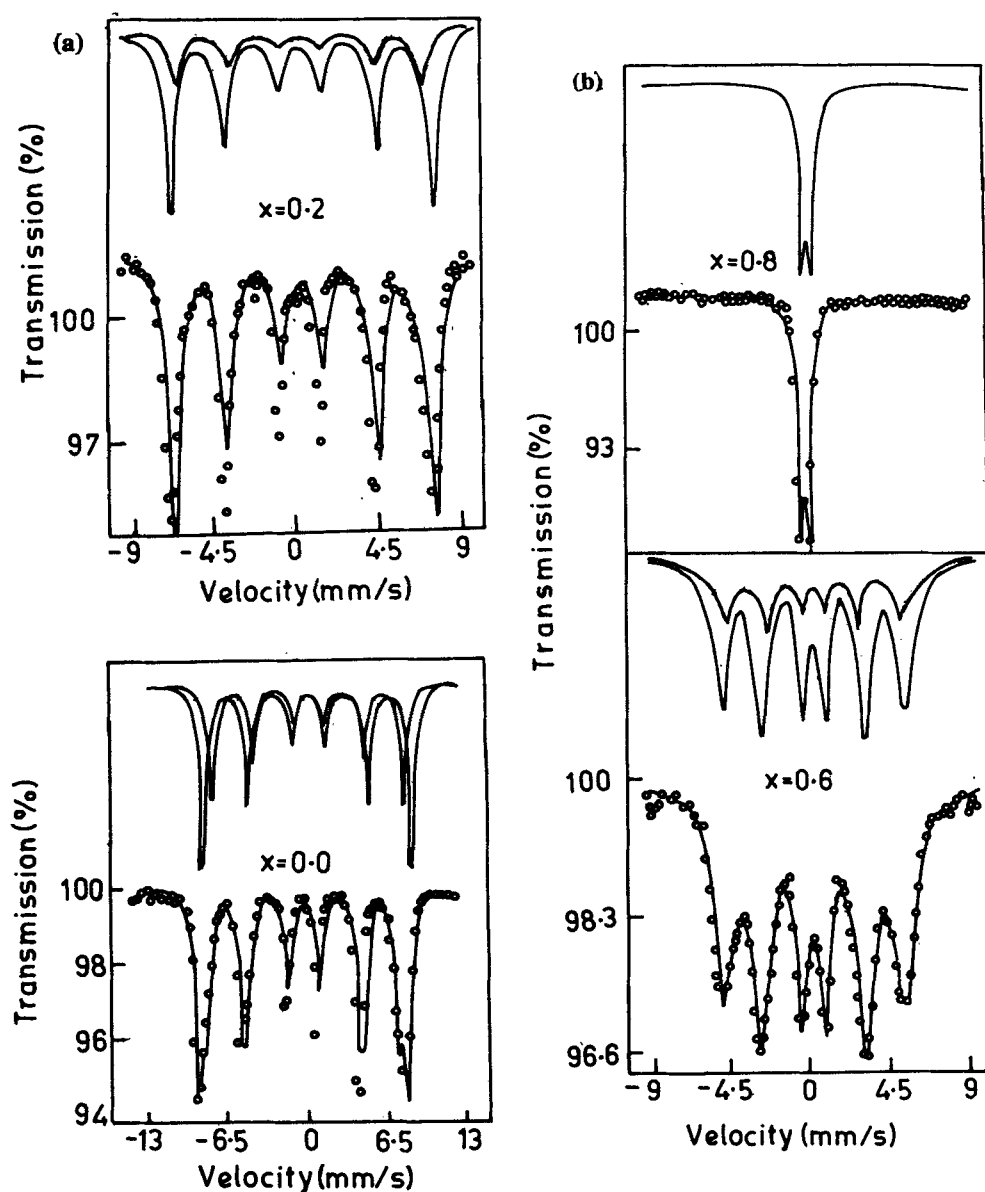


Figure 3. Typical Mössbauer spectra of $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ taken at 300 K for (a) $x=0.0$, 0.2 and (b) 0.6, 0.8.

$$\mu(x) = \frac{|H_{n_B}(x)|}{|H_{n_B}(0)|} M_B(x) - \frac{|H_{n_A}(x)|}{|H_{n_A}(0)|} M_A(x), \quad (5)$$

where $\mu(x)$ is the magnetic moment per formula unit (3), and $|H_{n_B}(x)|$ and $|H_{n_A}(x)|$ are magnitudes of the average nuclear magnetic fields for the B and A sites, respectively. It is assumed that the relative saturation magnetization at 300 K is equal to $\mu(x)/\mu(0)$ at 12 K. The values of magnetic moment per formula unit $n_B^M(x) \approx \mu(x)$ as a function of x were determined from (5) using the values of nuclear magnetic fields at 12 K from Mössbauer results (table 2) and the same are displayed in figure 4 and table 2. There is a reasonable agreement amongst the values of $n_B^M(x)$ obtained from Mössbauer data, $n_B(x)$ from magnetization measurements and $n_B^N(x)$ calculated from Neel's model as shown in figure 4 for $x=0.0$ to 0.2 confirming collinear magnetic structure. Mössbauer data, $n_B^M(x)$, and $n_B^N(x)$ for $x \geq 0.3$ clearly differ from magnetization data, $n_B(x)$, indicating

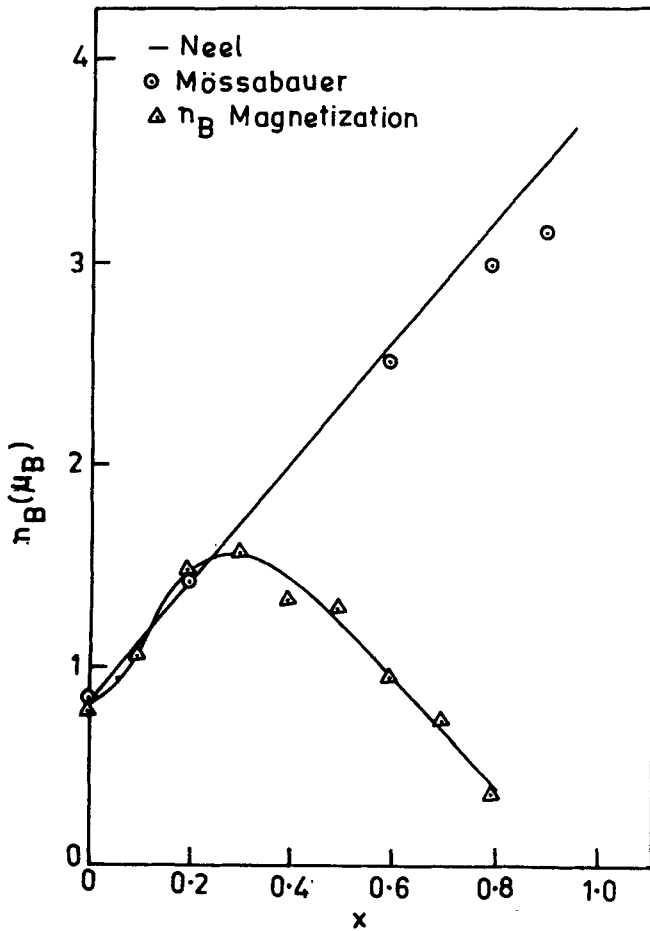


Figure 4. Variation of n_B (μ_B) with Mn concentration at 300 K for $Mg_{1+x}Mn_xFe_{2-2x}O_4$. The dots with circles represent the magnetic moment n_B^M obtained from Mössbauer data and the triangles represent data from magnetization measurements. The solid line corresponds to Neel's model.

that significant canting exists on B-site suggesting magnetic structure to be non-collinear.

Adopting the procedure of Kulkarni and Patil (1982), the condition for the existence of YK angle can be found for $x > 0.3$, and the molecular field acting on various ions in the present system are given by

$$\begin{bmatrix} H_A(\text{Fe}) \\ H_{B1}(\text{Mn}) \\ H_{B1}(\text{Fe}) \\ H_{B2}(\text{Mn}) \\ H_{B2}(\text{Fe}) \end{bmatrix} = \begin{bmatrix} \lambda_{AA} & \alpha & \beta & \alpha & \beta \\ \alpha & \gamma' & \epsilon' & \gamma & \epsilon \\ \beta & \epsilon' & \delta' & \epsilon & \delta \\ \alpha & \gamma & \epsilon & \gamma' & \epsilon' \\ \beta & \epsilon & \delta & \epsilon' & \delta' \end{bmatrix} \times \begin{bmatrix} (0.92-x)m_A(\text{Fe}) \\ 1/2(x)m_{B1}(\text{Mn}) \\ 1/2(1.08-x)m_{B1}(\text{Fe}) \\ 1/2(x)m_{B2}(\text{Mn}) \\ 1/2(1.08-x)m_{B2}(\text{Fe}) \end{bmatrix}, \quad (6)$$

where $H_A(\text{Fe})$ is the molecular field acting on the Fe^{3+} ion on the A-site due to all other ions, etc and

$$\begin{aligned} |m_A(\text{Fe})| &= |m_{B1}(\text{Fe})| = |m_{B2}(\text{Fe})| = 5\mu_B, \\ |m_{B1}(\text{Mn})| &= |m_{B2}(\text{Mn})| = 3\mu_B, \\ m_{B1} \cdot m_A &= -|m_{B1}| |m_A| \cos \alpha_{YK}, \\ m_{B2} \cdot m_A &= -|m_{B2}| |m_A| \cos \alpha_{YK}, \\ m_{B1} \cdot m_{B2} &= |m_{B1}| |m_{B2}| \cos 2\alpha_{YK}. \end{aligned}$$

The molecular field constants are related to the following sets of interactions

$$\begin{aligned} \lambda_{AA} &= A(\text{Fe}) - A(\text{Fe}), \\ \alpha &= A(\text{Fe}) - B_i(\text{Mn}), \\ \beta &= A(\text{Fe}) - B_i(\text{Fe}), \\ \gamma &= B_i(\text{Mn}) - B_2''(\text{Mn}), \\ \gamma' &= B_i(\text{Mn}) - B_i(\text{Mn}), \\ \delta &= B_1(\text{Fe}) - B_2(\text{Fe}), \\ \delta' &= B_i(\text{Fe}) - B_i(\text{Fe}), \\ \epsilon &= B_1(\text{Fe}) - B_2(\text{Mn}), \\ \epsilon' &= B_i(\text{Fe}) - B_i(\text{Mn}), \end{aligned}$$

where $i = 1, 2$.

The part of the interaction energy involving YK angles may be written as

$$\begin{aligned} E(\text{YK}) &= [15(0.92-x)\alpha\alpha + 25(0.92-x)(1.08-x)\beta] \\ &\quad \times \cos \alpha_{YK} - 1/4 [9x^2\gamma + 25(1.08-x)^2\delta + 30x \\ &\quad \times (1.08-x)\epsilon] \cos 2\alpha_{YK}. \end{aligned} \quad (7)$$

The energy is minimum for $\sin \alpha_{\text{YK}} = 0$, which corresponds to Neel's configuration or for

$$\cos \alpha_{\text{YK}} = \frac{15x(0.92 - x)\alpha + 25(0.92 - x)(1.08 - x)\beta}{9x^2\gamma + 25(1.08 - x)^2\delta + 30x(1.08 - x)\epsilon}, \quad (8)$$

which corresponds to the situation where the YK ordering is feasible. Here, for example the molecular field constant is related to the exchange integral J_α by

$$\alpha = \frac{2Z_{\text{AB}_i} J[\text{A}(\text{Fe}) - \text{B}_i(\text{Mn})]}{N_{\text{B}_i} g_{\text{A}} g_{\text{B}_i} \mu_{\text{B}}^2},$$

where Z_{AB_i} , the number of B_i nearest neighbour to A, N_{B_i} , the number of ions per formula unit, μ_{B} , the Bohr magneton and g the Lande's splitting factor.

On physical grounds, it is expected that J'_γ , J'_δ , J'_ϵ will be close to J_γ , J_δ and J_ϵ , respectively, but they are not necessarily equal.

The molecular field constants can be obtained from the observed variation of the saturation magnetization with Mn concentration. The exchange constants so obtained should be consistent with the existing data on similar ferrimagnetic systems. Kulkarni and Patil (1982) have found $J_\alpha = -5.25$ K for the $\text{Cu}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ system, in the case of $\text{Cu}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$, J_α is the interaction between $\text{A}(\text{Fe}^{3+})$ and $\text{B}_i(\text{Cu}^{2+})$, in the present case it corresponds

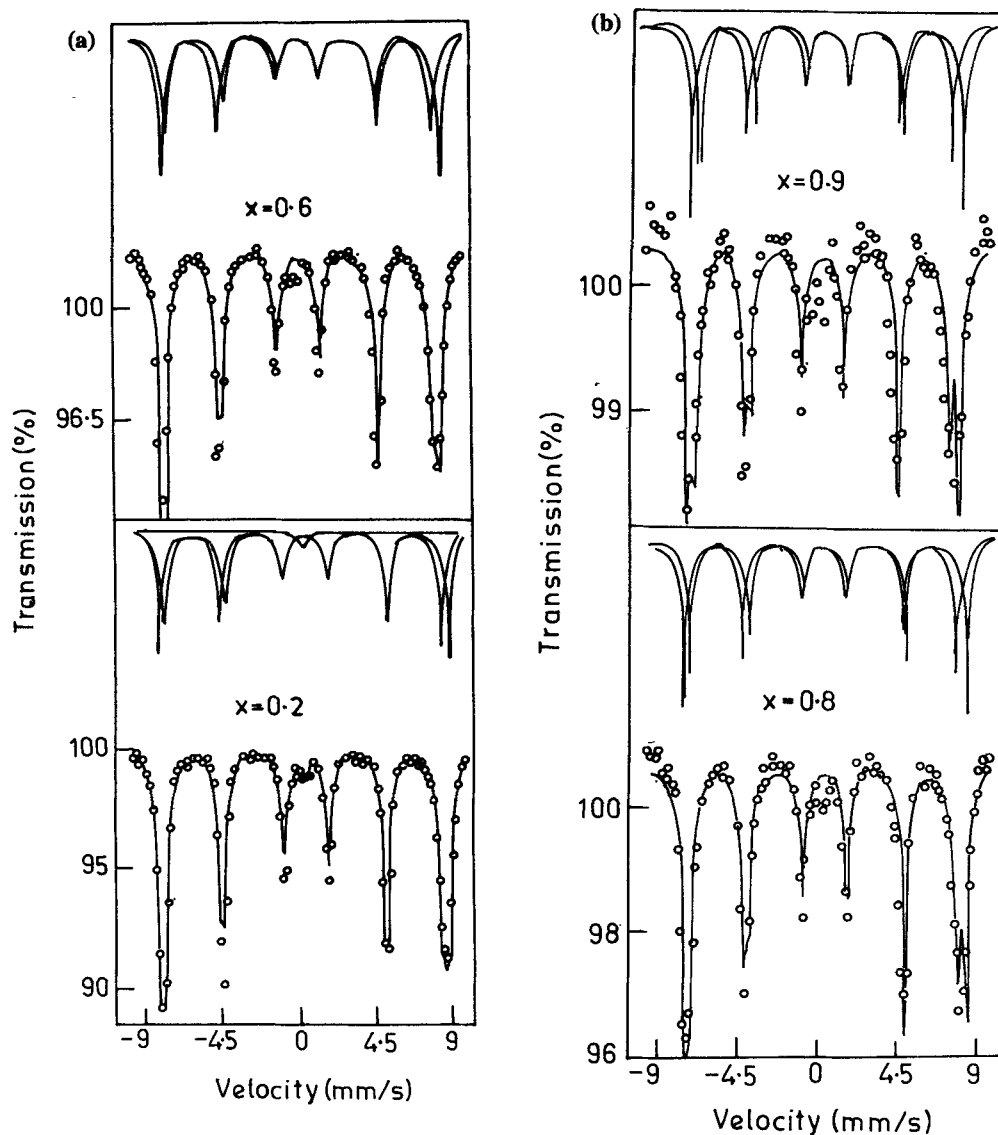


Figure 5. Typical Mössbauer spectra of $\text{Mg}_{1+x}\text{Mn}_x\text{Fe}_{2-2x}\text{O}_4$ taken at 12 K for (a) $x=0.2, 0.6$ and (b) $0.8, 0.9$.

to the interaction between A(Fe^{3+}) and B₁(Mn^{4+}). Assuming J_α is approximately equal to the product of the moments of the ions, one gets $J_\alpha(Fe^{3+} - Cu^{2+}) = -5$ K and $J_\alpha(Fe^{3+} - Mn^{4+}) = -15$ K. The value of $J_\alpha = -5$ K compares reasonably well with $J_\alpha = -5.25$ K value of Kulkarni and Patil (1982) for $Cu_{1-x}Zn_xFe_2O_4$. So $J_\alpha = -14$ K and $J_\beta = -13$ K values are accepted for $Mg_{1+x}Mn_xFe_{2-2x}O_4$. In a similar fashion the values of J_δ , J_γ and J_ϵ have been estimated as $J_\delta = -10$ K, $J_\gamma = -4.5$ K and $J_\epsilon = -7.5$ K. The values of the exchange constants used in calculating Yafet-Kittel angles, α_{YK} from (8) are listed in table 3 along with α_{YK} values.

The experimental values of α_{YK} have been obtained from the observed variation of n_B with x using the relation

$$n_B(x) = M_B(x) \cos \alpha_{YK} - M_A(x). \quad (9)$$

These values have also been given in table 3. There is a satisfactory agreement between the experimental and theoretical values of α_{YK} . Thus the observed variations of the saturation magnetization has been explained on the basis of the existence of Yafet-Kittel angles on the B-site spins. This suggests that A-B and B-B super-exchange interactions are comparable in strength.

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References

- Bhargava S C and Zeeman N 1980 *Phys. Rev.* **B21** 1717
 Brand R A, Georges-Gilbert H, Hubsch J L and Heller J A 1985 *J. Phys.* **F15** 1987
 Buerger M J 1960 *J. crystal structure analysis* (New York: John Wiley)
 Chakravarthy R, Madhav Rao L, Paranjpe S K, Kulshreshta S K, Sopen A K and Howells W S 1988 *J. Phys. (Paris) Colloq.* **49** C8-1111
 Dormann J L 1980 *J. Phys. (Paris) Colloq.* **41** 175
 Dormann J L and Reneudin P 1980 *Proc. int. conf. on ferrites*, Japan p. 156
 Dormann J L and Nogues M 1990 *J. Phys. Condense Matter* **2** 1223
 Dormann J L, Harfaoui M E I, Nogues M and Love J 1987 *J. Phys.* **C20** L161
 Goodenough J B 1963 *Magnetism of the chemical bond* (New York: John Wiley)
 Jani N N, Trivedi B S, Joshi H H and Kulkarni R G 1997 *Hyperfine interactions* (Netherlands: J. C. Baltzer AG, Science Publishers) **110** pp 227-237
 Jotania R B, Upadhyay R V and Kulkarni R G 1992 *IEEE Trans. magn.* **28** 1889
 Kulkarni R G and Patil V U 1982 *J. Mater. Sci.* **17** 843
 Muralidharan K, Srivastava J K, Moratha V R and Vijayaraghavan R 1985 *J. Phys.* **C18** 5897
 Neel L 1950 *C. R. Acad. Sci. Paris* **230** 375
 Radhakrishnamurthy C, Likhite S D and Sastry N P 1971 *Philos. Mag.* **23** 503
 Satyamurthy N S, Natera M G, Yousuf S I, Begum R J and Srivastava C M 1969 *Phys. Rev.* **181** 4412