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Review

# Magnetic properties of B and AB-spinels $Zn_{1-x}M_xFe_2O_4$ (M = Ni, Mg) materials

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ABSTRACT

The magnetic properties of diluted spinels $Zn_{1-x}M_xFe_2O_4$ (M = Mg, Ni) systems have been studied by mean field theory, probability law and high-temperature series expansions method in the range $0 \leq x \leq 1$. The exchange interactions are calculated by the first and second theory, respectively. The magnetic phase diagrams of $Zn_{1-x}M_xFe_2O_4$ (M = Mg, Ni) are calculated by the high-temperature series expansions, combined with the Padé approximants method. The critical exponent associated with the magnetic susceptibility ($\gamma$) is deduced.



**Contents**



## 1. Introduction

The $AB_2O_4$ type spinel ferrite shows various magnetic properties such as ferromagnetic and antiferromagnetic behaviours depending on the composition and cation distribution. For instance, bulk zinc ferrite $ZnFe_2O_4$ is known to possess a normal spinel structure and is an antiferromagnetic material [1], where the $Zn^{2+}$ ion always prefers to occupy A-site and all the $Fe^{3+}$ ions occupying B-sites are antiparallelly coupled. In contrast, $NiFe_2O_4$ is an inverse spinel that $Ni^{2+}$ ion always prefers to occupy B-site and two $Fe^{3+}$ ions occupy A-site and B-site separately [2]. Due to the super-exchange interactions between the spins of inter-sublattices (A–O–A and B–O–B) and intra-sublattices (A–O–B) via oxygen anions, two $Fe^{3+}$ ions are aligned oppositely on A- and B-sites while the $Ni^{2+}$ are parallel to $Fe^{3+}$ in B-sites, showing the long-range ferromagnetic order. More-

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over, ferromagnetic $MgFe_2O_4$ is a mixed inverse spinel that the $Mg^{2+}$ and $Fe^{3+}$ ions can easily occupy both of the A- and B-site.

The case of a ferrimagnetic spinels system $MFe_2O_4$ (M = Mg, Ni) with two magnetic sublattices A and B is particularly interesting because they may exhibit particular disordered magnetic ions in different sites. The magnetic structures depend upon the types of magnetic ions residing on the tetrahedral (A) and octahedral (B) sites and the relative strengths of the inter- ($J_{AB}$) and intra-sublattice interactions ($J_{AA}$, $J_{BB}$). Generally all the three exchange interactions $J_{AA}$, $J_{BB}$ and $J_{AB}$ are negative. Further, when all the metal ions (cations) are magnetic, usually the inter-sublattice interaction $J_{AB}$ is the strongest with $|J_{AB}| \gg |J_{BB}| > |J_{AA}|$. Thus, $J_{AB}$ renders the undiluted spinels as ferrimagnetic with A-site moments aligned antiparallel to the B-site moments keeping the AA and the BB bonds unsatisfied. The determination of critical exponents is an important aspect of the theoretical description and experimental characterisation of magnetic materials [3]. The exchange interactions $J_1(x)$ and $J_2(x)$ of the diluted $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ systems have been obtained by using the mean field theory in the pure case. The probability distribution function adapting the nature of dilution problem is applied for the two systems to calculate the exchange interaction $J_{AB}(x)$ in the range $0 \le x \le 1$. The Padé approximant (P.A.) [4] analysis of the high-temperature series expansion (HTSE) of the correlation length has been shown to be a useful method for the study of the critical region [5,6]. We have used this technique to determine the magnetic phase diagrams of $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ in the range $0 \le x \le 1$.

The series expansions of the susceptibility $\chi(T)$ have been derived to the seventh-order in the reciprocal temperature for spinels lattices including both nearest-neighbouring (nm) and next-nearest-neighbouring (nnn) interactions in the Heisenberg model [7]. In the antiferromagnetic behaviour we have used the results given in Ref [8]. We have applied this method to the spinel systems $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ to estimate the critical temperature and the critical exponent $\gamma$ associated with the magnetic susceptibility $\chi(T)$.

## 2. Theory method

### 2.1. Calculation of the exchange integrals

#### 2.1.1. Mean field approximation

Starting from the well-known Heisenberg model, the Hamiltonian of the system is given by:

$$H = -2\sum_{i,j} J_{ij}\vec{S}_i\vec{S}_j \tag{1}$$

where, $J_{ij}$ is the exchange integral between the spins situated at sites $i$ and $j$. $\vec{S}_i$ is the atomic spin of the magnetic ion located on the $i$th site. The factor "2" in Eq. (1) arises from the fact that, when summing over all possible pairs $\langle ij\rangle$ exchange interactions, we count each pair twice. In this work, we consider the nearest-neighbouring (nn) and next-nearest-neighbouring (nnn) interactions $J_1$ and $J_2$, respectively. In the case of spinels structure containing the magnetic moment only in the octahedral sublattice, the mean field approximation leads to a simple relations between the critical temperature $T_N$ and the paramagnetic Curie Weiss-temperature $\theta_p$, respectively, and the considered two exchange integrals $J_1$ and $J_2$.

Following, the method of Holland and Brown [9], the expressions of $T_N$ and $\theta_p$ describing the $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ systems are:

$$T_N = \frac{2S(S+1)}{3k_B}[-4J_1 + 2J_2] \tag{2}$$

$$\theta_p = \frac{2S(S+1)}{3k_B}[12J_1 + 6J_2] \tag{3}$$

where $k_B$ is the Boltzmann's constant and $S = 3/2$ is the spin of $Fe^{3+}$ ions.

To determine $J_1(x)$ and $J_2(x)$ in the whole range concentration for the $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ systems, we have used the experimental values of $T_N$ and $\theta_p$ obtained by magnetic measurement [10–13]. We have deduced the values of exchange integrals $J_1(x)$ and $J_2(x)$ for the $Zn_{1-x}Mg_xFe_2O_4$ and for the $Zn_{1-x}Ni_xFe_2O_4$ systems in the pure case.

#### 2.1.2. Probability law

The authors [14,15] in the last work, used the probability law to calculate the exchange interactions. In this work, we have applied the probability law in the diluted spinels $Zn_{1-x}M_xFe_2O_4$ (M = Mg, Ni) systems, only the random placement of the ions A and B leads to the spatial fluctuations of the signs and magnitudes of the super-exchange interaction between the magnetic ions A and B. Due to the nature of dilution problem we choose a probability law permitting us to determine exchange integral $J_{AB}(x)$ for each concentration $x$. The exchange integral of the opposite pure compound $AB_2X_4$ of the bound random spinel is denoted $J_{AB}$. The occupation probability $p(i)$ of the two ions A or B induced in the interaction is $p(i) = C_n^i x^{n-i}(1-x)^i$, where $n$ is the total number of lattice sites inside a sphere with the volume $4/3\pi R_i^3$ ($R_i$ denotes the distance between the sites $i$ and $j$, $n$ is the number of cations the $P$th coordination sphere around a given cation chosen as the central one, for this structure $n = 3$), while $i$ varies from 0 to 3. The exchange integral for such an occupation is assumed to be: $J_{AB}^i = \left(J_A^{n-i}J_B^i\right)^{1/n}$. The expression obtained is:

$$\begin{aligned} J_{AB}(x) &= \sum_{i=0}^{3} C_3^i x^{3-i}(1-x)^i \left(J_A^{3-i}J_B^i\right)^{1/3} \\ J_{Y-Fe}(x) &= x^3 J_{Y-Fe} + 3x^2(1-x)\left(J_{Y-Fe}{}^2 J_1\right)^{(1/3)} \\ &\quad + 3x(1-x)^2\left(J_{Y-Fe}J_1{}^2\right)^{(1/3)} + (1-x)^3 J_1 \end{aligned} \tag{4}$$

Y = Ni or Mg, ou $J_1 = J_{Fe-Fe} = -1.166$ K, $J_2 = -0.333$ K, in $ZnFe_2O_4$ obtained by Eqs. (2) and (3). $J_{AB}$ correspond to the exchange interactions of the opposite pure systems $AB_2X_4$.

$Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ are a diluted spinels $MgFe_2O_4$ and $NiFe_2O_4$ systems, respectively, with the values of the exchange integrals: $J_{Mg-Fe} = -22$ K for $MgFe_2O_4$ [16] and $J_{Ni-Fe} = -30$ K for $NiFe_2O_4$ [17]. The exchange integrals $J_{Fe-Fe}$, $J_{Mg-Mg}$ and $J_{Ni-Ni}$ are negligible in the $MgFe_2O_4$ and $NiFe_2O_4$ systems.

### 2.2. High-temperature series expansions

In order to deduce the expression of the susceptibility of the system with two sublattices, the Hamiltonian of the Heisenberg with extern field $h_{ex}$ may be put in the form:

$$\begin{aligned} H = &-2J_{AA}\sum_{\langle i,i'\rangle}\vec{S}_i\vec{S}_{i'} - 2J_{BB}\sum_{\langle j,j'\rangle}\vec{\sigma}_j\vec{\sigma}_{j'} \\ &- 2J_{AB}\sum_{\langle i,j\rangle}\vec{S}_i\vec{\sigma}_j - \mu_B h_{ex}\left(g_A\sum_i S_i^z - g_B\sum_j \sigma_j^z\right) \end{aligned} \tag{5}$$

where $\vec{S}$ and $\vec{\sigma}$ are spin vectors of magnitudes $\vec{S}^2 = S(S+1)$ and $\vec{\sigma}^2 = \sigma(\sigma+1)$ in sublattice A and B, respectively. $g_A$ and $g_B$ are the corresponding gyromagnetic factors and $\mu_B$ is the Bohr magneton. $h_{ex}$ is an external magnetic field ($z$ direction) introduced in order to provide an easy determination of the magnetic susceptibility. The first summation is over all spin pairs nearest-neighbours in sublattice

A, the second is over all spin pairs nearest-neighbours in sublattice B and the third is between all spin pair nearest-neighbours in A and B. $J_{AA}$, $J_{BB}$ and $J_{AB}$ are the intra- and the inter-sublattice exchange interactions between neighbouring spins.The magnetisation of the ferrimagnetic spinels systems is:

$$M = \mu_B \left( g_A \sum_i \left\langle S_i^z \right\rangle + g_B \sum_j \left\langle \sigma_j^z \right\rangle \right) \quad (6)$$

After computing the first derivative of magnetisation $\chi = (\partial M/\partial h_{ex})_{h_{ex}\to 0}$, we obtained the general expression of susceptibility for the collinear normal ferrimagnetic spinel as follows:

$$\chi = \frac{\mu_B^2}{3k_BT} \left( N_A g_A^2 \bar{S}^2 + N_B g_B^2 \bar{\sigma}^2 + g_A^2 \sum_{i \neq i'} \left\langle \vec{S}_i \vec{S}_{i'} \right\rangle + g_B^2 \sum_{j \neq j'} \left\langle \vec{\sigma}_j \vec{\sigma}_{j'} \right\rangle - 2 g_A g_B \sum_{i,j} \left\langle \vec{S}_i \vec{\sigma}_j \right\rangle \right) \quad (7)$$

where $N_A$ and $N_B$ are, respectively, the number of ion and the spin value of each type of spin.Finally, we obtain simple form

$$\chi = \frac{\mu_B^2}{3k_BT} \left( N_A g_A^2 \bar{S}^2 + N_B g_B^2 \bar{\sigma}^2 + N_A g_A^2 \gamma_{AA} + N_B g_B^2 \gamma_{BB} - 2 N_B g_A g_B \gamma_{BA} \right) \quad (8)$$

Following the procedure in [18–22], we compute the expressions of spin correlation functions $\gamma_{AA}$, $\gamma_{BB}$ and $\gamma_{AB}$ in terms of powers of $\beta$ and mixed powers of $J_1 = 2J_{BB}\bar{\sigma}^2$, $J_2 = 2J_{AB}\bar{S}\bar{\sigma}$ and $J_3 = 2J_{AA}\bar{S}^2$.The correlations functions $\gamma_{AA}$, $\gamma_{BB}$ and $\gamma_{AB}$ may be expressed as follows:

$$\begin{aligned} \gamma_{AA} &= \bar{S}^2 \sum_{q=1}^{7} \sum_{m=0}^{q} \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} a(m,n,p,q) J_1^m J_2^n J_3^p \beta^q \\ \gamma_{BB} &= \bar{\sigma}^2 \sum_{q=1}^{7} \sum_{m=0}^{q} \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} b(m,n,p,q) J_1^m J_2^n J_3^p \beta^q \\ \gamma_{BA} &= \bar{S}\bar{\sigma} \sum_{q=1}^{7} \sum_{m=0}^{q} \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} c(m,n,p,q) J_1^m J_2^n J_3^p \beta^q \end{aligned} \quad (9)$$

Nonzero coefficients $a(m,n,p,q)$, $b(m,n,p,q)$ and $c(m,n,p,q)$ up to order 7 on $\beta$ are given in Ref [7]. We use the powerful P.A. method [4], to estimate the critical temperature. In this method, the Curie temperature is determined by locating the singularities in the P.A. method to the HTSE of the magnetic susceptibility. The excellent reviews of these methods are available in Refs [23,24].

The simplest assumption that one can make concerning the nature of the singularity of the magnetic susceptibility $\chi(T)$ is that the neighbourhood of the critical temperature the above the following functions exhibit the asymptotic behaviour:

$$\chi(T) \propto (T - T_C)^{-\gamma} \quad (10)$$

The usual approach is to compute the series for the logarithmic derivative of $\chi(T)$,

$$\frac{d}{dT} \log[\chi(T)] \approx \frac{-\gamma}{T - T_C} \quad (11)$$

**Table 1**
The exchange integrals $J_{AB}$ of $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ as a function of dilution $x$.

| $x$ | $Zn_{1-x}Ni_xFe_2O_4$ | $Zn_{1-x}Mg_xFe_2O_4$ | |
|---|---|---|---|
| | $\left\lvert \frac{J_{AB}}{k_B}(K) \right\rvert$ [17] | $\left\lvert \frac{J_{AB}}{k_B}(K) \right\rvert$ (present work) | $\left\lvert \frac{J_{AB}}{k_B}(K) \right\rvert$ (present work) |
| 1 | 30.00 | 30 | 22 |
| 0.9 | – | 24.43 | 18.13 |
| 0.8 | – | 19.60 | 14.74 |
| 0.7 | – | 15.45 | 11.80 |
| 0.6 | – | 11.93 | 9.29 |
| 0.5 | – | 8.99 | 7.16 |
| 0.4 | – | 6.58 | 5.38 |
| 0.3 | – | 4.64 | 3.92 |
| 0.2 | – | 3.13 | 2.75 |
| 0.1 | – | 1.99 | 1.85 |
| 0 | 1.166 | 1.166 | 1.166 |

as this function has a simple pole $T_C$ and should be well represented by Padé approximants $[M, N]$. The exponent $\gamma$ is then re-estimated from the approximates to:

$$(T - T_C) \frac{d}{dT} \log[\chi(T)] \quad (12)$$

evaluated at $T = T_{FerriM}(T_N)$.

A P.A. $[M, N]$ to a magnetic susceptibility $\chi(T)$ is a rational fraction $P_M/Q_N$, with $P_M$ and $Q_N$, polynomials of order $M$ and $N$ in $\beta = 1/k_BT$, such that: $\chi(\beta) \approx P_M/Q_N + O(\beta^{M+N+1})$, $M$ and $N$ are the degree of the $P$ and $Q$ polynomials, respectively. Estimates of $T_{FerriM}(T_N)$ and $\gamma$ for $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ have been obtained using the P.A. method [4]. The simple pole corresponds to $T_{FerriM}(T_N)$ and the residues to the critical exponents $\gamma$. The obtained central value of $\gamma$ is: $\gamma = 1.36 \pm 0.02$ and $\gamma = 1.38 \pm 0.02$.

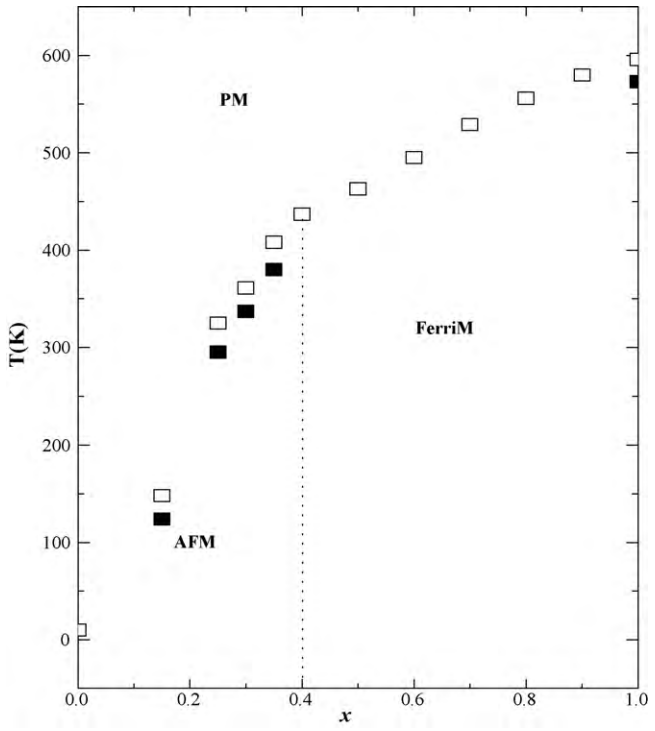


**Fig. 1.** The magnetic phase diagram of the $Zn_{1-x}Ni_xFe_2O_4$ systems. The open squares and the solid squares represent the results given by: HTSE method and magnetic measurements [11–13], respectively.

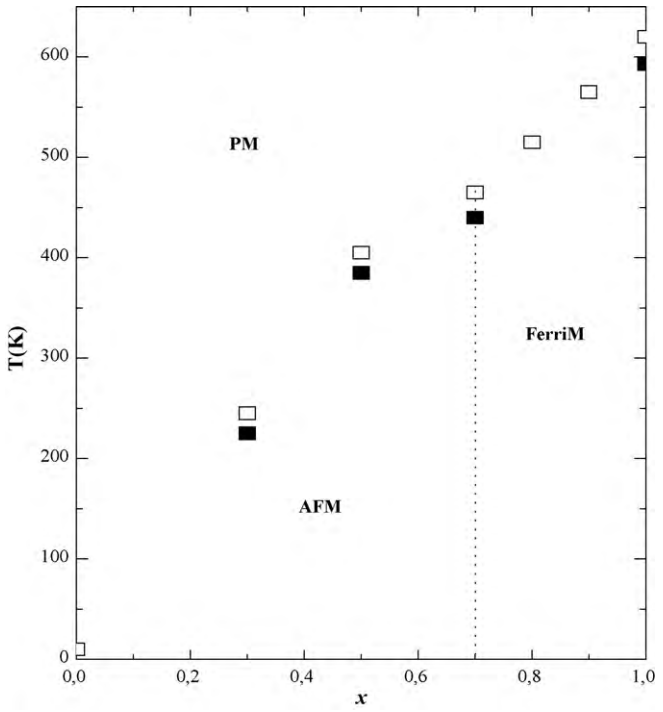


**Fig. 2.** The magnetic phase diagram of the $Zn_{1-x}Mg_xFe_2O_4$ systems. The open squares and the solid squares represent the results given by: HTSE method and magnetic measurements [11–13], respectively.

## 3. Discussions and conclusions

In present work, we have used the mean field theory to calculated the exchange interactions $J_1(x)$ and $J_2(x)$ of the $ZnFe_2O_4$ spinel. The probability distribution function adapting to the nature of dilution problem has been applied for the two $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ systems to calculate the exchange interaction $J_{AB}(x)$ in the range $0 \leq x \leq 1$ (see Table 1). From Table 1, on can see that the exchange integral decreases in absolute value when $x$ decreases in the two systems. In these spinels oxides the predominant exchange interaction is super-exchange between cations through the intervening oxygen (Zn–O–Fe, Zn–O–Zn or Fe–O–Fe). The main factors govern the strength of this antiferromagnetic interaction. In the $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ materials, the exchange integral $J_{AB}$ is the largest because the angle Zn–O–Fe is closest to 180° [11].

By applying the HTSE method to the magnetic susceptibility $\chi(T)$, we have estimated the critical temperature $T_{FerriM}(T_N)$ for each dilution $x$ in the $Zn_{1-x}Mg_xFe_2O_4$ and in $Zn_{1-x}Ni_xFe_2O_4$ systems. Several thermodynamic phases may appear for the $Zn_{1-x}Ni_xFe_2O_4$ including the ferrimagnetic (FerriM) ($0.4 < x \leq 1$) and antiferromagnetic (AFM) ($0 \leq x < 0.4$) and paramagnetic (PM) phases (see Fig. 1). In $Zn_{1-x}Mg_xFe_2O_4$ we find the several magnetic phase including the ferrimagnetic (FerriM) ($0.7 < x \leq 1$) and antiferromagnetic (AFM) ($0 \leq x < 0.7$), paramagnetic (PM) phases (see Fig. 2). In the two figures (Figs. 1 and 2), we have included, for comparison, the theoretical results obtained with the results obtained by magnetic measurement [11–13]. From these figures one can see good agreement between the theoretical phase diagram and experimental results. In the other hand, the sequence of [$M$, $N$] P.A. to series of $\chi(T)$ has been evaluated. The value of critical exponent $\gamma$ associated with the magnetic susceptibility $\chi(T)$, have been estimated. By examining the behaviour of these P.A., the convergence was found to be quite rapid. This procedure was repeated for series up to orders 6 and 5. Between the orders 6 and 7, the analysis of the series is not affected significantly. The approximants are well converged and estimates are accurate to high precision 1%. When the number of terms decreases from 6 to 5, the analysis of the series shows that estimates of increase by 2%. For short series $n \leq 4$ the accuracy in the calculation is not expected to be high. Estimates of the critical exponents associated with magnetic susceptibility for the $Zn_{1-x}Mg_xFe_2O_4$ and $Zn_{1-x}Ni_xFe_2O_4$ systems are found to be $\gamma = 1.36 \pm 0.02$ and $\gamma = 1.38 \pm 0.02$.

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