# The magnetic properties of diluted $CoFe_2O_4$ nanomaterials

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The magnetic properties of $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ are studied using mean-field theory and the probability distribution law to obtain the saturation magnetization, the coercive field, the critical temperature, and the exchange interactions with different values of $D$ (nm) and $x$. High-temperature series expansions (HTSEs) combined with the Padé approximant are used to calculate the critical temperature of $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$, and the critical exponent associated with magnetic susceptibility is obtained.

**Keywords:** $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$, probability law, saturation magnetization, coercive field, high-temperature series expansions

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## 1. Introduction

Ferrites are a group of oxides that have a general chemical formula $MFe_2O_4$, where $M$ is a divalent metal ion such as Zn, Cd, Cu, Ni, Co, Mg or Fe.[1] Mixed ferrites can be produced by mixing two or more kinds of $M$ ions.[2] $Zn_{0:5}Ni_{0:5}Fe_2O_4$ is an example of a mixed ferrite and it is one of the compounds studied in the present work. Ferrites exhibit a variety of useful and interesting properties. In this section we discuss briefly the synthesis, structure, magnetic properties and applications of ferrites. A brief account of the motivation of the present work is also given. The A site ions have four neighbouring oxygen ions forming a tetrahedron, while the B site ions are surrounded by six oxygen ions forming an octahedron.[3] We therefore discuss tetrahedral (A) and octahedral (B) sites. Ferrites can be represented by the chemical formula $AB_2O_4$, where A and B denote metal cations on the tetrahedral (A) and octahedral (B) sites, respectively. The spinel structure can be 'normal' or 'inverse', depending on the distribution of cations on the interstitial sites. In a normal spinel, all the tetrahedral (A) sites are occupied by divalent ions, while the octahedral (B) sites are occupied by trivalent ions. Cadmium ($CdFe_2O_4$) and zinc ($ZnFe_2O_4$) ferrites are examples of normal spinels,[4–6] where all the $Cd^{2+}$ and $Zn^{2+}$ ions are distributed on the A sites and the $Fe^{3+}$ ions occupy the B sites. In an inverse spinel the trivalent ions are distributed equally between the A and B sites, and all the divalent ions occupy B sites. Copper ($CuFe_2O_4$) and nickel ($NiFe_2O_4$) ferrites are examples of inverse spinels.[7,8] Half of the $Fe^{3+}$ ions occupy A sites and the other half occupy B sites with $Cu^{2+}$ or $Ni^{2+}$ cations. Since the magnetic moments of the ions on A sites are antiparallel to those on B sites in ferrites, the magnetization of an inverse spinel is due to divalent $M$ ions.[2] Most ferrites are partly normal or partly inverse.[2] The degree of inversion appears to have a remarkable effect on the magnetic properties. This is influenced by the sample preparation method and sintering temperature.[2,3] Cations also have site preferences for A or B sites, depending on their sizes and electronic configurations. Those with smaller sizes prefer to occupy the smaller tetrahedral (A) sites, while larger cations prefer roomier octahedral (B) sites.[2] $Zn^{2+}$ and $Cd^{2+}$ ions have been observed to show a preference for A sites, where their 4s, p and 5s, p electrons, respectively, form covalent bonds with six 2p electrons of oxygen ions.[9] In general, these are based on their favourable magnetic, electrical, mechanical or chemical properties. Sometimes two or more of the above properties are critical for certain applications. Ni–Zn substituted mixed

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ferrites are soft ferrimagnetic materials with low coercivity, high resistivity values, and small eddy current losses.[10,11] These properties make them excellent core materials for power transformers in electrical and telecommunication applications. They are also used in high frequency devices for radio frequency applications.[12,13] Manganese ferrites are widely used in electronic applications such as transformers, choke coils, noise filters, and recording media.[14] They are also used as catalyst and humidity sensors.[15] Multilayer chip inductors can also be made from ferrites. These are useful as surface mounting devices for the miniaturization of electronic devices such as cellular phones and video cameras.[16] The use of small and compact power supplies for computers, microprocessors and video cassette recorders has also led to an increased demand for ferrites with high switching frequencies and better material performance.[17] Recently, there has been a renewed interest in the study of nanosized ferrites. The reasons for this are potential applications in nanoscience and technology.[18] This has been driven by the quest for new products and the fundamental understanding of material properties when the material size approaches atomic scale. Cobalt ferrite ($CoFe_2O_4$) has high coercivity and moderate magnetization.[19] These properties, along with its physical stability, make $CoFe_2O_4$ nanomaterials applicable for magnetic recording applications such as audio, videotape, and high density recording disks.[20] Magnetic nanomaterials are also relevant to modern technologies such as the contrast enhancement of magnetic resonance imaging and high-density data storage, and nanomaterials can be used as magnetic carriers for the delivery of medicine to specific parts of the body.[21,22] The drug capsules can be thermally opened at a predetermined site within the body. The heating effect of nanomagnetic materials in response to alternating magnetic fields may also be useful in destroying tumor cells.[23]

In the present work, the expression of the critical temperature and the exchange interactions are given. According to Néel's two sublattice model of ferrimagnetism, the magnetic moment per formula unit in $\mu_B$, $M(x)$ is expressed as $M(x,D) = M_B(x,D) - M_A(x,D)$. The obtained values are presented in Table 1, where $M_B$ and $M_A$ are the B- and the A-sublattice magnetic moments in $\mu_B$, respectively. We present in Figs. 1 and 2 the variations in coercive field and saturation magnetization versus the size ($D$) of $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ nanomaterials, respectively. The theoretical saturation magnetization versus $x$ is given in Fig. 3, and the values of exchange interactions $J_{AB}$, $J_{BB}$, and $J_{AA}$ are given in Table 2 by using a probability distribution law for diluted $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ nanomaterials.

The Padé approximant[24] analysis of the high-temperature series expansions (HTSEs) of magnetic susceptibility has been shown to be a useful method of studying the critical region.[25,26] We have used this technique to determine the critical temperature $T_C$ and the critical exponent $\gamma$ associated with the magnetic susceptibility $\chi(T)$. The series expansions of the susceptibility $\chi(T)$ have been derived to the seventh-order in the reciprocal temperature for spinel lattices including both nearest-neighbouring (nn) and next-nearest-neighbouring (nnn) interactions in the Heisenberg model.[27] We have used this method to estimate the values of critical exponent $\gamma$ associated with the magnetic susceptibility for the $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ nanomaterials.

**Table 1.** The magnetic parameters of nanomaterials $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ with different values of $D$ (nm).

| Ferrite compositions | $D$/nm | $T_C$ (exp)[38] | $T_C$ (MFT) | $T_C$ (HTSE) | $H_C$[38] | $M_S$ (theor.) |
|---|---|---|---|---|---|---|
| $Co_{0.8}Zn_{0.2}Fe_2O_4$ | 37 | 300 | 320 | 314 | 961 | 37.69 |
| $Co_{0.4}Zn_{0.6}Fe_2O_4$ | 35 | 290 | 310 | 298 | 779 | 18.68 |
| $Co_{0.2}Zn_{0.8}Fe_2O_4$ | 39 | 175 | 190 | 185 | 688 | 9.3 |

**Table 2.** The exchange interactions of nanomaterials $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ with different values of $D$ (nm).

| Cation distribution | $-J_{AB}$(x) | $-J_{BB}$(x) | $-J_{AA}$(x) |
|---|---|---|---|
| $(Fe)_A(ZnFe)_BO_4$ | 6.1 | 12.2 | 6.1 |
| $(Co_{0.2}Fe_{0.8})_A(Zn_{0.2}Fe_{1.2})_BO_4$ | 7.85 | 17.56 | 4.4 |
| $(Co_{0.4}Fe_{0.6})_A(Zn_{0.6}Fe_{1.4})_BO_4$ | 8.12 | 23.92 | 4.2 |
| $(Co_{0.6}Fe_{0.4})_A(Zn_{0.4}Fe_{1.6})_BO_4$ | 6.9 | 31.23 | 5.47 |
| $(Co_{0.8}Fe_{0.2})_A(Zn_{0.2}Fe_{1.8})_BO_4$ | 4.19 | 39.52 | 8.24 |
| $(Co)_A(Fe_2)_BO_4$ | 0 | 48.8 | 12.5 |

# 2. Theory

## 2.1. Calculation of the exchange integrals by mean field theory

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

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

where $J_{ij}$ is the exchange integral between the spins situated at sites $i$ and $j$, and $\boldsymbol{S}_i$ is the spin operator localized at site $i$. The expression of the critical temperature for diluted $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ nanomaterials is given by

$$\begin{aligned} T_C(\mathrm{K}) = {} & \frac{2}{3}\frac{1}{2}(72)^{0.5}\big[S_{Fe^{3+}}S_{Co^{2+}}(1-x)xJ(Fe^{3+}\text{–}Co^{2+}) \\ & + S^{3+}_{Fe}S^{3+}_{Fe}(1-x)(1+x)J(Fe^{3+}\text{–}Fe^{3+})\big] \\ & + (72)^{0.5}\big[(1+x)(1+x)S^{3+}_{Fe}S^{3+}_{Fe} \\ & \times J(Fe^{3+}\text{–}Fe^{3+})\big]. \end{aligned} \tag{2}$$

(i) ***Probability law***

In recent work, the authors[28] used the probability law to calculate the exchange integrals. In this work, we have applied the probability law in the diluted spinel systems $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$. Only random placements of the A and B ions lead 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 which allows us to determine exchange integral $J_{AB}(x)$ for each concentration $x$. The exchange integral of the opposite pure compound $AB_2X_4$ ($J_{AA}$ or $J_{BB}$) of the bound random spinel is denoted by $J_{AB}$. 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 $\frac{4}{3}\pi R_i^3$ ($R_i$ denotes the distance between sites $i$ and $j$, $n$ the number of cations, and the P-th refers to the coordination sphere around a given cation chosen as the central one). The exchange integral for such an occupation is assumed to be $J^i_{AB} = (J^{n-i}_A J^i_B)^{1/n}$. The expressions obtained are

$$\begin{aligned} J_{AA(x)} = {} & \frac{1}{4}\big[2(1-x)(1-x)J_{AA}(Fe^{3+}\text{–}Fe^{3+}) \\ & + 2x^2 J_{AA}(Co^{2+}\text{–}Fe^{3+})\big], \\ J_{AB}(x) = {} & \frac{1}{2}\big[(1-x)(1+x)J_{AB}(Fe\text{–}Fe) \\ & + x(1-x)J_{AB}(Co^{2+}\text{–}Fe^{3+})\big], \\ J_{BB}(x) = {} & \big[(1+x)^2 J_{BB}(Fe^{3+}\text{–}Fe^{3+})\big], \\ & S_{Co^{2+}} = 1, \quad S_{Fe^{3+}} = 5/2, \\ & J(Fe^{3+}\text{–}Co^{2+}) = -25.0\ \mathrm{K}, \\ & J(Fe^{3+}\text{–}Fe^{3+}) = -12.2\ \mathrm{K}, \\ & J(Co^{2+}\text{–}Co^{2+}) = -18.9\ \mathrm{K}.[29] \end{aligned}$$

(ii) ***Scaling law***

The results found in Ref. [30] comply with the analytic expression for $T_C$ being made on the basis of a mean-field $T_C$ examination, given as

$$T_C(D) = T_C(\infty)[Z_0 + 2Z_1\cos(\pi/D+1)]/(Z_0+2Z_1),$$

where $Z_0 = 4$ and $Z_1 = 4$ are the number of nearest neighbours to a lattice point in the same layer and the number of nearest neighbours in one of the adjacent layers for a cubic lattice (spinel). $D$ is the size of the $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ nanomaterial. The obtained values are given in Table 1.

## 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 external field $h_{ex}$ may be put in the form

$$\begin{aligned} H = {} & -2J_{AA}\sum_{\langle i,i'\rangle}\boldsymbol{S}_i\boldsymbol{S}_{i'} - 2J_{BB}\sum_{\langle j,j'\rangle}\boldsymbol{\sigma}_j\boldsymbol{\sigma}_{j'} \\ & - 2J_{AB}\sum_{\langle i,j\rangle}\boldsymbol{S}_i\boldsymbol{\sigma}_j \\ & - \mu_B h_{ex}\Big(g_A\sum_i S_i^z - g_B\sum_j \sigma_j^z\Big), \end{aligned} \tag{3}$$

where $\boldsymbol{S}$ and $\boldsymbol{\sigma}$ are the spin operators of ions in sublattices A and B, respectively; $g_A$ and $g_B$ are the corresponding gyromagnetic factors; the symbol $\langle\cdots\rangle$ denotes the summation over nearest neighbours; and $J_{AA}$, $J_{BB}$, and $J_{AB}$ are the intra- and inter-sublattice exchange interactions in the ferrimagnetic spinels.

The susceptibility of the collinear normal ferrimagnetic spinel is as follows:

$$\begin{aligned} \chi = {} & \frac{\mu_B^2}{3k_BT}(N_A g_A^2\boldsymbol{S}^2 + N_B g_B^2\boldsymbol{\sigma}^2 - g_A^2\sum_{i\neq i\backslash}\langle\boldsymbol{S}_i\boldsymbol{S}_{i\backslash}\rangle \\ & - g_B^2\sum_{j\neq j\backslash}\langle\boldsymbol{\sigma}_j\boldsymbol{\sigma}_{j\backslash}\rangle - 2g_Ag_B\sum_{i,j}\langle\boldsymbol{S}_i\boldsymbol{\sigma}_j\rangle), \end{aligned} \tag{4}$$

where ($N_A$, $N_B$) and ($S = 1$, $\sigma = \frac{5}{2}$) are the numbers of the ion and the spin value of each type of spin, respectively.

Finally, we obtain a simple form as

$$\begin{aligned} \chi = {} & \frac{\mu_B^2}{3k_BT}(N_A g_A^2\bar{S}^2 + N_B g_B^2\bar{\sigma}^2 - N_A g_A^2\gamma_{AA} \\ & - N_B g_B^2\gamma_{BB} - 2N_B g_A g_B\gamma_{BA}). \end{aligned} \tag{5}$$

Following the procedure in Refs. [31]–[37], we compute the expressions of the spin correlation functions $\gamma_{AA}$, $\gamma_{BB}$, and $\gamma_{AB}$ in terms of powers of $\beta = 1/(k_BT)$ ($k_B$ is the Boltzmann constant) and mixed powers of

$J_1 = 2J_{\rm BB}\bar{\sigma}^2$, $J_2 = 2J_{\rm AB}\,\bar{S}\bar{\sigma}$, and $J_3 = 2J_{\rm AA}\bar{S}^2$, with

$$\bar{S} = \sqrt{S(S+1)} \quad \text{and} \quad \bar{\sigma} = \sqrt{\sigma(\sigma+1)}. \tag{6}$$

The correlation functions $\gamma_{\rm AA}$, $\gamma_{\rm BB}$, and $\gamma_{\rm AB}$ may be expressed as follows:

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

where 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. [27].

The simplest assumption that one can make concerning the nature of the singularity of the magnetic susceptibility $\chi(T)$ is that at the neighbourhood of the critical point, the above function exhibits the asymptotic behaviour:

$$\chi(T) \propto (T - T_{\rm C})^{-\gamma}. \tag{8}$$

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

$$\frac{\rm d}{{\rm d}T} \log[\chi(T)] \approx \frac{-\gamma}{T - T_{\rm C}}, \tag{9}$$

as this function has a simple pole $T_{\rm C}$ and should be well represented by Padé approximants $[M,\, N]$. The exponent $\gamma$ is then re-estimated from the approximant

$$\gamma = \lim_{T \longrightarrow T_{\rm C}} (T - T_{\rm C}) \frac{\rm d}{{\rm d}T} \log[\chi(T)] \tag{10}$$

evaluated at $T = T_{\rm C}$.

Estimates of $T_{\rm C}$ and $\gamma$ for $(\mathrm{Co}_x\mathrm{Fe}_{1-x})_{\rm A}(\mathrm{Zn}_{1-x}\mathrm{Fe}_{1+x})_{\rm B}\mathrm{O}_4$ have been obtained using the Padé approximant (see Table 1). The simple pole corresponds to $T_{\rm C}$ and the residues to the critical exponent $\gamma$. The obtained central value of $\gamma$ in the ordered region is $\gamma = 1.39 \pm 0.01$.

## 3. Discussion

In this work, critical temperature values are obtained by mean-field theory, and the exchange interactions $J_{\rm AA}$(x), $J_{\rm AB}(x)$, and $J_{\rm BB}(x)$ are also obtained. The saturation magnetization and the coercive field are obtained with different $D$ (nm) nanomaterials of $(\mathrm{Co}_x\mathrm{Fe}_{1-x})_{\rm A}(\mathrm{Zn}_{1-x}\mathrm{Fe}_{1+x})_{\rm B}\mathrm{O}_4$. The expression of $H_{\rm C}$ is given by the formula $H_{\rm c} = a + b/D$, where $a$ and $b$ are constants. In Fig. 1, we have plotted the coercive field $H_{\rm C}$ versus $1/D$. The constants $a$ and $b$ are found to be $-4.390 \pm 0.919$ and $197.499 \pm 34.906$ Oe (1 Oe=79.5775 A/m), respectively. According to Neel's two-sublattice model of ferrimagnetism, the magnetic moment per formula unit in $\mu_{\rm B}$, $M(x)$ is expressed as: $M(x) = M_{\rm B}(x) - M_{\rm A}(x)$. Figures 2 and 3 show the variation in saturation magnetization $M_{\rm S}$ with size $(D)$ of $(\mathrm{Co}_x\mathrm{Fe}_{1-x})_{\rm A}(\mathrm{Zn}_{1-x}\mathrm{Fe}_{1+x})_{\rm B}\mathrm{O}_4$ nanoparticle and dilution $x$, respectively.

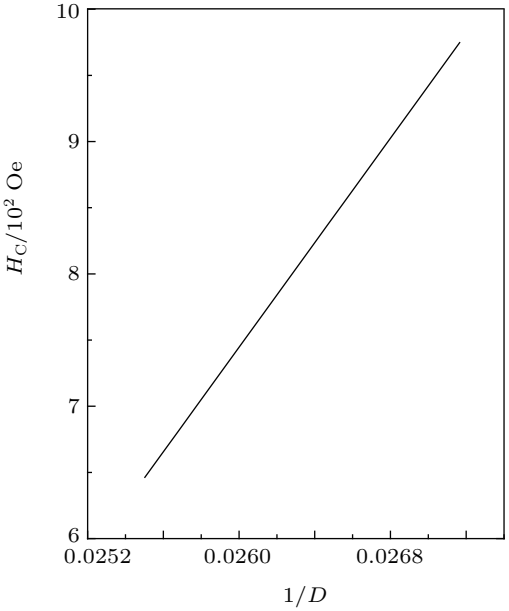



**Fig. 1.** The coercive field $H_{\rm C}$ versus $x$.

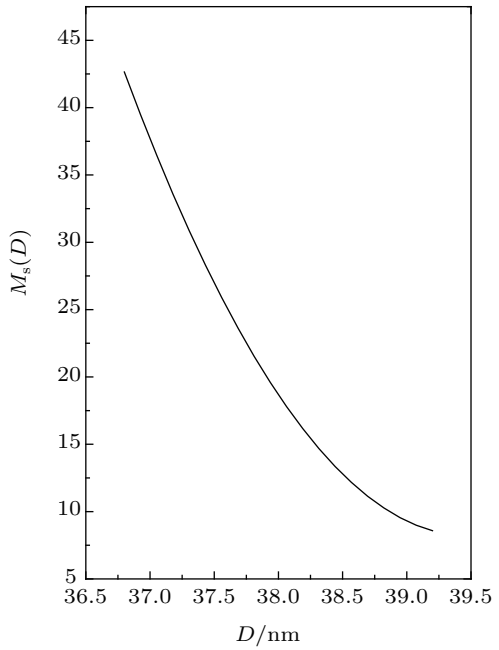



**Fig. 2.** The saturation magnetization $M_{\rm s}$ versus $D$.

The HTSE extrapolated with Padé approximant turns out to be a convenient method for providing a valid estimation of the critical temperature of the real system. By applying this method to the magnetic susceptibility $\chi(T)$, we estimate the critical temperature $T_{\rm C}$ for each dilution $x$ in the

$(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ systems (see Table 1). Several thermodynamic phases may appear, including the ferrimagnetic phase (FEM) ($0 \le x \le 1$) and paramagnetic (PM) ($0 \le x \le 1$). On the other hand, the value of critical exponent $\gamma$ associated with the magnetic susceptibility $\chi(T)$ is estimated. The sequence of $[M, N]$ Padé approximant to the series of $\chi(T)$ is evaluated. By examining the behaviour of these Padé approximants, the convergence is found to be quite rapid. The estimate of the critical exponents associated with magnetic susceptibility for the $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ systems indicates $\gamma = 1.39 \pm 0.01$. On the other hand, the value of critical exponent $\gamma$ associated with the magnetic susceptibility $\chi(T)$ is estimated for several $[M, N]$ Padé approximants. The convergence is extremely good and from the elements near to and on the diagonal of the Padé approximant $[M, N]$. We estimate the central value of the critical exponent to be $\gamma = 1.39 \pm 0.01$.

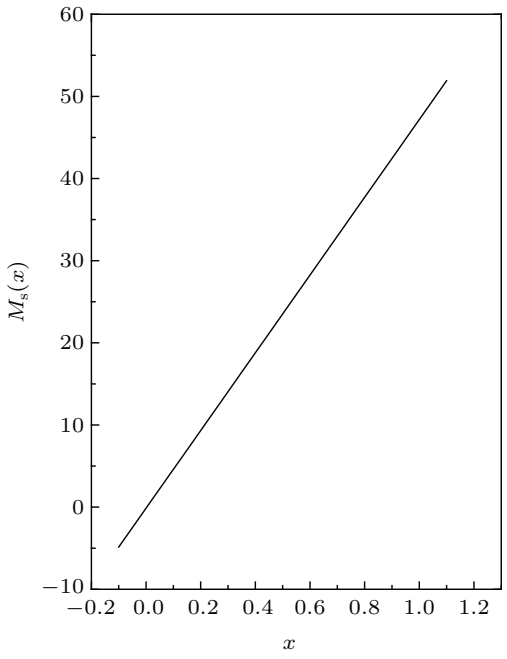



**Fig. 3.** The saturation magnetization $M_s$ versus $x$.

# 4. Conclusions

The intersublattice A–B superexchange, the intrasublattice B–B, and the intrasublattice A–A superexchange interactions $J_{AA}(x)$, $J_{AB}(x)$, and $J_{BB}(x)$, respectively, in $(Co_xFe_{1-x})_A(Zn_{1-x}Fe_{1+x})_BO_4$ are obtained using the probability distribution law (see Table 2). The exchange $J_{BB}$ is strong compared with $J_{AB}$ and $J_{AA}$ for all values of $x$. The values of critical temperature are obtained by mean-field theory (see Table 1). The variations in coercive field $H_C$ versus $1/D$ are given for different values of $x$. $H_C$ decreases with increasing $D$. The values of saturation magnetization $M_s$ are given for different values of $x$ and $D$. $M_s$ increases with increasing $x$, but decreases with increasing $D$.

HTSEs combined with the Padé approximant are obtained for different values of $x$ and $D$. The critical temperature values obtained by HTSE and those obtained by mean-field theory are compared with each other (see Table 1). The critical exponent $\gamma$ values associated with magnetic susceptibility $\chi(T)$ are estimated and comparable to those given by the three-dimensional Heisenberg model.

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