AN ANALYSIS OF THE SPIN STRUCTURE OF Zn—Ni FERRITES IN VIEW OF THE EXPERIMENTAL DATA

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The results of neutron diffraction, Mössbauer Effect (ME) measurements as well as the data on magnetization and Curie point of $Zn_xNi_{1-x}Fe_2O_4$ are in terms of the Localized Canting Model (LCM). It is shown that LCM can account for the dependence of magnetization on Zn content with almost constant ratios for the exchange parameters J_{ij} over the range of x; $0 \leqslant x \leqslant 0.9$. It is found that the dependence of the average canting angle on the external magnetic field, observed in ME, could be explained by LCM when the J_{ij} are assumed to decrease with increasing x.

1. Introduction

The spin structure of Zn–Ni ferrites has been the subject of a considerable attention in the recent years. In 1962, Ishikawa [1] starting from the concept of superparamagnetic clusters expressed in the terms of the molecular field approximation has explained the character of the dependence of low temperature magnetization $\mu(x)$ vs zinc content. In this approach, the value of $\mu(x)$ for $x > 0.5$ appeared to depend crucially on the size distribution of the clusters whereas the spins inside the clusters and the matrix have been assumed to form the collinear ferrimagnetic array. In 1969 however, Satya Murthy et al. [2] have reported the results of a neutron diffraction study of a powdered $Zn_xNi_{1-x}Fe_2O_4$ system from which they infered the existence of a Yafet–Kittel (YK) type of spin structure. The presence of a long range magnetic order with canted spins in the B sublattice was manifested by the weak (200) reflection in the diffraction pattern taken below the so-called Y–K transition temperature, T_{YK}.

Further confirmation of spin canting in the powdered Zn–Ni system was deduced from ME measurements on ^{57}Fe nuclei by applying a magnetic field, H_{ext}, paralle lto the gamma ray beam [3–6]. The authors cited above have observed the non-zero intensities of $\Delta m = 0$ transitions (lineas 2 and 5) for compositions with $x > 0.5$ and have found that these intensities rise with increasing Zn content. Since $\Delta m = 0$, the intensity does

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not vanish ionic spins of Fe^{3+} are not parallel (or antiparallel) to the gamma-ray beam, therefore the presence of 2 and 5 lines in the spectra has been interpreted as direct evidence and a measure of canting angles between B site spins. The interpretations proposed in the papers [3–6] differ generally in that Daniels et al. [3] and Leung et al. [4] regard their data as a confirmation of Y–K structure, whereas Petitt [5] and Clark et al. [6] relate the non-zero $\Delta m = 0$ intensities to the local spin configurations. In all ME studies mentioned above with the exception of [6], the conclusion about spin canting was derrived on the basis of measurements performed with a unique value of the external field applied to the given sample. In [6], two magnitudes of H_{ext} have been used and it was noticed that the canting angle decreases with increasing field for all compositions x in the range $0.6 \leqslant x \leqslant 0.95$. In our previous paper [7], we reported our preliminary ME measurements in $Zn_xNi_{1-x}Fe_2O_4 (x = 0.7)$ in the presence of various external fields ranging between 5 and 60 kOe. The changes of the average angle between the B site of Fe^{3+} ionic moments and H_{ext} were interpreted in terms the so called Localized Canting Model (LMC), [8], extended by us to the case of a three sublattice system in the presence of an external field. However, the numerical values of the exchange parameters given in [7] are not valid due io an error in the computer program.

In the present paper we analyze the applicability of LCM in a quantitative description of various magnetic properties of the $Zn_xNi_{1-x}Fe_2O_4$ system. The various kinds of information accessible from ME and neutron diffraction measurements are also discussed.

2. *Experimental basis*

The following magnetic properties observed experimentally heve been taken into account in our analysis of the spin structure of the Zn–Ni system:

1. The dependence of low temperature saturation magnetization $\mu(x)$ and Curie point $T_c(x)$ on the zinc content x as given in literature by Guillaud et al. [9], Fig. 1 and Leung et al. [4], Fig. 2 — respectively.

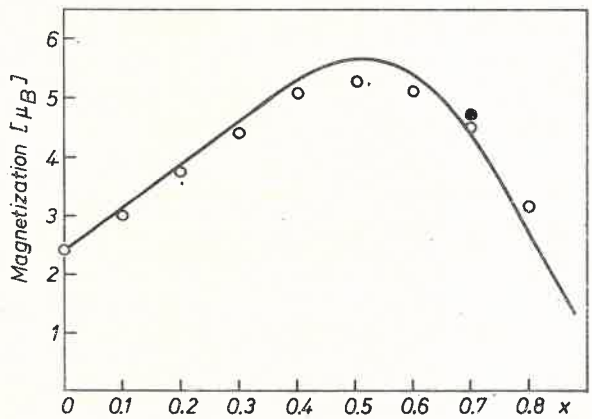

Fig. 1. Experimental and calculated curves from LCM of saturation magnetization of $Zn_xNi_{1-x}Fe_2O_4$ at 4.2 K vs x values; ○ experimental from Guillaud [9], ——— LCM(a) for: $\delta_1 = 0.155$, $\delta_2 = \delta_3 = \delta_4 = 0$, ● LCM(b) for: $\delta_1 = 0.15$, $\delta_2 = 0.75$

2. The features of neutron diffraction and ME patterns observed by us for powdered $Zn_xNi_{1-x}Fe_2O_4$ ($x = 0.7$). Below, we shall comment in more detail on some aspects of the latter type of data.

In Fig. 3, two neutron diffraction patterns are shown: the lower, obtained by us for $x = 0.7$ at 78 K and the upper reproduced for comparison with [2] as obtained for $x = 0.75$ at 300 K. It is seen that contrary to [2] as in our case, there is no (200) reflection which could manifest the existence of a long range Y–K type of spin canting sublattice.

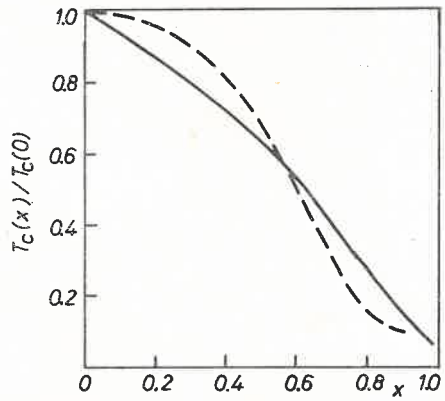

Fig. 2. Experimental and calculated curves from LCM of the relative Curie point $T_c(x)/T_c(0)$ of $Zn_xNi_{1-x}Fe_2O_4$ vs x values; ——— experimental from Leung [4], — — — LCM(a) for $\delta_1 = 0.155$, $\delta_2 = \delta_3 = \delta_4 = \delta_5 = 0$

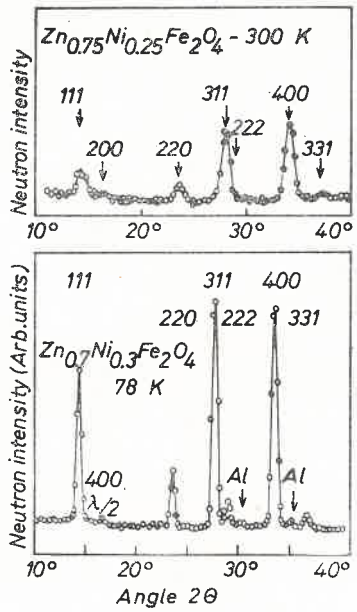

Fig. 3. Neutron diffraction patterns: the lower one was obtained in this work for $Zn_xNi_{1-x}Fe_2O_4$ ($x = 0.7$) at 78 K, the upper reproduced from Satya, Murthy et al. [2] for $x = 0.75$ at 300 K

According to our estimation, the trace of reflections observed at $2\theta \simeq 16°$ originates from the contamination of the second order reflection from the (400) plane. This means that the eventual canting of spins in the B sublattice may occur in a random local way.

Fig. 4 shows the ME spectra taken of the same sample at 4.2 K in the presence of an applied magnetic field parallel to the γ-ray beam. The intensities of the external fields were 5, 10, 20, 30, 40 and 60 kOe. It is seen that the rising magnetic field separates the

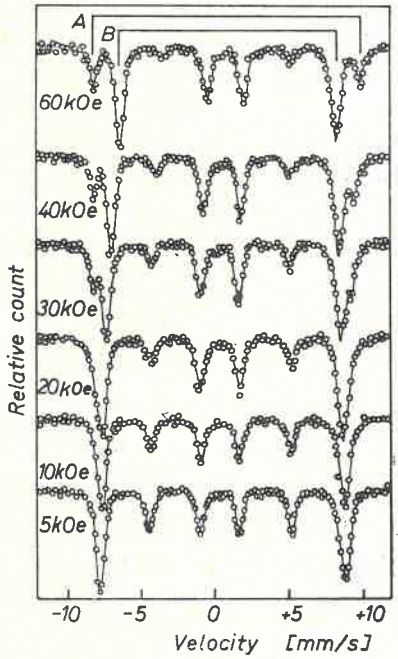

Fig. 4. Mössbauer spectra of $Zn_xNi_{1-x}Fe_2O_4$ ($x = 0.7$) at 4.2 K with external fields 5, 10, 20, 30, 40, 60 kOe parallel to the γ-ray beam. The solid curves are least square fits

Zeeman sextets belonging to A and B ion Fe^{3+} sites and gradually quenchs the intensities of $\Delta m = 0$ transitions, i.e. lines 2 and 5.

In order to get quantitative information about the spin structure of the system, the double hyperfine pattern corresponding to A and B sites has been fitted to the experimental spectra by means of a routine computer procedure. In the program, the areas under the lines have been expressed by appropriate combinations of the $\overline{\cos^2\psi_i}$ function, where ψ_i — is the angle between the ith Fe^{3+} ionic moment and H_{ext}. The cation distribution was assumed to follows the formula: $(Zn_xFe_{1-x})\,[Fe_{1+x}Ni_{1-x}]O_4$. It appeared that when $\cos^2\psi_A$ and $\cos^2\psi_B$ are put into the programme as variable parameters, the best fit to the experimental spectra in the fields $H_{ext} \geqslant 30$ kOe is obtained for $\overline{\cos^2\psi_A} = 1.0 \pm 0.12$ and χ^2 — ranging between 1.7—2.03. Therefore, in the subsequent computer fit we have set the $\overline{\cos^2\psi_A}$ term equal to unity leaving the $\overline{\cos^2\psi_B}$ term as a variable parameter. The best fit obtained in such a way is plotted in Fig. 4 where the solid line represents the computed

spectra and dots the experimental points. The values of $\overline{\cos^2\psi_B}$ as a function of H_{ext} are shown in Fig. 5.

In order to verify to what extent the values of $\cos^2\psi_B$, derived in such a way, could be changed, if in reality, a fraction of the B site moments is inverted by the spin-flip process with respect to the resultant magnetization of the B — sublattice, the following test has been performed. It was assumed that for a given x there is a trial cation distribution following the formula:

$$(Zn_xFe_{1-x+\delta})\,[Ni_{1-x}Fe_{1+x-\delta}]O_4.$$

This means that in the presence of H_{ext}, a fraction δ of the B site moments is seen in the Mössbauer pattern as if it would belong to the A — sublattice, which is the case spin-flip takes place. It appeared, however, that $\overline{\cos^2\psi_B}$ vs H_{ext} computed with such a trial cation distribution is the same as the values derived previously with a normal cation distribution

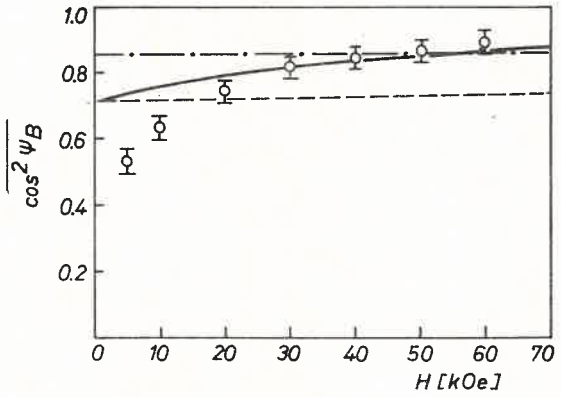

Fig. 5. Experimental and calculated curves from LCM for $\overline{\cos^2\psi_B}$ at 4.2 K for $Zn_xNi_{1-x}Fe_2O_4$ ($x = 0.7$) vs external magnetic field; ○ ME experiment; ——— LCM(a) for $J_{21} = -4.5$, — — — LCM(a) for $J_{21} = -50.5$ $\delta_1 = 0.155$, $\delta_2 = \delta_3 = \delta_4 = 0$; —•—•— LCM(b) for $J_{21} = -49.6$, $\delta_1 = 0.15$, $\delta_2 = 0.75$

within the computational error ± 0.04, if δ does not exceeds 10% of total amount of Fe^{3+} ions in the B sublattice. Since there is no reason to expect that the fraction $\delta/(1+x)$ is greater than the probability $P(6, x)$, that a given Fe^{3+}(B) ion is surrounded exclusively by Zn ions in the A sublattice, which for $x = 0.7$ equals 0.118, we shall assume hereinafter that the values of $\cos^2\psi_B$ in Fig. 5 are determined with sufficient accuracy. It should be emphasized that we regard the $\overline{\cos^2\psi_B}$, but not $\overline{\cos\psi_B}$, as a quantity directly accessible from ME transitions. This results from the lack of a long-range Y–K structure, the existence of which did not effect our neutron diffraction measurements, the noncollinearity of spins may occur only in a random local way and hence $\overline{\cos\psi_B} \neq (\overline{\cos^2\psi_B})^{1/2}$. The importance of this inequality, disregarded in [4] and [6] is illustrated best by the following example.

The value of $\overline{\cos\psi_B}$ predicted by LCM-a (see Section 3) for $x = 0.7$, $H_{ext} = 0$ is 0.64 whereas $(\overline{\cos^2\psi_B})^{1/2} = 0.84$.

3. *Localized Canting Model of $Zn_xNi_{1-x}Fe_2O_4$ in the presence of an external magnetic field*

The Localized Canting Model worked out by Rosencwaig [8] for two sublattice ferrimagnets is a mathematical representation of the idea proposed by Geller [10] according to which, in a diamagnetically substituted ferrimagnet, there occurs random spin canting in the sublattice opposite to that in which the substitution takes place.

Below, we outline the procedure leading to the expressions for the quantities represented in LCM in the three sublattice Zn–Ni system in the presence of an external magnetic field H_{ext}.

Two versions of LCM are considered: LCM-a in which it is assumed that Fe^{3+} and Ni^{2+} moments in the B site form on the average, identical local canting angles, $\psi_B(Fe) = \psi_B(Ni)$ and LCM-b. This is the extreme case of the assumption that $\psi_B(Fe) \neq \psi_B(Ni)$.

a. The case of indentical canting angles in Ni(B) and Fe(B) sublattices LCM-a versions

In the external magnetic fields higher than those required for ferrimagnetic domain alignment, the molecular field originating from the Fe(A) sublattice and acting on a chosen B site moment is parallel to the external magnetic field, whereas the molecular fields from Fe(B) and Ni(B) sublattices form an average an angle $(\pi - \bar{\theta}_c)$ with H_{ext} and an angle $\bar{\theta}_c$ between themselves. This configuration of fields leads to a local canting of a chosen B site moment with an angle ψ_B such that $\cos\psi_B$ is given by:

$$\cos\psi_B = \frac{(H_{21}+H_{ext})-(H_{22}+H_{23})\,\overline{\cos\theta_c}}{[(H_{22}+H_{23})^2+(H_{21}+H_{ext})^2-2(H_{22}+H_{23})(H_{21}+H_{ext})\,\overline{\cos\theta_c}]^{1/2}}, \qquad (1)$$

where $H_{ij} = \dfrac{1}{q_i\mu_B} n_{ij}J_{ij}S_j$ represents the molecular field originating from the j-th sublattice and experience by the i-th site ion. Indices 1, 2, 3 correspond to Fe(A), Fe(B) and Ni(B) respectively, n_{ij} represents the number of nearest neighbors on the j-th sublattice that interact with the i-th ion, J_{ij} is the exchange parameter and S_j is the spin of j-th ion. The value of $\overline{\cos\theta_c}$, which reflects the influence of the rest of the lattice is to be computed from the minimum energy condition. The potential energy of such a spin system in the presence of an external field is given by

$$V = -\tfrac{1}{2}\sum M_iH_i - \sum M_iH_{ext}. \qquad (2)$$

Here, $M_i = g_i\mu_Bq_iS_i$ is a magnetic moment of the i-th sublattice, where q_i is the number of i-th ions per unit formula $q_1 = q_3 = 1-x$, $q_2 = 1+x$, $H_i = \sum_j H_{ij}$ is the resultant molecular field acting on the i-th site ion. The state of lowest energy has the configuration for which $\delta V/\partial\theta_c = 0$. Using this condition one finds from (2) after some algebra that

$$\cos\theta_c = \frac{q_2n_{21}J_{21}(S_1S_2)+q_3n_{31}J_{31}(S_1S_3)+q\mu_B(q_2S_2+1.2q_3S_3)H_{ext}}{2[q_2n_{22}J_{22}S_2^2+2q_2n_{23}J_{23}(S_2S_3)+q_3n_{33}J_{33}S_3^2]}, \qquad (3)$$

under the simplifying assumption that on the average $q_i n_{ij} = q_j n_{ji}$. Since there exists chemical disorder in the Zn–Ni system, the Zn^{2+}, Fe^{3+} ions in the tetrahedral sites and Fe^{3+}, Ni^{2+} ions in octahedral sites are distributed at random. Therefore, to evaluate the average value of $\overline{\cos\theta_c}$ one has to average expression [3] over all possible values of n_{ij}. Defining $P(N_{iK}, n_{ij})$ as a probability that of N_{iK} equivalent neighboring sites there is n_{ij} of j-th ions we get

$$\overline{\cos\theta_c} = \sum_{n_{21},n_{31},n_{22},n_{33}} P(N_{2A}, n_{21})P(N_{3A}, n_{32})P(N_{2B}, n_{22})P(N_{3B}, n_{33}) \cos\theta_c, \tag{4}$$

where N_{iK} are: $N_{1A} = 4$, $N_{2A} = N_{3A} = 6$, $N_{nB} = N_{3B} = 6$, $N_{1B} = 12$ and $n_{23} = N_{2B} - n_{22}$.

In our calculation we assume after [8] that $P(N_{iK}, n_{ij})$ is a simple binominal distribution function depending on the Zn content x. The same averaging produce has to be adopted in order to compute the averages connected with a local canting angle ψ_B.

Thus

$$\overline{\cos^2\psi_B} = \sum_{n_{21},n_{22}} P(N_{2A}, n_{21})P(N_{2B}, n_{22}) \cos^2\psi_B \tag{5}$$

and

$$\overline{\cos\psi_B} = \sum_{n_{21},n_{22}} P(N_{2A}, n_{21})P(N_{2B}, n_{22}) \cos\psi_B, \tag{6}$$

where $\cos\psi_B$ is given by (1).

Now, according to the cation distribution $(Zn_xFe_{1-x})[Ni_{1-x}Fe_{1+x}]O_4$ the magnetization of the system can be expressed in the form

$$\mu(x) = [(1+x)\mu_{Fe} + (1-x)\mu_{Ni}]\overline{\cos\psi_B} - (1-x)\mu_{Fe}, \tag{7}$$

where μ_{Fe} and μ_{Ni} are the ionic magnetic moments of Fe^{3+} and Ni^{2+} is assumed here to have 5 μ_B and 2.4 μ_B respectively. The dependence of the relative Curie point is to be computed from a low of corresponding states expressed in [8] by

$$\frac{T_c(x)}{T_c(0)} = \frac{[(\bar{S}+1)/\bar{S}]x}{[\bar{S}(+1)\bar{S}]0} \frac{(-V)x}{(-V)0}. \tag{8}$$

Here $S = \frac{1}{N}\sum_{i=1}^{3} q_i S_i$ where $N = 7$ is the number of ions per formula unit in Zn–Ni ferrite. The potential energy $V(x)$ is to be found from (2) taken with $H_{ext} = 0$.

b. The case of parallel Ni(B)–Fe(A) and canted Fe(B) sublattice LCM-b version

When the molecular fields originating from the Fe(A) and Ni(B) sublattices and acting on a chosen B site moment are both parallel to H_{ext}, whereas the molecular field from Fe(B) makes on the average the angle $(\pi - \bar{\theta})$, with respect to H_{ext}, then the local canting angle ψ_B of the B moment is such that

$$\cos\psi_B = \frac{(H_{21}+H_{23}+H_{ext}) - H_{22}\overline{\cos\theta_c}}{[H_{22}^2 + (H_{21}+H_{23}+H_{ext})^2 - 2H_{22}(H_{21}+H_{23}+H_{ext})\cos\theta_c]} \tag{9}$$

and the average canting angle $\bar{\theta}_c$ computed from (2) is such that

$$\cos\theta_c = \frac{n_{21}J_{21}S_1 + n_{23}J_{23}S_3 + g\mu_B H_{ext}}{2n_{22}J_{22}S_2}. \tag{10}$$

Using the same procedure for averaging as in case (a) we can express the magnetization $\mu(x)$ in the form

$$\mu(x) = [(1+x)\mu(\mathrm{Fe})]\,\overline{\cos\psi_B} - (1-x)\,[\mu(\mathrm{Fe}) + \mu(\mathrm{Ni})]. \tag{11}$$

4. Result and discussion

Rosencwaig [8] has shown that LCM is capable of predicting the substitution dependence of magnetization at 0K and the Curie temperature of a singly and doubly substituted YIG-system. The only adjustable parameters of the theory were the ratios of the intrasublattice to intersublattice exchange integrals. Applying the same model to the three sublattice case, Grill [11] et al. were able to explain several magnetic properties of substituted YbIG systems. In this last case a theoretical fit to the experimental results was obtained using the exchange integrals of pure YbIG. J_{ij} were assumed the same for the substituted samples.

It is therefore interesting to verify whether the LCM expressed in the form presented in Section 3 applies also to Zn–Ni ferrite. Of particular interest is to answer the question whether the properties discussed in Section 2 could be describe using the same set of J_{ij} parameters for all Zn-contents.

Using expression derived in Section 3, the magnetization $\mu(x)$ and relative Curie temperature $T_c(x)/T_c(0)$ as function of x and $\overline{\cos^2\psi_B}$ vs H_{ext} for $x = 0.7$ have been evaluated with various sets of J_{ij} parameters by means of a computer program. The best fit to the experimental data was obtained with

$$\delta_1 = \frac{J_{22}}{J_{21}} = 0.155; \quad \delta_2 = \frac{J_{23}}{J_{21}} = \delta_3 = \frac{J_{13}}{J_{21}} = \delta_4 = \frac{J_{33}}{J_{21}} = \delta_5 = \frac{J_{11}}{J_{21}} = 0$$

and $J_{21} = -4.5$ in $[\mathrm{KK_B}]$ units. The ratio δ_5 is not represented in $\mu(x)$ and $\overline{\cos^2\psi_B}$.

The results are presented in graphical form in Fig. 1, Fig. 2 and Fig. 5.

It is seen that with these J_{ij}, the LCM in its version a — i. e. with identical canting angles of Fe(B) and Ni(B) moments accounts well for $\mu(x)$, reproduces roughly the character of the dependence of $T_c(x)/T_c(0)$ on x and gives a fair fit of $\overline{\cos^2\psi_B}$ vs H_{ext}. Obviously, the theoretical values of this last quantity may be compared with the experimental data only in the range of the applied fields in which a domain alignment process is over i. e. for $x \geqslant 30$ kOe. This is because the LCM predicts the effect of the applied field on the spin structure but not on the domain magnetization process. However, the low values of J_{21} and zero value of J_{13} exchange parameters remain questionable. For instance, the absolute Curie point T_c computed with these exchange parameters for the $NiFe_2O_4$

structure as a solution of the molecular field equations

$$m_i T_c - \frac{S_i+1}{3S_i K_B} g_i \mu_B S_i H_i = 0 \tag{12}$$

equals 75 K instead of about 850 K. Taking the value $J_{21} = -50.5$ and the ratios $\delta_m (m = 1, \ldots, 5)$ as before, one obtains the proper value of $T_c = 850$ K and unchanged fit of $\mu(x)$ and $T_c(x)/T_c(0)$ as compare to those given in Fig. 1 and Fig. 2 since these quantities do not depend on the absolute but on the relative values of the exchange parameters. However, the evident discrepancy arises between the calculated and experimental $\overline{\cos^2\psi_B}$ vs H_{ext} as is seen in Fig. 5. Firstly, the calculated $\overline{\cos^2\psi_B}$ essentially does not depend on H_{ext} between 0 and 60 kOe, which means that intensities of these fields are too small to affect the canted spin structure of the system. Secondly, the values of theoretical $\overline{\cos^2\psi_B}$ are smaller than experimental ones in $H_{ext} \geqslant 30$ kOe which means in turn LCM-a predicts the canting angles greater than actual non-collinearity angles observed in ME measurements.

Let us now consider the results of a more realistic assumption, that the moments of Fe^{3+}(B) and Ni^{2+}(B) do not form on the average the identical canting angles.

Since it is not possible to express in an algebraic form the angular relations represented by LCM in the presence of two different local angles ψ_B(Fe) and ψ_B(Ni) we shall consider only the extreme case of this model, i. e. LCM-b, see Section 3. Namely, we shall assume that all Ni(B) moments are inverted with respect to the resultant magnetization of the system and are parallel to two moments of the Fe(A) sublattice. It may be readily seen that such an assumption makes sense only for $x \geqslant 0.6$ since within the range $0.2 < x < 0.6$ the computed $\mu(x)$ would always be smaller than the experimental values even if Fe(B) moments are assumed to form a collinear array.

Using expressions pertinent to LCM-b the value of magnetization for $x = 0.7$ and $\overline{\cos^2\psi_B}$ vs H_{ext} have been evaluated with different sets of J_{ij} parameters. In addition the Curie point of $NiFe_2O_4$ have been computed from (12).

The proper $T_c = 850$ K for $NiFe_2O_4$ and resonably close to the experimental value of magnetization μ for $x = 0.7$ were obtained with $J_{21} = -49.6$, $\delta_1 = 0.15$, $\delta_2 = 0.75$, $\delta_3 = 0.75$, $\delta_4 = \delta_5 = 0$. Here, only δ_1, and δ_2 affects $\mu(x)$ and $\overline{\cos^2\psi_B}$. The calculated $\overline{\cos^2\psi_B}$ vs H_{ext} is shown in Fig. 5 by the upper dashed line. It is seen that once again, the $\overline{\cos^2\psi_B}$ does not depend on H_{ext}.

However, unlike the LCM-a with $J_{21} = -50.5$ K, the LCM-b predicts the constant $\overline{\cos^2\psi_B} = 0.85$ to which the experimental values approach asymptotically with increasing applied field. These facts would not be in contradiction if the increase of $\overline{\cos^2\psi_B}$ with increasing H_{ext} observed in ME was caused by the domain alignment process rather than by an effect of the applied field on a canted spin structure. However, this second process has been confirmed by the fact that for $H_{ext} \geqslant 30$ kOe, the Fe(A) moments are parallel to the applied field. Therefore, we must recognize the prediction of LCM-b shown in Fig. 5 as a failure.

From the studies presented above the following conclusion could be drawn:

1. The complementary, neutron diffraction and ME measurements enable a determination of the type of non-collinearity of spin array in mixed ferrimagnets. In our case of $Zn_xNi_{1-x}Fe_2O_4$ for $x = 0.7$ it appeared that the canting of the B site moments has a short range, local character.

2. LCM applied by us to $Zn_xNi_{1-x}Fe_2O_4$ is capable of accounting for the dependence of magnetic moment of Zn content with constant ratios of exchange parameters.

3. In order to account for the dependence of canting angle on the magnitude of the applied field, one has to use the exchange parameters of about one order of magnitude smaller than those given for $NiFe_2O_4$ [12, 13]. This means that exchange parameters decrease strongly with increasing Zn-content.

4. A detailed, quantitative description of the low temperature properties of the Zn–Ni system requires systematic data on $\overline{\cos^2\psi_B}$ vs H_{ext} for various x, the ME observation of ^{61}Ni nuclei and the numerical solution of LCM for different angles ψ_B(Fe) and ψ_B(Ni).

5. The confirmation of spin canting in the Zn–Ni system is not in contradiction with the presence of superparamagnetic effects observed in ME at elevated temperatures and large Zn-content as it was established in our previous studies reported in [14].

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