Mössbauer study of $Ni_{0.25}Zn_{0.75}Fe_2O_4$. II. Noncollinear spin structure

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Mössbauer spectra of $Ni_{0.25}Zn_{0.75}Fe_2O_4$ at low temperatures ($T < 100$ K) in the absence of an external magnetic field show a rapid decrease in the magnetic splitting and an increase in linewidths with the increase in temperature. This has been concluded to be due to the low value of the transition temperature at which the noncollinear structure changes into an antiparallel arrangement of ionic moments at A and B sites. The dependence of the Yafet-Kittel angle on the external magnetic field can be used to determine the exchange constants between Fe^{3+} and its neighboring ions. The splitting of the B-site spectrum into components with and without appreciable noncollinearity of ionic moments, however, introduces uncertainty in the determination of the exchange constants. The experimental data support a localized-canting model of spins.

I. INTRODUCTION

The determination of the saturation values of hyperfine magnetic fields and Yafet-Kittel angles, and of the natural linewidths in the Mössbauer spectra of mixed ferrites has been conventionally made[1–16] from the spectra taken in the range of temperature from 4.2 to 20 K. This has been believed to be reasonable because the above quantities are expected to show little change in this temperature range owing to the comparatively large value of the Néel temperatures ($T/T_N \sim 0.1$).

In earlier studies on $Co_{0.3}Zn_{0.7}Fe_2O_4$ and $Ni_{0.25}Zn_{0.75}Fe_2O_4$, however, an anomalous decrease in the magnetic splitting and an increase in relaxation line broadening was observed[17–20] when the temperature was raised to 79 K, $T/T_N \sim 0.25$. The zero-field spectra at the low temperatures could be very satisfactorily fitted with theoretically simulated ionic-spin-relaxation spectra, also in the presence of the anomalous effect. The analysis revealed[17–20] an anomalous increase in the value of the s parameter of the relaxation spectrum as the temperature increased to 78 K. The application of an external magnetic field of 12.3 kG reduced[18] this anomalous behavior but was insufficient to reveal the origin of such a behavior. Furthermore, the most interesting temperature region from 4.2 to 78 K was not investigated.

In the present study of $Ni_{0.25}Zn_{0.75}Fe_2O_4$, Mössbauer spectra have been obtained in the temperature range from 4.2 to 300 K, with greater emphasis on the region up to 100 K, in the presence of longitudinal magnetic fields up to 80 kG. The features of the experimental spectra related to the spin fluctuations have been discussed in the preceding paper,[21] hereinafter called Paper I. It was also concluded in Paper I that the anomalous decrease in the magnetic splitting due to the increase in temperature at $T < 100$ K is not due to the presence of superparamagnetism. The present analysis reveals that the transition temperature at which the noncollinear structure changes into an antiparallel arrangement of ionic moments at A and B sites, T_{YK}, is much lower in comparison to the Néel temperature, T_N, and is responsible for the sharp decrease in magnetic splitting due to the increase in temperature below 80 K.

The dependence of the Yafet-Kittel angle[22] α_{YK} on the external magnetic field is dependent on the exchange interactions of the Fe^{3+} ion with the neighboring ions. The uncertainty in the near-neighbor configuration of the ions which possess the experimentally observed noncollinearity, however, leads to uncertainties in the values of the exchange constants derived from the field dependence of α_{YK}. The experimental data seem to support a localized-canting model.[23]

II. EXPERIMENTAL RESULTS AND ANALYSES

The experimental arrangement and other relevant details have been described in Paper I. Mössbauer spectra in the temperature range from 4.2 to 77 K obtained before polarizing the absorber in any way by the external magnetic field have been shown in Fig. 1. The internal fields at Fe nuclei at A and B sites are equal at 4.2 K. The spectra in the temperature range from 4.2 to 30 K show anomalous increases in linewidths but have little overlap between the lines. A least-squares fit to a sum of six Lorentzian lines shows that the Δ_{34} (Δ_{ij} refers to the separation

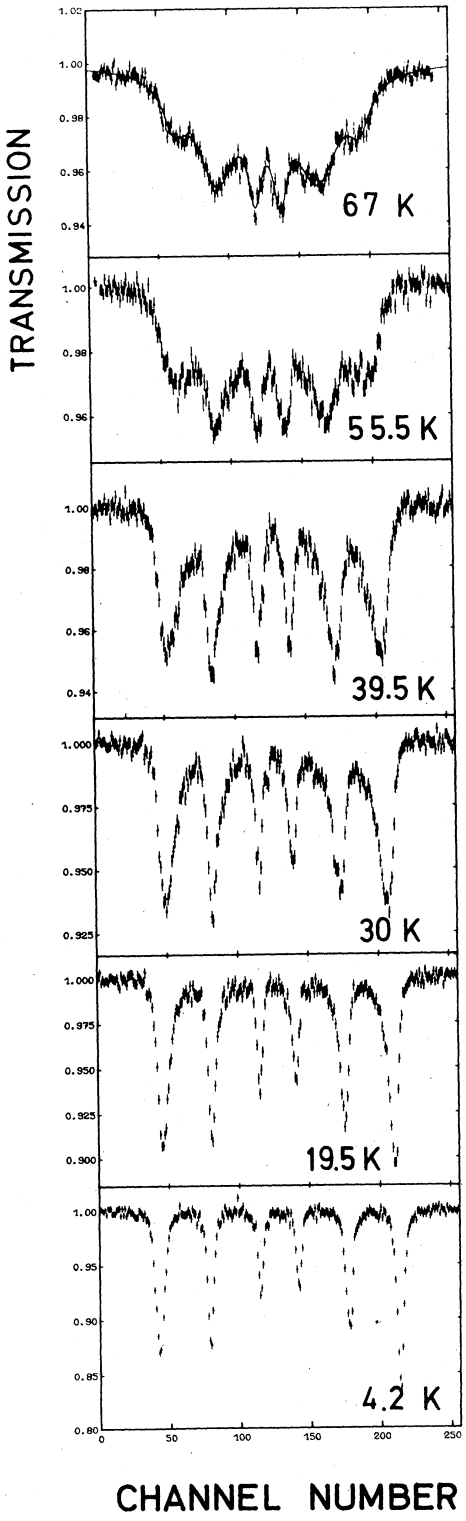

FIG. 1. Mössbauer spectra of $Ni_{0.25}Zn_{0.75}Fe_2O_4$ in absence of an external field. The change in velocity per channel (ΔV) is 0.097 mm/sec.

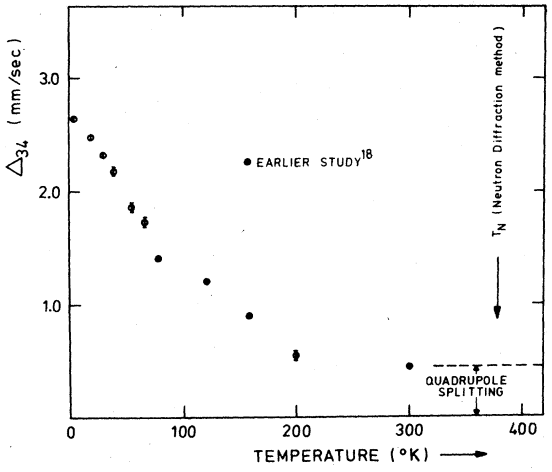

FIG. 2. Temperature dependence of the inner lines separation, in absence of an external magnetic field. The values obtained in the temperature range from 77 to 300 K in an earlier study (Ref. 18) using the same sample have been included for completeness.

between ith and jth lines) decreases and the linewidths increase anomalously with the increase in temperature. This procedure yields an average of the temperature dependences of Δ_{34} of A and B components. It is, however, reasonable to conclude that if the behavior of one of the two components is more normal, the behavior of the other component should be more anomalous. The anomalous behavior of Δ_{34} can be divided into three regions (Fig. 2). In the first region, Δ_{34} decreases from 2.64 mm/sec at 4.2 K to 2.17 mm/sec at 39.5 K which means a decrease of 91 kG due to an increase in temperature of only 35 K. The decrease expected on the basis of the Brillouin curve for a sublattice consisting of ions with $S=\frac{5}{2}$ and $T_N=375$ K is less than 2 kG for an increase in temperature by 35 K in this temperature range. This experimentally observed rate of decrease implies that the magnetic splitting should collapse below 200 K although the Néel temperature is 375 K (as shown by the neutron-diffraction measurement[24]). In the second range of temperature which is well characterized by the behavior in the range from 80 to 160 K, the Δ_{34} decreases[18] from 1.36 mm/sec (267.25 kG) at 80 K to 0.86 mm/sec (169 kG) at 160 K. This rate of decrease is nearly half of the rate in the first region. At still higher temperature, i.e., above 200 K, the rate of decrease[18] of Δ_{34} becomes much slower and inappreciable at 300 K.

In the second phase of the experiment, the absorber was first polarized by an application of a longitudinal field of 80 kG at 4.2 K and subsequently the spectra were obtained in the fields of 0, 15, 22, 30, 50, 75, and 80 kG at 4.2 K (Fig. 3). The intensities of the second and the fifth lines increase with the decrease of the magnetic field.

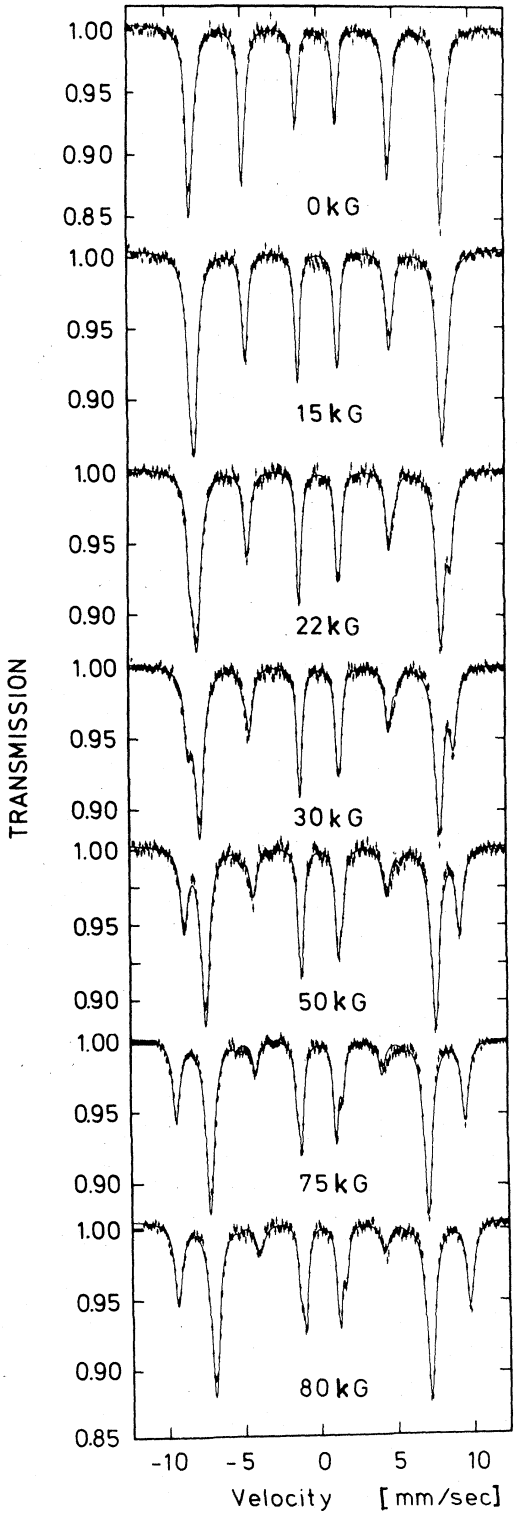

FIG. 3. Mössbauer spectra at 4.2 K in the presence of a longitudinal field of 80 kG. The solid curves have been obtained using the least-squares method described in the text.

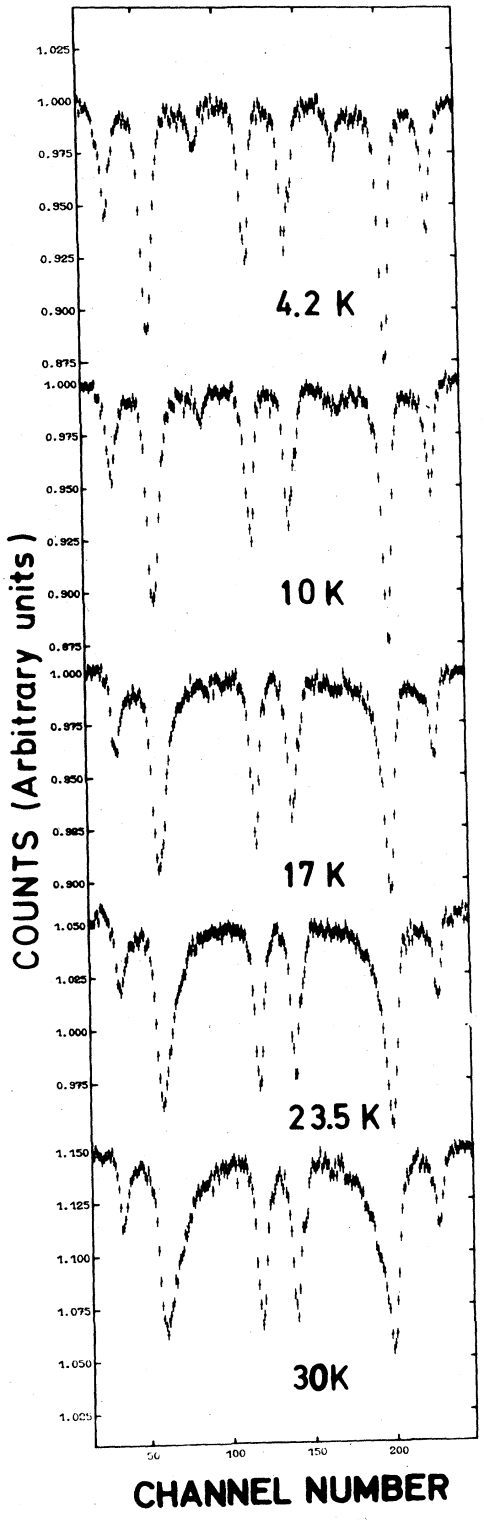

FIG. 4. Mössbauer spectra at low temperatures in the presence of a longitudinal field of 80 kG. $\Delta V = 0.097$ mm/sec.

The spectra obtained in the presence of a longitudinal field of 80 kG at temperatures in the range from 4.2 to 40 K have been shown in Fig. 4. The remarkable feature of these spectra is the sharp decrease in the intensities of the lines corresponding to the $\Delta m = 0$ nuclear transitions in the component spectrum of the ions at B sites even when the temperature increases from 4.2 to 30 K. This is evident even from a visual comparison of the spectra at 4.2 and 23.5 K in Fig. 4.

A. Dependence of Yafet-Kittel angle on external magnetic field

The experimental spectra shown in Fig. 3 were fitted with two six-line patterns using a least-squares method. It was assumed that the line widths and the intensities of pairs of corresponding lines in a component spectrum are equal. It was found necessary to constrain the widths of the lines corresponding to $\Delta m = 0$ nuclear transitions to reasonable values. The widths and the intensities of two lines which have large overlap were not allowed to vary independently. They were treated alternately as free parameter.

The value of H_{int}^{B} derived from this fitting procedure depends mainly on the positions of the outer lines. The fitting in the regions of the lines corresponding to $\Delta m = 0$ nuclear transitions was unsatisfactory in all the spectra shown in Fig. 3. Consequently, the positions of the lines corresponding to $\Delta m = 0$ transitions have been determined by fitting these two lines separately with Lorentzian lines. The line separation Δ_{25} thus determined corresponds to a larger value of hyperfine magnetic field than the value of H_{int}^{B} obtained from the fitting procedure described above in which it was mainly dependent on Δ_{16}. This thus shows that the component spectrum B consists of at least two components B_1 and B_2. (This is shown very clearly by spectra at higher temperatures describe in Paper I.)

The experimental data show that B_1 contributes to the outer lines of the B component but not appreciably to the lines correpsonding to $\Delta m = 0$ transitions. The hyperfine field and the Yafet-Kittel angles derived from Δ_{25} using the relation

$$\alpha_{\text{YK}} = \arccos\left(\frac{H_{\text{eff}}^2 - H_{\text{int}}^2 - H^2}{2H_{\text{int}}H}\right) \tag{1}$$

have been included in the Table I along with the other results of the above analysis. Furthermore, the spectrum in the presence of 80 kG has been unfolded into three components A, B_1, and B_2 assuming $\alpha_{\text{YK}}^{B_1} = 0$ and $\alpha_{\text{YK}}^{B_2} \neq 0$ (Fig. 1 of Paper I). The relative intensities of the outer lines and the lines corresponding to $\Delta m = 0$ transitions in the component spectrum B_2 thus determined has been used to calculate α_{YK} using the relation,

$$\alpha_{\text{YK}} = \arcsin\left(\frac{6A_{2,5}}{4A_{1,6} + 3A_{2,5}}\right)^{1/2} . \tag{2}$$

Here A refers to the area of the absorption line and the subscripts i,j refer to ith and jth lines. The value thus obtained shows good agreement with the value determined using Δ_{25} and the relation (1), given in Table I.

III. THEORETICAL CONSIDERATIONS

A. Dependence of Yafet-Kittel angle on the external magnetic field

The Yafet-Kittel model[22] of the noncollinear spin structure has been discussed extensively in Ref. 25. A simple extension of the model gives the dependence of the Yafet-Kittel angle (α_{YK}) on the external magnetic field, H. The A and B sublattices are subdivided into two ($A^{1,2}$) and four ($B^{1,2,3,4}$) sublattices, respectively, such that the intrasublattice exchange interactions are much weaker than the intersublattice exchange interactions.[22,25] Consequently, spins on any of the six sublattices have no tendency to orient at an angle with respect to each other but can make angle with respect to spins on the other sublattices. The magnetic fields at the A^i and B^i sublattices can be written

$$\vec{H}_{A^i} = -[\lambda_{A^iA^j}\vec{\mu}_A + \lambda_{AB}\vec{\mu}_B + (\lambda_{A^iA^i} - \lambda_{A^iA^j})\vec{\mu}_{A^i}] + \vec{H} , \tag{3}$$

$$\vec{H}_{B^i} = -[\lambda_{AB}\vec{\mu}_A + \lambda_{B^iB^j}\vec{\mu}_B + (\lambda_{B^iB^i} - \lambda_{B^iB^j})\vec{\mu}_{B^i}] + \vec{H} , \tag{4}$$

where $\lambda_{ij} = (2J_{ij}n_{ij}/g^2\mu_B^2)$, μ_B represents the Bohr magneton and J_{ij} is positive for antiferromagnetic exchange. In the following we consider the case when the noncollinearity appears on B sublattice only. It follows from Eq. (4) that $[\vec{H} - (\lambda_{AB}\vec{\mu}_A + \lambda_{B^iB^j}\vec{\mu}_B)]$ should be parallel to $\vec{\mu}_{B^i}$. Similar expression for $\vec{H}_{B^j}$ shows that $[\vec{H} - (\lambda_{AB}\vec{\mu}_A + \lambda_{B^iB^j}\vec{\mu}_B)]$ should be parallel to $\vec{\mu}_{B^j}$ also. As $\vec{\mu}_{B^i}$ and $\vec{\mu}_{B^j}$ are noncollinear,

$$\vec{H} = \lambda_{AB}\vec{\mu}_A + \lambda_{B^iB^j}\vec{\mu}_B . \tag{5}$$

Equations (3)–(5) show that the magnetic fields $\vec{H}_{A^i}$ and $\vec{H}_{B^i}$ and consequently the magnetic energy is not changed when the orientations of the sublattices B^i are changed, provided the resultant $\vec{\mu}_B$ remains unaltered. Thus the substitution

$$\vec{\mu}_{B'} = \vec{\mu}_{B^1} + \vec{\mu}_{B^3} ,$$

$$\vec{\mu}_{B''} = \vec{\mu}_{B^2} + \vec{\mu}_{B^4} ,$$

TABLE I. Results of the analyses of the spectra shown in Fig. 3. Absorber: $Ni_{0.25}Zn_{0.75}Fe_2O_4$; Temperature: 4.2 K.

External Field (kG)	H^A_{eff} (kG)	H^B_{eff} (kG)	$H^{B_2}_{eff}$ (kG)	$\alpha^{B_2}_{YK}$ (deg)	$\lvert\Delta_{12}-\Delta_{56}\rvert$ (mm/sec) *A* site	*B* comp.	$\Gamma^A_{1,6}$ (mm/sec)	$\Gamma^B_{1,6}$ (mm/sec)	$\Gamma^{B_2}_{2,5}$ (mm/sec)	$\Gamma^B_{3,4}$ (mm/sec)	$\frac{A^B_{1,6}}{A^A_{1,6}}$	$\frac{A^B_{3,4}}{A^A_{3,4}}$
80	595.3 ±0.4	439.7 ±0.2	448 ±2	31	0.055 ±0.015	0.011 ±0.007	0.59 ±0.02	0.66 ±0.01	0.5 ±0.1	0.435 ±0.02	2.37	2.83
75	589.9 ±0.5	444.9 ±0.3	456 ±2	35.5	0.04 ±0.02	0.010 ±0.008	0.58 ±0.02	0.65 ±0.01	0.65 ±0.1	0.40 ±0.02	2.45	2.36
50	563.9 ±0.6	470.8 ±0.2	479.5 ±2	43	0.05 ±0.02	0.0055 ±0.008	0.55 ±0.03	0.65 ±0.01	0.6 ±0.1	0.42 ±0.02	2.60	3.15
30	544.0 ±0.6	491.4 ±0.3	501.5 ±1	62	0.055 ±0.015	0.0115 ±0.007	0.555 ±0.03	0.66 ±0.015	0.64 ±0.07	0.44 ±0.02	2.88	3.60
22	534.3 ±0.7	500.0 ±0.4			0.03 ±0.02	0.019 ±0.008	0.45 ±0.04	0.68 ±0.02		0.42[c]	3.88	2.82
15	526.5 ±0.6	506.4 ±0.2			0.00 ±0.02	0.0195 ±0.007	0.555[c]	0.66[c]		0.42[c]	3.40	2.92
0	515.0[c]	514.9 ±0.45			0.03[c]	0.0105[c]	0.63[c]	0.68[c]		0.42[c]		

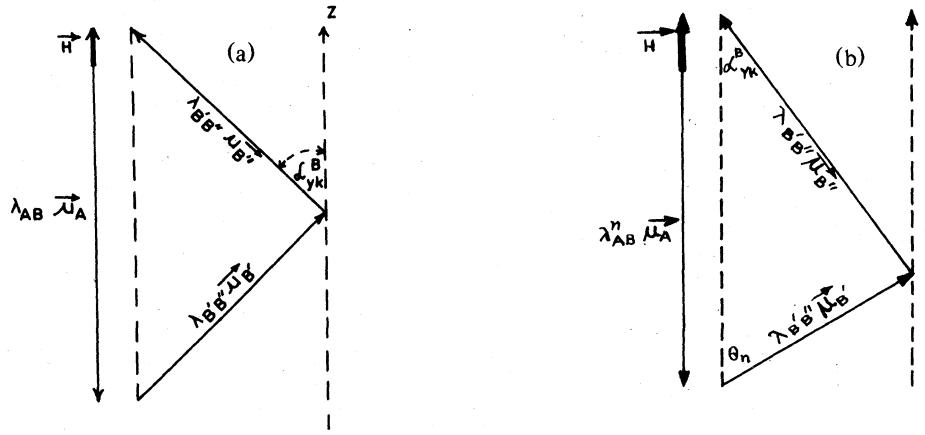

FIG. 5. (a) Schematic representation of Eq. (6) leading to the result (7). (b) Schematic representation of the relationship between θ_n and α_{YK} in the localized canting model described in the text.

does not change the magnetic exchange energy. The experimental observations also show that the B sublattice splits into two sublattices, only, which implies that $\vec{\mu}_{B^1}$ and $\vec{\mu}_{B^3}$ and also $\vec{\mu}_{B^2}$ and $\vec{\mu}_{B^4}$ are collinear in pairs. Thus Eq. (5) can be written

$$\vec{H} = \lambda_{AB}\vec{\mu}_A + \lambda_{B'B''}(\vec{\mu}_{B'} + \vec{\mu}_{B''}) , \tag{6}$$

which gives [Fig. 5(a)]

$$\cos\alpha^B_{YK} = \frac{H + \lambda_{AB}\mu_A}{2\mu_{B'}\lambda_{B'B''}} . \tag{7}$$

This relation shows that $\cos\alpha_{YK}$ is linearly dependent on the value of the external field, H. An experimental determination of $\cos\alpha_{YK}$ vs H provides the values of λ_{AB} and $\lambda_{B'B''}$.

B. Localized canting model of non-collinear spin structure

The Yafet-Kittel model of the noncollinear spin structure was extended by Rosencwaig[23] to include the effect of the local differences in the occupancy of the neighboring sites by the magnetic and nonmagnetic ions on the collinearity. The model known as the localized canting model has been further extended by Piekoszewski *et al.*[12] to include the effect of the external field on α_{YK}. The relations obtained by Rosencwaig[23] and Piekoszewski *et al.*[12] can be easily obtained using the procedure outlined above to derive Eq. (7). The Yafet-Kittel angle θ_n formed by an ion possessing near-neighbor configuration n is related to the average α^B_{YK} (as determined by the neutron-diffraction method) by the relation [Fig. 5(b)]

$$\cos\theta_n = \frac{(\lambda^n_{AB}\mu_A + H) - \lambda_{B'B''}\mu_{B'}\cos\alpha^B_{YK}}{[(\lambda^n_{AB}\mu_A + H)^2 + (\lambda_{B'B''}\mu_{B'})^2 - 2(\lambda^n_{AB}\mu_A + H)(\lambda_{B'B''}\mu_{B'})\cos\alpha^B_{YK}]^{1/2}} ,$$

where

$$\cos\alpha^B_{YK} = \sum_n p(n)\cos\theta_n$$

and $p(n)$ represents the probability that a B-site ion possess near-neighbor configuration n. This expression shows that B-site ions with no magnetic neighbor ($\lambda^n_{AB} = 0$) make an angle $(\pi - \alpha^B_{YK})$ in the absence of the external field.

C. Transition temperatures T_{YK} and T_N

In the absence of the external field the condition (5) for the appearance of noncollinearity in the B sublattice gives on substitution in Eqs. (3) and (4)

$$\vec{H}_{A^i} = -\left[\frac{1}{2}(\lambda_{A^iA^i} + \lambda_{A^iA^j}) - \frac{\lambda^2_{AB}}{\lambda_{B'B''}}\right]\vec{\mu}_A , \tag{8}$$

$$\vec{H}_{B'} = -(\lambda_{B'B'} - \lambda_{B'B''})\,\vec{\mu}_{B'} . \tag{9}$$

This shows that in the presence of noncollinear spin structure, the A and the two B sublattices behave[25] like independent ferromagnetic lattices with Curie temperatures

$$T^A_C = \left[\frac{\lambda^2_{AB}}{\lambda_{B'B''}} - \frac{1}{2}(\lambda_{A^iA^i} + \lambda_{A^iA^j})\right]\frac{g^2\mu_B^2 S_A(S_A+1)}{3k}$$

and

$$T_C^{B',B''} = (\lambda_{B'B''} - \lambda_{B'B'}) \frac{g^2 \mu_B^2 S_B (S_B + 1)}{3k},$$

respectively. The striking result is the low value of $T_C^{B'}$ in the presence of noncollinearity which is independent of the nature of ions at A sites in $Ni_xZn_{1-x}Fe_2O_4$ and the presence of the external magnetic field. This result is obtained without making any approximation. The faster decrease of the B-site moment continues until the noncollinear spin structure changes[25] into the antiparallel spin structure at a temperature T_{YK} [α_{YK} becomes zero in Eq. (7) at T_{YK}]. T_{YK} is dependent on the concentration of Zn^{2+} ions due to the occurence of λ_{AB} in Eq. (7). The effect of the small value of $T_C^{B'}$ on the initial temperature dependence of sublattice magnetization of B' will be clearly observable if $\lambda_{AB}\mu_A$ is low, i.e., the concentration of Zn^{2+} ions is large. The presence of H which augments $\lambda_{AB}\mu_A$ in Eq. (7) shows that an increase in H is equivalent to a decrease in the concentration of Zn^{2+} and a lowering of T_{YK}. In the range of temperature from T_{YK} to T_N, the temperature dependences of the A and B sublattices are no longer mutually independent.

IV. DISCUSSION

A. Effect of external magnetic field on noncollinear spin structure

In the region of large external fields ($H > 30$ kG), the magnetic polarization occurs mainly due to the dependence of α_{YK} on the external magnetic field, unlike at smaller H where the domain alignment is primarily responsible for the observed polarization effect on the spectra.[12,26] This is also borne out by the negligible value of α_{YK}^A at high H (>30 kG). The variation in noncollinearity of ions at B sites due to the variation in n_{AB} from site to site is only partly taken into consideration in the analysis of the experimental spectra shown in Fig. 3. The component spectrum due to ions at B sites has been split into two components B_1, with $\alpha_{YK} = 0$, and B_2, with $\alpha_{YK} \neq 0$. The values of $\alpha_{YK}^{B_2}$, given in Table I, thus represents an average of the orientations of ions with θ_n in the range $0 < \theta_n < 90°$. The slope of $\cos\alpha_{YK}^{B_2}$ vs H curve in the region from 50 to 80 kG is 0.044 which gives $J_{B'B''} = 0.55$ K on using Eq. (7) and substituting $n_{B'B''} = 6$. As the slope is independent of n_{AB}, the value of $J_{B'B''}$ thus obtained is accurate. The value of the intercept of $\cos\alpha_{YK}^{B'}$ vs H curve on Y axis is, however, dependent on n_{AB} and thus is sensitive to the statistical variation in n_{AB} from site to site. If we neglect this spread, the intercept ($=-0.53$) obtained from $\cos\alpha_{YK}^{B_2}$ vs H curve in the region of $H > 50$ kG, gives $J_{AB} = 16J_{B'B''}$, using $n_{AB} = 1.5$ and $n_{B'B''} = 6$. It is, however, reasonable to expect that exclusion of B_1 component in obtaining the $\cos\alpha_{YK}^{B_2}$ vs H curve implies that B_2 spectra correspond to $n_{AB} < 1.5$ (the average value). Thus we conclude $J_{AB} > 16J_{B'B''}$ which is in satisfactory agreement with results of neutron-diffraction measurement. The above analysis does not take into consideration the effect of the presence of Ni^{2+} in place of a part of Fe^{3+} on B sites which means $J_{B'B''}$ obtained above is an average of Fe-Fe and Fe-Ni superexchanges with appropriate weight factors (1 : 0.15).

B. Interpretation of temperature dependence of Δ_{34} at low temperatures

It was shown in Paper I that the anomalous decrease in the magnetic splitting with the increase in temperature at $T < 100$ K is not due to the presence of superparamagnetic fluctuations. A remarkable experimental observation which provides a clue to the interpretation of the low-temperature behavior of Δ_{34} is the disappearance of the lines corresponding to $\Delta m = 0$ nuclear transitions in the B_2 component even when the temperature is raised to 30 K, in the field of 80 kG (Fig. 4). This shows that $\alpha_{YK}^{B_2}$ decreases rapidly with the increase in temperature from 4.2 to 30 K.

As described in Sec. III, the molecular fields at A, B', and B'' sites in the presence of noncollinearity are given by Eqs. (8) and (9). Thus initially A and the two B sublattices behave like ferromagnetic lattices with Curie temperatures T_C^A and $T_C^{B'}$, respectively. The relation (9) shows that the initial magnetization of the ions at B sites forming noncollinear spin arrangement should show a temperature dependence independent of α_{YK} and the nature of ions at A site, i.e., λ_{AB}, and corresponding to a much lower T_C than T_N. As discussed in Sec. III, the effect is expected to be more pronounced when the diamagnetic substitution is large as in the present case. As 87.5% of the Fe^{3+} ions lie on the B sublattice and a large fraction of them are expected to form noncollinear spin structure, the sharper decrease in Δ_{34} as T increases from 4.2 to 40 K is understandable. At a temperature T_{YK} lower than T_N, the noncollinear configuration of spin changes[25] to collinear arrangement of spins on A and B sites when $T_C^A > T_C^{B'}$, as in the present case. In the range of temperature between T_{YK} and T_N the magnetization of A and B sublattices are mutually dependent through λ_{AB} and corresponds to the Néel temperature T_N. The application of an external field reduces the noncollinearity but the fields at the B sublattices remain unchanged if $\alpha_{YK} \neq 0$. This interpretation explains the temperature and field dependences at lower temperatures, but cannot explain the collapse of the magnetic splitting at $T \sim 250$ K ($< T_N$) in absence of the external field.

Thus as mentioned earlier, whereas the collapse of the splitting at temperature (~250 K) lower than T_N show presence of fluctuation effect like superparamagnetism, in addition to the ionic spin relaxation, the sharp decrease of Δ_{34} at low temperatures appears to be due to $T_{YK} << T_N$. This is well supported by theoretical considerations. The effect of the external field is twofold: (i) to suppress the fluctuation which causes the collapse of the magnetic splitting at $T \sim 250$ K; (ii) to decrease α_{YK}^{B}. Ions at B sites for which α_{YK} is reduced to zero and the fluctuation effects are suppressed due to the application of the external field show a normal temperature dependence of $\langle S_z \rangle$. This is in agreement with the characteristics of A and B_1 components. On the other hand, B-site ions for which $\alpha_{YK} \neq 0$ and only superparamagnetic fluctuation effects are suppressed by the external field would show a faster decrease of Δ_{34}, independent of the strength of the external field, upto T_{YK}, as are the experimentally observed characteristics of the B_2 component. This explanation is also consistent with the field dependence of the shape of the B_2 component spectrum at 100 K and the rapid fall in the intensities of the lines corresponding to $\Delta m = 0$ nuclear transitions as the temperature increases from 4.2 to 30 K, in presence of 80 kG.

V. CONCLUSIONS

Whereas the assumption of the presence of superparamagnetic effect satisfactorily explains the collapse of magnetic splitting at $T \sim 250$ K ($< T_N$), it is found to be an incorrect explanation for the anomalous decrease of Δ_{34} at lower temperatures ($T < 100$ K). It has been concluded that the low-temperature behavior of Δ_{34} arises from $T_{YK} << T_N$, where T_{YK} is the temperature at which the noncollinear spin arrangement transforms into a configuration of antiparallel spins. Below T_{YK} the temperature dependences of the magnetizations of the B' and B'' sublattices are theoretically expected to correspond to a low value of the Curie temperature $T_C^{B'} << T_N$ and to be independent of the nature of the ions at A sites and of the external field, in good agreement with the experimental results. The temperature dependence of $\langle S_z \rangle$ corresponding to the B_2 component which shows noncollinearity of spins in 80 kG is not different from the behavior of Δ_{34} in the absence of the external field (Fig. 4 of Paper I) and the intensities of the lines corresponding to $\Delta m = 0$ transitions in this component spectrum becomes negligible even at 30 K.

The analysis has been made assuming the presence of only two components, $B_1(\alpha_{YK} = 0)$ and $B_2(\alpha_{YK} \neq 0)$, in the B-site spectrum. The experimental data are not suitable for testing the localized canting model[24] in greater details. It is also not clear if the larger relative intensity of the A component than is given by the cation distribution,

$$(Zn_{0.75}Fe_{0.25})[Ni_{0.25}Fe_{1.75}]O_4 ,$$

(here the parentheses indicate cations at A sites and the square brackets indicate cations at B sites) is due to B-site ions with no magnetic neighbors contributing to the A component (Table I). The thickness correction is not larger than 10% for any of the lines in the presence of the external field. The present study shows that the complicated behavior of spin moments are best studied using Mössbauer spectroscopy. The anomalous decrease in Δ_{34} at low temperatures shows the necessity of measurement at lower temperature for the determination of saturation values of hyperfine magnetic fields and Yafet-Kittel angle and natural linewidths in the magnetically split spectrum.

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