Local magnetic structure and anomalies of magnetic hyperfine interactions of the ^{57}Fe and ^{119}Sn nuclei in lithium iron aluminates

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An investigation was made of the Mössbauer spectra of the ^{57}Fe and ^{119}Sn nuclei in ordered lithium iron aluminates $Li_{0.5}Fe_{2.5-x}Al_xO_4$:Sn with compositions in the range $0.8 \leqslant x \leqslant 1.0$ (near the compensation point). The results indicated a local canted (noncollinear) magnetic structure in which the canting angle was a function of temperature and aluminum concentration. Hyperfine magnetic fields with a complex distribution $P(H^{Sn})$ were observed at the tin nuclei. Anomalies of the temperature dependence of the $P(H^{Sn})$ distribution were attributed to changes of the various contributions made to the H^{Sn} field because of transformation of the local canted magnetic structure into a collinear one.

1. INTRODUCTION

A special feature of the magnetic properties of the $Li_{0.5}Fe_{2.5-x}Al_xO_4$ system is an anomalous temperature dependence of the magnetization.[1] In the case of pure Li ferrite the direction of magnetization σ_s coincides with the direction of the magnetic moment of the octahedral B sublattice. As iron is replaced with aluminum, the "normal" temperature dependence $\sigma_s(T)$ of the Q type (in accordance with the Néel classification) transforms into a P-type curve at $x = 0.6$, whereas at $x = 0.8$ it becomes an N-type dependence with the compensation point at $T_c \approx 150$ K. Further replacement of iron with aluminum (up to $x = 1.0$) restores the "normal" temperature dependence of the magnetization but the direction of the net magnetization is now determined by the tetrahedral A sublattice.

At low aluminum concentrations the A sublattice contains only the iron ions. As x is increased, some of the Al ions are displaced to the tetrahedral sites. The cation distribution and the Néel temperature depend on the conditions during preparation of these materials and, therefore, different authors report somewhat different values.[1-3] Investigations of the dependences $\sigma_s(H)$ and neutron diffraction data support indirectly the possibility of formation of a canted (noncollinear) magnetic structure in the range $x > 0.8$ (Refs. 4 and 5). The observed anomalies of the Mössbauer spectra of the ^{57}Fe nuclei found at certain iron concentrations[6,7] have been interpreted on the basis of a relaxation model.

We investigated the Mössbauer spectra of ordered lithium aluminum ferrite spinels (lithium iron aluminates) in the aluminum concentration range $0.8 \leqslant x \leqslant 1.0$ where compensation points T_c had been found. In addition to a study of the Mössbauer spectra of the ^{57}Fe nuclei, we used the method of a diamagnetic nuclear probe in the form of ^{119}Sn by introducing Sn^{4+} tin impurity ions into the ferrite lattice. Several magnetically inequivalent positions were found and these were associated with the immediate environment effects; there were also local regions with a canted spin structure. We determined the distribution functions of the magnetic fields $P(H^{Sn})$ at the tin nuclei. We observed anomalies of the temperature dependence of the parameters of the $P(H^{Sn})$ distribution at temperatures close to the compensation point.

2. EXPERIMENTAL METHOD

A system of tin-doped $Li_{0.5}Fe_{2.5-x}Al_xO_4$:Sn ferrites ($\sim$0.01 at. % Sn; $x = 0.8$, 0.9. 0.955, and 1.0) was synthesized by the ceramic technology using a method of simultaneous deposition of hydroxides. X-ray diffraction confirmed that the samples with the cubic spinel structure consisted of a single phase. The values of the unit cell parameters a_0 and the Néel temperatures T_N obtained in the present study for the $Li_{0.5}Fe_{2.5-x}Al_xO_4$:Sn system were as follows:

x	0,80	0,90	0,955	1,00
a_0, Å	8.219(3)	8.206(3)	8.208(3)	8,213(3)
T_N, K	601(3)	580(3)	567(3)	563(3)

An ordered distribution of univalent and trivalent cations in the ratio 1:3 was found at the octahedral positions (this was similar to the distribution found in pure lithium ferrite $Li_{0.5}Fe_{2.5}O_4$).

The Mössbauer spectra were determined for the ^{57}Fe and ^{119}Sn nuclei in the temperature range 80–650 K. The gamma radiation sources were ^{57}Co in a Cr matrix and ^{119m}Sn in the compound $BaSnO_3$.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. Spectra of the ^{57}Fe nuclei

The Mössbauer spectra of the ^{57}Fe nuclei recorded in the range $T < T_N$ (Fig. 1) exhibited a "paramagnetic" doublet in the central part of the spectrum against the background of the hyperfine magnetic splitting. In contrast to Ref. 7, the intensity of the paramagnetic doublet far from T_N did not vary with temperature or with the external magnetic field. We therefore concluded that this doublet appeared not because of relaxation, but because of the presence of the iron ions excluded from participation in the exchange interaction.

3.1.1. Magnetically disordered Fe^{3+} ions. We used the cation distribution of Ref. 2 to calculate for each sample the fraction of the iron ions with a weaker exchange coupling. In the case of the ordered structure of the spinel the immediate environment of an iron ion in the A sublattice consists of B ions distributed as follows:

$$3Li^+ + (9-n)Fe^{3+} + nAl^{3+},$$

where $n = 0, 1, \ldots, 9$. Then, the probability $P(m)$ that an

Fe^{3+} (A) ion has m magnetic neighbors in the B sublattice is

$$P_9(m)=C_m^9 k_B^{9-m}(1-k_B)^m, \qquad (1)$$

where k_B is the fraction of the nonmagnetic ions in the B sublattice. Consequently, in the case of the Fe^{3+} (B) ion the probability of the presence of m magnetic neighbors in the A sublattice is

$$P_6(m)=C_m^6 k_A^{6-m}(1-k_A)^m, \qquad (2)$$

where k_A is the fraction of the nonmagnetic ions in the A sublattice.

Figure 2 shows the experimentally determined (at $T = 300$ K) relative intensities of the paramagnetic doublet, proportional to the number of the Fe^{3+} ions which did not participate in the magnetic ordering and the corresponding calculated values. Our calculations were carried out for three cases: *a*) $m = 0$; *b*) $m \leqslant 1$; *c*) $m \leqslant 2$. A good agreement between the experimental and calculated data in case *b* indicated that the paramagnetic doublet exhibited by our samples could be explained satisfactorily by the Gilleo model,[8] i.e., on the assumption that only the Fe ions with at least two adjacent-sublattice magnetic ions in the immediate cation environment can be magnetically ordered.

3.1.2. Magnetically inequivalent cation positions. The different environments of the iron ions in the two sublattices give rise to a complex hyperfine splitting pattern because of the superposition of the subspectra due to the Fe^{3+} ions at specific local environments.

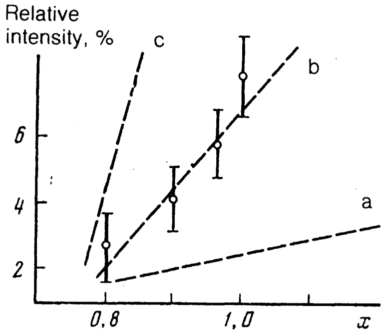

FIG. 2. Experimental (points) and calculated (dashed lines) values of the number of the iron ions that do not participate in the magnetic ordering. The calculated dependences are plotted for $m = 0$ (a), $m \leqslant 1$ (b), and $m \leqslant 2$ (c).

The parameters of the individual components were determined by analyzing the spectra of all the samples recorded at temperatures 80 and 300 K in accordance with a special program. In the initial approximation the statistical regularization method[9] was used to reduce the individual lines in the spectrum to zero width. Then, the individual lines in the spectrum were resolved into 6–8 peaks subject to the following assumptions.:

1) the ratio of the intensities of the individual subspectra is described by Eqs. (1) and (2);

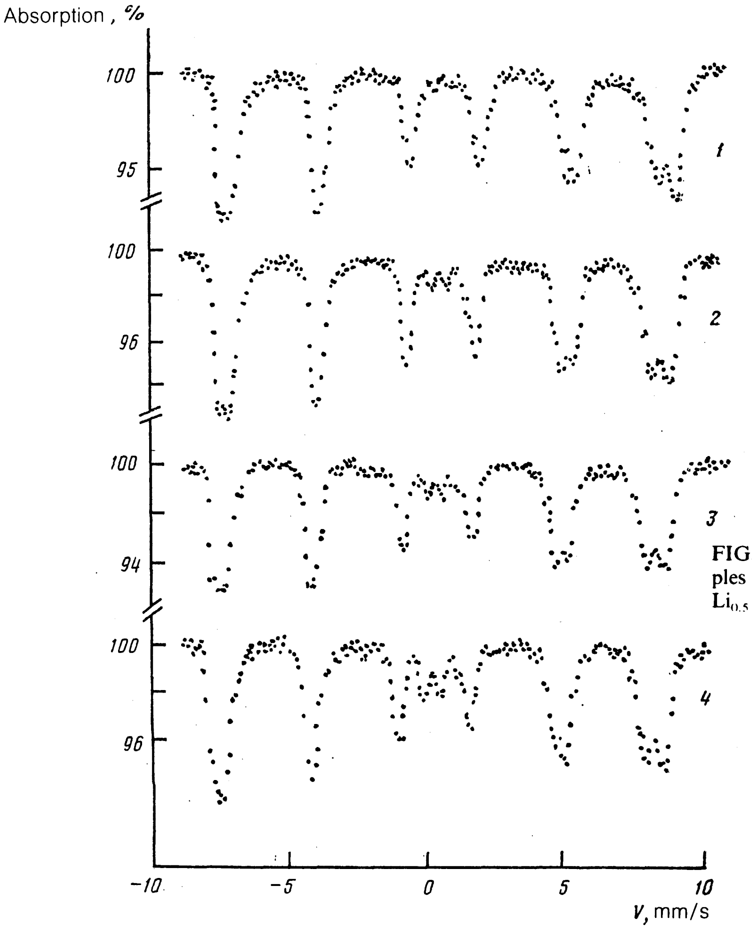

FIG. 1. Mössbauer spectra of the ^{57}Fe nuclei at 80 K, recorded for samples with different aluminum concentrations in the $Li_{0.5}Fe_{2.5-x}Al_xO_4$:Sn system: 1) $x = 0.8$; 2) 0.9; 3) 0.955; 4) 1.0.

TABLE I. Parameters of the Mössbauer spectra of the ^{57}Fe nuclei of iron ions with different magnetic environments in $Li_{0.5}Fe_{1.5}Al_{1.0}O_4$:Sn ferrite.

T, K	A sublattice			B sublattice		
	m	$H\pm3$, kOe	$\Delta E\pm0,02$, mm/s	m	$H\pm3$, kOe	$\Delta E\pm0,02$, mm/s
80	7	516	−0.52	6	529	−0,26
	6	505	−0.44	5	516	−0,25
	5	492	−0.38	4	505	−0,16
	4	481	−0.27	3	493	−0.07
	3	469	−0.15			
	2	450	0			
	0; 1		0.55			
300	7	460	0,04	6	481	−0,28
	6	452	−0,2	5	464	−0,26
	5	445	−0.25	4	445	−0,12
	4	424	−0.23	3	423	−0.07
	3	398	−0,2			
	2	360	−0,05			
	0; 1		0.6			

2) the isomeric shifts are the same for the paramagnetic doublet and the magnetically split part of the spectrum;

3) the cation distribution is the same as that postulated in Ref. 2.

This analysis enabled us to resolve the experimental spectra into the components representing the iron ions with different magnetic environments. The parameters of the iron ions for the $x = 1.0$ case are listed in Table I.

It is clear from this table that the replacement of one $Fe^{3+}(A)$ ion with an $Al^{3+}(A)$ ion in the immediate environment of the B iron ion reduces the local magnetic field $H(B)$ at the nucleus by an amount ΔH amounting to 12–13 kOe at 80 K and 17–22 kOe at 300 K. These values are in agreement with the results reported for the Li–Zn and Li–Ga systems in Refs. 10 and 11. Removal of one Fe^{3+} ion from the immediate environment of $Fe^{3+}(A)$ reduces the local magnetic field $H(B)$ by ~ 13 kOe, which is close to the results reported for the Li–Ti system in Ref. 12. It should be pointed out that at temperatures $T \neq 0$ the value of ΔH should consist mainly of two contributions: firstly, due to the hyperfine magnetic field transferred from the Fe^{3+} ion via oxygen to the central cation (indirect hyperfine interaction field H_{ind}) and, secondly, the temperature-dependent contribution due to the weakening of the exchange interaction on removal of one of the magnetic neighbors of the central ion (equivalent to lowering of the Néel point).

The values of the parameters (Table I) obtained by resolving the spectra of the samples with $x = 0.8$, 0.9, and 0.955 agreed within the limits of the experimental error with those found for $x = 1.0$ if we allowed for the intensities of the individual components. The average values of the fields $\bar{H} = \Sigma P_i H_i$ [here, P_i is the ratio of the area under the ith subspectrum to the total area under the spectrum; P_i represents the probabilities $P(m)$ in Eqs. (1) and (2)], calculated from the data of Table I, are as follows:

$$\bar{H}_A=419 \text{ kOe}, \quad \bar{H}_B=452 \text{ kOe at } 300 \text{ K};$$

$$\bar{H}_A=481 \text{ kOe}, \quad \bar{H}_B=506 \text{ kOe at } 80 \text{ K}.$$

The temperature dependences of the average magnetic hyperfine fields for the A and B sublattices were used to find the Néel temperatures T_N for all the samples (see Sec. 2).

3.1.3. Quadrupole splitting. The quadrupole splitting of the paramagnetic doublet ΔE_p was 0.55 ± 0.01 mm/s at room temperature, in agreement with the values calculated using the model of point charges.[13]

It is worth noting the behavior of the quadrupole shift in the spectra determined below the magnetic ordering temperature. Table I gives the values of ΔE obtained when the spectra were resolved into the components representing different local environments. The weighted average $\overline{\Delta E_A}$ was -0.26 mm/s at 80 K and -0.17 mm/s at 300 K. This behavior of the quadrupole shift can be explained as follows.

The magnetic moment of each domain in lithium aluminum spinels is directed along a specific axis of the $\langle 111 \rangle$ type (which is the easy magnetization axis). The principal electric-field gradient (EFG) axis is just as likely to be oriented along one of the four directions of the $\langle 111 \rangle$ type. Then, in the absence of an external magnetic field each sublattice should have two types of sites differing in respect of the angle α between the EFG axis and the magnetization: for one-quarter of the Fe^{3+} ions we have $\alpha_1 = 0$, whereas for the remaining three-quarter of Fe^{3+}, we have $\alpha_2 = 70°32'$.

Since $\Delta E = \frac{1}{2}\Delta E_0(3\cos^2\alpha - 1)$, where ΔE_0 is the true value of the quadrupole shift and ΔE is the observed value, it follows that in the case of the Mössbauer spectrum one-quarter of the ions is characterized by $\Delta E_1 = \Delta E_0$, whereas in the case of the remaining three-quarters we have $\Delta E_2 = -\Delta E_0/3$. Consequently, the Mössbauer spectrum of such a sample should exhibit two six-line components (sextets), one with positive values of ΔE and the other with negative. In the case of a real spectrum of a polycrystalline sample the pattern is averaged out and we have

$$\overline{\Delta E}=0,25\Delta E_0+0,75(-\Delta E_0/3)=0.$$

When the magnetic moment of the iron ion is tilted from the $\langle 111 \rangle$ axis by an angle θ, the positive part of $\overline{\Delta E}$ decreases and the negative one increases. Therefore, the average value of $\overline{\Delta E}$ becomes negative and its absolute value increases on increase in θ.

Our experimental data thus allow us to draw the following conclusions.

1. The deviation of the average value of $\overline{\Delta E}$ from zero demonstrates that the magnetic moments of the iron ions are

not collinear, i.e., a canted magnetic structure occurs in both sublattices.

2. The reduction in the average value of $\overline{\Delta E}$ on increase in temperature means that the canting angle decreases on increase in temperature, i.e., that thermal vibrations destroy the canted magnetic structure.

3. The difference between the values of ΔE for different local environments (see Table I) indicates that the angle θ varies with the number of the magnetic neighbors.

3.1.4. Magnetic structure. Interesting results were obtained when the Mössbauer spectra were recorded at $T = 300$ K in the presence of an external magnetic field $H_0 = 20$ kOe directed at right-angles to the wave vector $\mathbf{k}_\gamma$ of the gamma rays. It is known that the ratio of the intensities I_i of the lines in the Mössbauer spectrum of a magnetically ordered crystal are in the ratio

$$I_1 : I_2 : I_3 = 3 : q : 1, \qquad q = \frac{4 \sin^2 \beta}{1 + \cos^2 \beta}, \tag{3}$$

where β is the angle between the magnetic moment and $\mathbf{k}_\gamma$. Therefore, the ratio of the intensities of the resonance lines can provide information on the magnetic structure of a crystal.

We shall now consider the two cases when this can be done.

1. In the first case the magnetic moments of the iron ions lie within a cone which makes an angle θ_0 with an external magnetic field $\mathbf{H}_0$ directed perpendicularly to the vector $\mathbf{k}_\gamma$ (Fig. 3). We then have $\cos \beta = AO/OC = \sin \theta \cos \varphi$.

We shall now calculate the average value of $\overline{\cos^2 \beta}$ inside a cone with a vertex angle θ_0:

$$\overline{\cos^2 \beta} = \frac{\int_0^{2\pi} \int_0^{\theta_0} \cos^2 \beta \sin \theta \, d\theta \, d\varphi}{\int_0^{2\pi} \int_0^{\theta_0} \sin \theta \, d\theta \, d\varphi} = \frac{2 - \cos \theta_0 - \cos^2 \theta_0}{6}. \tag{4}$$

Two known special cases follow from Eq. (4):

i) a collinear magnetic structure characterized by $\mathbf{H}_0 \perp \mathbf{k}_\gamma$, $\theta_0 = 0$ and $q = 4$;

ii) an analog of a polycrystalline sample or a homogeneous distribution of the magnetic moments on a sphere: $\theta_0 = \pi$ and $q = 2$. It should be noted that when the magnetic moments are distributed on a hemisphere ($\mathbf{H}_0 \perp \mathbf{k}_\gamma$, $\theta_0 = \pi/2$), we again obtain $q = 2$ as in the case of a polycrystalline sample when $\mathbf{H}_0 = 0$.

2) In the second case all the magnetic moments have the same fixed angle $\theta_0 \neq 0$ (helical magnetic structure). We then have

$$\overline{\cos^2 \beta} = \sin^2 \theta_0 \frac{\int_0^{2\pi} \cos^2 \varphi \, d\varphi}{2\pi} = {}^1\!/_2 \sin^2 \theta_0. \tag{5}$$

In particular, if $\theta_0 = \pi/2$ (when the magnetic moments lie in the XY plane, spiral structure), we find that $\overline{\cos^2 \beta} = \frac{1}{2}$, $\overline{\sin^2 \beta} = \frac{1}{2}$, and $q = 4/3$.

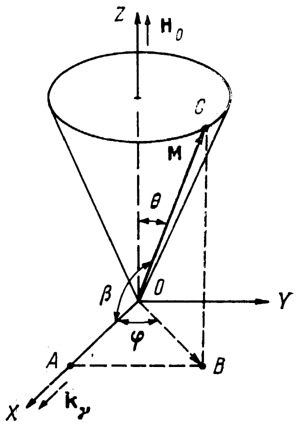

FIG. 3. Mutual orientations of the vectors representing the magnetic moment $\mathbf{M}$ of an iron ion, an external magnetic field $\mathbf{H}_0$, and the wave vector $\mathbf{k}_\gamma$ of gamma rays.

Our experiments in an external magnetic field at $T = 300$ K showed that the values of q found from the ratio of the areas under the Mössbauer lines were approximately equal for all the samples and amounted to 3.8 ± 0.1. Using Eqs. (3)–(5), we then calculated the angle θ_0 for the two cases discussed above.

In case 1, when the angle of tilt of the magnetic moments from the Z axis varies continuously from 0 to θ_0, we obtain $\theta_0 = (18 \pm 5)^\circ$. In case 2, when the angle of tilt of all the magnetic moments relative to the Z axis is the same and equal to θ_0, we obtain $\theta_0 = (13 \pm 4)^\circ$.

These results agree well with the neutron diffraction data reported in Ref. 5, where a sample of $\mathrm{Li_{0.5}Fe_{1.5}Al_{1.0}O_4}$ was investigated at $T = 300$ K and a helical structure with a cone vertex half-angle $\theta = (14 \pm 6)^\circ$ was observed in the B sublattice.

Clearly, in the case of our samples the state was mixed, representing contributions of the variants 1 and 2, because the angle θ varied from 0 to a certain maximum value, depending on the number of the nonmagnetic neighbors in the immediate cation environment. However, we found that this variation is not continuous but discrete, i.e., for each type of the local environment (and there are seven in the B sublattice) there is a specific angle θ. The average value of the angle θ depends on the statistical weight of each local environment configuration and, consequently, it varies with the aluminum concentration. Our value for the average angle θ agrees with the binomial nature of the distribution of the $\mathrm{Al^{3+}}$ ions in the lattice.

This result is not in conflict either with the "spin semiglass" structure proposed by Villain.[14] In the case of this structure the z components of the magnetic moments of the $\mathrm{Fe^{3+}}$ ions at different inequivalent positions are parallel to the external field or to the easy magnetization axis, forming a long-range canted magnetic order, whereas the components normal to the field are distributed at random resembling a spin glass configuration.

3.2. Spectra of the ^{119}Sn nuclei

The Mössbauer spectra of the ^{119}Sn nuclei were found to be split by the magnetic hyperfine interaction in all the samples and they represented strongly broadened sextets with a

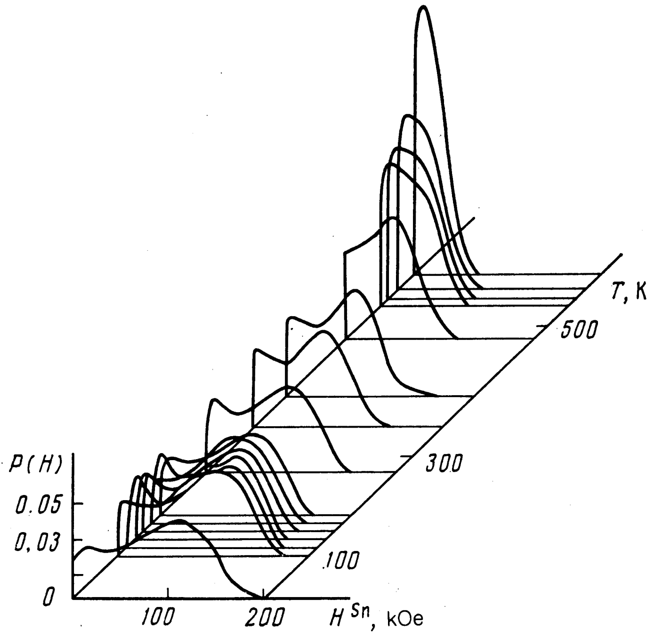

FIG. 4. Distribution functions of hyperfine magnetic fields H^{Sn} obtained at different temperatures in the ferrite $Li_{0.5}Fe_{1.7}Al_{0.8}O_4$:Sn.

single central peak. The isomeric shift δ of the central paramagnetic peak was equal to the isomeric shift of the magnetically split part of the spectrum and amounted to 0.2–0.3 mm/s (relative to SnO_2) at room temperature. The intensity of this peak increased on increase in temperature and on increase in the aluminum content.

3.2.1. Magnetic hyperfine fields at the ^{119}Sn nuclei. The method of statistical regularization[9] was used by us to find the distribution functions $P(H^{Sn})$ of the magnetic fields at the tin nuclei at various temperatures. The nature of the distribution $P(H^{Sn})$ shown in Fig. 4 demonstrated a considerable scatter of the magnetic field intensities at the tin nuclei within the range $0 \leqslant H^{Sn} \leqslant 200$ kOe. It is worth noting a major change in the $P(H^{Sn})$ distribution with temperature and also a considerable "zero-field" contribution to $P(H^{Sn})$.

A detailed analysis of the function $P(H^{Sn})$ was made on the basis of the temperature dependences of the following quantities (Fig. 5): the average field $\overline{H}^{Sn}$, given by the ratio

$$\overline{H}^{Sn} = \sum P(H_i)H_i \Big/ \sum P(H_i)$$

the fields H_1^{Sn} and H_2^{Sn} corresponding respectively to the maximum and minimum of the distribution function $P(H^{Sn})$ (Fig. 5a), and the zero-field contribution $P(0)$ (Fig. 5b). Extrapolation of the $\overline{H}^{Sn}(T)$ curves to 0 K in accordance with the law $H \propto T^{-3/2}$ gives $\overline{H}^{Sn}(0) = 82 \pm 3$ kOe for all the compositions in the range $0.8 \leqslant x \leqslant 1.0$. It is clear from Fig. 5 that the quantities H_1^{Sn}, H_2^{Sn}, and $P(0)$ have singularities in the temperature range 150–200 K.

3.2.2. Temperature anomalies of H^{Sn}. Usually the tin ions occupy the B sites in the spinel structure. It is shown in Ref. 15 that the field H^{Sn} at the tin nuclei in spinels is primarily due to two contributions: a negative one from the intersublattice indirect A–O–B exchange interaction [indirect transfer of the hyperfine magnetic field along the Fe(A)–O–Sn(B) chain] and a positive one due to an intrasublattice direct B–B exchange [due to a direct Sn(B)–Fe(B) overlap of the orbitals of the magnetic and diamagnetic ions]. The competition between these two contributions determines

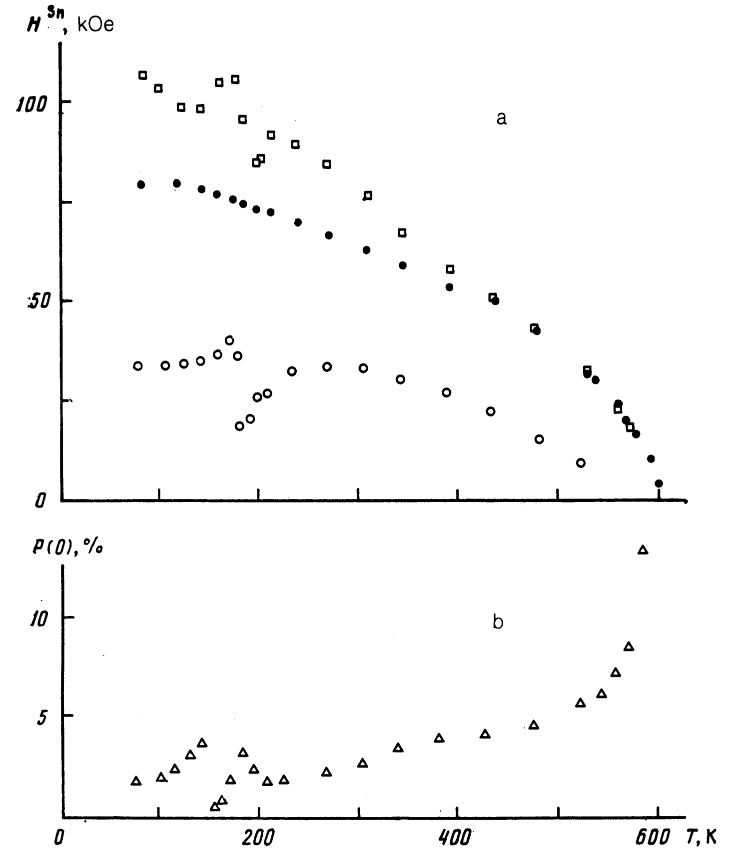

FIG. 5. Temperature dependences of different parameters of the distribution function. a: ● $\overline{H}^{Sn}$; □) H_1^{Sn}; ○) H_2^{Sn}. b: Zero-field contribution $P(0)$.

largely the temperature dependences of the fields at the tin nuclei in spinels.

The temperature dependences of the magnetic moments of the sublattices cannot by themselves give rise to anomalies of $H^{\rm Sn}$ at the compensation point T_c if there is no external magnetic field. Anomalies of the temperature dependences of the distribution parameters $H_1^{\rm Sn}$, $H_2^{\rm Sn}$, and $P(0)$ may be associated only with a rapid variation of the contributions made to the field $H^{\rm Sn}$ by some group of iron ions with a specific local environment. Clearly, the main reason for such anomalies is a change in the angular structure of the magnetic moments of the $\rm Fe^{3+}$ ions in the A and B sublattices. It is shown in several papers (see, for example, Refs. 5, 16, and 17) that in the case of dilute ferrite spinels an increase in temperature reduces the canting angle and at some temperature T_i the canted structure may "collapse" to form a linear one. The point T_i can be regarded as the magnetic phase transition. In the case of a magnetically dilute system in the presence of local regions with different magnetic environments the temperature of such a transition to the collinear magnetic structure will be different for two different groups of atoms so that the transition itself will be smeared out.

In general, such a transition is unrelated to the compensation point and the range of temperatures where the anomalies of $H^{\rm Sn}$ are observed is accidentally close to T_c. However, it is shown in Ref. 6 that in the case of the spinel structure the actual compensation point can be a consequence of the temperature dependences of the canting angles of the sublattice moments.

A detailed investigation of the changes in the distribution $P(H^{\rm Sn})$ due to variation of temperature confirms that the canted structure is transformed into a collinear one. If we consider the function representing the difference between two distributions $P(H^{\rm Sn})$ for a pair in samples with the investigated concentrations x_1 and x_2 but at the same temperature, then in the collinear magnetic structure case we can easily determine theoretically such a function using the binomial distribution of cations around the $\rm Fe^{3+}$ (A) ion and ignoring the B–B interaction. Figures 6 and 7 show the calculated experimental functions of these differences plotted for different compositions and temperatures. We can see that the experimental curve approaches the calculated one only at high temperatures. It therefore follows that the transition from the canted to the collinear structure occurs at these temperatures and it demonstrates once more that the canting angle decreases on increase in temperature.

It is particularly interesting to consider the origin of the zero-field contribution to $P(H^{\rm Sn})$. We shall assume that this contribution is created by two sources: the tin ions with only aluminum ions and/or magnetically disordered iron ions in the adjacent sublattice, and the tin ions surrounded by magnetically ordered $\rm Fe^{3+}$ ions the contributions of which to $H^{\rm Sn}$ due to the A–O–B and B–B exchange interactions balance each other out. The former mechanism is confirmed by the rise of $P(0)$ on increase in the aluminum concentration in the samples. The second mechanism is clearly the reason for the temperature anomalies of $P(0)$.

4. CONCLUSIONS

Our investigations demonstrated that some of the $\rm Fe^{3+}$ ions in $\rm Li_{0.5}Fe_{2.5-x}Al_xO_4$:Sn ferrite spinels are in a magne-

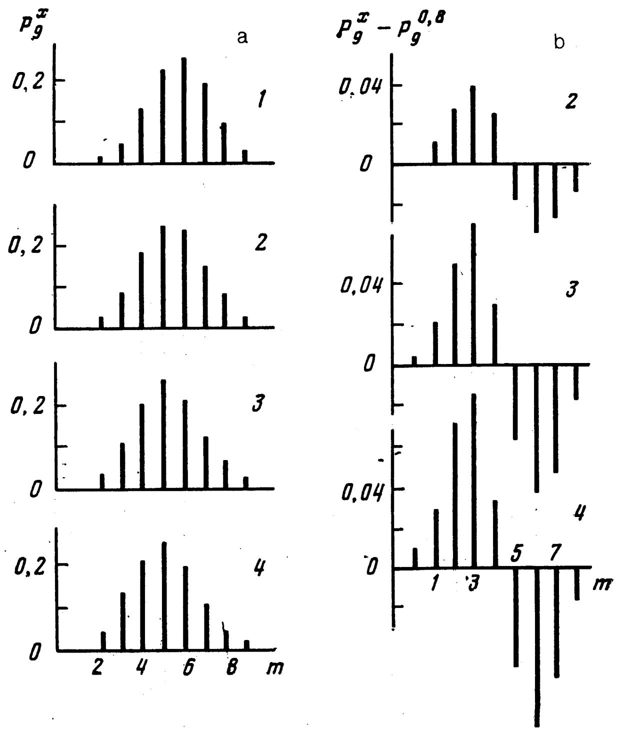

FIG. 6. Distribution functions of the probabilities $P(m)$ obtained for different values of x (a) and the differences between these distributions for pairs of concentrations (b); 1) $x = 0.8$, 2) 0.9, 3) 0.955, 4) 1.0.

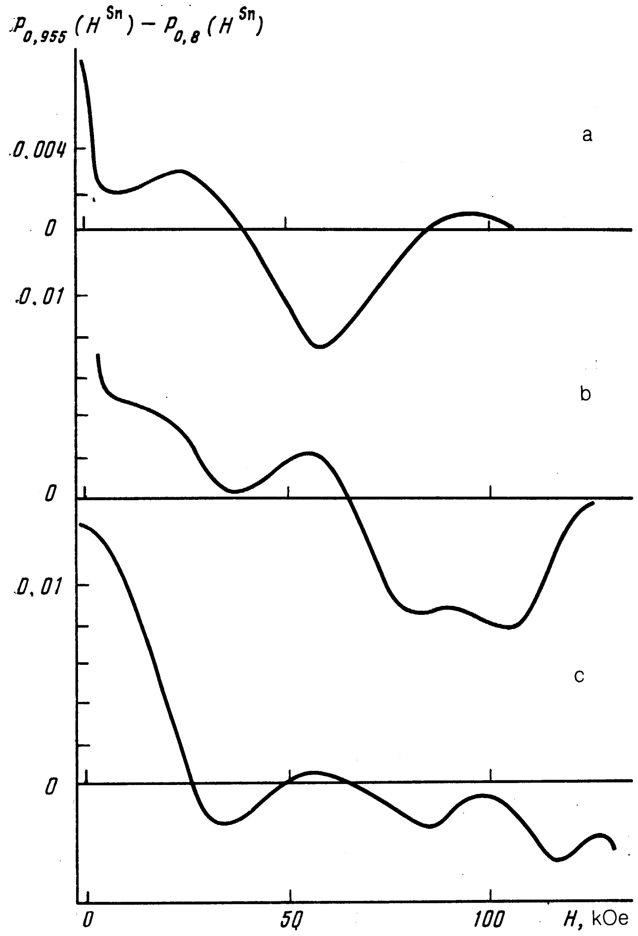

FIG. 7. Experimental curves representing the difference between the distribution functions $P(H^{\rm Sn})_{0.955} - P(H^{\rm Sn})_{0.8}$ at different temperatures: a) $T = 400$ K; b) 200 K; c) 80 K.

tically disordered state at temperatures $T < T_N$. An estimate of the number of these ions confirms the validity of the model of dilute magnetic materials proposed by Gilleo.[8] The Fe^{3+} ions are located at magnetically inequivalent positions, differing in respect of the local environment. We determined the fields H_{ind}, corresponding to a reduction in the magnetic field at the ^{57}Fe nuclei when the nearest Fe^{3+} ion was replaced with Al^{3+}.

The investigated compounds exhibited a canted magnetic structure and it was found that the canting angle θ was a function of temperature and of the magnetic environment. Local regions with a canted structure were distributed at random throughout a crystal and did not form a magnetic lattice. The nature of this canted magnetic structure was determined in greater detail and the average values of the canting angles θ were found for two variants of the structure.

Hyperfine magnetic fields were observed at the ^{119}Sn nuclei and they were characterized by a complex distribution $P(H^{\text{Sn}})$ which depended on temperature and on the aluminum concentration. Temperature anomalies of the dependence $P(H^{\text{Sn}})$ were attributed to a competition of the contributions made to the field H^{Sn} by the magnetic moments of the A and B sublattices and by the transformation of the canted magnetic structure into a collinear one.

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