ELECTRICAL AND MAGNETIC PROPERTIES

Magnetic Phase Transitions in the Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ Alloy: Method of Random Fields of Exchange Interaction

V. I. Belokon'[a] and O. I. Dyachenko[b], *

[a] *Institute of High Technologies and Advanced Materials, Department of Theoretical Physics and Intelligent Technologies, Far Eastern Federal University, Aiax, Russkii Island, Vladivostok, Primorskii Krai, 690922 Russia*
[b] *Institute of High Technologies and Advanced Materials, Department of General and Experimental Physics, Far Eastern Federal University, Aiax, Russkii Island, Vladivostok, Primorskii Krai, 690922 Russia*
**e-mail: dyachenko.oi@dvfu.ru*

Abstract—The magnetic properties of the Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ alloy were studied using the method of random fields of exchange interaction. The critical concentrations, at which a transition from a ferrimagnetic state to a spin-glass state ($x = 0.66$) with a subsequent transition to an antiferromagnetic state ($x = 0.99$), are observed. A magnetic phase diagram, which compares the calculation results with the experiment, was constructed.

INTRODUCTION

Complex compounds based on ferrites and chromites of transition metals with a spinel structure are one of the promising objects of research in condensed matter physics due to a combination of important technical characteristics. For example, the FeCr$_2$S$_4$ compound is known as a ferrimagnet with a resistivity close to that of a semiconductor [1–3], the temperature of magnetic ordering is one of the highest among magnetic semiconductors, and this alloy belongs to multiferroics [4–6] and perovskite manganites [7–9]. The use of these materials opens up great opportunities in various fields of science and technology.

As one of the components of the Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ compound, the FeCr$_2$S$_4$ alloy is a normal cubic spinel with a magnetic moment of $1.6\mu_B$. The magnetic interaction of ions Cr^{3+} with each other is predominantly ferromagnetic. In turn, the interaction of ions Fe^{2+} with ions Cr^{3+} is antiferromagnetic. As follows from experimental studies, the magnetic moment is $4.2\mu_B$ for ion Fe and $2.9\mu_B$ for ion Cr. Saturation magnetization is achieved at $1.6\mu_B$ (2Cr^{3+}–Fe^{2+}) with the implementation of ferrimagnetic ordering. A flat peak in the temperature dependence of magnetization curves, which could be a consequence of the reentrant spin glass state, was observed at 60 K. Another component of the Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ alloy is the Zn$_x$Cr$_2$S$_4$ antiferromagnet ($T_N = 18$ K). This compound has a complex magnetic structure and is characterized by the coexistence of two magnetic phases below the ordering temperature. One of them has a helical structure at temperatures below 15.5 K while the other phase is a collinear antiferromagnet at temperatures below 12 K [10].

As follows from the experimental study [11], the Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ compound can be in ferrimagnetic, spin-glass, and antiferromagnetic states depending on the zinc concentration.

In an attempt to further study the magnetic properties of Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ and its components, we correlate the experimental data with our results obtained by the method of random fields of exchange interaction [12–15].

The method of random fields of exchange interaction is a promising method for studying critical phenomena in various magnetic materials, including alloys with various concentrations of exchange-interacting ions. Most approaches to describing the magnetism of disordered and quasi-disordered media use the assumption of a random distribution of exchange integrals in the Hamiltonian of spin-spin interaction. In this case, the parameters of the distribution function must be consistent with experimental data. Within the framework of the Ising model, this method makes it possible to determine the distribution function of random fields of exchange interaction, the parameters of which are consistent with each other and calculated using the law of interaction of spins (or

magnetic moments of particles, clusters, grains, etc.). This approach also makes it possible to relate the possibility of phase transitions to the concentration of "ferromagnetic" atoms and to calculate critical concentrations.

METHOD OF RANDOM FIELDS OF EXCHANGE INTERACTION

The method of random interaction fields is a method of molecular (effective) field, which is considered as a random variable [12]. The Heisenberg Hamiltonian of a system of interacting particles (ions) has the form:

$$H = -\sum_{i<k} J_{ik} S_i S_k - g\mu_B H_l \sum_i S_i, \qquad (1)$$

where S_i are the components of the spin vector, g is the Lande multiplier, H is the external magnetic field, and μ_B is the Bohr magneton. Since $g\mu_B S_i$ is the magnetic moment of ion m, the classical Hamiltonian can be written as follows:

$$H = -\sum_{i<k} J_{ik} m_i m_k - H_l \sum_i m_i. \qquad (2)$$

The summation in the first term is carried out over all pairs of particles. In the Ising model, the magnetic moment can have only two possible orientations: $+m$ (up) and $-m$ (down). If we assume the possibility of replacing exchange-interacting particles at some lattice sites with "frozen-in" "non-magnetic" impurities with a density of $1 - p$, we obtain a model with dilution across sites. Term $m_i J_{ik} m_k$ can be considered as the energy of magnetic moment m_i in the exchange interaction field of $H_{ik} = J_{ik} m_k$. The molecular field theory (exchange interaction field) considers an individual ion of $m_i \equiv m$ interacting with its environment. If $H = 0$, then

$$H = -\sum_i m_i \sum_k J_{ik} m_k = -\sum_i m_i \sum_k H_{ik} = -\sum_i m_i H_i, \quad (3)$$

where $H_i = \sum_k J_{ik} m_k$ is the total field of exchange interaction on m_i, $\varphi_k = H_{ik}$, and the i index is omitted.

The dependence of the exchange integral on the distance determines the number of terms that should be considered in the sum. This sum is a random variable, the distribution density of which, in some approximation, as shown in our studies [12−15], has the form:

$$W(H, M) = \frac{1}{\sqrt{\pi}B} \exp\left\{-\frac{[H - H_0 M]^2}{B^2}\right\}, \qquad (4)$$

where the average value of $\langle H \rangle = H_0 M$ and the dispersion of $2\sigma^2 = B^2$ are expressed in terms of the p concentration of exchange-interacting particles, the φ_k effective field of exchange interaction produced by an atom with number k, and configurationally and thermodynamically averaged magnetic moment M as follows:

$$H_0 = p\sum_k \varphi_k, \quad B^2 = 2p\left[1 - M^2 p\right]\sum_k \varphi_k^2. \qquad (5)$$

The equation determining the dependence of average magnetic moment M on temperature and the atomic concentration has the form:

$$M = \int \tanh\left[\frac{m_0 H}{kT}\right] W(H, M)\,dH, \qquad (6)$$

where m_0 is the magnetic moment of the atom. $W(H, M)$ is a "smeared" δ-function.

Then, equation (6) will be as follows:

$$M = \frac{1}{\sqrt{\pi}B} \int \tanh\left(\frac{m_0(H + H_0 M)}{kT}\right) \exp\left(-\frac{H^2}{B^2}\right) dH. \quad (7)$$

Simple estimates can be obtained by replacing the Gaussian distribution function with approximate function $f(H)$:

$$f(H) = \begin{cases} 0, & H > B, \quad H < -B, \\ \dfrac{1}{2B}, & -B < H < B. \end{cases} \qquad (8)$$

Examples of the numerical solution of equation (7) with exact and approximate functions, from which it follows that near the phase transition points, where the M values are small, the error in the calculations is insignificant, are given in [13].

For small M values, in this case, we have that

$$M = \frac{1}{2B} \int \tanh\left(\frac{m_0(H + H_0 M)}{kT}\right) dH. \qquad (9)$$

When expanding in a series in terms of a small parameter:

$$\tanh\left(\frac{m_0 H}{kT} + \frac{m_0 H_0 M}{kT}\right) = \tanh(x + y) = \tanh x$$
$$+ (\tanh(xy))' + + \frac{1}{2!}\left(\tanh(xy^2)\right)'' + \frac{1}{3!}\left(\tanh(xy^3)\right)'''. \quad (10)$$

After integrating this expression with allowance for parity $(\tanh(x))'$ and $(\tanh(x))'''$ for M^2, we obtain:

$$M^2 = \frac{3\left(\dfrac{H_0}{B}\tanh\left(\dfrac{m_0 B}{k_B T}\right) - 1\right)}{\dfrac{H_0^3}{Bk^2 T^2}\left(\tanh\left(\dfrac{m_0 B}{kT}\right) - \left(\tanh\left(\dfrac{m_0 B}{kT}\right)\right)^3\right)}. \quad (11)$$

The condition for the appearance of non-zero M is:

$$\frac{H_0}{B}\tanh\left[\frac{m_0 B}{kT}\right] > 1. \qquad (12)$$

An approximate equation for the Curie point is:

$$\frac{H_0}{B}\tanh\left[\frac{m_0 B}{kT_c}\right] = 1. \tag{13}$$

It should be noted that since B depends on M^2, then immediately near the Curie point, we use the B value at $M = 0$.

Obviously, $\frac{H_0}{B}$ must be greater than unity. The, the $\frac{H_0}{B} = 1$ condition determines critical concentration p_c of exchange-interacting ions. In the case of a crystalline ferromagnet and the interaction between particles of only the first coordination sphere, $\varphi_k = f = \text{const}$. Hence,

$$\gamma = \frac{H_0}{B} = \frac{p_c z f}{f\sqrt{2p_c z}}, \tag{14}$$

where z is the number of nearest neighbors. For the case of direct exchange, the $\frac{H_0}{B} < 1$ condition means the disappearance of the percolating cluster. At

$$\frac{H_0}{B} < 1 \tag{15}$$

and T below the paramagnetic Curie point, ordering of the cluster glass type is possible.

As was already noted above, in the case of direct exchange, i.e., exchange only with nearest neighbors, the critical concentration value is close to the values at which the percolating cluster disappears in the percolation theory. Accordingly, long-range order disappears, but short-range order still remains in the clusters. When moving to the molecular field theory, which does not distinguish between long- and short-range order, the Curie point is the temperature of destruction and short-range order as well. As was shown in our study [17], the inverse magnetic susceptibility goes to infinity at the Curie point and at a temperature above the Curie point asymptotically tends to the value that follows from the molecular field theory. The temperature of destruction of short-range order is called the paramagnetic Curie point, which is described in detail in the monograph [18].

At $B \to 0$, to determine the paramagnetic Curie point, we obtain the expression: $\frac{m_0 H}{kT_c} = 1$ that corresponds to the molecular field theory. In turn, the $\frac{H_0}{B}$ ratio for simple lattices does not depend on the exchange integral and is determined only by the z number of nearest neighbors. If the concentration of interacting ions is less than $\rho_c = \frac{2}{z}$, $\frac{H_0}{B} < 1$, the thermodynamically and configurationally average value of magnetic moment M goes to zero. Since $\frac{2}{z}$ are close to
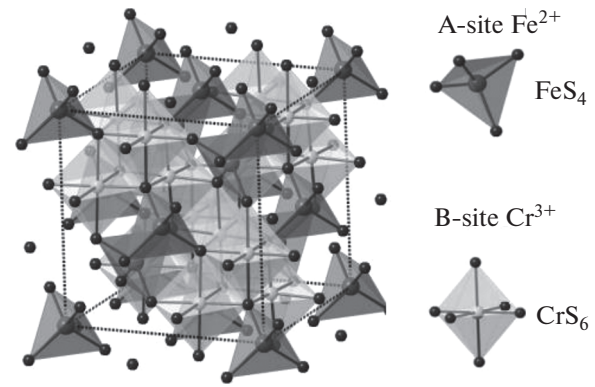
Fig. 1. Scheme of the crystal structure of $FeCr_2S_4$. Fe^{2+}, and Cr^{3+} ions are located at the center of tetrahedral and octahedral S^{2-} cells.

the critical values of the percolation theory, the equality of $M = 0$ can be interpreted as the destruction of the percolating cluster and the transition to the cluster glass phase.

As was shown in our study [14], for a two-sublattice ferromagnet, the expression defining Curie point T_c is more complicated, but in the absence of intra-sublattice interactions, $\rho_c = \frac{2}{z_1}\frac{2}{z_2}$, where z_1 and z_2 are the nearest neighbors of ions from neighboring sublattices.

TYPES OF MAGNETIC ORDER IN THE ALLOY $Fe_{1-x}Zn_xCr_2S_4$

As an example of how random interaction fields work, we can consider the $Fe_{1-x}Zn_xCr_2S_4$ alloy. Its component $FeCr_2S_4$ belongs to the well-known spinel family with the AB_2X_4 formula, where A is a divalent metal cation (Ni^{2+}, Fe^{2+}, Mn^{2+}, Mg^{2+}, Zn^{2+}), B is a trivalent metal cation (Al^{3+}, U^{3+}, Cr^{3+}, Fe^{3+}, Mn^{3+}), and X is anion (F^-, Cl^-, CN^-, Se^{2-}, S^{2-}, Te^{2-}). The spinel crystal has an *fcc* lattice, at the nodes of which sulfur anions forming a dense cubic three-layer packing are located. The spinel structure has two different cationic sublattices: the tetrahedral or A-sublattice (1/8 positions are filled) and the octahedral or B-sublattice (1/2 positions are filled). In normal spinels, Fe^{2+} and Cr^{3+} cations occupy correspondingly tetrahedral A- and octahedral B-interstitial sites (see Fig. 1) [16].

The tetrahedral Fe^{2+} ion in $FeCr_2S_4$ is surrounded by 12 nearby octahedral Cr^{3+} ions. In turn, the octahedral Cr^{3+} ion is surrounded by six nearest tetrahedral Fe^{2+} ions [11]. The number of active neighbors of Cr^{3+} through sulfur ions is four. In turn, three Fe^{2+} ions can be associated with 12 Cr^{3+} ions, i.e., in fact, a chromium ion is affected by three iron ions and three chromium ions. The interaction of chromium ions with each other, as well as chromium ions with iron ions, is
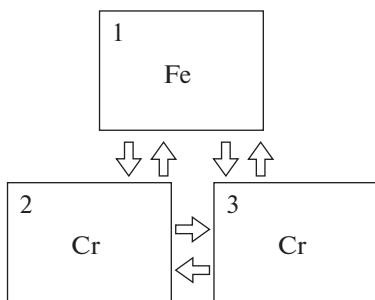
Fig. 2. Scheme of the arrangement of chromium ions in $FeCr_2S_4$.

an indirect exchange through sulfur ions and so it can be assumed that in the case of a significant excess of the intensity of the Fe−Cr exchange interaction over Cr−Cr, the critical value of the density of Fe^{2+} ions is

$$\rho_c = \frac{2}{z_1}\frac{2}{z_2} = \frac{2}{3}\frac{2}{4} = 0.33.$$

The exchange interaction in the Fe sublattice is weak and is suppressed by the much stronger AFM exchange of Fe^{2+}−Cr^{3+}, which leads to ferrimagnetic ordering with interacting Cr^{3+} and Fe^{2+} sublattices at $T = T_N \sim 180$ K [1].

The arrangement of Fe and Cr ions in the $Fe_{1-x}Zn_xCr_2S_4$ alloy is shown in Fig. 2. Arrows indicate interactions between groups of ions. Cr ions are divided into two groups since with a 100% content of non-magnetic Zn ions in the $Fe_{1-x}Zn_xCr_2S_4$ alloy, only the Cr−Cr interaction, which is usually described in terms of two-sublattice models, remains.

Thus, in our model, following formulas (5), for the $Fe_{1-x}Zn_xCr_2S_4$ compound (see Fig. 2), we can write the moments of the distribution function in the following form:

$$H_{21} = z_1 m_1 J_{21}(1-x); \quad H_{31} = z_1 m_1 J_{31}(1-x);$$
$$H_{13} = z_2 m_3 J_{13}; \quad H_{12} = z_2 m_2 J_{12};$$
$$H_{23} = z_1 m_3 J_{23}; \quad H_{32} = z_1 m_2 J_{32};$$
$$B_1 = \sqrt{2\left(z_2(M_2 j_{12})^2 + z_2(m_3 J_{13})^2\right)}; \quad (16)$$
$$B_2 = \sqrt{2\left(z_1(1-x)(m_1 J_{21})^2 + z_1(m_3 J_{23})^2\right)};$$
$$B_3 = \sqrt{2\left(z_1(1-x)(m_1 J_{13})^2 + z_1(m_2 J_{32})^2\right)}.$$

Here, H_{ij} are the fields acting on the ions of the ith group from the ions of the jth group, J_{ij} is the exchange integrals between the ions of groups i and j, B_1 is the dispersion, which determines the fields acting on the Fe ions of the first group from the Cr ions belonging to the second and third groups, B_2 is the dispersion, which determines the fields acting on Cr ions of the second group from Fe ions belonging to the first group and Cr ions of the third group, B_3 is the dispersion, which determines the fields acting on Cr ions of the third group from Fe ions belonging to the first group and Cr ions of the second group, magnetic moments of chromium ions from the third and second groups are $m_3 = m_2 = 3\mu_B = 3 \cdot 927 \times 10^{-26} = J/T$, a magnetic moment of an iron ion from the first group is $m_1 = 4\mu_B = 4 \times 927 \times 10^{-26} = J/T$, $k = 1.38 \times 10^{-23}$ J/K is the Boltzmann constant, $z_1 = 3$, $z_2 = 4$, and x is the degree of substitution of Fe ions ($p = 1 - x$).

The M_1, M_2, M_3 relative magnetic moments corresponding to each group (Fig. 2) can be calculated using the formulas of the random field theory [12]:

$$M_1 = \frac{1}{2B_1}\int_{-B_1}^{B_1} \tanh\left[\frac{m_1}{kT}(H_1 + M_2 H_{12} + M_3 H_{13})\right]dH_1;$$
$$(17)$$
$$M_2 = \frac{1}{2B_2}\int_{-B_2}^{B_2} \tanh\left[\frac{m_2}{kT}(H_2 + M_3 H_{23} + M_1 H_{21})\right]dH_2;$$
$$M_3 = \frac{1}{2B_3}\int_{-B_3}^{B_3} \tanh\left[\frac{m_3}{kT}(H_3 + M_1 H_{31} + M_2 H_{32})\right]dH_3.$$

In a first approximation in M_k, near the phase transition point, the magnetization takes the form:

$$M_1 = \frac{1}{B_1}\tanh\left[\frac{m_1 B_1}{kT}\right](M_2 H_{12} + M_3 H_{13});$$
$$M_2 = \frac{1}{B_2}\tanh\left[\frac{m_2 B_2}{kT}\right](M_1 H_{21} + M_3 H_{23}); \quad (18)$$
$$M_3 = \frac{1}{B_3}\tanh\left[\frac{m_3 B_3}{kT}\right](M_1 H_{31} + M_2 H_{32}).$$

The determinant of the system of equations relative to M_k is equal to:

$$\begin{vmatrix} 1 & -\dfrac{H_{12}}{B_1}\tanh\left[\dfrac{m_1 B_1}{kT}\right] & -\dfrac{H_{13}}{B_1}\tanh\left[\dfrac{m_1 B_1}{kT}\right] \\[2mm] -\dfrac{H_{21}}{B_2}\tanh\left[\dfrac{m_2 B_2}{kT}\right] & 1 & -\dfrac{3}{B_2}\tanh\left[\dfrac{m_2 B_2}{kT}\right] \\[2mm] -\dfrac{H_{31}}{B_3}\tanh\left[\dfrac{m_3 B_3}{kT}\right] & -\dfrac{H_{32}}{B_3}\tanh\left[\dfrac{m_3 B_3}{kT}\right] & 1 \end{vmatrix}. \quad (19)$$
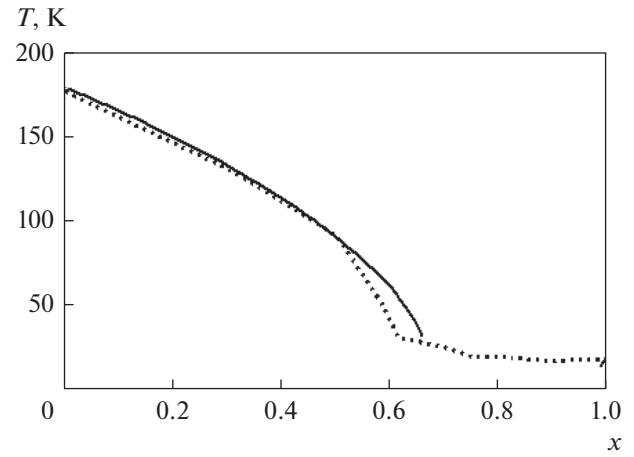
At $T = T_N$, the determinant becomes zero.
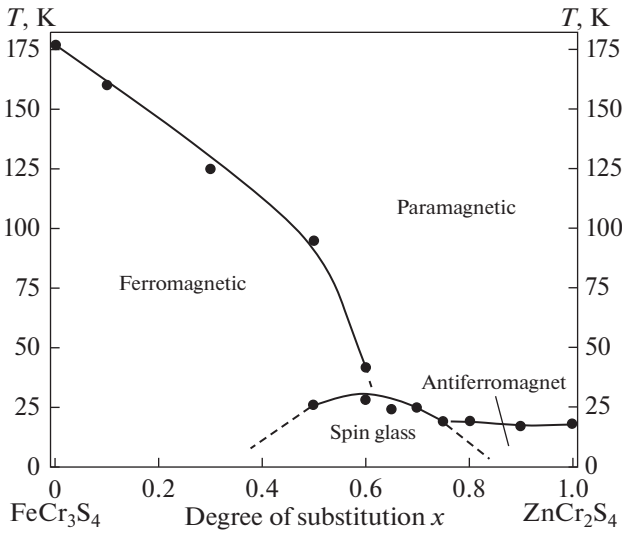
Fig. 3. Magnetic phase diagram of the FeCr$_2$S$_4$–ZnCr$_2$S$_4$ system [11].

Fig. 4. Magnetic phase diagram of the FeCr$_2$S$_4$–ZnCr$_2$S$_4$ system. The dotted line is experimental data [11], and the solid line is the calculation results.

Now we can find the value of exchange integral $J_{23} = J_{32}$ responsible for the interaction between chromium ions of the second and third groups at the experimental Neel temperature of the Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ antiferromagnet at $x = 1$. As follows from Fig. 3 [11], $T_N = 18$ K.

In this case, magnetic Fe^{2+} ions are replaced only by non-magnetic Zn^{2+} ions since they have the same ionic radius (0.83 Å) and so $J_{21} = J_{12} = J_{13} = J_{31} = 0$, then the value of the determinant has the form:

$$1 - \frac{1}{B_2 B_3} H_{23} H_{32} \tanh\left[\frac{B_2 m_2}{kT}\right] \tanh\left[\frac{B_3 m_3}{kT}\right] = 0. \quad (20)$$

When substituting formulas (16) into equation (20), we obtain: $J_{23} = J_{32} = -1.5 \times 10^{23} \, T^2/J$.

As can be seen in Fig. 3, the Neel temperature of the Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ ferrimagnet at $x = 0$ is $T_N = 180$ K. The value of the determinant for the ferrimagnet in this case is:

$$1 - \frac{H_{12} H_{21}}{B_1 B_2} \tanh\left[\frac{m_1 B_1}{kT}\right] \tanh\left[\frac{m_2 B_2}{kT}\right] - \frac{H_{13} H_{31}}{B_1 B_3} \tanh\left[\frac{m_1 B_1}{kT}\right] \tanh\left[\frac{m_3 B_3}{kT}\right]] - \frac{H_{23} H_{32}}{B_2 B_3} \tanh\left[\frac{m_2 B_2}{kT}\right] \tanh\left[\frac{m_3 B_3}{kT}\right]$$
$$- \frac{H_{12} H_{23} H_{31}}{B_1 B_2 B_3} \tanh\left[\frac{m_1 B_1}{kT}\right] \tanh\left[\frac{m_2 B_2}{kT}\right] \tanh\left[\frac{m_3 B_3}{kT}\right] - \frac{H_{13} H_{21} H_{32}}{B_1 B_2 B_3} \tanh\left[\frac{m_1 B_1}{kT}\right] \tanh\left[\frac{m_2 B_2}{kT}\right] \tanh\left[\frac{m_3 B_3}{kT}\right] = 0. \quad (21)$$

From equation (21), the value of the integral of the exchange interaction between iron and chromium ions can be found: $J_{21} = J_{12} = J_{13} = J_{31} = -6.5 \times 10^{23} \, T^2/J$.

Then, by changing the x concentration of zinc ions from 0 to 1 with allowance for the obtained exchange integrals using formula (21), it is possible to plot the dependence of temperature on the x concentration for the Fe$_{1-x}$Zn$_x$Cr$_2$S$_4$ alloy (Fig. 4). As can be seen in the plot, for concentrations x from 0 to 0.66, the compound is in a ferrimagnetic state and the curve that we obtained is consistent with the experimental data. For higher concentrations, the determinant has no solution, which corresponds to spin-glass ordering, and at $x = 0.99$, the alloy becomes an antiferromagnet (which also follows from our calculations).

If we strive $B_1 \to 0$, $B_2 \to 0$, $B_3 \to 0$, i.e., we move to the molecular field theory, and calculate the paramagnetic Curie point from equation (21), we obtain a graph (Fig. 5), from which it can be seen that, in the x
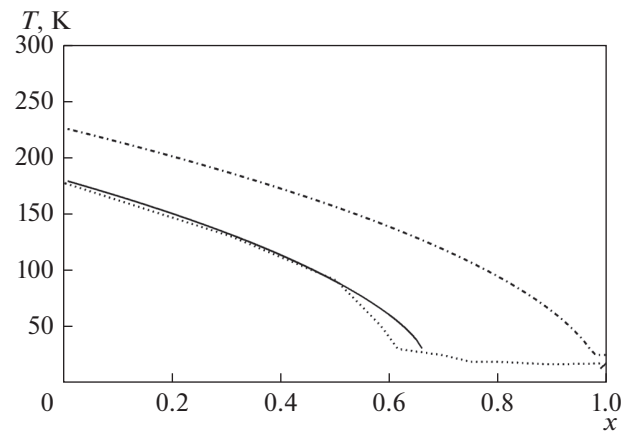
Fig. 5. Magnetic phase diagram of the FeCr$_2$S$_4$-ZnCr$_2$S$_4$ system. The dotted line is experimental data [11], the solid line is the calculation results for the Néel point, and the dotted line with the dot is the calculation results for the paramagnetic point.

interval from 0.66 to 0.99, the long-range order is destroyed, but short-range order is still present.

CONCLUSIONS

Thus, the method of random interaction fields makes it possible to sufficiently accurately describe the dependence of the temperature of the $Fe_{1-x}Zn_xCr_2S_4$ phase transition on the concentration of iron ions and determine the critical concentration corresponding to the transition to the cluster glass state.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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