Current Perspectives

The combination of the random interaction fields' method and the Bethe–Peierls method for studying two-sublattice magnets

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ABSTRACT

In this paper, the concentration magnetic phase transitions are described using the random interaction fields' method in combination with the Bethe–Peierls method. This approach allows us to take into account the correlation between magnetic moments and obtain a more accurate solution for the calculation of the Curie point in comparison with the use of the Kikuchi method, which is considered as the most accurate method in the molecular field theory. As an example, we study the dependence of the Curie point on the concentration of titanium in titanomagnetite. This material is one of the typical natural ferrimagnetics.

1. Introduction

The different variants of the method of the molecular field [1–4] are often used to describe magnetic properties, because the exact solutions to the task connected with the magnetic phase transitions for 3-D lattices are absent. As a rule, these methods give us the Curie point as higher than in reality and do not allow us to describe the concentration of magnetic phase transitions. Earlier, we proposed the random interaction fields' method [5,6] which allows us to describe such transitions, but it also gives us the Curie point as higher than in reality. It is connected with the absence of the accounting of the correlation between the magnetic moments. In this work, in the framework of the random interaction fields' method, we use the idea proposed by Bethe–Peierls [1]. The comparison of the Curie point calculated using the methods of the molecular field is only possible for plane lattice, for which the exact solution was proposed by Onsager [3,7]. It turned out that the method for constructing the distribution function of random interaction fields allows us to obtain new results in the framework of the idea of the molecular field compared with the methods

proposed by Bethe Peierls, Bragg-Williams, Onsager and Kikuchi:

1. This method can be applied to any types of exchange interactions [5,8,9].
2. It allows us to describe the concentration of magnetic phase transitions [10–13].
3. There is a possibility of the account of the variation of the Curie point by reason of the diffusion of magnetic particles [14].

In this paper, we show a way to extend this method to the two-sublattice magnetics. As an example, we study the dependence of the Curie point on the concentration of titanium in titanomagnetite.

2. Random interaction fields' method and Bethe–Peierls method: combination

As known in the framework of the Ising model, the exact solution to the phase transition is obtained only in the case of spin chain. Onsager's solution is also obtained on a plane lattice for direct interaction, from which follows the Curie temperature $T_c = 2.27$ in dimensionless variables (magnetic moment μ, the exchange integral J and the Boltzmann constant k_B equal to unity). In dimensionless variables, in this case the Curie point, $T_c = z$, where z is the number of nearest neighbors. For a plane lattice, $z = T_c = 4$

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(Bragg–Williams method [2]), which is significantly higher than the exact value $T_c=2.27$. The random interaction fields' method developed in our works [5,6] allows us to calculate the density distribution of the molecular field $W(H)$:

$$W(H) = \frac{1}{\sqrt{\pi}B} \exp\left(-\frac{[H - MH_0]^2}{B^2}\right),$$ (1)

where

$$H_0 = p \sum_k \varphi_k,$$ (2)

$$B^2 = 2p[1 - M^2 p] \sum_k \varphi_k^2.$$ (3)

Here, φ_k is the molecular field formed on the selected atom placed at the origin with the magnetic moment with the number k, p is the density of atoms capable of exchange interaction ('ferromagnetic' atoms), and M is the relative magnetic moment per atom after the thermodynamic and configurational averaging.

From the equation

$$M = \int \tanh\left[\frac{\mu H}{kT}\right] W(H)\, dH$$ (4)

it is possible to obtain the condition for the occurrence of a spontaneous magnetic moment,

$$\frac{H_0}{B} \tanh\left[\frac{\mu B}{kT_c}\right] = 1,$$ (5)

where T_c is the Curie point. Consequently, a nonzero value of T_c is possible if $H_0/B > 1$. For direct interaction, when $\varphi_k = J$ for the nearest neighbors and $\varphi_k = 0$ for the other particles, the magnitude $H_0/B > 1$ leads to the condition $p > 2/z$. Thus, the magnitude $p_c = 2/z$ is a critical concentration at which there is a possibility of a spontaneous magnetic moment.

For indirect exchange, which is carried out through the conduction electrons, different types of ordering can be observed: ferromagnetism ($H_0/B > 1$), antiferromagnetism ($H_0/B < -1$) and spin glass state ($H_0/B < 1$).

Note that Eq. (5) determines the Curie point $T_c=3.2$; it is also significantly higher than the exact value. It is connected with the absence of the accounting correlation between the magnetic moments. In the method of the accounting correlation proposed by Bethe and Peierls [1] in the first approximation, the essence of the method is 'immersion' of the magnetic moment and its nearest neighbors (for which the correlation is taken exactly) into the effective field created with other lattice atoms. The partition function S for $z+1$ magnetic moments is the sum of two terms – S_+ and S_- corresponding to the direction of the central spin 'up' and 'down'. The effective field h is considered upward. Then

$$S_+ = \left(\alpha \exp\left[\frac{1}{T}\right] + \frac{1}{\alpha}\exp\left[-\frac{1}{T}\right]\right)^z,$$ (6)

$$S_- = \left(\alpha \exp\left[-\frac{1}{T}\right] + \frac{1}{\alpha}\exp\left[\frac{1}{T}\right]\right)^z,$$ (7)

where $\alpha = \exp[h/T]$. The exchange integral, the magnetic moment and the Boltzmann constant are equal to unity. The relative magnetic moment of the central atom is determined as

$$\mu = \frac{S_+ - S_-}{S},$$ (8)

and for the 'peripheral atom'

$$\mu = \frac{1}{S}\frac{\alpha}{z}\frac{\partial}{\partial\alpha}(S_+ + S_-).$$ (9)

The equality $\mu_0 = \mu_1$ determines the dependence of the effective field h on the temperature. It is easy to show that if $h \to 0$, this equation leads to the equation for the determination of the temperature $T_c \ll 1$, below which $h \neq 0$ is possible:

$$(z - 1)x^2 + 1 = zx,$$ (10)

where $x = \tanh[1/T_c]$, as follows:

$$\tanh\left[\frac{1}{T_c}\right] = \frac{1}{z - 1}.$$ (11)

This result can easily be converted into a form obtained in the framework of the 'quasi-chemical method'. For $z=4$, it follows the Curie point $T_c=3$. It turns out that for the temperature $T > 0$, the effective field is $h > z - 1$. It gives reason to assume that the correlation leads to an effective decrease of the exchange integral, which is approximately $(z - 1)/z$ times. Then the corresponding dimensionless Curie point calculated in Eq. (5) for the standard lattices ($z = 4, 6, 8, 12$) is equal to $(2.31; 4.33; 6.33; 10.33)$. In particular, the result for $z=4$ is closer to the exact solution than that using the method of Kikuchi, which is considered as the most accurate method in the molecular field theory and requires a significant computational work ($T_c = 2.42$), as seen in Tables 1 and 2.

Discrepancies in our data with the results presented in the papers [15,16] explain the different conditions during the Monte Carlo simulation, such as a different number of Monte Carlo steps and the size of systems.

3. Two-sublattice magnets

If there are two sublattices, in accordance with the results presented in [14], we can write the distribution function of random interaction fields on the atoms of the first sublattice as

$$W(H_1) = \frac{1}{\sqrt{\pi}B_1} e^{-(H_1 - M_1 H_{011} - M_2 H_{012})^2/B_1^2},$$ (12)

where

$$B_1^2 = 2p_1(1 - M_1^2 p_1) \sum_k \varphi_{11k}^2 + 2p_2(1 - M_2^2 p_2) \sum_l \varphi_{12l}^2,$$ (13)

$$H_{011} = p_1 \sum_k \varphi_{11k},$$ (14)

$$H_{012} = p_2 \sum_l \varphi_{12l}.$$ (15)

The distribution function of random interaction fields on a selected atom of the second sublattice is

Table 1
Comparison of the Curie points obtained numerically and analytically.

Number of the nearest neighbors z	Curie point (analytical solution)	Curie point (numerical solution)
4	2.31	2.27
6	4.33	4.35
8	6.33	5.6
12	10.33	9.4

Table 2
Comparison of the Curie points obtained using different methods.

Method	Curie point for $z=4$
Bragg Williams	4
Bethe Peierls	2.89
Kikuchi	2.42
Our method	2.31
Onsager	2.27

$$W(H_2) = \frac{1}{\sqrt{\pi} B_2} e^{-(H_2 - M_2 H_{022} - M_1 H_{021})^2 / B_2^2}, \tag{16}$$

where

$$B_2^2 = 2p_2(1 - M_2^2 p_2) \sum_l \varphi_{22l}^2 + 2p_1(1 - M_1^2 p_1) \sum_k \varphi_{21k}^2, \tag{17}$$

$$H_{022} = p_2 \sum_l \varphi_{22l}, \tag{18}$$

$$H_{021} = p_1 \sum_k \varphi_{21k}, \tag{19}$$

index k numbers the atoms of the first type, and l numbers the atoms of the second type.

Here, $\varphi_{11} = m_1 J_1$, m_1 is the magnetic moment per atom in the first sublattice, and J_1 is the exchange integral for the first sublattice atoms. Then $\varphi_{12} = m_2 J_{21}$, m_2 is the magnetic moment per atom in the second sublattice, and $J_{21} = J_{12}$ are the exchange integrals for atoms of different sublattices. For $\varphi_{21} = m_1 J_{12}$, $\varphi_{22} = m_2 J_2$, notation is similar.

$$\alpha_1 - \beta_1 = M_1, \tag{20}$$

$$\alpha_2 - \beta_2 = M_2. \tag{21}$$

Here, φ_{12k} is the exchange 'field' created by the atom number k of the second sublattice on the selected atoms of the first sublattice and placed at the origin, α is the probability of finding the atom oriented 'up', β is the probability of finding the atom oriented 'down' and p is the concentration of ferromagnetic particles. M_1 is the relative magnetic moment for the atoms of the first sublattice, and $M_1 m_1 = \langle m_1 \rangle$ is the average magnetic moment of the atom of the first sublattice. For the magnetic moment M_2 notation is similar. Then, the average moment per atom in the system is determined as $\bar{m} = (\langle m_1 \rangle + \langle m_2 \rangle)/2$.

For the direct exchange,

$$B_1^2 = 2p_1(1 - M_1^2 p_1)(z_1 m_1^2 J_1^2) + 2p_2(1 - M_2^2 p_2)(z_2 m_2^2 J_{12}^2), \tag{22}$$

z_1 is the number of the nearest neighbors of an atom of the first sublattice, z_2 is the number of the nearest neighbors of an atom of the second sublattice, $H_{011} = p_1 z_1 J_1$, $H_{012} = p_2 z_2 J_{12}$,

$$B_2^2 = 2p_2(1 - M_2^2 p_2)(z_2 m_2^2 J_2^2) + 2p_1(1 - M_1^2 p_1)(z_1 m_1^2 J_{21}^2), \tag{23}$$

where $H_{022} = p_2 z_2 J_2$, $H_{021} = p_1 z_1 J_{21}$.

Substantial simplification of the equations can be achieved by replacing the distribution function of the random interaction fields on the 'rectangular function' [6]. Then,

$$M_1 = \frac{1}{B_1} \int_{-B_1}^{B_1} \tanh\left[\frac{m_1}{kT}(H_1 + M_1 H_{011} + M_2 H_{012}) \right] dH_1, \tag{24}$$

$$M_2 = \frac{1}{B_2} \int_{-B_2}^{B_2} \tanh\left[\frac{m_2}{kT}(H_2 + M_2 H_{022} + M_1 H_{021}) \right] dH_2, \tag{25}$$

We introduce the change of variables $(m_1/kT)H_1 = x_1$, $(m_1/kT)H_{011} = h_{011}$, $(m_1/kT)H_{012} = h_{012}$, $m_1 B_1/kT = b_1$, $(m_2/kT)H_2 = x_2$, $(m_2/kT)H_{022} = h_{022}$, $(m_2/kT)H_{021} = h_{021}$, $m_2 B_2/kT = b_2$, and then the equation for the magnetization takes the form:

$$M_1 = \frac{1}{2b_1} \int_{-b_1}^{b_1} \tanh[x_1 + M_1 h_{011} + M_2 h_{012}] \, dx_1, \tag{26}$$

at high temperatures and $M_1 \ll 1$, $M_2 \ll 1$:

$$M_1 = \frac{\tanh(b_1)}{b_1}(h_{011} M_1 + h_{012} M_2) - \frac{1}{3} \frac{\tanh(b_1)}{b_1 \cosh^2(b_1)}(h_{011} M_1 + h_{012} M_2)^3, \tag{27}$$

$$M_2 = \frac{\tanh(b_2)}{b_2}(h_{022} M_2 + h_{021} M_1) - \frac{1}{3} \frac{\tanh(b_2)}{b_2 \cosh^2(b_2)}(h_{022} M_2 + h_{021} M_1)^3, \tag{28}$$

From Eqs. (24) and (25) by integration, we obtain

$$\begin{cases} M_1 = \frac{kT}{2B_1 m_1} \ln \frac{\cosh\left[\frac{m_1}{kT}(M_1 H_{011} + M_2 H_{012} + B_1) \right]}{\cosh\left[\frac{m_1}{kT}(M_1 H_{011} + M_2 H_{012} - B_1) \right]}; \\ M_2 = \frac{kT}{2B_2 m_2} \ln \frac{\cosh\left[\frac{m_2}{kT}(M_2 H_{022} + M_1 H_{021} + B_2) \right]}{\cosh\left[\frac{m_2}{kT}(M_2 H_{022} + M_2 H_{021} - B_2) \right]}. \end{cases} \tag{29}$$

The asymptotic behavior M_1 and M_2 at $M_1 \to 0$ and $M_2 \to 0$ expressed by Eqs. (27) and (28), and in equations for B_1 (13) and B_2 (17), it is possible to drop out M_1^2 and M_2^2. In the opposite case at $M_1, M_2 \to 1$, $T \to 0$ the distribution function of the random interaction fields $W(H)$ tends toward δ-function; therefore, Eqs. (24) and (25) become

$$\begin{cases} M_1 = \tanh\left[\frac{m_1}{kT}(M_1 H_{011} + M_2 H_{012}) \right]; \\ M_2 = \tanh\left[\frac{m_2}{kT}(M_2 H_{022} + M_1 H_{021}) \right]. \end{cases} \tag{30}$$

It should be noted that in the case of the equivalence sublattices and negative value H_{012} the solution of the system $M_1 = -M_2$ is antiferromagnet.

By solving Eqs. (27) and (28), we can obtain the following systems in the zero approximation:

$$\begin{cases} M_1 = \frac{\tanh(b_1)}{b_1}(h_{011} M_1 + h_{012} M_2); \\ M_2 = \frac{\tanh(b_2)}{b_2}(h_{022} M_2 + h_{021} M_1). \end{cases} \tag{31}$$

These homogeneous equations are solvable when the determinant is equal to zero:

$$\left(\frac{\tanh(b_1)}{b_1} h_{011} - 1 \right)\left(\frac{\tanh(b_2)}{b_2} h_{022} - 1 \right) - h_{012} h_{021}\left(\frac{\tanh(b_1)}{b_1} \frac{\tanh(b_2)}{b_2} \right) = 0. \tag{32}$$

By expressing the magnetic moment M_2 through the magnetic moment M_1 (in the zero approximation), we obtain the equation for the determination of the magnetic moment M_1, which has a nonzero solution in case of the change of the sign '+' on the sign

'$-$' and the sign '$-$' on the sign '$+$' of the determinant. The first change in the sign corresponds to the appearance of a nonzero magnetic moment in the sublattice with a large exchange interaction (in this case M_1). The second change of the sign in the absence of the interaction between the sublattices would mean the emergence of the magnetic moment M_2. If there is a weak interaction, the magnetic moment M_2 occurs simultaneously with the magnetic moment M_1 and the second change in the sign corresponds to a sharp growth of the vector M_2 due to the interaction J_{22}. With a strong interaction between sublattices the exchange integral J_{22} is weak and Eq. (32) has only one solution to the temperature of the phase transition T_c, which corresponds to the appearance of the vector M_1.

By substituting the values for h_{011}, h_{012}, h_{021} and h_{022} we obtain the following equation for the determinant:

$$\left(\frac{H_{011} \tanh\left(\frac{m_1 B_1}{kT_C} \right)}{B_1} - 1 \right) \left(\frac{H_{022} \tanh\left(\frac{m_2 B_2}{kT_C} \right)}{B_2} - 1 \right)$$
$$- \frac{H_{012} H_{021}}{B_1 B_2} \tanh\left(\frac{m_1 B_1}{kT_C} \right) \tanh\left(\frac{m_2 B_2}{kT_C} \right) = 0. \tag{33}$$

When the concentration of 'ferromagnetic' atoms p_1 in the first sublattices and the concentration of 'ferromagnetic' atoms p_2 in the second sublattices are reduced, the temperature of the phase transition T_c is also reduced. At the temperature $T_c \rightarrow 0$, we obtain the following expression:

$$\left(\frac{H_{011}}{B_1} - 1 \right) \left(\frac{H_{022}}{B_2} - 1 \right) - \frac{H_{012} H_{021}}{B_1 B_2} = 0. \tag{34}$$

The above equation allows us to estimate the critical concentrations p_1 and p_2, below which the ordering in sublattices and the system as a whole is impossible ($T_c = 0$).

Fig. 1 was obtained using Eq. (34). The phase diagram defines the area of the concentrations p_1 and p_2 at which ordering in a two-sublattice magnet material with the specified parameters $m_1 = 1$, $m_2 = 1.2$, $J_1 = 1$, $J_2 = 2$, $J_{12} = -0.5$ is possible.

4. Titanomagnetite

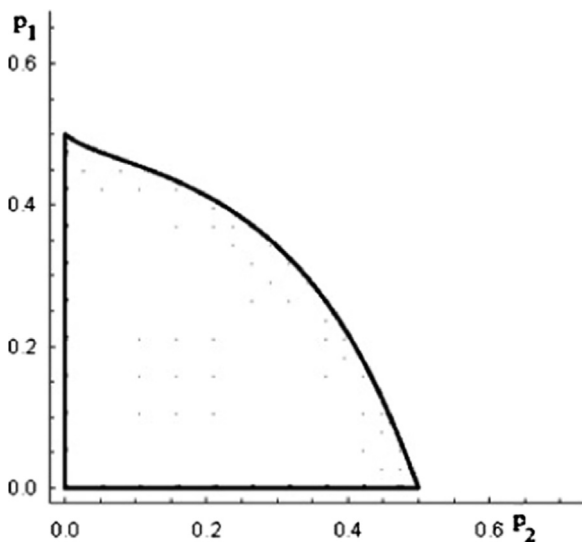The approach formed above can be applied to titanomagnetite.

Fig. 1. The phase diagram defines the area of the concentrations p_1 and p_2 at which ordering in a two-sublattice magnet material with the specified parameters $m_1 = 1$, $m_2 = 1.2$, $J_1 = 1$, $J_2 = 2$, $J_{12} = -0.5$ is possible.

Magnetite has an inverse spinel structure: $(Fe^{3+})[Fe^{2+}Fe^{3+}]O_4$. In octahedral positions (Fe^{3+}), there are only cations with a valence of $3+$. In tetrahedral sites $[Fe^{2+}Fe^{3+}]$ there are twice more cations with a valence of $2+$ or $3+$.

The magnitude Ms of the cation $Fe^{3+}(3d5)$ equals $5\mu_B$, and the cation $Fe^{2+}(3d6)$ – $4\mu_B$ (it is connected with the number of uncompensated electrons in the 3d-electron shell). Titanium ions replace Fe^{3+} sites in tetrahedral positions and make the transition Fe^{3+} in Fe^{2+} in octahedral positions. Thus, actually there are three sublattices: Fe^{3+} in octahedral positions and Fe^{3+} with Fe^{2+} in the tetrahedral positions. Negative exchange interaction is observed between the sublattices 1–2 and 1–3.

Due to the fact that there is no interaction between sublattices, the system of Eqs. (30) and (31) can be easily solved:

$$\begin{cases} M_1 = \frac{\tanh(b_1)}{b_1} (h_{012} M_2 + h_{013} M_3); \\ M_2 = \frac{\tanh(b_2)}{b_2} h_{021} M_1; \\ M_3 = \frac{\tanh(b_3)}{b_3} h_{031} M_1. \end{cases} \tag{35}$$

From Eq. (27):

$$\frac{\tanh\left(\frac{m_1 B_1}{kT} \right)}{\frac{m_1 B_1}{kT}} (h_{012} M_2 + h_{013} M_3)$$

$$- \frac{1}{3} \frac{\tanh\left(\frac{m_1 B_1}{kT} \right)}{\frac{m_1 B_1}{kT} \cosh^2\left(\frac{m_1 B_1}{kT} \right)} (h_{012} M_2 + h_{013} M_3)^3 = M_1. \tag{36}$$

We substitute M_2 and M_3 from the system of Eqs. (35) in Eq. (36). Then

$$M_1^2 = \frac{\dfrac{\tanh\left(\frac{m_1 B_1}{kT} \right)}{\frac{m_1 B_1}{kT}} \left(\dfrac{\tanh\left(\frac{m_2 B_2}{kT} \right)}{\frac{m_2 B_2}{kT}} H_{021} \frac{m_2}{kT} H_{012} \frac{m_1}{kT} + \dfrac{\tanh\left(\frac{m_3 B_3}{kT} \right)}{\frac{m_3 B_3}{kT}} H_{031} \frac{m_3}{kT} H_{013} \frac{m_1}{kT} \right) - 1}{\dfrac{1}{3} \dfrac{\tanh\left(\frac{m_1 B_1}{kT} \right)}{\frac{m_1 B_1}{kT} \cosh^2\left(\frac{m_1 B_1}{kT} \right)} \left(\dfrac{\tanh\left(\frac{m_2 B_2}{kT} \right)}{\frac{m_2 B_2}{kT}} H_{021} \frac{m_2}{kT} H_{012} \frac{m_1}{kT} + \dfrac{\tanh\left(\frac{m_3 B_3}{kT} \right)}{\frac{m_3 B_3}{kT}} H_{031} \frac{m_3}{kT} H_{013} \frac{m_1}{kT} \right)^3}. \tag{37}$$

The temperature at which the numerator is zero is the Curie point:

$$\frac{\tanh\left(\frac{m_1 B_1}{kT} \right)}{\frac{m_1 B_1}{kT}} \left(\frac{\tanh\left(\frac{m_2 B_2}{kT} \right)}{\frac{m_2 B_2}{kT}} H_{021} \frac{m_2}{kT} H_{012} \right.$$
$$\left. \frac{m_1}{kT} + \frac{\tanh\left(\frac{m_3 B_3}{kT} \right)}{\frac{m_3 B_3}{kT}} H_{031} \frac{m_3}{kT} H_{013} \frac{m_1}{kT} \right) = 1, \tag{38}$$

where $H_{021} = p_2 z_2 m_2 J_{12}$, $H_{021} = p_1 z_1 m_1 J_1 2$, $H_{013} = p_3 z_3 m_3 J_{13}$, $H_{031} = p_1 z_1 m_1 J_{13}$, $B_1 = \sqrt{2 p_2 z_2 (m_2 J_{12})^2 + 2 p_3 z_3 (m_3 J_{13})^2}$, $B_2 = \sqrt{2 p_1 z_1 (m_1 J_{12})^2}$,
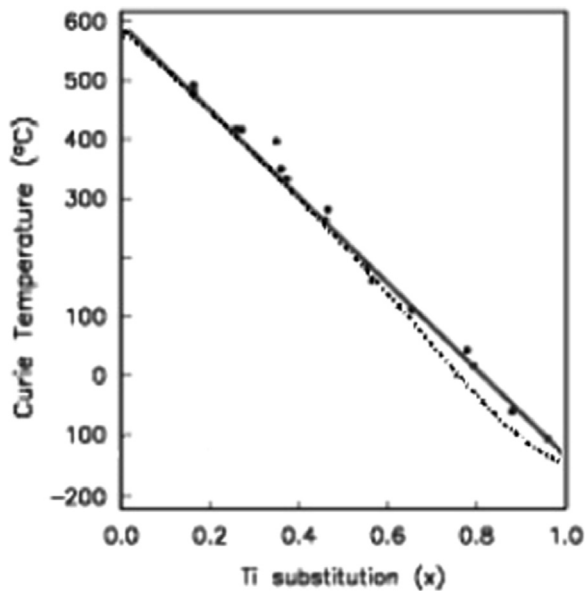
Fig. 2. The dependence of the Curie temperature on the titanium concentration. The solid curve was obtained experimentally [17]; the dashed curve was obtained using the random interaction fields' method.
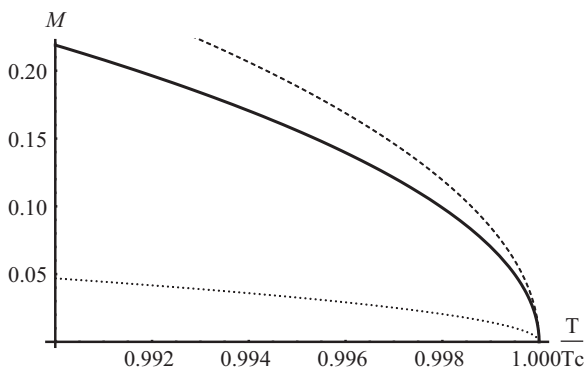
Fig. 3. Graph of dependence M_1 on T/T_c in the immediate vicinity of T_c is shown by means of the black line, $M_2 + M_3$ on T/T_c by means of the dashed line and $(M_2 + M_3) - M_1$ on T/T_c by means of the dotted line at $p=1$.

$B_3 = \sqrt{2p_1 z_1 (m_1 J_{13})^2}$.

The extreme case is considered to find the exchange integral J_{13} when the iron atoms are completely absent in sublattice 2, and in sublattices 1 and 3 there are only the ions Fe^{2+}. It is ulvöspinel. Using the Curie temperature of ulvöspinel ($-150\,°C$) and Eq. (5), we obtain the exchange integral $J_{13} = 3.85 \cdot 10^{24}$. At a known value J_{13} and the Curie point of magnetite ($580\,°C$), the exchange integral $J_{12} = 1.66 \cdot 10^{25}$ is calculated.

Fig. 2 shows the dependence of the Curie temperature on the titanium concentration. The solid curve was obtained experimentally [17]; the dashed curve was obtained using the random interaction fields' method. Graph of dependence M_1 on T/T_c in the immediate vicinity of T_c is shown in Fig. 3 by means of the black line, $M_2 + M_3$ on T/T_c by means of the dashed line and $(M_2 + M_3) - M_1$ on T/T_c by means of the dotted line at $p=1$.

5. Conclusion

1. The random interaction fields' method got further development: the accounting of the correlation using the Bethe–Peierls method allowed us to significantly improve the assessment of the Curie temperature for the standard lattices. In particular, for a flat square lattice result was obtained $T_c=2.31$, while the exact solution of Onsager gave us the temperature $T_c=2.28$, and the most accurate, but extremely cumbersome approximate solution of Kikuchi based on the method of the molecular field – $T_c = 2.42$.
2. The two-sublattice magnetic materials with two types of atoms in the sublattices, with different values of ferromagnetic exchange interactions between sublattices, and with the presence of antiferromagnetic interactions between sublattices, have been studied.
3. The theoretical magnetic phase diagram in the plane $p_1 - p_2$, where p_1 and p_2 are the concentration of the exchange interaction of atoms of the first and second sublattices' two-dimensional two-sublattice magnet with specific values of the parameters in the framework of the Ising model, was plotted.
4. The dependence of the Curie temperature on the concentration of titanium in titanomagnetite was investigated. The plots obtained using the random interaction fields' method have turned out in quantitative and qualitative agreement with the graph obtained by means of the experiment.

Acknowledgments

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