



Research article

Magnetic phase transition in an amorphous alloy: The theory of random fields of exchange interaction

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ABSTRACT

In this study, an attempt was made to sequentially calculate the Curie temperature of iron-containing alloys based on the theory of random fields of exchange interaction. This method makes it possible to determine the conditions for the occurrence of ferromagnetism in an amorphous alloy depending on the concentration of exchange-interacting ions, their Holschmidt radius, and the type of crystal lattice of the transition metal.

1. Introduction

In recent years, in condensed matter physics, a research direction related to the production of amorphous metal alloys, whose 70%–85% of composition contains a transition metal and 15%–30% has metalloid, has received a significant development. Among such materials, amorphous alloys based on iron and cobalt are the most promising in terms of practical use and studying the structural features of an amorphous condensed state. The structure of such alloys is characterized by the absence of a strict periodicity inherent in the crystalline structure in the arrangement of atoms, ions, and molecules. The absence of long-range ordering in the arrangement of atoms leads to the realization of a set of physical properties that cannot be obtained in a solid state with a crystalline structure [1–3].

The first amorphous or glassy alloy $\text{Au}_{80}\text{Si}_{20}$, quenched directly from the melt, was obtained by a group of scientists led by Professor P. Duwez in 1960 [4]. It was not stable at room temperature and therefore not suitable for future research. For the next decade there was no progress in this area [5], except for another work by P. Duwez [6] on the preparation and study of the Pd-Si alloy. The main results of the experiment were as follows: (1) the obtained amorphous Pd-Si system is stable at room temperature; the crystallization temperature is about 400; (2) Pd is a transition metal and can be replaced with any other transition metal. The emergence of methods for obtaining continuous amorphous ribbons (in the 1970s, prior to that only small amorphous plates could be obtained [5]) and the possibility of their application to many systems of metal alloys [7] provoked great interest from various scientific groups. The development of rapid solidification technology [8,9] has introduced a rich spectrum of alloys with outstanding properties. Their lasting scientific and technological impact

can be assessed by the constant influx of scientific research devoted to fundamental questions, discovery of new materials, and improvement of technological processes. To date, the achievements of the rapid quenching process include the following: obtaining thin magnetically soft amorphous ribbons, preparing and manufacturing precursors for nanocrystalline alloys, and creating bulk amorphous alloys.

Despite many publications on the research topic [10,11], there are gaps in the explanation of some magnetic properties of amorphous metal alloys, including the behaviour of the Curie point depending on the concentration and type of metalloids. From general considerations, the Curie point of an amorphous iron-based alloy should be lower than that of a crystalline analogue since the number of neighbours in the first and second coordination spheres, which make the main contribution to the field of exchange interaction in the bcc lattice, is greater than in objects with random close packing, for which the average coordination number is $z \approx 12$ [1]. In this study, an attempt was made to sequentially calculate the Curie temperature of iron-containing alloys based on the theory of random fields of exchange interaction [12,13].

We considered the magnetic properties of amorphous alloys based on the following assumptions:

1. For crystalline iron, the field of exchange interaction was created by ions of the first and second coordination spheres. The exchange integrals were determined using the Bethe–Slater curve. The action of the remaining ions was screened by the nearest ones and was not considered.
2. Ions of the first and second coordination spheres in amorphous iron were arranged randomly and occupied a volume determined by their average density.

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2. Distribution density of interaction fields

Following [14] we considered a system of interacting particles randomly distributed over the volume. The projection of the field H_i on the z axis (the axis of symmetry in the Ising model), created at the origin by one arbitrary particle located at a point with coordinate r_i with a magnetic moment \mathbf{m}_i , can be determined by means the law:

$$H_i = \varphi(r_i, \mathbf{m}_i). \quad (1)$$

Given the known distribution of particles over r_i and \mathbf{m}_i , the distribution density of the interaction field on a particle located at the origin of coordinates is a δ -function of the form:

$$\delta \left[H_i - \sum_i \varphi(r_i, \mathbf{m}_i) \right] \quad (2)$$

In turn, the density of the random distribution of particles over the volume V is determined as follows:

$$\begin{aligned} & \frac{dV_1}{V} \cdot \frac{dV_2}{(V-V_0)} \cdot \dots \cdot \frac{dV_N}{(1-NV_0)} = \\ & = \frac{1}{V^N \left(1 - \frac{V_0}{V}\right) \left(1 - \frac{2V_0}{V}\right) \dots \left(1 - \frac{NV_0}{V}\right)} = \\ & = \frac{1}{V^N \left(1 - \frac{V_0}{V} - \frac{2V_0}{V} \dots - \frac{NV_0}{V}\right)} = \\ & = \frac{1}{V^N \left(1 - \frac{V_0}{V} (1 + 2 \dots + N)\right)} = \frac{1}{V^N \left(1 - \frac{N^2 V_0}{2V}\right)} = \\ & = \frac{1}{V^N} \left(1 + \frac{N^2 V_0}{2V}\right) = \frac{1}{V^N} \left(1 + \frac{N}{2} \frac{V_0}{V}\right)^N = \frac{1}{v^N}, \end{aligned} \quad (3)$$

where the relation $(dV_{(i+1)}/V_i)$ is the probability of the particle, with number $(i+1)$ located in the volume element dV_{i+1} at the point with coordinate \mathbf{r}_{i+1} . Magnitude V_0 is own volume particles. The value V determines the volume of the sample. Magnitude N is the number of particles. Here, it is considered that $(NV_0/V) \ll 1$.

The distribution of particles over magnetic moments is determined by the relation $\prod_{i=1}^N \tau(\mathbf{m}_i) d\mathbf{m}_i$. Here, $\tau(\mathbf{m}_i)$ is the distribution density of particles according to the value m_i and the direction of the magnetic moment \mathbf{m}_i : $\vartheta = 0$, $\vartheta = \pi$, where the angle ϑ determines the orientation of \mathbf{m}_i with respect to the z axis. The distribution over the angles φ is uniform with a density of $1/(2\pi)$. Accordingly, $\tau(\mathbf{m}_i)$ can be written as follows:

$$\tau(\mathbf{m}_i) = \frac{1}{m_i^2} \delta(m_i - m_0) \frac{1}{\sin \vartheta} [\alpha_i \delta(\vartheta) + \beta_i \delta(\vartheta - \pi)] \frac{1}{2\pi}, \quad (4)$$

$$\int \tau(\mathbf{m}_i) m_i^2 dm_i \sin \vartheta_i dV_i d\varphi = 1, \quad (5)$$

where α_i is relative probability of upward direction of the magnetic moment, β_i is relative probability of downward direction of the magnetic moment, magnitude m_0 is the magnetic moment per particle, $\alpha_i + \beta_i = 1$.

In the field H_i , the equilibrium values α_i and β_i are defined as follows:

$$\alpha_i = \frac{\exp\left(\frac{m_i H_i}{k_B T}\right)}{\exp\left(\frac{m_i H_i}{k_B T}\right) + \exp\left(-\frac{m_i H_i}{k_B T}\right)}, \quad (6)$$

$$\beta_i = \frac{\exp\left(-\frac{m_i H_i}{k_B T}\right)}{\exp\left(\frac{m_i H_i}{k_B T}\right) + \exp\left(-\frac{m_i H_i}{k_B T}\right)}. \quad (7)$$

where magnitude k_B is the Boltzmann constant and magnitude T is the absolute temperature.

The relative magnetic moment per particle with number i can be determined by the difference $(\alpha_i - \beta_i)$.

The task of determining the thermodynamically and configurationally averaged value of the magnetic moment M is greatly simplified if values α_i and β_i are replaced by their average values:

$$\alpha_i = \alpha, \quad \beta_i = \beta. \quad (8)$$

In this case, the equation for the determination of the particle's average relative magnetic moment will have the following form:

$$M = \int (\alpha - \beta) W(H, \alpha, \beta) dH. \quad (9)$$

Considering the probability of particle distribution over volume and magnetic moment, the distribution density of the random interaction field H can be represented as

$$W(H) = \frac{1}{v^N} \iint \delta\left(H - \sum_{i=1}^N \varphi_i\right) \prod_{i=1}^N \tau(\mathbf{m}_i) d\mathbf{m}_i dV_i. \quad (10)$$

The characteristic function

$$A(\rho) = \int W(H) \exp(i\rho H) dH, \quad (11)$$

is defined as follows:

$$\begin{aligned} A(\rho) &= \frac{1}{v^N} \int \exp\left(i\rho \sum_{i=1}^N \varphi_i\right) dH \cdot \\ &\cdot \iint \delta\left(H - \sum_{i=1}^N \varphi_i\right) \prod_{i=1}^N \tau(\mathbf{m}_i) d\mathbf{m}_i dV_i, \end{aligned} \quad (12)$$

Considering the normalization conditions (5), the characteristic function $A(\rho)$ can be rewritten in the following form:

$$A(\rho) = \left(1 - \frac{n}{N} \int (1 - \exp(i\rho\varphi)) \tau(\mathbf{m}_i) d\mathbf{m}_i dV_i\right)^N, \quad (13)$$

where value $n = N/v$ is the "effective" number of particles per unit volume. In the limit $N \rightarrow \infty$, we have

$$A(\rho) \rightarrow \exp(-F(\rho)), \quad (14)$$

$$W(H) = \int_{-\infty}^{\infty} \exp(-F(\rho)) \exp(-i\rho H) d\rho, \quad (15)$$

$$\begin{aligned} F(\rho) &= n \int dV \int (1 - \exp(i\rho\varphi)) \tau(\mathbf{m}) d(\mathbf{m}) = \\ &= \alpha I_- + \beta I_+, \end{aligned} \quad (16)$$

where $I_{\mp} = n \int_V (1 - \exp(\mp i\rho\varphi)) dV$.

Here, the change in the sign of the field is considered when the magnetic moment rotates from direction α (up) to direction β (down). The structure of $F(\rho)$ is such that the main contribution to the integral Eq. (16) comes from the values of $F(\rho)$ close to zero. Indeed,

$$F(\rho) = i(\alpha - \beta) H_0 \rho - \frac{B^2}{4} \rho^2 + \dots \quad (17)$$

Restricting ourselves to the first three terms in the expansion of the exponential from formula (11), we obtain the following:

$$A(\rho) = \exp\left(-i(\alpha - \beta) H_0 \rho - \frac{B^2}{4} \rho^2\right). \quad (18)$$

Then we have

$$W(H) = \frac{1}{\sqrt{\pi} B} \exp\left(-\frac{(H - H_0(\alpha - \beta))^2}{B^2}\right), \quad (19)$$

where

$$H_0 = n \int \varphi(\mathbf{r}) dV, \quad B^2 = 2n \int \varphi^2(\mathbf{r}) dV. \quad (20)$$

Similar relationships for crystalline ferromagnets are as follows:

$$H_0 = p \sum \varphi_k, \quad B^2 = 2p(1 - m^2 p) \sum \varphi_k^2, \quad (21)$$

where p is the concentration of exchange-interacting ions at the sites of the crystal lattice [12,13]. Near the Curie point, $B^2 \approx 2p \sum \varphi_k^2$.

Thus, the main characteristics of the distribution function H_0 and B are interconnected through the interaction law $\varphi(\mathbf{r})$. As for the exchange interaction of two particles, its energy can be determined as follows: $E_{ij} = -m_i m_j J_{ij} = -m \cdot m J(r_{ij}) = -m \varphi(r_{ij})$.

3. Self-consistent equation for the average relative magnetic moment

Obviously, in the state of thermodynamic equilibrium, the average value of the relative magnetic moment in the Ising model ($M = \alpha - \beta$) is obtained by averaging over the Gibbs distribution and configurations:

$$M = \int \tanh\left(\frac{mH}{k_B T}\right) W(H) dH. \quad (22)$$

Thus, Eq. (22) will have the following form:

$$M = \frac{1}{\sqrt{\pi B}} \int_{-B}^B \tanh\left(\frac{m(H+H_0 M)}{k_B T}\right) \exp\left(-\frac{H^2}{B^2}\right) dH. \quad (23)$$

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

$$f(H) = \begin{cases} 0, & H > B, H < -B, \\ \frac{1}{2B}, & -B < H < B. \end{cases} \quad (24)$$

The article [14] gives examples of the numerical solution of Eq. (23) with exact and approximate functions, from which it follows that near the phase transition points, where the values M are small, the error in the calculations is insignificant. For small values M , in this case we obtain the following:

$$M = \frac{1}{2B} \int_{-B}^B \tanh\left(\frac{m(H+H_0 M)}{k_B T}\right) dH. \quad (25)$$

When expanded in a series in terms of a small parameter $\frac{mH_0 M}{k_B T}$,

$$\begin{aligned} \tanh\left(\frac{mH}{k_B T} + \frac{mH_0 M}{k_B T}\right) &= \tanh(x+y) \\ &= \tanh x + (\tanh(xy))' + \frac{1}{2!} (\tanh(xy^2))'' \\ &+ \frac{1}{3!} (\tanh(xy^3))''' . \end{aligned} \quad (26)$$

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

$$M^2 = \frac{3\left(\frac{H_0}{B} \tanh\left(\frac{mB}{k_B T}\right) - 1\right)}{\frac{H_0^3}{B k_B^2 T^2} \left(\tanh\left(\frac{mB}{k_B T}\right) - \left(\tanh\left(\frac{mB}{k_B T}\right)\right)^3\right)}. \quad (27)$$

The Curie point is determined by the relation

$$\frac{H_0}{B} \tanh\left(\frac{mB}{k_B T_c}\right) = 1. \quad (28)$$

Obviously, the relation H_0/B must be greater than 1. And the condition $(H_0/B) = 1$ determines the critical concentration p_c of exchange-interacting ions. In the case of a crystalline ferromagnet and interaction between particles of only the first coordination sphere, $\varphi_k = f = const$. From here,

$$\gamma = \frac{H_0}{B} = \frac{p_c z f}{f \sqrt{2p_c} z} = 1, \quad (29)$$

$$p = \frac{2}{z}, \quad (30)$$

where z is the number of nearest neighbours.

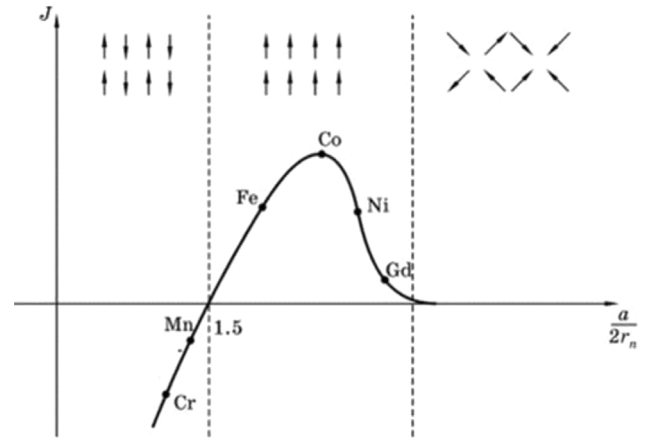


Fig. 1. The Bethe–Slater dependence [16] of the exchange integral J on the ratio of the distance between ion a to the diameter of the unfilled shell $2r_n$.

4. Magnetic phase transition in an amorphous alloy

The greatest difficulty for calculating the exchange interaction fields in an amorphous alloy is the calculation of the exchange integral J between neighbouring atoms (ions) as a function of the distance between them. Perhaps the only exact result was obtained when calculating the exchange interaction energy of an ionized hydrogen molecule [15]. The exchange energy, up to sign, is proportional to the exchange integral and has the form

$$E_0 \sim \frac{1}{r_0} \left(1 - \frac{2}{3} r_0^2\right) \exp(-r_0), \quad (31)$$

where $r_0 = \frac{a}{c}$, c is the radius of the first Bohr orbit, and a is the distance between the nuclei. In general, electrons' interaction energy with each other (overlapping of electron shells) is positive, and the interaction energy of nuclei and electrons is negative. Therefore, a positive value of J will be favoured by an increase in the ratio of the distance between ions in a crystal to the radius of electron shell r_n , although the absolute value of J should decrease in this case. Or the atoms of a ferromagnet must be far enough apart.

As a rule, ferromagnetic materials belong to the group of transition elements that have an unfilled electron s-shell, that is, an electron shell with a large orbital number and a total spin moment that is not equal to zero. Fig. 1 shows the Bethe–Slater dependence [16] of the exchange integral J on the ratio of the distance between ion a to the diameter of the unfilled shell $2r_n$, which qualitatively correctly reflects the dependence of the exchange integral on the distance. Ferromagnetic elements Fe, Co, and Ni have the highest value of the exchange integral. Based on such a scheme, it is possible to explain not only the ferromagnetism of Fe, Co, and Ni but also the antiferromagnetism of alloys, and so forth.

We tried to approximate the Bethe–Slater curve based on formula (31). Assuming the dependence of the exchange integral on the ratio of the distance between ions a to the diameter of the unfilled shell $2r_n$ in the form:

$$J(x) = \frac{e^{-2x}(-1 + \frac{4x^2}{9})}{x}, \quad (32)$$

where $x = a/(2r_n) = a/d$. As can be seen in Fig. 2, function (32) is in good agreement with the Bethe–Slater.

Crystalline iron has a body-centred cubic (bcc) lattice, 2 atoms per cell. Atoms are located at the vertices of the cube, and one atom is in the centre of its volume. A small difference in the distances to the first and second coordination spheres allows us to assume that six ions of the second coordination sphere should also be included among the nearest neighbours, considering the corresponding exchange integral. It is assumed that higher-order coordination spheres do not make a

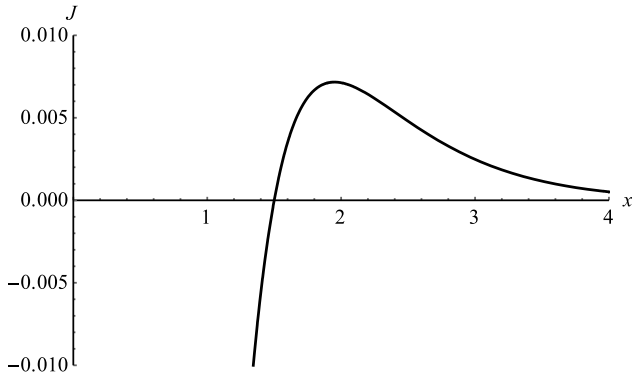


Fig. 2. Dependence of the exchange integral J on the ratio of the distance between ion a to the diameter of the unfilled shell $2r_n$ ($x = \frac{a}{2r_n} = \frac{a}{d}$). The results for function (32) are shown.

significant contribution to the exchange interaction with the central atom. Let us find from Eq. (21) near the phase transition point values H_0 and B for pure iron, limiting ourselves to two coordination spheres, taking into account that for the first coordination sphere $x_1 = \frac{\sqrt{3}}{2} \frac{a_0}{d} = 1.678$ and the second coordination sphere $x_2 = \frac{\sqrt{4}}{2} = \frac{a_0}{d} = 1.936$, the lattice parameter for iron $a_0 = 2.866 \cdot 10^{-10}$ m and the ion diameter for iron $d = 148 \cdot 10^{-12}$ m.

$$H_0 = A \left(8 \cdot \frac{e^{-2x_1} \left(-1 + \frac{4x_1^2}{9} \right)}{x_1} + 6 \cdot \frac{e^{-2x_2} \left(-1 + \frac{4x_2^2}{9} \right)}{x_2} \right), \quad (33)$$

$$B = A \sqrt{16 \left(\frac{e^{-2x_1} \left(\frac{4x_1^2}{9} - 1 \right)}{x_1} \right)^2 + 12 \left(\frac{e^{-2x_2} \left(\frac{4x_2^2}{9} - 1 \right)}{x_2} \right)^2}.$$

Here, parameter A gives the Bethe–Slater function the dimension of exchange field strength. Then, knowing the Curie temperature of pure iron $T_c = 1043$ K, the magnetic moment in Bohr magnetons $m = 2.2\mu_B$, $\mu_B = 927 \cdot 10^{-26}$ J T⁻¹ by formula (28), considering the values from formula (33), we determine $A = 88032$ T. The effective number of nearest neighbours $z = \left(\frac{H_0}{B} \right)^2 \frac{z}{p} = 13.64$. In [17], the curves of dependence $I_s/I_{s0} = f(T)$ of spinning tapes based on Fe are presented. One of these curves, corresponding to iron content of 70%, is shown in Fig. 3.

Using the approach described above, consider an amorphous alloy containing iron in an amount of 70% of the total composition. The volume of the crystal cell in accepted units is . The volume occupied by two ions is, where is the Holschmidt radius and $r = 74 \cdot 10^{-12}$ m is the iron ion radius. The volume fraction occupied by ions is defined as $\frac{NV_0}{V} = \frac{4.93}{7.26} = 0.68$. Thus, the effective density based on formula (3) can be found as $n = 0.7 \frac{NV_0}{V} \left(1 + 0.7 \frac{NV_0}{2V} \right) = 0.59$.

Using formula (20), we determine the moments of the distribution function by integrating over volume:

$$H_0 = A \cdot n \int \varphi(r) dV =$$

$$= 8802 \cdot 0.59 \cdot 4\pi \int_{1.677}^2 e^{-2x} \left(-1 + \frac{4x^2}{9} \right) x dx = 489 \text{ T},$$

$$B = A \sqrt{2n \int \varphi^2(r) dV} = \quad (34)$$

$$= 8802 \sqrt{0.59 \cdot 8\pi \int_{1.677}^2 \left(e^{-2x} \left(-1 + \frac{4x^2}{9} \right) \right)^2 dx} = 241 \text{ T}.$$

The integration limits were selected from $r_{min} = \frac{r_g}{r} = 1.677$ to r_{max} , where the upper limit r_{max} can be found, assuming that 14 ions with a

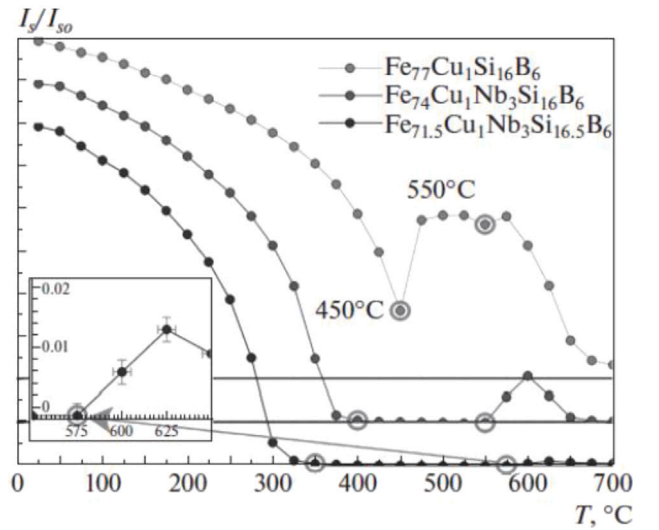


Fig. 3. The curve of dependence $I_s/I_{s0} = f(T)$ of spinning tapes based on Fe [17].

filling density of 0.68 should be placed in the resulting volume:

$$0.68 \cdot \frac{4}{3} \pi (r_{max}^3 - r_{min}^3) = 14. \quad (35)$$

From Eq. (35), it follows that $r_{max} = 2$. From Eqs. (28) and (34), one can determine the Curie temperature of an amorphous alloy with an iron concentration of 70% and a magnetic moment of iron ions, $m = 1.9\mu_B$. We considered that the average magnetic moment of bulk iron changes to $1.9\mu_B$ when passing from a crystalline compound to its amorphous counterpart [18]. Substituting into Eq. (28) the values obtained from formula (34), as well as the calculated parameter A , we find that the Curie temperature in the case of an amorphous alloy was $T_c \approx 570$ K, which is consistent with the result shown in Fig. 3.

It is easy to see that by replacing the concentration by 74% we can obtain a temperature of 645 K from our formulas, which also corresponds to the experimental data.

Note that our approach practically does not consider the effect of metalloids, which contribute to the amorphization of alloys. Therefore, the result obtained can be considered approximate. Using the values H_0 and B obtained by us, we can estimate the “effective number of nearest neighbours” z of iron ions for an amorphous alloy using the ratio $z = \left(\frac{H_0}{B} \right)^2 \frac{z}{p}$. Then, $z = 13.9$, which also agrees with the result obtained experimentally in [17].

5. Conclusion

Thus, the method of random fields of exchange interaction makes it possible to determine the conditions for the occurrence of ferromagnetism in an amorphous alloy depending on the concentration of exchange-interacting ions, their Holschmidt radius, and the type of crystal lattice of the transition metal. From Eq. (28), it follows that the occurrence of ferromagnetism is possible only under the condition $\frac{H_0}{B} > 1$. From a physical point of view, this can be interpreted as the emergence of a leaky cluster. If $0 < \frac{H_0}{B} < 1$, then a cluster spin glass state type structure appears. When $\frac{H_0}{B} < 0$, antiferromagnetic ordering is possible. The Curie point can significantly depend on the magnetic moment of iron ions, which in turn, is determined by the concentration and type of metalloids that make up the alloy. The data known to us indicate that in alloys with an iron ion concentration of 70%–80%, the magnetic moment per atom ranges from $1.2\mu_B$ to $1.9\mu_B$. The effective number of the nearest neighbours at an iron ion concentration of 70% during amorphization turns out to be approximately 13.9, which corresponds to the experimental data.

CRedit authorship contribution statement

V. Belokon: Conceptualization, Methodology, Software. **R. Lapenkov:** Writing – review & editing, Investigation. **O. Dyachenko:** Data curation, Writing – original draft, Investigation.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Dyachenko Olga reports was provided by Far eastern federal. Dyachenko Olga reports a relationship with Far Eastern Federal University that includes: employment.

Data availability

No data was used for the research described in the article.

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