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Role of transition metals in modifying the exchange interaction and anisotropy of amorphous Fe-Si-B alloys

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Abstract: Comprehensive studies of the temperature-dependent properties of amorphous Fe-Co-Si-B-Mn alloys have been conducted using differential scanning calorimetry and induction vibromagnetometry. It is established that the investigated alloys exhibit a two-stage crystallization process; the presence of cobalt in the composition leads to a non-monotonic character of the thermomagnetic curves. The phase transition temperatures, including the Curie temperature, were determined. The spin-wave stiffness constant was calculated using a modified Bloch's law. The research results are important for understanding the effect of cobalt doping on the magnetic properties of iron-based amorphous alloys.

Keywords: amorphous alloys, exchange interaction, random magnetic anisotropy model

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Роль переходных металлов в модификации обменного взаимодействия и анизотропии аморфных сплавов Fe-Si-B

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Аннотация. Проведены комплексные исследования температурно-зависимых свойств аморфных сплавов Fe-Co-Si-B-Mn с использованием методов дифференциальной

сканирующей калориметрии и индукционной вибромагнитометрии. Установлено: исследованные сплавы имеют двухступенчатый процесс кристаллизации; присутствие кобальта в составе приводит к немонотонному характеру термомагнитных кривых. Определены температуры фазовых переходов, в том числе температура Кюри. Расчет спин-волновой константы жесткости проведен с использованием модифицированного закона Блоха. Результаты исследования важны для понимания влияния легирования кобальтом на магнитные свойства аморфных сплавов на основе железа.

Ключевые слова: аморфные сплавы, обменное взаимодействие, модель случайной магнитной анизотропии

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Introduction

In recent decades, research on amorphous metal alloys has become one of the priorities of modern materials science due to their unique physical properties and a wide range of potential applications in various fields of technology [1–3]. Special attention is paid to studying the effect of alloying elements on the magnetic and structural characteristics of amorphous alloys, which opens up new prospects for creating functional materials with specified properties [4, 5].

The purpose of this work is a comprehensive study of the magnetic properties and thermal stability of a series of amorphous Fe-Co-Si-B-Mn alloys with variable cobalt content.

Materials and Methods

Ferromagnetic Fe-Co-Si-B-Mn alloys in the form of ~20 microns thick ribbons obtained by rapid quenching from a melt on a rotating copper disk in an argon atmosphere were studied [6–8]. The analysis of the structure of the samples at the atomic level was carried out using the X-ray diffractometer "KOLIBRI" of JSC "Burevestnik", Ka(Cu) = 1.54 Å, the X-ray profiles were obtained from the free side of the spinning tapes. Alloy compositions $Fe_{78}B_{13}Si_9$, $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$, $Co_{74}Fe_3Mn_3B_{15}Si_5$ determined roentgenofluorescence method (Shimadzu spectrometer EDX-7000, standard qualitative and quantitative analysis using a fixed element – boron). The analysis of the structural relaxation processes in the studied samples was carried out on a differential scanning calorimeter "DSC 404 F1 Pegasus". The measurements were carried out in the temperature range from 300 K to 973 K, and the heating rate was 10 K/s. The magnetic characteristics of the alloys were studied using an induction vibration magnetometer and a PPMS 9T Quantum Design installation.

Results and Discussion

X-ray diffraction analysis of Fe-Co-Si-B-Mn samples with varying degrees of cobalt and manganese doping confirmed their amorphous state, Fig. 1, *a*. The diffraction profiles of the studied samples are characterized by a wide diffuse maximum at an angle of $2\theta \approx 45^\circ$, corresponding to an amorphous matrix with a short-range order. With an increase in the cobalt content, a decrease in the radius of the first coordination sphere is observed, which indicates a decrease in structural disorder, Table 1.

When studying the thermal properties of amorphous Fe-Co-Si-B-Mn alloys by differential scanning calorimetry (DSC), a two-stage crystallization process was recorded for all

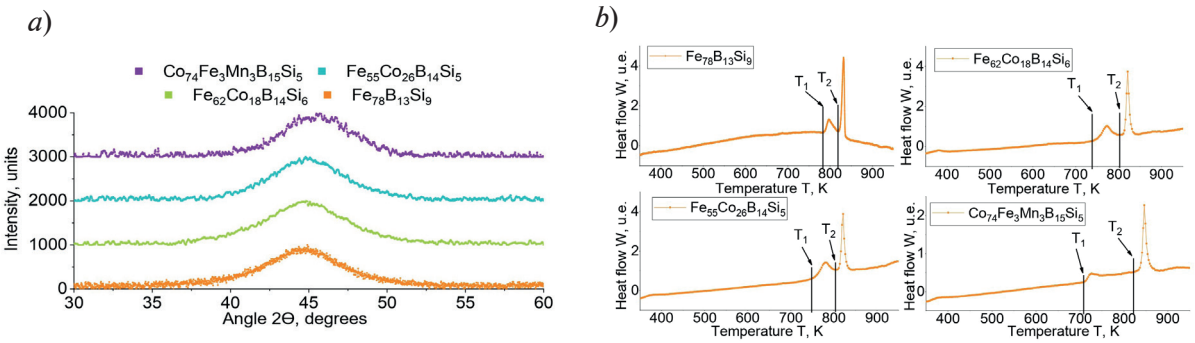

Fig. 1. X-ray diffraction profiles (*a*) and thermograms (*b*) of amorphous alloys $Fe_{78}B_{13}Si_9$, $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$, $Co_{74}Fe_3Mn_3B_{15}Si_5$

samples (Fig. 1, *b*). The first stage is characterized by the precipitation of α-(Fe, Co) nanocrystals Si [11], the second and final one is the crystallization of an amorphous matrix into metallides (FeB, Co_2B).

The complex nature of the structural relaxation of amorphous alloys $Fe_{78}B_{13}Si_9$, $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$, and $Co_{74}Fe_3Mn_3B_{15}Si_5$ was confirmed by an analysis of the temperature dependences of saturation magnetization in Fig. 2, *a*, and the Curie temperatures were determined. Alloy $Fe_{78}B_{13}Si_9$ is characterized by the classical behavior of magnetization with increasing temperature for amorphous alloys [9]. Which made it possible to directly determine the Curie temperature T_C, Table 1. Upon further heating, a paramagnetic plateau is observed before the crystallization of the alloy begins, and the nonmonotonic behavior of saturation magnetization makes it possible to determine the Curie temperature of the crystalline phases. Note: for amorphous alloys $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$, and $Co_{74}Fe_3Mn_3B_{15}Si_5$, the saturation magnetization does not decrease to zero with increasing temperature, so the temperature of the onset of crystallization of these alloys with the loss of magnetic phases is below the Curie temperature T_C for the amorphous state, Fig. 2, *a*.

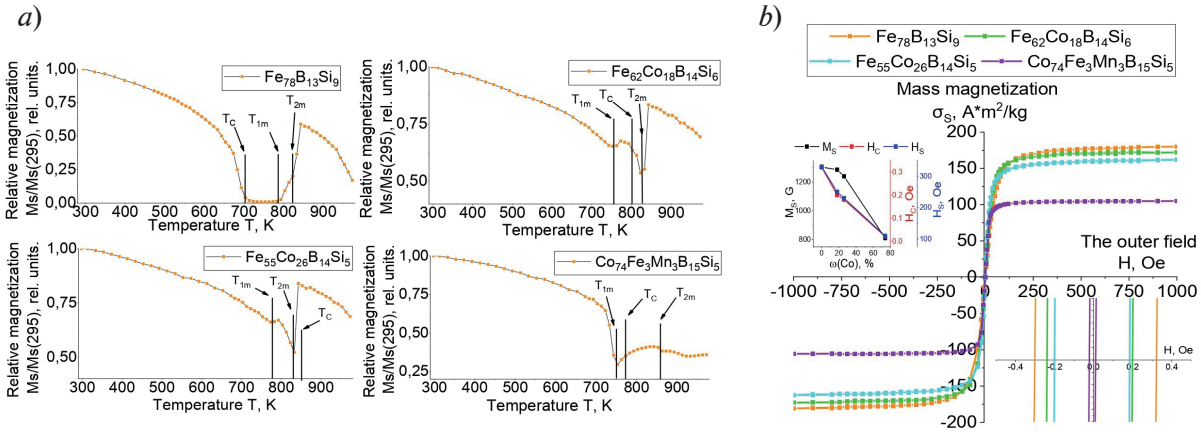

Fig. 2. Dependence of saturation magnetization on temperature (*a*) and hysteresis curves (*b*) for amorphous alloys $Fe_{78}B_{13}Si_9$, $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$, $Co_{74}Fe_3Mn_3B_{15}Si_5$

The determination of phase transition temperatures by the DSC method made it possible to correlate them with the characteristics obtained from thermomagnetic curves for $Fe_{78}B_{13}Si_9$, $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$, and $Co_{74}Fe_3Mn_3B_{15}Si_5$ alloys. Table 1. The difference is due to a complex, multi-stage crystallization mechanism in multicomponent systems, in which non-magnetic phases can also occur in Fe and Co transition metal alloys. Note that the addition of cobalt and an increase in its concentration in the alloy leads to an increase in the Curie temperature, which is associated, as mentioned below, with an increase in the exchange interaction constant. The decrease in T_C for $Co_{74}Fe_3Mn_3B_{15}Si_5$ (to 772 K) is primarily attributed to the significantly reduced iron content (3 at.%), which is the main ferromagnetic component in this

Table 1

Position 2θ, radius of the first coordination sphere R_1, phase transition temperatures T_1, T_2, released energy ΔH, obtained from DSC and vibromagnetometry methods T_{1m}, T_{2m}, T_C

Samples	2θ, deg.	R_1, Å	T_1, K	T_2, K	T_{1m}, K	T_{2m}, K	T_C, K
$Fe_{78}B_{13}Si_9$	44.66	2.49	782	819	787	824	701
$Fe_{62}Co_{18}B_{14}Si_6$	44.78	2.48	734	806	743	821	802
$Fe_{55}Co_{26}B_{14}Si_5$	44.96	2.47	744	804	774	832	861
$Co_{74}Fe_3Mn_3B_{15}Si_5$	45.54	2.44	702	828	748	855	772

system. The presence of Mn with its negative exchange interaction integral further contributes to this reduction, Table 1.

An analysis of the hysteresis curves showed that with an increase in the cobalt content in the amorphous alloys of the Fe-Si-B system, there is a consistent decrease in the saturation magnetization of the M_S and the coercive force, Fig. 2, *b*, Table 2. The alloy $Co_{74}Fe_3Mn_3B_{15}Si_5$ demonstrates the lowest H_C value, which makes it a promising candidate for use in devices operating in alternating magnetic fields [10]. However, its reduced M_S limits its use in highly inductive applications. The $Fe_{62}Co_{18}B_{14}Si_6$ alloy demonstrates the most balanced properties.

Studies of the saturation magnetization dependence on temperature in the range of 50 K–300 K, at a field strength of 1000 Oe (Fig. 3, *a*) revealed differences in the behavior of the samples. For the Fe-Si-B alloy, a standard increase in magnetization is observed with a decrease in temperature; alloys with cobalt showed an abnormal bending of the curves in the region of 150 K.

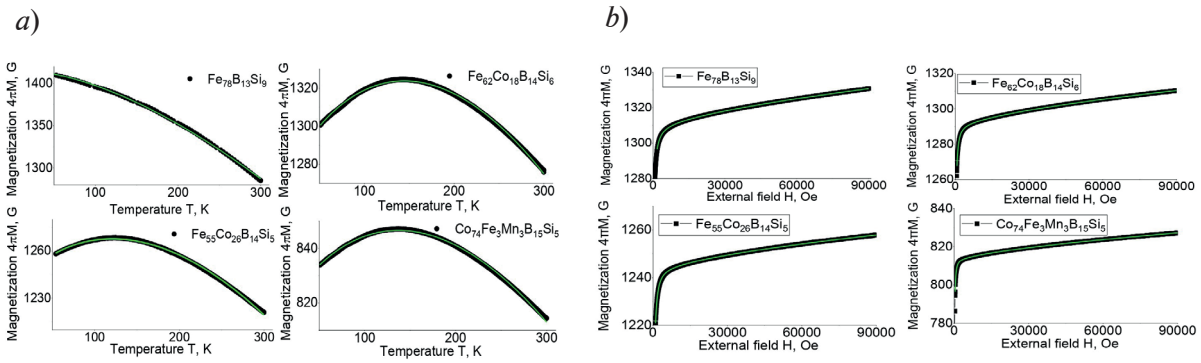

Fig. 3. Dependence of saturation magnetization on temperature in the range (50–300) K (*a*) and dependence of magnetization on the external magnetic field within the limits (*b*) for amorphous alloys $Fe_{78}B_{13}Si_9$, $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$, $Co_{74}Fe_3Mn_3B_{15}Si_5$. The green line shows the approximation curve

To analyze the low-temperature dependences, a modified Bloch function was used, taking into account the Holstein-Primakoff correction [11]:

$$\left(M_S(T) = M_S(0)\cdot\left(1 - B\cdot T^{(3/2)}\right) + C\cdot f_{HP}\cdot T\right), \tag{1}$$

where M_S is the saturation magnetization, B is the Bloch constant, and C is the approximation parameter, f_{HP} is the Holstein-Primakov function

Based on the Bloch constant obtained for amorphous alloys $Fe_{78}B_{13}Si_9$, $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$, $Co_{74}Fe_3Mn_3B_{15}Si_5$, Table 2, the spin-wave stiffness constant D [12] was calculated:

$$D = \frac{k_B}{4\pi}\left(\frac{2.612 g\mu_B\mu_0}{M_0 B}\right)^{\frac{2}{3}},\ A = \frac{D\cdot M_S}{2g\mu_B}, \tag{2}$$

where μ_B is the Bohr magneton, g is the Lande multiplier, and k_B is the Boltzmann constant. These characteristics B and D allowed us to obtain the values of the exchange interaction constant A for the studied alloys, Table 2.

With an increase in the cobalt concentration in the Fe-Si-B matrix, an increase in the spin-wave stiffness constant D is observed, Table 2 and the exchange interaction constant A. In the $Co_{74}Fe_3Mn_3B_{15}Si_5$ alloy, the negative sign of the Mn exchange interaction compensate the increase in the exchange interaction due to cobalt.

Table 2

Parameters of approximation by Bloch's law and exchange interaction constants of the studied amorphous alloys

Samples	$M_S(0)$, G	H_S, Oe	H_C, Oe	B, 10^{-5} K$^{-3/2}$	D, meV·Å^2	A, 10^{-7} erg/cm
$Fe_{78}B_{13}Si_9$	1416	330	0.32	2.48	84.9	4.77
$Fe_{62}Co_{18}B_{14}Si_6$	1257	250	0.2	6.01	80.5	4.46
$Fe_{55}Co_{26}B_{14}Si_5$	1233	230	0.18	4.30	102	5.44
$Co_{74}Fe_3Mn_3B_{15}Si_5$	812	110	0.02	5.69	112	3.90

The analysis of the parameters of random magnetic anisotropy for the amorphous alloys under study was performed based on the dependences of magnetization on the external field in the range from 0 to 90 kOe at room temperature, Fig. 3, *b*.

To analyze the obtained dependences, the classical law of approximation of magnetization to saturation was supplemented by the first term of the Holstein-Primakov function. The modified approximation equation has the following form:

$$M = M_S \cdot \left(1 - \frac{(\alpha \cdot H_a)^2}{H^{\frac{1}{2}}\left(H^{\frac{3}{2}} + H_L^{\frac{3}{2}}\right)}\right) + \chi H + A\sqrt{H}, \quad (3)$$

where H_a is the field of local anisotropy of magnetic correlations; H_L is the correlation field, α is the symmetry coefficient, χ is the high-field magnetic susceptibility, and $\sqrt{H}$ is the first term of the Holstein−Primakov function.

From the obtained approximation parameters, the values of the local anisotropy Klocal and the length of the magnetic correlations L_C are determined according to the following formulas [13], Table 3.

$$K_{local} = \frac{H_a \cdot M_S}{2}, \; L_C = \sqrt{\frac{2A}{M_S \cdot H_L}}. \quad (4)$$

Table 3

Parameters of the approximation of the curves in Fig. 3, *b* using the law of approximation of magnetization to saturation, the constant of random local magnetic anisotropy K_{local}, the length of magnetic correlations L_C for the studied amorphous alloys

Samples	H_a, Oe	M_S, G	H_L, Oe	K_{local}, 10^6 erg/cm^3	L_C, nm
$Fe_{78}B_{13}Si_9$	3428	1304	1917	5.7	8.9
$Fe_{62}Co_{18}B_{14}Si_6$	2016	1286	779	3.3	14.1
$Fe_{55}Co_{26}B_{14}Si_5$	2831	1240	1037	4.5	12.4
$Co_{74}Fe_3Mn_3B_{15}Si_5$	1110	811	309	1.2	34.4

Conclusions

It is shown that magnetically soft amorphous alloys $Fe_{78}B_{13}Si_9$, $Fe_{62}Co_{18}B_{14}Si_6$, $Fe_{55}Co_{26}B_{14}Si_5$ and $Co_{74}Fe_3Mn_3B_{15}Si_5$ in the range of temperatures studied are characterized by a two-stage process of transition to an equilibrium state. The effect of cobalt content on thermal stability shows a non-monotonic behavior. While some compositions exhibit decreased crystallization temperatures, others demonstrate increased thermal stability, as evidenced by T_1 and T_2 values in Table 1. This behavior indicates a complex interplay of structural and compositional factors, which is associated with accelerated structural relaxation and reduced thermal stability of the amorphous phase; there is a decrease in saturation magnetization (~ 800 Gs), as well as a significant decrease in coercive force (0.02 Oe). The $Co_{74}Fe_3Mn_3B_{15}Si_5$ fast-quenched alloy has the minimum H_C, but the lowest M_S value, which limits its use in devices with high magnetic induction. The Curie temperature, T_C ~700 K., has been experimentally determined for the amorphous $Fe_{78}B_{13}Si_9$ alloy. The addition of Co to the basic matrix is accompanied by a decrease in the crystallization temperature and an increase in T_C for the amorphous state.

An analysis of the low-temperature dependence of the magnetization of the studied alloys revealed an abnormal bend in the temperature range of about 150 K. This feature may be due to the specifics of the structure of amorphous alloys, where the variability of interatomic distances is observed, which has a complex effect on the exchange interaction, affecting not only the quantitative characteristics, but also the sign of the exchange integral. The formation of regions with antiferromagnetic exchange interaction is a key factor determining the characteristic feature of low-temperature magnetic dependences in the form of an inflection of the magnetization curve. The calculations performed showed an increase in the spin-wave stiffness constant and an increase in the correlation length for the Co-enriched alloys studied.

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