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Physical Properties of High-Cobalt Amorphous Alloys

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The paper presents the results of a comprehensive study of electrical, thermoelectric, galvanomagnetic and magnetic characteristics in a wide range of temperatures and concentrations to identify the crystallization process of high-cobalt amorphous alloys of $Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$ and $Co_{71.67}Fe_{5.7}Ni_{11.9}Si_{8.23}B_{2.5}$ systems, as well as the effect of crystallization on electrical, galvanomagnetic and magnetic properties.

Key words: Hall coefficient, electrical resistivity, thermo-EMF coefficient, saturation magnetization, amorphous alloys, magnetic ordering, crystallization.

У роботі наведено результати комплексного дослідження електричних, термоелектричних, гальваномагнетних та магнетних характеристик у широкому інтервалі температур та концентрацій для виявлення процесу кристалізації висококобальтових аморфних стопів систем $Co_{59,64}Fe_{5,78}Ni_{23,8}Si_{8,23}B_{2,5}$ та $Co_{71,67}Fe_{5,7}Ni_{11,9}Si_{8,23}B_{2,5}$, а також впливу кристалізації на електричні, гальваномагнетні та магнетні властивості.

Ключові слова: Голлів коефіцієнт, питомий електроопір, коефіцієнт термо-ЕРС, намагнетованість насичення, аморфні стопи, магнетне упорядкування, кристалізація.

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1. INTRODUCTION

Resistivity and thermo-EMF coefficient are measured by the contact method. To study the Hall effect in amorphous alloys and initial crystalline alloys, the method of alternating current and alternating magnetic fields of different frequencies (70 Hz and 50 Hz, respectively) is used. To obtain information on the nature of the structural transformations occurring in amorphous alloys, the temperature dependence of the saturation magnetization of amorphous alloys is investigated on a vibration magnetometer.

The relationship of the anomalous Hall coefficient with saturation magnetization and electrical resistivity is shown.

2. EXPERIMENTAL RESULTS

Partial replacement of cobalt with nickel leads not only to a shift in the temperature range of structural and magnetic orderings, but also changes the absolute values of electrical resistivity ρ, thermo-EMF S and Hall R_S coefficients, magnetization I_S, DTA (differential thermal analysis). In this case, the behaviour of the temperature dependences of physical properties changes significantly even within the amorphous state.

The temperature dependences of the electrical resistivity ρ, the thermo-EMF coefficient S, the Hall coefficient R_S, the saturation magnetization I_S and the DTA curve of the $Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$ alloy are shown in Fig. 1.

A characteristic feature of the $Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$ alloy is a high electrical resistivity, a low thermal resistance coefficient (TRC), which is $\partial\rho/\partial T = 2.27\cdot10^{-10}$ Ω·m/K in the range of 300–630 K. In the range of 630–740 K, the specific electrical resistance ρ practically does not change. At $T = 750$ K, the TRC sign changes, and in the temperature range of 800–900 K, the value of ρ remains constant and is equal to $\rho = 1.65\cdot10^{-6}$ Ω·m. In the range of 900–1050 K, an even greater decrease is observed from $\rho = 1.65\cdot10^{-6}$ Ω·m to the minimum value $\rho = 1.56\cdot10^{-6}$ Ω·m at $T = 1050$ K. Cooling the crystallized sample leads to a decrease in ρ according to the linear-parabolic law in the temperature range 1050–450 K, and in the range of 450–300 K according to the linear law with TRC $\partial\rho/\partial T = 12.6\cdot10^{-10}$ Ω·m/K.

The DTA method is used to determine the crystallization temperature of amorphous alloys upon heating. With continuous heating at a rate of 20 K/min, three regions of heat release are found, which correspond to 800–820–840 K for the $Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$ alloy.

The thermo-EMF coefficient S of the amorphous state takes values from 2.0 μV/K to 4.0 μV/K, and that of the crystallized sample from 3.0 μV/K to 9.0 μV/K. $S(T)$ dependence of the crystallized sample

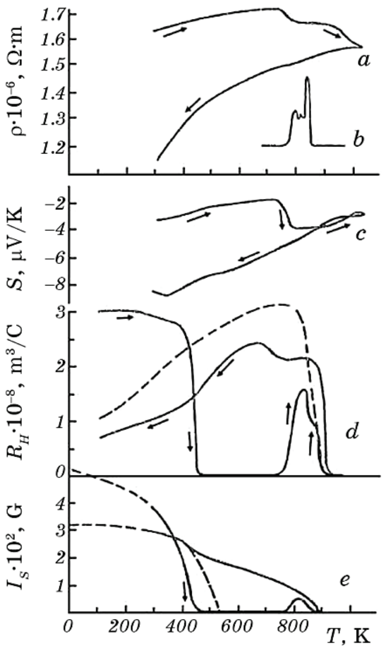

Fig. 1. Temperature dependences of electrical resistivity ρ (*a*), DTA (*b*), thermo-EMF coefficient S (*c*), Hall coefficient R_H (*d*), saturation magnetization I_S (*e*) of the $Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$ alloy.

passes through a minimum at $T = 350$ K.

Hall coefficient R_H in the range of 100–250 K does not depend on temperature, it has a positive sign and is equal to $3.0 \cdot 10^{-8}$ m^3/C. A further increase in temperature leads to a monotonic decrease in the value of R_H, which drops to zero at 450 K. Then an increase in temperature does not change the zero value of R_H up to 750 K. A further increase in temperature leads first to an increase in R_H to a value of $1.6 \cdot 10^{-8}$ m^3/C at $T = 830$ K, and then again to a stepwise decrease to zero at 1000 K. Cooling the crystallized sample first leads to an increase in R_H with maximum values at 840 K and 670 K equal to $2.2 \cdot 10^{-8}$ m^3/C and $2.45 \cdot 10^{-8}$ m^3/K, respectively, and then to a decrease to $R_H = 0.7 \cdot 10^{-8}$ m^3/C at $T = 100$ K. At a temperature equal to $T = 430$ K, the slope of the $R_H = f(T)$ curve changes, *i.e.* in the range of 650–430 K $\partial R_H/\partial T = 5.0 \cdot 10^{-11}$ m^3/(C·K), and in the range of 430–100 K $\partial R_H/\partial T = 1.8 \cdot 10^{-11}$ m^3/(C·K) (Fig. 1). The dependence $R_H = f(T)$ for an individual initial crystalline sample (dashed line) is close to $R_H = f(T)$ for the crystallized sample, but with the only difference that it is smoother.

The temperature dependence of the saturation magnetization $I_S = f(T)$ of the $Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$ alloy also has an anomalous char-

TABLE 1. Values of physical properties of Co–Fe–Ni–Si–B alloys on the concentration of nickel.

Sample composition, state	$\rho\cdot10^{-6}$, $\Omega\cdot$m	I_S, G	T_C, K (amorphous)	T_C, K (crystalline)
$Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$	1.6	3.6	450	910
$Co_{71.67}Fe_{5.7}Ni_{11.9}Si_{8.23}B_{2.5}$	1.05	4.9	570	1000

acter. For the amorphous state, the value of I_S decreases from 398 G at 300 K to zero at 480 K. With further heating of the sample, the value of I_S increases, reaching a maximum value of 55 G at 300 K. At this kinks are observed on the curve $I_S = f(T)$ at 910 and 440 K.

Analysis of the curves $R_H = f(T)$ and $I_S = f(T)$ shows that the Curie temperature T_C of the amorphous and crystallized phases is 450 and 910 K, respectively [1]. The results of studies of $Co_{71.67}Fe_{5.7}Ni_{11.9}Si_{8.23}B_{2.5}$ alloy (with a lower Ni content) are shown in the following Table 1.

It can be seen from the table that a decrease in Ni and an increase in Co in the alloy decreases the absolute value of ρ, increases the magnetization and Curie points of the amorphous and crystalline phases.

Indeed, the magnetic moments per atom of Fe, Co and Ni, respectively, are 2.6, 1.6, and 0.6 μ_B. With a decrease in the Ni concentration in amorphous alloys based on Co–Fe, the electrical resistivity decreases, the magnetization and the Curie point of the amorphous and crystalline phases increase. It is known that in ferromagnets the Hall resistance is the sum of the normal, proportional to magnetic induction B, and the anomalous, proportional to magnetization I_S, parts.

In this case, the Hall resistance has the form:

$$\rho_H = \frac{U_H d}{J} = R_0 B + 4\pi I_S R_S, \tag{1}$$

where J is the current through the sample, d is the thickness of the sample.

In the region $T/T_C < 1$, the magnetization and the Hall coefficient are measured. In view of the fact that $B = 4\pi I_S$, it follows $R_S = \rho_H/4\pi I_S$, where ρ_H is the Hall resistance. Thus, using the temperature dependences of the saturation magnetization $I_S(T)$ in the temperature range $T < T_C$, the temperature dependence of the anomalous Hall coefficient $R_S(T)$ is calculated.

3. RELATIONSHIP BETWEEN PHYSICAL PROPERTIES

It is found in [2, 3] that in the scattering of conduction electrons by magnetic inhomogeneities, including spin waves, the temperature dependences of R_S are proportional to the magnetic contribution to the total resistance. It is shown in [4, 5] that the temperature dependence of the anomalous Hall coefficient not only depends on scattering by spin waves, but also on the scattering of electrons by phonons. Therefore, a comparison is made of the temperature dependence of R_S on I_S^2 and ρ.

Figure 2 shows the dependence of the anomalous Hall coefficient R_S on the square of spontaneous magnetization (upper graph) and the relationship between the temperature-dependent part of the anomalous Hall coefficient $\Delta R_S/R_S$ and the temperature-dependent part of the electrical resistivity $\Delta\rho/\rho$ (lower graph) of the $Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$ alloy in amorphous and crystalline states.

It follows from the graph that in a certain temperature range there is a linear relationship between R_S and the square of spontaneous magnetization, which can be represented as:

$$\Delta R_S = R_S(T) - R_S(T_\mathrm{H}) = \alpha\left[I_S^2(T_\mathrm{H}) - I_S^2(T)\right], \tag{2}$$

where $R_S(T)$ is the anomalous Hall coefficient for the alloy at a certain

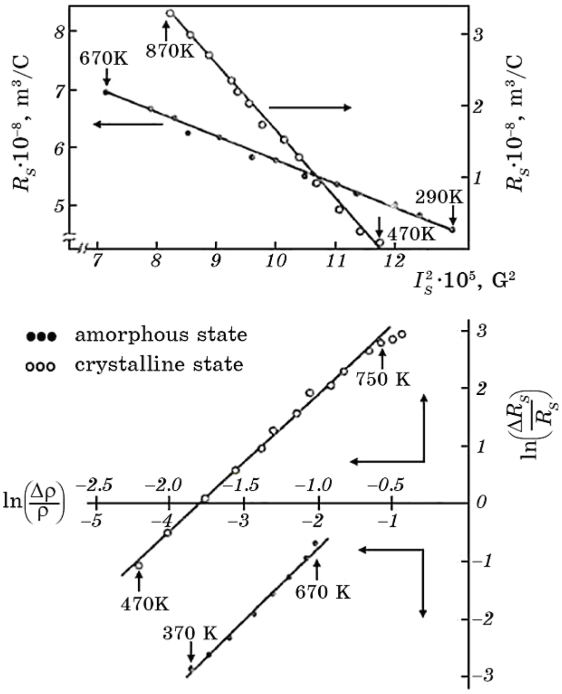

Fig. 2. Dependence of the spontaneous Hall coefficient R_S on the square of spontaneous magnetization I_S^2 (upper graph) and the relationship between the anomalous Hall coefficient R_S and the electrical resistivity ρ (lower graph) of the $Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$ alloy in the amorphous (•••) and crystalline (○ ○ ○) states.

temperature ($T < T_C$) with the corresponding magnetization $I_S(T)$. $R_S(T_H)$ is anomalous Hall coefficient of the same alloy at the initial (room) temperature with magnetization $I_S(T_H)$. Equation (2) characterizes the ferromagnetic fraction of the anomalous Hall coefficient in a certain temperature range, depending on the saturation magnetization. Table 2 shows the values of α calculated using equation (2) (the angle of the $R_S(T)$ and $I_S^2(T)$ dependence for the amorphous and crystalline state).

From Table 2 and Fig. 2 it can be seen that the dependence of R_S on is I_S^2 linear, but in the amorphous state R_S depends on I_S^2 less strongly than in the crystalline state. There is a linear relationship between the anomalous Hall coefficient R_S and the phonon part of the resistivity $\Delta\rho/\rho$, which can be represented as

$$\frac{\Delta R_S}{R_S} = \frac{R_S(T) - R_S(T_H)}{R_S(T_H)} = \beta\left(\frac{\Delta\rho}{\rho}\right)^n. \quad (3)$$

For the studied alloys, the dependence of $\ln(\Delta R_S/R_S)$ on $\ln(\Delta\rho/\rho)$ in the amorphous state is weaker than in the crystalline state, *i.e.* the β coefficient of the amorphous phase is less than that of the crystalline phase. In a certain temperature range, where linear dependences are observed, the degree $n \approx 1$. This indicates that in the kinetic properties of amorphous alloys, phonons and spin waves play a less important role than in transport properties, in comparison with the corresponding crystalline alloys. The absolute value of the anomalous Hall coefficient is usually much higher in metals and alloys in the amorphous state than in the crystalline state. This is probably a direct consequence of the high electrical resistivity in the amorphous state.

It is shown in [3, 6, 7] that for crystalline alloys with asymmetric scattering of spin-polarized *d*-like electrons, the temperature dependence of R_S is described by the relation:

$$R_S = a\rho + b\rho^2, \quad (4)$$

where ρ is the total resistance.

TABLE 2. Values of the coefficient α of equations (2) in amorphous and crystalline states.

Sample composition	Amorphous state		Crystalline state	
	Temperature range T, K	α, 10^{-13} m^3/(C·G^2)	Temperature range T, K	α, 10^{-13} m^3/(C·G^2)
$Co_{59.64}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$	100–300	2.09	100–500	15.02
$Co_{71.67}Fe_{5.7}Ni_{11.9}Si_{8.23}B_{2.5}$	100–300	9.00	100–600	16.5

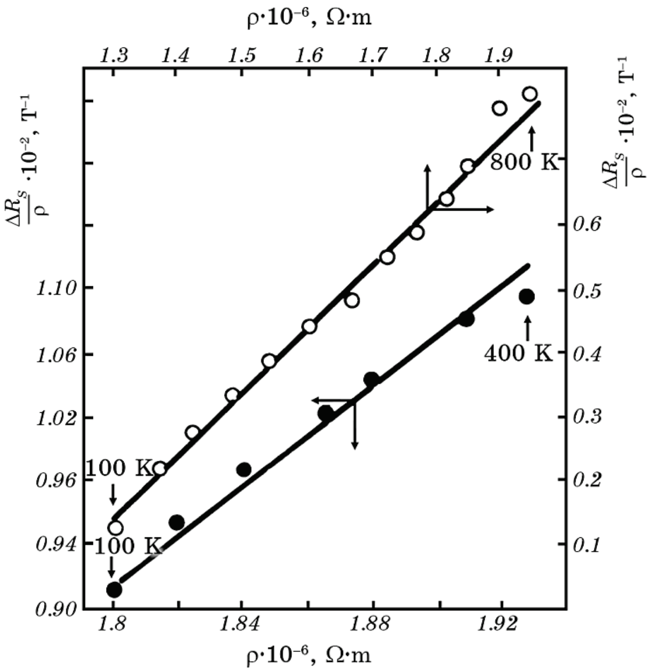

Fig. 3. Dependence of R_S/ρ on the electrical resistivity ρ of the $Co_{71.67}Fe_{5.7}Ni_{11.9}Si_{8.23}B_{2.5}$ alloy in amorphous (•••) and crystalline (○ ○ ○) states.

In [8] it is found that the same relation holds for amorphous alloys, and, as a rule, for highly resistive amorphous alloys, the first term in (4) is less than the second.

We are the first to check the correctness of the theory of the anomalous Hall effect of amorphous ferromagnets [8]. For example, Fig. 3 shows the dependence of R_S/ρ on ρ for $Co_{71.67}Fe_{5.7}Ni_{11.9}Si_{8.23}B_{2.5}$ alloy in the amorphous and crystalline states. It can be seen that the dependence (4) is really well satisfied in the temperature range $T < T_C$.

Table 3 shows the temperature ranges that are fulfilled (4) and the

TABLE 3. Coefficients a and b calculated from Equation (4) for the investigated alloys in the amorphous and crystalline states.

Sample composition	Phase	Temperature range T, K	$a\cdot10^{-2}$, $m^2/(\Omega\cdot C)$	$b\cdot10^{-2}$, $m/(\Omega^2\cdot C)$
$Co_{59.69}Fe_{5.78}Ni_{23.8}Si_{8.23}B_{2.5}$	Amorphous	100–200	–0.014	4.67
	Crystalline	100–400	–0.11	13.17
		500–750	–0.95	75.67
$Co_{71.67}Fe_{5.7}Ni_{11.9}Si_{8.23}B_{2.5}$	Amorphous	100–400	–0.089	6.39
	Crystalline	100–800	–0.049	4.024

numerical values of the coefficients a and b of equation (4).

It is also shown that the ratio R_S and ρ continues to retain during various annealing of both amorphous samples and their crystalline analogues.

Thus, the experimental results obtained in this work are well explained within the framework of the theories developed in [7, 8].

4. CONCLUSIONS

1. A comprehensive study of the temperature dependences of electrical resistivity, thermo-EMF coefficient, and anomalous Hall coefficient, saturation magnetization of amorphous alloys based on metals of the iron group with metalloids in amorphous and crystalline states is carried out.

2. For the investigated alloys in the amorphous and crystalline states, relations are established linking the anomalous Hall coefficient R_S with the saturation magnetization I_S and the temperature-dependent part of the electrical resistivity $\Delta\rho/\rho$ in the form

$$\begin{gathered} \Delta R_S = \alpha\left[I_S^2(T_{\mathrm{H}}) - I_S^2(T)\right], \\ \frac{\Delta R_S}{R_S} = \beta\left(\frac{\Delta\rho}{\rho}\right)^n, \end{gathered} \tag{5}$$

which are carried out in a wide temperature range.

Moreover, the coefficients α and β for the amorphous state of the alloy are smaller than for the crystalline state, which indicates that the phonon contribution and spin waves play a relatively less important role in the kinetic properties of the amorphous state.

The anomalous Hall effect is much more pronounced in alloys in the amorphous state than in the crystalline state, which is a direct consequence of the high electrical resistance in the amorphous state.

3. It is shown that for high-cobalt alloys at the temperature range $T < T_{\mathrm{C}}$, the dependence $R_S = a\rho + b\rho^2$ (ρ is the resistivity) is fulfilled, where the first term is less than the second, which confirms the theory of the anomalous Hall effect of amorphous alloys by Vedyaev and Granovsky.

4. The kinetics and mechanism of magnetic and structural transformations in amorphous alloys based on metals of the iron group with metalloids have been investigated.

It is found that structural transformations occurring in alloys in a certain temperature range lead to changes in physical properties.

It is shown that the dependences $\rho = f(T)$, $S = f(T)$, $R_{\mathrm{H}} = f(T)$, $I_S = f(T)$ are due to the crystallization of the alloys and changes in the short-range order structure.

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