

PACS numbers: 74.43. – f,Kj, 61.43.Er

## RELATIONSHIP OF THE HALL COEFFICIENT WITH SOME PARAMETERS IN AMORPHOUS AND CRYSTALLINE FERROMAGNETS

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*For the series of alloys based on iron metal group the temperature dependences of the Hall coefficient, electrical resistance, saturation magnetization in the amorphous and crystalline states are investigated. Within the Kondorsky-Vedyaev-Granovski theory framework the relationship between anomalous Hall coefficient  $R_s$ , electrical resistivity  $\rho$  and saturation magnetization  $J_s$  was determined. The change mechanism for some physical parameters of amorphous alloys was revealed both in the case of the heat treatment and for the transition to the crystalline state as well.*

**Keywords:** HALL EFFECT, AMORPHOUS METAL ALLOYS, MAGNETIZATION, ELECTRICAL RESISTIVITY, FERROMAGNETIC ALLOYS.

*(Received 19 April 2010, in final form 30 April 2010)*

### 1. INTRODUCTION

The Hall effect is the most sensitive to the magnetic state among the kinetic properties of amorphous metal alloys (AMA). However, in spite of a certain success in the development of the anomalous Hall effect (AHE) theory in amorphous ferromagnets [1-3], information about the experimental study of the galvanomagnetic properties of AMA is rather scanty and contradictory [4-17]. Meanwhile, investigation of the temperature dependence of the Hall effect in amorphous ferromagnets allows to estimate the sign and concentration of the charge carriers, determine the relationship between anomalous Hall coefficient  $R_s$ , electrical resistivity  $\rho$  and saturation magnetization  $J_s$ , and experimentally verify the efficiency of the Kondorsky-Vedyaev-Granovski theory [1-3, 5, 10] as well. This gives the possibility to obtain information about the nature of kinetic phenomena in AMA depending on their composition and structure peculiarities, and also to reveal the change mechanism for some physical parameters both during the heat treatment and at the transition to the crystalline state.

The aim of the present work is to study the temperature dependences of the Hall coefficient  $R_H$ , electrical resistivity  $\rho$  and saturation magnetization  $J_s$  in ferromagnetic alloys based on iron metal group in amorphous and crystalline states and to determine the relationship between these parameters.

## 2. SUBJECTS AND METHODS OF RESEARCH

As the subjects of research we used binary and multi-component amorphous alloys in the form of strips of the thickness of 30  $\mu\text{m}$  and of the width of 12 mm obtained in the same conditions by the quenching from liquid state based on the technique described in [13]. Alloy amorphism was controlled by X-ray analysis using diffractometer DRON-2.0 in  $\text{Fe}_{K\alpha}$ -radiation [13]. Measurements of the temperature dependences of the electrical resistivity  $\rho$  and the Hall coefficient  $R_{\text{H}}$  in the range of 77-1000 K were performed on rectangular samples  $4 \times 12$  mm cut from the central part of amorphous strip using four-probe technique [9-10]. The effective Hall coefficient  $R_{\text{H}}$  was determined by the formula:

$$R_{\text{H}} = \frac{U_{\text{H}}d}{IH} = \frac{R_0B + 4\pi J_s R_s}{H}, \quad (1)$$

where  $U_{\text{H}}$  is the voltage drop;  $d$  is the sample thickness;  $I$  is the current passing through the sample into the magnetic field  $H > H_s$ ;  $R_0B$  is the term corresponding to the usual Hall effect, which follows from the action of the Lorentz force on the charge carriers;  $4\pi J_s R_s$  is the characteristic contribution for ferromagnets connected with the spin-orbit interaction;  $J_s$  is the saturation magnetization.

Since the Hall resistivity  $\rho_{\text{H}}$  can be written in the form

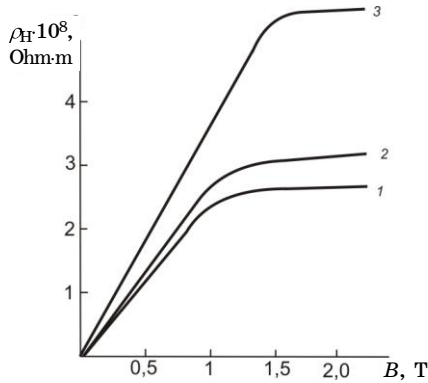
$$\rho_{\text{H}} = R_0B + 4\pi R_s J_s, \quad (2)$$

investigation of the dependence  $\rho_{\text{H}} = f(B)$  allows to separate experimentally the normal  $R_0$  and the anomalous  $R_s$  components of the Hall coefficient:  $R_0$  is determined from the slope of the curve  $\rho_{\text{H}} = f(B)$  at  $B > 4\pi J_s$  and  $R_s$  is found from extrapolation of the curve  $\rho_{\text{H}} = f(B)$  until it intersects with the ordinate axis at  $B = 0$ . Temperature dependences of the saturation magnetization were studied using the vibration magnetometer [10].

## 3. EXPERIMENTAL RESULTS AND DISCUSSION

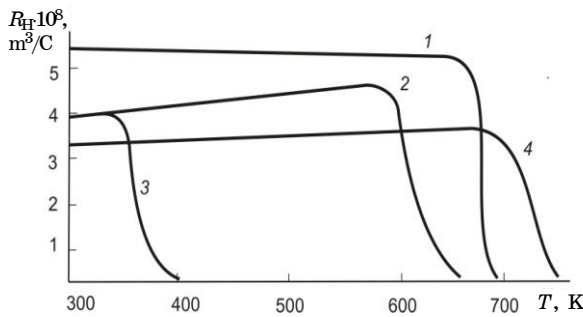
Typical dependences of the Hall resistivity  $\rho_{\text{H}}$  on the external magnetic field for some AMA based on iron are presented in Fig. 1. Note, that partial substitution of iron by nickel decreases the value of the Hall resistivity and shifts the point of inflection on the curves  $\rho_{\text{H}}(B)$  to the region of weaker magnetic fields. For most of AMA based on iron metal group the coefficient  $R_s > 0$  and the normal Hall coefficient  $R_0$  weakly depends on the temperature and almost two orders of magnitude less than  $R_s$ . Thus, at  $T = 300$  K for amorphous  $\text{Fe}_{40}\text{Ni}_{40}\text{P}_{14}\text{B}_6$  alloy the normal component  $R_0 = 6,6 \cdot 10^{-10}$   $\text{m}^3/\text{C}$  and the anomalous component  $R_s = 3,4 \cdot 10^{-8}$   $\text{m}^3/\text{C}$ , and for  $\text{Fe}_{60}\text{Co}_{20}\text{Si}_3\text{B}_{12}$  alloy  $R_0 = 1,1 \cdot 10^{-11}$   $\text{m}^3/\text{C}$  and  $R_s = 4,1 \cdot 10^{-8}$   $\text{m}^3/\text{C}$ .

Charge carrier concentrations for AMA based on iron metal group calculated in the free electron model approximation at 300 K are in the range of  $(8-20) \cdot 10^{27}$   $\text{m}^{-3}$  that coincides with the results of [6, 7]. Radius of the Fermi sphere for most of AMA is  $k_{\text{F}} = (0,16-0,18)$   $\text{nm}^{-1}$ .



**Fig. 1** – Dependence of the Hall electrical resistivity  $\rho_H$  on the induction of the external magnetic field at 300 K:

- 1 –  $\text{Fe}_{80}\text{B}_{20}$ ;
- 2 –  $\text{Fe}_{40}\text{Ni}_{40}\text{P}_{14}\text{B}_6$ ;
- 3 –  $\text{Fe}_{40}\text{Ni}_{40}\text{B}_{20}$



**Fig. 2** – Temperature dependence of the Hall constant of amorphous alloys:

- 1 –  $\text{Fe}_{80}\text{B}_{20}$ ;
- 2 –  $\text{Fe}_{40}\text{Ni}_{38}\text{Mo}_4\text{B}_{18}$ ;
- 3 –  $\text{Fe}_{60}\text{Ni}_{69.8}\text{Si}_{4.7}\text{B}_{3.5}$ ;
- 4 –  $\text{Fe}_{60}\text{Co}_{20}\text{Si}_8\text{B}_{12}$

Temperature dependences in Fig. 2 show that  $R_H$  has the positive sign and depending on the composition it changes in the range of  $(3-6) \cdot 10^{-8} \text{ m}^3/\text{C}$ , and the coefficient  $\partial R_H / \partial T$  for AMA is positive and equal to  $(1-3) \cdot 10^{-11} \text{ m}^3/(\text{C} \cdot \text{K})$ . Amorphous  $\text{Fe}_{80}\text{B}_{20}$  alloy is the exception since  $\partial R_H / \partial T < 0$  for it. It is significant that for crystalline alloys based on iron metal group the absolute values of  $R_s$  are less and  $\partial R_H / \partial T > 0$  are somewhat greater than those for AMA of the same composition and are in the range of  $(4-7) \cdot 10^{-11} \text{ m}^3/(\text{C} \cdot \text{K})$ . Such difference in the values of  $R_s$  and  $\partial R_H / \partial T$  is connected [3] with the enhancement of the spin-orbit interaction as a result of the disorder in the atomic structure of AMA.

Within the diffraction model the authors of [18] obtained an expression for the temperature dependence of the coefficient  $R_s(T)$  of multi-component amorphous ferromagnetic alloys based on  $d$ -metals at  $T < T_C$ . Improper spin-orbit interaction of the spin-polarized  $s$ - or  $d$ -type carriers with localized  $d$ -electrons and scattering on structural (concentration) and magnetic inhomogeneities were considered to be responsible for AHE. Calculation of  $R_s$  was performed starting from the Boltzmann kinetic equation by the iteration method in the small parameter  $\lambda = \tau/t \ll 1$ , where  $\tau$  and  $t$  are the relaxation times of elastic and inelastic scattering processes, respectively. Temperature dependence  $R_s(T)$  has the form

$$R_s = R_s^{(0)} + \alpha(T/T_C)^{3/2} + \beta(T/T_C) + \gamma(T/T_C)^2, \quad (3)$$

where  $\alpha$ ,  $\beta$ ,  $\gamma$  are the coefficients depending on triple and paired correlation function of atom arrangement.

In contrast to crystalline alloys, coefficients  $\alpha$ ,  $\beta$ ,  $\gamma$  in Eq. (3) for AMA have a complex dependence on structural factors. Temperature dependence of the Hall coefficient  $R_H$  for AMA can be a consequence of the carrier activation and hopping (if the Fermi level is close to the mobility edge for  $d$ -electrons), of the temperature dependence of  $s$ - $d$ -hybridization and probability of the interband transitions, difference in the temperature dependences of the mobilities of different charge carriers and other reasons [13]. At  $T < 300$  K temperature dependence of  $R_H(T)$  in nanocrystalline systems can be connected with the presence of the fine crystalline phase. This effect is immaterial at high temperatures where difference in values of  $R_H$  and  $\rho$  for the crystalline and amorphous states [13] decreases. The main reason of the difference in temperature dependences of AHE in the amorphous and crystalline states for alloys of the same composition is the difference in the scattering mechanisms conditioned by the peculiarity of the short-range order systems. Phonon scattering, extrinsic scattering and scattering caused by the thermal spin fluctuations contribute to the total electrical resistivity  $\rho$  in ferromagnetic metals.

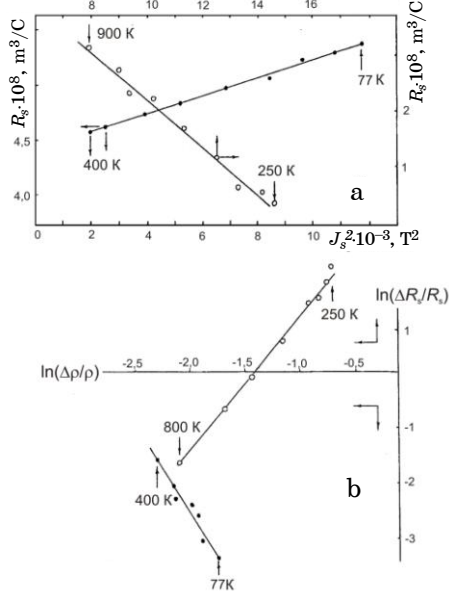
We also note that the effective magnetic moment in AMA calculated per atom of magnetoactive components (Fe, Co, Ni) has somewhat larger values than for crystalline samples. This indicates the formation of the superparamagnetic clusters in AMA during fast cooling from the liquid state. Increase in the local magnetic moments in multi-component nanocrystalline ferromagnets with the decrease in the cooling rate of a melt indicates the increase in the tendency to the formation of the superparamagnetic inhomogeneities [10].

It is established for amorphous ferromagnetic Fe-Co-Si-B and Fe-Co-Mo-B systems that with the increase in the metalloids content in alloys the absolute values of  $R_H$  increase and the quantity  $\partial R_H / \partial T$  decreases. With the increase in the iron content in alloys of these systems the values of  $R_s$  and  $\rho_H$  increase. Substitution of iron by nickel in Fe-Co-Si-B and Fe-Co-Mo-B systems shifts the point of inflection on the curves  $\rho_H = f(B)$  to the region of weaker magnetic fields.

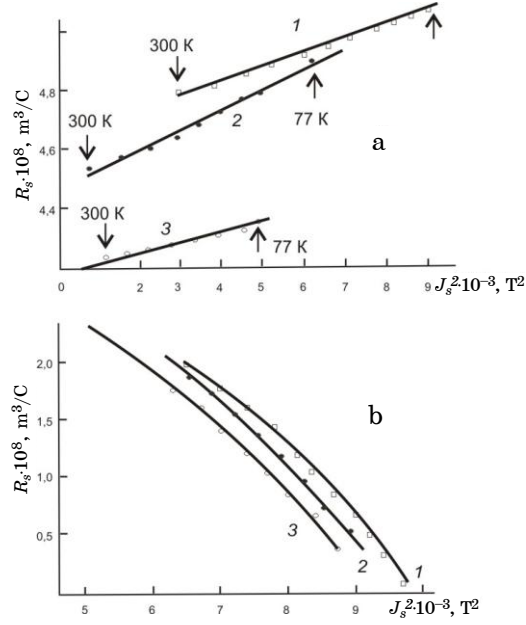
The authors of Ref. [12] studied the Hall effect in amorphous ferromagnetic alloys of Fe-B system with the boron content from 14 to 20 wt %. It is established that in the alloy with the boron content of 17,5 wt % (where the invar properties are most pronounced) at  $T = 300$  K the value of  $R_H$  is equal to  $8,9 \cdot 10^{-8}$  m<sup>3</sup>/C. With the annealing temperature rise, the Hall coefficient  $R_H$  practically does not change until the Curie temperature. At  $T \approx T_c$  the decrease in the value of  $R_H$  by an order of magnitude is observed. These results allow to suggest that the electronic and magnetic states of amorphous alloys of Fe-B systems are the same as those for classical invar alloys of Fe-Ni system.

With the increase in the chromium content in the range of 8-15 wt % in amorphous alloys of Fe-Cr-B system the tendency to decrease in the values of  $R_s$  and  $\rho_H$  is observed. In amorphous alloys of Co-P system with the rise in the phosphorus content within 4-12 wt % the increase in the values of  $R_s$  and  $\rho_H$  is observed. In this case the point of inflection on the curves  $\rho_H = f(B)$  is shifted to the region of weaker magnetic fields [10].

In Figs. 3, 4 we present the typical dependences of the AHE coefficient on the squared saturation magnetization  $J_s$  and show the relationship between the temperature-dependent part of  $R_s$  and  $\rho$  in amorphous and crystalline states of other multi-component ferromagnets based on iron metal group.



**Fig. 3** – Dependences  $R_s = f(J_s^2)$  (a) and  $\ln(\Delta\rho/\rho) = f(\Delta R_s/R_s)$  (b) for  $\text{Fe}_{68.5}\text{Co}_8\text{Cr}_8\text{Si}_5\text{B}_{11}$  alloy in the amorphous ( $\bullet$ ) and crystalline ( $\circ$ ) states



**Fig. 4** – Dependence of the anomalous Hall coefficient  $R_s$  on the squared magnetization  $J_s^2$  for amorphous (a) and crystalline (b) alloys: 1 –  $\text{Fe}_{77}\text{Cr}_8\text{B}_{15}$ ; 2 –  $\text{Fe}_{75}\text{Cr}_{10}\text{B}_{15}$ ; 3 –  $\text{Fe}_{73}\text{Cr}_{12}\text{B}_{15}$

Similar behavior of the dependences  $R_s = f(J_s^2)$  and  $\ln(\Delta\rho/\rho) = f(\Delta R_s/R_s)$  is observed for highly cobalt AMA [13] as well. It follows from the analysis of these dependences that in a certain temperature range, which is typical for each alloy, the linear dependence between  $R_s$  and squared magnetization  $J_s^2$  holds, and it can be represented in the form

$$\Delta R_s = R_s(T) - R_s(T_H) = \alpha_R [J_s^2(T_H) - J_s^2(T)], \quad (4)$$

where  $R_s(T)$  and  $J_s(T)$  are the AHE coefficient and saturation magnetization at  $T < T_C$ , respectively;  $R_s(T_H)$  and  $J_s(T_H)$  are the values of the AHE coefficient and saturation magnetization at the fixed temperature  $T_H < T_C$ . Eq. (4) characterizes the influence of the ferromagnetic contribution to the value of the coefficient  $R_s$  depending on the saturation magnetization  $J_s$ .

In Table 1, we present values of the coefficients  $\alpha_R$  from Eq. (4) for some ferromagnetic alloys in the amorphous and crystalline states. It is seen that the coefficients  $\alpha_R$  are less in the amorphous state than in the crystalline one for the same alloy compositions. This indicates that the relationship between  $R_s$  and  $J_s^2$  is weaker in the amorphous state than in the crystalline one (see Fig. 4). Decrease in the value of  $R_s$  with the increase of the chromium content can be explained by the antiferromagnetic chromium nature, and as a result the monotonous decrease of the saturation magnetization  $J_s$  occurs. Increase in the Cr content in AMA of  $\text{Fe}_{85-x}\text{Cr}_x\text{B}_{15}$  system is accompanied by the decrease in the effective magnetic moment that implies about the formation of the magnetic clusters with antiparallel spin orientation [10].

**Table 1** – The values of the coefficient  $\alpha_R$  for some alloys in the amorphous and crystalline states

Alloy	Amorphous state		Crystalline state	
	Temperature range, K	$\alpha_R \cdot 10^{13}$ , $\text{m}^3/(\text{C}\cdot\text{K})$	Temperature range, K	$\alpha_R \cdot 10^{13}$ , $\text{m}^3/(\text{C}\cdot\text{K})$
$\text{Fe}_{58}\text{Co}_{20}\text{Si}_{12}\text{B}_{10}$	100...550	1,49	450...850	3,73
$\text{Fe}_{40}\text{Ni}_{38}\text{Mo}_4\text{B}_{18}$	300...400	1,36	300...450	2,01
$\text{Fe}_{66,9}\text{Ni}_{24,7}\text{Si}_{4,9}\text{B}_{3,5}$	350...600	1,02	300...600	0,96
$\text{Fe}_{44,2}\text{Ni}_{44,2}\text{Mo}_{7,7}\text{B}_{3,9}$	300...450	2,06	300...450	2,29
$\text{Co}_{86,35}\text{Fe}_{6,15}\text{Si}_{4,9}\text{B}_{2,6}$	350...700	1,34	300...750	1,93
$\text{Co}_{71,7}\text{Fe}_{5,7}\text{Ni}_{11,9}\text{Si}_{8,2}\text{B}_{2,5}$	100...350	9,02	100...350	12,95
$\text{Co}_{59,7}\text{Fe}_{5,8}\text{Ni}_{23,8}\text{Si}_{8,2}\text{B}_{2,5}$	100...350	2,09	100...500	15,02
$\text{Co}_{84,35}\text{Fe}_{5,8}\text{Si}_{7,4}\text{B}_{2,45}$	300...450	3,66	300...500	4,55

A weaker dependence  $\ln(\Delta\rho/\rho) = f[\ln(\Delta R_s/R_s)]$  in the amorphous state in comparison with the crystalline one indicates that the contribution of the phonon scattering mechanism in amorphous alloys is considerably weaker than in crystalline ones of the same composition. Other effects [13] probably influence the carrier scattering mechanism in ferromagnetic AMA.

In ferromagnetic alloys the relationship between  $R_s$  and reduced electrical resistance  $\Delta\rho/\rho$  is also observed and can be represented in the form [2, 3]

$$\Delta R_s/R_s = \beta(\Delta\rho/\rho)^m. \quad (5)$$

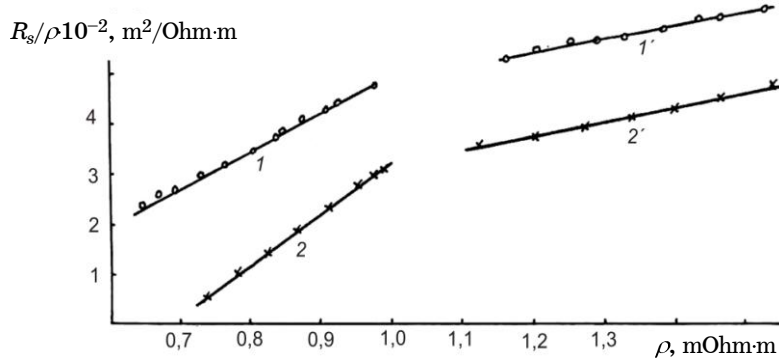
Coefficients  $m$  and  $\beta$ , for example, for  $\text{Co}_{86,4}\text{Fe}_{6,1}\text{Si}_{4,9}\text{B}_{2,6}$  alloy are equal to 0,47 and 2,17, respectively, in the amorphous state and 1,47 and 4,78 in the crystalline state. Other alloys based on iron metal group have the values of  $m$  and  $\beta$  close to the mentioned above. This indicates that the AHE in the studied AMA based on iron metal group appears considerably weaker in the amorphous state than in the crystalline one.

To determine the contribution of the phonon scattering mechanism to the temperature dependences  $R_s(T)$  and  $\rho(T)$ , the fulfillment of the relation

$$\Delta R_s(T) = a\rho + b\rho^2 \quad (6)$$

was verified [1-3]. In (6), the first term is defined by the asymmetric carrier scattering and the second one is connected with the mechanism of the lateral displacement of the carriers under the action of the spin-orbit interaction.

Fig. 5 illustrates the typical dependences  $R_s/\rho = f(\rho)$  for alloys based on iron metal group in the amorphous and crystalline states. The values of the coefficients  $a$  and  $b$  are represented in Table 2. For other alloys based on iron metal group the coefficients  $a$  and  $b$  have close values, and the value of the first term in Eq. (6) is substantially less than that of the second one.



**Fig. 5** – Dependences  $R_s/\rho = f(\rho)$  for  $\text{Co}_{84,35}\text{Fe}_{5,8}\text{Si}_{7,4}\text{B}_{2,45}$  ( $\times$ ) and  $\text{Fe}_{60}\text{Co}_{20}\text{Si}_8\text{B}_{12}$  ( $\circ$ ) alloys in the crystalline (1 and 2) and amorphous (1' and 2') states

**Table 2** – The values of the coefficients  $a$  and  $b$  for some alloys in the amorphous and crystalline states

Alloy	Amorphous state			Crystalline state		
	Temperature range, K	$a \cdot 10^2$ , $\text{m}^2/(\text{Ohm}\cdot\text{C})$	$b \cdot 10^4$ , $\text{m}/(\text{Ohm}^2\cdot\text{C})$	Temperature range, K	$a \cdot 10^2$ , $\text{m}^2/(\text{Ohm}\cdot\text{C})$	$b \cdot 10^4$ , $\text{m}/(\text{Ohm}^2\cdot\text{C})$
$\text{Fe}_{60}\text{Co}_{20}\text{Si}_8\text{B}_{12}$	300...620	0,033	5,91	470...870	0,044	6,66
$\text{Fe}_{40}\text{Ni}_{38}\text{Mo}_4\text{B}_{18}$	300...450	0,683	48,0	300...450	0,038	7,13
$\text{Fe}_{66,9}\text{Ni}_{24,7}\text{Si}_{4,9}\text{B}_{3,5}$	300...550	0,068	6,7	500...800	0,128	11,2
$\text{Co}_{84,35}\text{Fe}_{5,8}\text{Si}_{7,4}\text{B}_{2,45}$	300...500	0,844	66,5	300...550	0,167	27,6
$\text{Co}_{59,7}\text{Fe}_{5,8}\text{Ni}_{23,8}\text{Si}_{8,2}\text{B}_{2,5}$	250...400	1,030	14,7	500...750	0,950	75,7
$\text{Co}_{71,7}\text{Fe}_{5,7}\text{Ni}_{11,9}\text{Si}_{8,2}\text{B}_{2,5}$	100...400	0,089	6,39	200...500	0,049	4,39

Proportionality  $R_s \sim \rho^2$  is the consequence of topological disorder in highly resistive amorphous alloys [3], and differences in the values of the coefficients  $a$  and  $b$  for the amorphous and crystalline states of ferromagnetic alloys indicates the partial transformation of the electron energy spectrum under amorphization. Analysis of the dependences  $R_s/\rho = f(\rho)$  also shows that the phonon contribution to the kinetic properties of AMA is less significant in the amorphous state than in the crystalline one. The main reason of the difference in dependences  $R_s(T)$  in crystalline and amorphous ferromagnetic alloys is the differences in the scattering mechanisms conditioned by the structure disorder [13].

Carrier scattering on magnetic inhomogeneities significantly influences the dependences  $R_s(T)$  for amorphous ferromagnets. The authors of [3] note that under the action of the spin-orbit interaction during the conduction electron scattering on impurity centers (and also on magnons, phonons, etc.) the asymmetry of the scattering probability appears, and it leads to the Hall “curling” of the conduction electrons. For  $d$ -type states in AMA not only the diffusion but also close to hopping charge transport is possible, and the electron autolocalization on virtual defects with the electron trap formation [13] as well. Therefore the temperature dependences  $R_s(T)$  of the most of

AMA are determined not only by their composition but also by the local magnetic and structure inhomogeneities, by the degree of heterogeneity of alloys and etc., which, in turn, depend on the technological conditions of their formation and thermal treatment.

#### 4. CONCLUSIONS

For amorphous ferromagnets based on iron metal group  $R_s > 0$  and depending on the chemical composition of alloys and modes of obtaining them, it is in the range of  $(1-6) \cdot 10^{-8} \text{ m}^3/\text{C}$ . During the transition from the amorphous to the crystalline state  $R_s$  decreases and  $\partial R_s / \partial T$  increases that indicates the reduction of the spin-orbit interaction. The normal Hall coefficient  $R_0$  depends weakly on the temperature and about two orders of magnitude less than  $R_s$ . Relationship between  $R_s$  and  $J_s^2$  in the amorphous state is weaker than in the crystalline one that is connected with the structure disorder. By that reason the phonon contribution to the kinetic properties of amorphous alloys is less significant than of crystalline ones. Temperature dependences  $R_s(T)$  and  $\rho(T)$  indicate the partial transformation of the electron energy spectrum at the transition from the amorphous to the crystalline state.

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