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MAGNETISM AND FERROELECTRICITY

Low-Temperature Properties of Magnetic $Ni_{50+x}Mn_{25-x+y}Ga_{25-y}$ Shape Memory Alloys

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Abstract—The effect of atomic disordering and deviation from the Ni_2MnGa stoichiometric composition on the low-temperature properties of alloys with magnetically controlled shape memory effect is studied. The specific features of the magnetic, galvanomagnetic, and electrical properties of alloys with magnetically controlled shape memory effect are discussed. The specific features of the magnetic, galvanomagnetic, and electrical properties of alloys $Ni_{50}Mn_{25}Ga_{25}$, $Ni_{54}Mn_{21}Ga_{25}$, and $Ni_{50}Mn_{28.5}Ga_{21.5}$ in the temperature range $2 \le T \le 80$ K under magnetic fields $H \le 12$ MA/m are studied.

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1. INTRODUCTION

Currently, investigations of alloys that are based on the stoichiometric Ni_2MnGa composition and exhibit the magnetically controlled shape memory effect are of great interest. Much attention has been given to revealing the specific features of their thermal, magnetic, and structural properties. The aim of those studies was to create materials with the best functional properties, namely, high reversible magnetostriction, high magnetocaloric effect, and the temperatures of martensitic (T_M) and magnetic (T_C) phase transitions close to room temperature. Purposeful search for such materials requires a knowledge of their electronic characteristics.

In this work, along with data on the crystal structure, we present the results of studying the magnetic, galvanomagnetic (Hall effect, magnetoresistance), and electrical properties of three $Ni_{50+x}Mn_{25-x+y}Ga_{25-y}$ alloys ($x = 0$, $y = 0$; $x = 4.0$, $y = 0$; and $x = 0$, $y = 3.5$) at low temperatures $T \ll (T_M, T_C)$. At these component concentrations, the magnetic and electronic parameters of the alloys are changed most significantly [1–6]. In this work, we also consider the influence of atomic disordering on the crystal structure and low-temperature properties of the alloys.

The $Ni_{50}Mn_{25}Ga_{25}$ and $Ni_{50}Mn_{28.5}Ga_{21.5}$ alloys were prepared by induction melting in an argon atmosphere. Then, the polycrystalline ingots were homogenized at temperatures of 1073–1173 K for 6–30 h. The preparation method and the structure of the $Ni_{51}Mn_{21}Ga_{25}$ alloy were described in [1–5]. All the alloys were atomically disordered by subsequent ultrarapid melt quenching (RMQ) at a rate of ~10^4–10^5 K/s on a rotating copper disc. The crystalline structure was characterized by high-resolution transmission electron microscopy using JEM-200CX and CM-30 microscopes and also using a DRON x-ray diffractometer. The magnetic properties were studied on a MPMS-9 apparatus (Quantum Design) for measuring physical properties in magnetic fields $H \le 7$ MA/m. The temperature dependences of the electrical resistivity $\rho(T)$ and magnetoresistance $\Delta\rho/\rho = [\rho(T, H) - \rho(T, 0)]/\rho(T, 0)$ were measured by a potentiometric method in the range $4 < T < 80$ K at $H = 0$ and 8 MA/m. The field dependences of the Hall resistivity $\rho_{xy}(H)$ and magnetoresistance $\Delta\rho/\rho(H)$ were measured at $T = 4.2$ K and $H \le 12$ MA/m.

2. CRYSTAL STRUCTURE

The electron microscopy and x-ray diffraction studies show that the alloys under consideration in the initial (as-cast) state and after RMQ disordering exhibit, at elevated temperatures $T > T_M$, an $L2_1$ austenitic fine-grained structure. The average grain size in the rapidly quenched alloys is ~0.5 μm, which is three orders of magnitude smaller than that in the initial as-cast alloys. Moreover, in samples obtained by RMQ, the densities of dislocations and vacancies are somewhat greater. The RMQ method we used does not produce a homogeneous amorphous state similar to that reported in [7].

When cooled near the temperature T_M, the alloys undergo a martensitic transformation. In this case, plates of tetragonal martensite have five-layered modulated structure 5*M* [2, 3]. Further cooling of the alloys

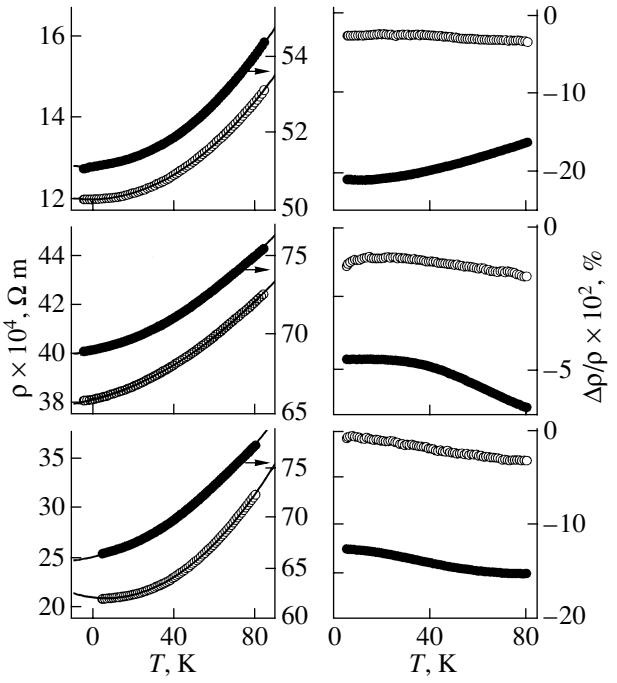

Fig. 1. Temperature dependences of the electrical resistivity and magnetoresistance of $Ni_{50+x}Mn_{25-x+y}Ga_{25-y}$ alloys: experimental data for $Ni_{50}Mn_{25}Ga_{25}$ (top panels), $Ni_{50}Mn_{28.5}Ga_{21.5}$ (middle panels), and $Ni_{54}Mn_{21}Ga_{25}$ (bottom panels). Open symbols are data for the initial (as-cast) alloys, solid symbols are data for rapidly quenched alloys, and solid lines are fits of the $\rho(T)$ dependence obtained using Eq. (1).

below T_M is accompanied by the martensite–martensite transition $5M \longrightarrow 7M$. At low temperatures $T \ll (T_M$ and $T_C)$, the alloys exhibit a martensitic $7M$ phase substructure, which is characterized by a parquet structure of pairwise-twinned crystals containing thin nanoscopic twins or stacking faults [1–6]. The latter defects cause the appearance of equidistant extra reflections corresponding to the long-period $7M$ structure. Moreover, below the temperature of the first martensitic transformation T_M, the alloys contain untwinned lamellar crystals of tetragonal (c/a = 1.2) martensite (NM martensite).

The x-ray diffraction data confirm, on the whole, the sequence of martensitic transformations $L2_1 \longrightarrow 5M + HM \longrightarrow 7M + HM$ characteristic of as-cast and rapidly quenched Ni_2MnGa-based alloys.

Table 1. Coefficients involved in the temperature dependence of the resistivity of the alloys studied

Alloy		H, MA/m	$\rho_0 \times 10^4$, Ω m	$a \times 10^6$, Ω m/K	$b \times 10^8$, Ω m/K^2
$Ni_{50}Mn_{25}Ga_{25}$	Initial	0	12.03	–0.786	5.2
		8	11.77	–0.65	4.764
	RMQ	0	51.15	–0.639	5.8
		8	42.23	–0.473	7.9
$Ni_{54}Mn_{21}Ga_{25}$	Initial	0	21.26	–2.594	19.38
		8	20.74	–3.205	18.74
	RMQ	0	66.3	4.637	11.9
		8	59.2	1.27	11.24
$Ni_{50}Mn_{28.5}Ga_{21.5}$	Initial	0	37.95	1.68	5.18
		8	37.48	1.945	4.403
	RMQ	0	68.83	1.892	8.185
		8	65.78	2.471	5.218

3. ELECTRICAL PROPERTIES

Figure 1 shows the temperature dependences of the resistivity of the $Ni_{50+x}Mn_{25-x+y}Ga_{25-y}$ alloys (x = 0, y = 0; x = 4, y = 0; and x = 0, y = 3.5) in the initial (as-cast) state and after rapid quenching of the samples measured in a zero magnetic field over the temperature range $4.2 \le T \le 80$ K. As is seen from this figure and Table 1, over the entire temperature range considered, the $\rho(T)$ dependences of the alloys under study are described, within the limits of the measurement error, by the following expression characteristic of ferromagnetic metals [8]:

$$\rho = \rho_0 + aT + bT^2, \quad (1)$$

where ρ_0 is the residual resistivity characterizing the charge carrier scattering by inhomogeneities of the Coulomb potential. The deviation of the alloy composition from the stoichiometry increases ρ_0 by a factor of about two when weakly magnetic Ni atoms replace strongly magnetic Mn atoms, and by a factor of approximately three when excess Mn atoms substitute for nonmagnetic Ga atoms.[1] A similar increase in ρ_0 is also caused by atomic disordering of the RMQ alloy samples studied (Table 1). The $\rho(T)$ measurements performed at H = 8 MA/m show that the change in ρ_0 caused by a magnetic field is more significant for the rapidly quenched alloys. It follows from all the experimental data that ρ_0 of the alloys studied is mainly dependent on the heterogeneity of their magnetic subsystem.

The coefficient b in Eq. (1) is mainly determined by the mechanism of electron–electron scattering, which is increased owing to transitions from the s to d band. However, in the alloys under study, the coefficient b is an order of magnitude higher than that usually observed in transition metals [10]. The maximum coefficient b is observed in the nickel-enriched alloy $Ni_{54}Mn_{21}Ga_{25}$. The square temperature dependence of the resistivity can be due to electron–magnon scattering, as it was established for ferromagnetic transition metals, for

[1] According to [9], the atomic moments of the components of the alloy studied are as follows: $\mu_{Mn} = 3.44\mu_B$, $\mu_{Ni} = 0.29\mu_B$, and $\mu_{Ga} \sim 0$.

example, in [11]. This conclusion is confirmed, in particular, by the experimentally observed decrease in the coefficient b in a magnetic field for practically all the alloys studied (Table 1).

The origin of the terms linear in temperature in Eq. (1) has been studied both theoretically and experimentally by many researchers [8]. In our case, it is unusual that the coefficient a in Eq. (1) is negative for the stoichiometric $Ni_{50}Mn_{25}Ga_{25}$ and nonstoichiometric $Ni_{54}Mn_{21}Ga_{25}$ alloys. However, the atomic disordering of the $Ni_{54}Mn_{21}Ga_{25}$ alloy causes the coefficient a to be positive. For the rapidly quenched $Ni_{54}Mn_{21}Ga_{25}$ alloy and the nonstoichiometric $Ni_{50}Mn_{28.5}Ga_{21.5}$ alloy (both as-cast and rapidly quenched), the coefficient $a > 0$. The coefficient a can change in sign if it is associated with scattering of conduction electrons by spin waves. In this case, the magnitude and sign of the coefficient a depend on the dispersion law for the conduction electrons. The observed change of the coefficient a in sign indicates a substantial transformation of the electron spectrum in the vicinity of the Fermi level, depending on the component concentrations and, as a result, the atomic disordering of the magnetic alloys studied.

It should be noted that the $\rho(T)$ dependences of the nonstoichiometric $Ni_{50}Mn_{28.5}Ga_{21.5}$ alloy at low temperatures $T < 40$ K $\ll (T_M, T_C)$ can also be described within the limits of the measurement error by the expression

$$\rho = \rho_0 + cT^{3/2}. \quad (2)$$

Such temperature dependences of the resistivity of ferromagnetic alloys of transition metals are usually associated with scattering of conduction electrons by thermal excitations in domain boundaries [8]. However, the measurements of $\rho(T)$ in an external magnetic field causing the domain boundaries in the $Ni_{50}Mn_{28.5}Ga_{21.5}$ alloy to disappear show an insignificant change in the coefficient of $T^{3/2}$. Moreover, Eq. (2) is not obeyed for other alloys studied in this work. Therefore, it can be concluded that the contribution to $\rho(T)$ due to conduction electron scattering by spin waves in domain boundaries is insignificant in the case considered.

The weak influence of an external magnetic field on the temperature dependences of the resistivity of these alloys also indicates that the coefficients a and b in Eq. (1) are mainly determined by the parameters of their electronic band structure near the Fermi surface. It is most likely that the coefficient a is indeed mainly determined by charge carrier scattering on spin waves, in which case even the sign of the coefficient a can change depending on the dispersion law of electrons [8]. The coefficient b is mainly due to scattering by static and dynamic heterogeneities of the alloys accompanied by the transfer of conduction s electrons to the d band.

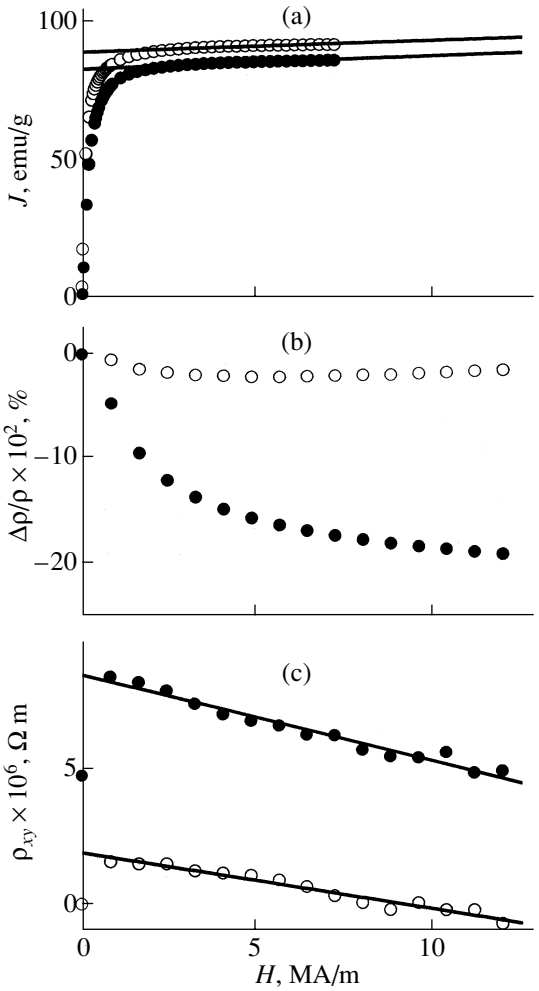

Fig. 2. Field dependences of (a) the magnetization, (b) magnetoresistance, and (c) Hall resistance of the $Ni_{50}Mn_{25}Ga_{25}$ alloy measured at $T \sim 4.2$ K. Open symbols are data for the initial (as-cast) alloys, solid symbols are data for rapidly quenched alloys, and solid lines are the fitting of the experimental data to Eqs. (3) and (5).

4. MAGNETIC AND GALVANOMAGNETIC PROPERTIES

The field dependences of the magnetic and galvanomagnetic properties of the $Ni_{50+x}Mn_{25-x+y}Ga_{25-y}$ alloys ($x = 0$, $y = 0$; $x = 4$, $y = 0$; and $x = 0$, $y = 3.5$) measured at $T = 4.2$ K are presented in Figs. 2–4. It is seen that the magnetization of all the samples varies almost linearly with magnetic field at $H \geq 2$ MA/m; i.e., this range corresponds to a "pseudo-paramagnetic process," for which the following relation is valid:

$$J(H) = J_S + \chi_P H. \quad (3)$$

Therefore, we can determine the spontaneous magnetization J_S and the magnetic susceptibility χ_P, which are necessary to analyze the galvanomagnetic properties of the alloys. It follows from Table 2 that J_S

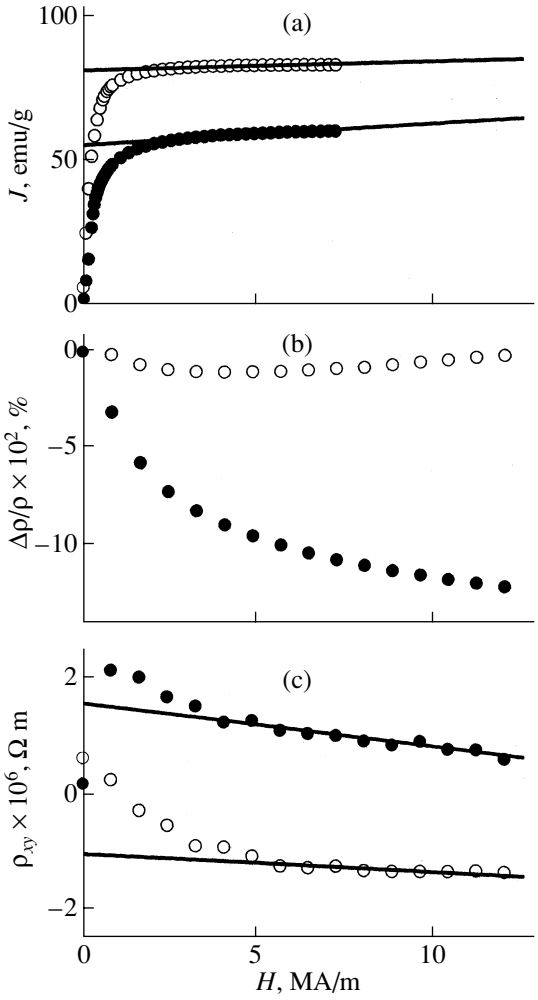

Fig. 3. Field dependences of (a) the magnetization, (b) magnetoresistance, and (c) Hall resistance of the $Ni_{54}Mn_{21}Ga_{25}$ alloy measured at $T \sim 4.2$ K. The designations are the same as in Fig. 2.

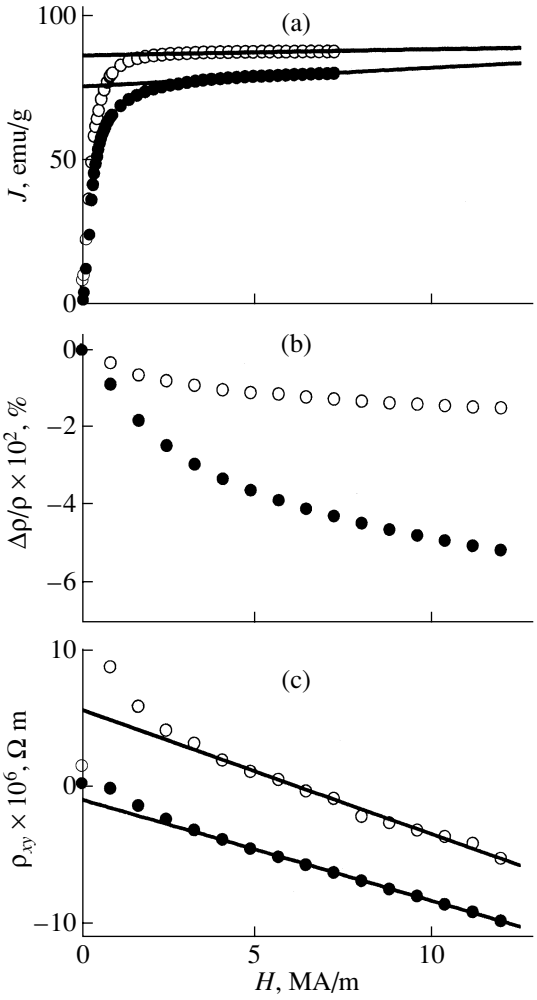

Fig. 4. Field dependences of (a) the magnetization, (b) magnetoresistance, and (c) Hall resistance of the $Ni_{50}Mn_{28.5}Ga_{21.5}$ alloy measured at $T \sim 4.2$ K. The designations are the same as in Fig. 2.

decreases insignificantly as the alloy composition deviates from the stoichiometry. It is natural in the case where less magnetic nickel atoms replace strongly magnetic manganese. However, in the case where nonmagnetic gallium is replaced by strongly magnetic manganese, the decrease in J_S can likely be explained by the occurrence of frustrated exchange bonds in the alloy leading to antiferromagnetic ordering of the magnetic moments of single manganese atoms occupying the gallium sites. Most likely, for the same reason, the spontaneous magnetization decreases with atomic disordering of the alloys due to RMQ. Accordingly, the susceptibility of the pseudo-paramagnetic process increases and J_S decreases with a deviation of the composition from stoichiometry and with atomic disordering (Table 2).

As follows from Figs. 2–4, the field dependences of the magnetoresistance of the alloys measured at $T =$ 4.2 K under conditions $\mathbf{H} \perp \mathbf{j}$ (transverse magnetoresistance)[2] are those typical of ferromagnets. In the pseudo-paramagnetic region ($H > 2$ MA/m), the $\Delta\rho/\rho(H)$ dependences are described by a quadratic polynomial practically for all the alloys. Indeed, using Eq. (3), we obtain [8]

$$\Delta\rho/\rho = J_S^2 - J^2 = -2\chi_P J_S H - \chi_P^2 H^2. \quad (4)$$

The only exclusion is the $\Delta\rho/\rho(H)$ dependence for the initial (as-cast) $Ni_{50}Mn_{25}Ga_{25}$ and $Ni_{54}Mn_{21}Ga_{25}$ alloys exhibiting a fairly low electrical resistance (Table 1). Apart from the negative magnetic component, these alloys are characterized by a substantial positive component of the magnetoresistance due to the Lorentz force causing the conduction electrons to spiral in a magnetic field. It follows from Fig. 1 that, in most of the cases under consideration, the negative quantity

[2] Here, **H** and **j** are the magnetic-field and current-density vectors, respectively.

Table 2. Composition and magnetic properties of the alloys studied

Alloy		J_S, A m^2/kg	$\chi_P \times 10^6$, m^3/kg	$R_0 \times 10^{14}$, m^3/A s	$R_S \times 10^{13}$, m^3/A s
$Ni_{50}Mn_{25}Ga_{25}$	Initial	90.1	0.466	–2.9	2.67
	RMQ	83.8	0.518	–9.16	12.7
$Ni_{54}Mn_{21}Ga_{25}$	Initial	81.6	0.351	0.31	–1.56
	RMQ	55.9	0.779	–3.49	3.52
$Ni_{50}Mn_{28.5}Ga_{21.5}$	Initial	86	0.26	–3.49	8.35
	RMQ	75.3	0.704	–0.054	–1.3

$\Delta\rho/\rho(T)$ increases with temperature, which is natural, because the Lorentz addition is suppressed due to the increased electrical resistance of the sample.

The field dependences of the Hall resistance $\rho_{xy}(H)$ are shown in Figs. 2–4. The Hall effect in transition metals is known [8] to have two components differing in origin. One component is the ordinary Hall effect due to the Lorentz force. Another component is the anomalous (spontaneous) Hall effect due to spin–orbit coupling, which can be determined in a two-band *s–d* model by both "intrinsic" *s–s* interaction and "extrinsic" interaction of the *s* spin with a *d* orbital. The former effect is mainly dependent on the specific features of the electronic band structure near the Fermi level E_F and, thus, varies only slightly with temperature and in a magnetic field. The latter effect, conversely, is mainly due to scattering of the conduction electrons and, thus, exhibits a strong temperature dependence. The two Hall components are characterized by the ordinary (R_0) and anomalous (R_S) Hall coefficients, respectively.

In order to describe the Hall effect in magnets, as a rule, one uses the standard expression [8]

$$\rho_{xy} = R_0 B + 4\pi R_S J = R_0^* H + R_S^* J_S. \quad (5)$$

Here, $B = H + (4\pi - N)J$ is the magnetic induction in the sample, where N is the demagnetization factor of the sample. For samples in the shape of a parallelepiped ~$(3 \times 10 \times 0.2)$ mm in size used in this study when measuring the Hall effect, we have $N \sim 12$. From the experimental $\rho_{xy}(H)$ curves, which are linear in the pseudoparamagnetic region, we determined the quantity $4\pi[R_S + (1 - N/4\pi)R_0]J_S$ by extrapolation to a zero magnetic field and the quantity $R_0^* = R_0 + 4\pi\chi_P R_S$ from the slope of the curve. With the values of J_S and χ_P determined from the magnetization data, we find the ordinary (R_0) and anomalous (R_S) Hall coefficients.

According to the theory of kinetic phenomena in metals (see, e. g., [8]), in a single-electron approximation, we have

$$R_0 = 1/nec$$

and, in the case of two types of Hall carriers (electrons and holes),

$$R_0 = (n_h\mu_h^2 - n_e\mu_e^2)/(n_h\nu_h + n_e\mu_e). \quad (6a)$$

Here, e is the electron charge; c is the velocity of light in free space; and n_i and μ_i are the density and mobility of ith-type carriers, respectively. At low temperatures (which are considered in this case), we can assume that the conduction electron scattering by heterogeneities of the magnetic subsystem is the main mechanism. Therefore, we have [8]

$$R_S = \pm\frac{\lambda_{\text{eff}}}{E_F}\frac{\rho_m}{J_S}, \quad (6b)$$

where the plus and minus signs correspond to the hole and electron conduction, respectively; λ_{eff} is the spin–orbit coupling constant; and ρ_m is the magnetic component of the resistivity, which is actually the quantity ρ_0 in this case.

It is seen from Figs. 2–4 that, at $H > 3$ MA/m, the Hall resistance of the alloys studied is practically a linear function of magnetic field. It is reasonable to assume that the slope of the $\rho_{xy}(H)$ dependences at $H <$ 3 MA/m is changed owing to the addition to the anomalous Hall coefficient due to changes in ρ_m and R_S, which vary with magnetic field in the absence of complete magnetic saturation, as considered, e.g., in [12]. Table 2 lists the values of the ordinary and anomalous Hall coefficients estimated from Eq. (5) at $H > 3$ MA/m. It is seen that the estimated ordinary Hall coefficient has values common for transition metals. The coefficient R_0 is negative for practically all the alloys studied. For the initial (as-cast) $Ni_{54}Mn_{21}Ga_{25}$ alloy, the experimental coefficient $R_0 > 0$, and the ordinary Hall coefficient for the rapidly quenched $Ni_{54}Mn_{21}Ga_{25}$ alloy is close to zero. Using Eqs. (6), we can conclude that the majority carriers in these alloys are *s* electrons. It agrees with the conclusions drawn from the measured thermopower of the alloys studied [1].

According to Table 2, the absolute values of the anomalous Hall constant R_S are one to two orders of magnitude higher than those of the ordinary Hall constant R_0. Thus, the Hall effect in the ferromagnetic alloys studied is mainly determined by its anomalous component. The coefficients R_S and R_0 are opposite in sign for practically all the alloys. Using Eqs. (6), it is natural to

relate the different signs of the coefficients R_0 and R_S to the sign of the spin–orbit coupling constant λ_{eff}.

5. CONCLUSIONS

Thus, we have shown that a deviation of the alloy composition from the stoichiometric $Ni_{50}Mn_{25}Ga_{25}$ produced by replacing strongly magnetic manganese atoms by weakly magnetic nickel atoms or by substituting magnetic manganese atoms for nonmagnetic gallium atoms not only increases the temperatures of the martensitic transitions but also enhances structural and magnetic heterogeneities of the alloys. Similar changes also occur in the disordered alloys obtained by rapid melt quenching. At low temperatures, however, all the alloys retain a 7*M*-type long-period crystalline structure and the long-range ferromagnetic order.

A change in the alloy composition from the stoichiometric $Ni_{50}Mn_{25}Ga_{25}$ and subsequent rapid melt quenching decrease the spontaneous magnetization and increase the magnetic susceptibility measured in high magnetic fields. The enhancement of the structural and, particularly, magnetic disorder is accompanied by an increase in the residual electrical resistance and a substantial change in the temperature-dependent part of $\rho(T)$. The temperature dependence of the electrical resistance of the alloy is determined by two components associated with electron–electron scattering and scattering of conduction electrons by spin waves.

The magnetoresistance also has two contributions, namely, a positive component due to the Lorentz force causing conduction electrons to spiral and a negative component due to the ordering of spin heterogeneities in a magnetic field. The latter contribution is dominant. The change in the magnitude and sign of the coefficient R_0 and the change in the temperature-dependent components of the electrical resistance due to the deviation of the alloy composition from the stoichiometry and to atomic disordering of the alloy are associated with a transformation of the electronic band structure. The behavior of the anomalous Hall coefficient is associated with changes both in the magnetic component of the resistance and the magnetization and in the spin–orbit coupling constant.

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