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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

Magnetotransport Properties of Thin $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ Films

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Abstract—The magnetic and magnetotransport properties of thin Heusler alloy $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ films deposited onto MgO(100) substrates are studied over a wide temperature range, which includes a martensitic transition (MT). For this composition, the MT is not accompanied by a magnetic phase transition, since the martensitic and austenitic phases are ferromagnets with similar magnetizations. The electrical resistivity does not undergo sharp changes during the MT. The magnetoresistance is negative, decreases in magnitude with increasing temperature in the range 100–250 K corresponding to the MT, and then increases to −1%. The field dependences of the Hall effect resistivity have the shape that is characteristic of homogeneous ferromagnetic alloys. The coefficients of the normal and anomalous Hall effects are determined. The anomalous Hall effect coefficient is shown to be described by the relation $R_s = \alpha\rho + \beta\rho^2$, where ρ is the electrical resistivity and the second term is lower than the first, which indicates an important role of the interference impurity–phonon scattering mechanism.

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1. INTRODUCTION

Heusler alloys have unique multifunctional properties important for a variety of practical applications, which determines the continuously increasing interest in these systems [1, 2]. Alloys of the Ni–Mn–X (X = Ga, In, Sb) family of a certain concentration composition undergo the martensitic transition (MT) from a high-temperature cubic phase (austenite) to a low-temperature phase with tetragonal distortions (martensite) at characteristic temperature T_M and vice versa at temperature T_A when the temperature increases, and this transition is accompanied by a temperature hysteresis. This transition is responsible for many properties, in particular, giant deformation [3], the giant magnetocaloric effect [4], the giant anomalous Hall effect (AHE) [5], and the magnetic shape memory effect [6]. Recently, skyrmions [7, 8] and the corresponding topological Hall effect [9] were found in Ni–Mn–Ga and Ni–Mn–In alloys. The MT is considered to be associated with the collective Jahn–Teller effect, in which the gain in energy due to a decrease in the lattice symmetry is compensated by the loss in the energy of the electronic subsystem. Although a number of experimental facts (see, e.g., [10]) and theoretical calculations (see, e.g., [11]) support this concept, there are data that contradict it. For example, the electron contribution to the heat capacity in Ni–Mn–In alloys, which is proportional to the density of states (DOS) at the Fermi level, changes slightly during the MT [12], the changes in the magnetooptical Kerr effect are also low [13], and the electron contribution to the magnetocaloric effect has not been found [4]. These facts indicate a rather complex and not fully understood nature of the rearrangement of the electronic structure during the MT. Magnetotransport effects, such as magnetoresistance and the Hall effect, are among the most effective indicators of phase transitions and changes in electronic and magnetic structures. These phenomena, especially the Hall effect with separated normal and anomalous components, were studied only for some Heusler alloy compositions, which undergo well-pronounced MT. A change in the carrier type during MT [14], a redistribution of the *d* spin-up and spin-down states [15], and strong antiferromagnetic correlations [16] were revealed. The study of the AHE, which is central in the group of spontaneous galvanomagnetic phenomena and a prominent representative of spin-dependent transport phenomena, in Heusler alloys is also of independent importance, since discussions about the role of various mechanisms in the formation of this effect are still ongoing [17]. For example, the authors of [18] recently stated that the AHE in Ni–Mn–Ga alloys is associated with skew scattering, and the authors of [14–16] showed that neither skew scattering nor the side jump mechanism or the intrinsic mechanism can explain the experimental data obtained for Ni–Mn–In–based alloys and the $Ni_{47.3}Mn_{30.6}Ga_{22.1}$ alloy.

In this work, the magnetic properties, the electrical resistivity, the magnetoresistance, and the Hall effect resistivity of thin $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ films are studied over a wide temperature range, including the MT region. The choice of this alloy composition, which belongs to the class of magnetic shape memory alloys, is determined by the following factors. First, the structural properties of this composition have been studied in detail [19, 20]. Second, the MT in these alloys is not as pronounced as that in Ni–Mn–In or Ni–Mn–Ga alloys, the transport properties of which were studied earlier. Moreover, the MT in this alloy occurs in the ferromagnetic phase; i.e., there are no sharp changes in the magnetization or resistance at MT, and we can say that this is an example of "hidden" MT. Third, the high Curie temperature due to the presence of cobalt and the MT temperature range above the liquid-nitrogen temperature allow us to study the AHE over a wide temperature range. We demonstrate that the behavior of magnetoresistance, Hall effect resistivity, and the coefficients of normal and anomalous Hall effects in Ni–Fe(Co)–Ga films differs radically from that in Ni–Mn–In and Ni–Mn–Ga alloys.

2. EXPERIMENTAL

2.1. Samples

Polycrystalline 1-μm-thick films of the $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ (at %) Heusler alloy were prepared by sputtering of an initial target onto MgO(001) substrates heated to a temperature of 773 K. The details of film preparation and the structural properties were described in [19, 20]. Based on structural measurements by X-ray diffraction analysis, X-ray photoelectron spectroscopy, magnetic circular dichroism, and ferromagnetic resonance [19, 20], we found that the forward and reverse MTs in the films occur without changing the type of magnetic ordering (i.e., MT occurs inside the ferromagnetic phase) and the films are characterized by three ferromagnetic phases, namely, a cubic $L21$-ordered austenitic phase, a tetragonal martensitic phase, and disordered cubic γ phase.

2.2. Magnetic and Transport Measurements

The magnetization measurements were performed on a Lake Shore VSM vibrating-sample magnetometer in the temperature range 80–400 K and magnetic fields up to 16 kOe. Below, we determine magnetization M as the magnetic moment of a Hall sample in the form of a 2 × 6 mm strip. A magnetic field was applied perpendicular to the thin-film sample plane, i.e., in the Hall geometry.

The magnetotransport properties were measured by the standard four-probe method in magnetic fields up to 21 kOe at 80–400 K. To exclude magnetization even effects in determining the Hall coefficients, the Hall effect resistivity measurements at each temperature were carried out for two opposite field and current directions.

3. RESULTS AND DISCUSSION

3.1. Magnetization

The temperature dependences of the magnetization $M(T)$ in magnetic fields of 50 Oe and 16 kOe are shown in Fig. 1a and correlate well with the results of earlier studies of these films (see Fig. 3 in [19]). The MT starts at 250 K (Fig. 1a), and the magnetization changes slightly and monotonously during the MT. The MT is completed at $T < 80$ K, which is also consistent with the results of [19]. The MT manifests itself only in weak magnetic fields. It should be noted that the magnetization at low temperatures and 16 kOe is almost twice as large as of that at 50 Oe, which indicates the presence of noncollinear structures in weak fields at low temperatures; these structures are caused by the competition of ferromagnetic and antiferromagnetic exchange interactions.

Figure 1b shows the field dependences of the magnetization. These data are used to determine the coefficients of the normal and anomalous Hall effects.

3.2. Electrical Resistivity and Magnetoresistance

Figure 2 shows the temperature dependence of the resistivity during heating and cooling.

The MT is very weak in the temperature dependence of the resistivity. The divergence of the curves upon heating and cooling occurs at approximately 250 K, i.e., at the same temperature as in the temperature dependence of the magnetization. We note the following two features of the electrical resistivity. First, it is low, less than 150 μmΩ cm at all temperatures; that is, these alloys are low-resistance despite the fact that they are polycrystalline, four-component, and multiphase. Second, the temperature coefficient of resistivity is almost constant over the entire temperature range.

Figure 3 shows the temperature dependence of the magnetoresistance MR = $(\rho(H) - \rho(H))/\rho(0)$.

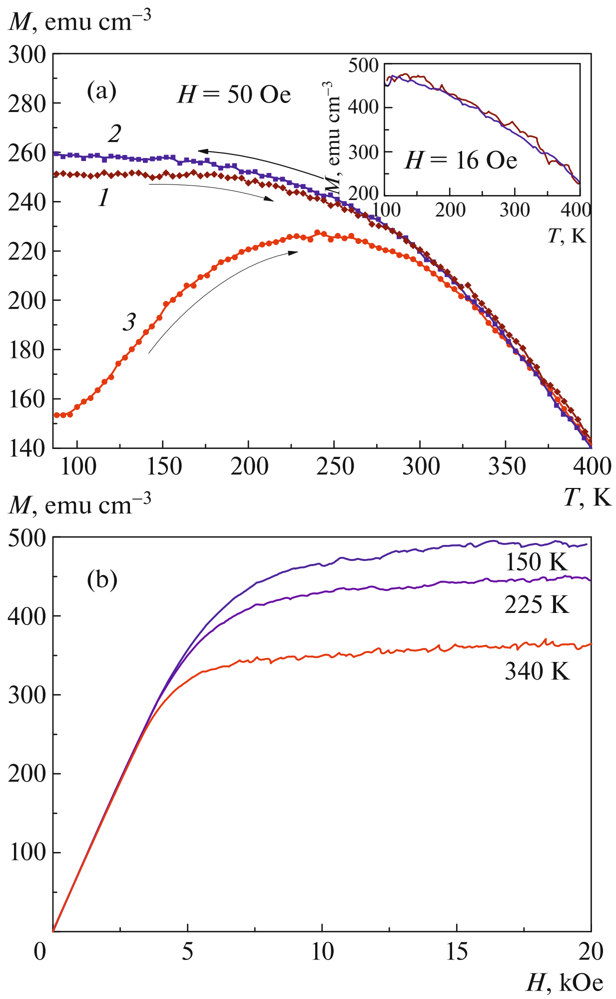

Fig. 1. (a) Temperature dependences of the magnetization of the thin $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ films upon (*1*) heating, (*2*) cooling in a magnetic field of 50 Oe (inset, the same at 16 kOe), and (*3*) heating after zero-field cooling. (b) Field dependences of the magnetization at temperatures of 150, 225, and 340 K.

The magnetoresistance is negative and lower than 1%: when the temperature increases, magnetoresistance first decreases and then increases above 250 K, demonstrating a wide minimum. This behavior is unusual, since the magnetoresistance in the vicinity of structural phase transitions usually has well-pronounced extremum in the transition region.

3.3. Hall Effect

Figure 4 shows the field dependences of the Hall effect resistivity ρ_H at temperatures of 150, 225, 340 K, i.e., in the vicinity of the MT and in the austenitic phase above the MT. The behavior of the Hall resistivity is typical of homogeneous ferromagnets rather than multiphase samples or Heusler alloys with a well-pronounced MT. The Hall resistivity first increases with

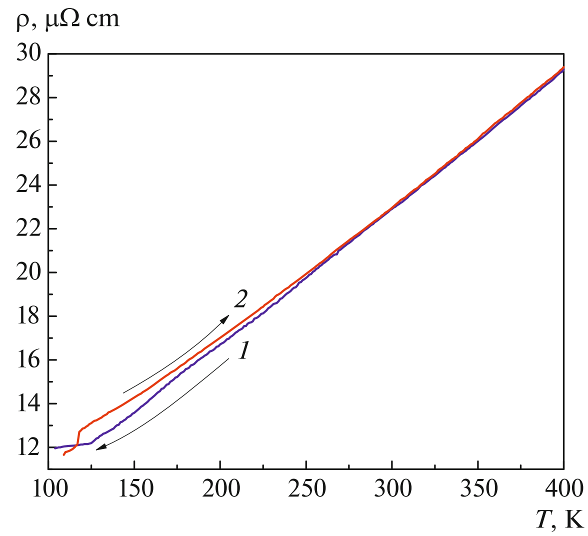

Fig. 2. Temperature dependence of the electrical resistivity of the $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ films upon (*1*) heating and (*2*) cooling.

the field, following the change in the magnetization, and then levels off in the same fields as the magnetization.

Let us begin a discussion of the experimental data with the electrical resistivity. The resistivity of the samples is approximately the same as for the crystalline alloys based on Ni and Fe [20]. The strong temperature dependence of the resistivity, which is determined by scattering by phonons and magnetic inhomogeneities, is close to linear. The phonon and magnetic contributions to the resistivity of ferromagnets are known to be linear in temperature in the temperature ranges above the Debye temperature and below the Curie temperature. For our composition, the Curie temperature is about 400 K (see Fig. 1a), and the Debye temperature, by analogy with the data obtained for Ni_2MnGa and Ni_2MnIn alloys (see Table 6 in [21]), can be estimated at 200–250 K. The most important fact is a very weak change in the resistivity at the MT. If the MT is associated with a change in the electronic structure, the density of states (DOS) at the Fermi level, $g(E_F)$, should change, which is confirmed by the theoretical calculations [22]. However, this does not manifest itself in the resistivity, which depends on the DOS, according to Mott's expression [23], as $|V^2|/g(E_F)^3$, where V is the scattering potential. Apparently, the change in the DOS is compensated by a change in the scattering potential. Unfortunately, the electronic structure calculations were performed only for $T = 0$; therefore, a direct comparison of the theory with the experiment, which does not take into account the temperature changes in the DOS, is impossible. Since the magnetization changes slightly, the transformation of the d states should be insignificant, which

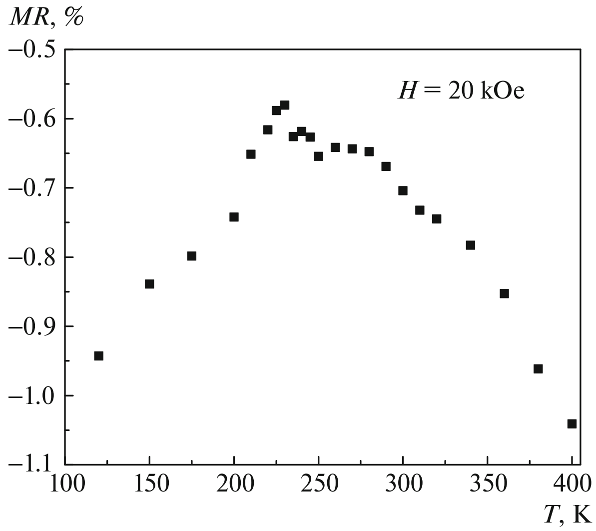

Fig. 3. Temperature dependence of the magnetoresistance of the $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ films.

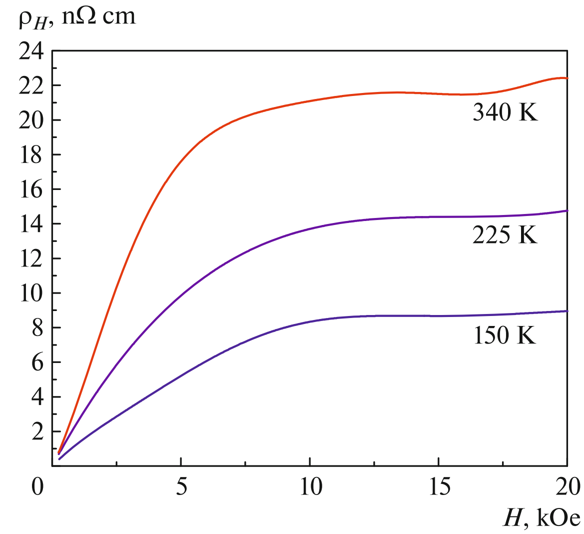

Fig. 4. Field dependences of the Hall resistivity of the $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ films at 150, 225, and 340 K.

means that the *s–d* scattering does not change fundamentally at the MT in the case under consideration.

The negative magnetoresistance is associated with the suppression of spin fluctuations and a magnetic disorder by a magnetic field. According to this mechanism, this contribution to the magnetoresistance increases when the Curie temperature (400 K) in the austenitic phase is approached, i.e., above 250 K. The decrease in the magnetoresistance at 100–250 K, i.e., in the MT region, looks unexpected. It is also noteworthy that the magnetoresistance at low temperatures is of the same order as that at high temperatures, although it should be much lower for magnon scattering. We attribute this behavior of the magnetoresistance to the presence of antiferromagnetic correlations leading to noncollinear local structures. The scattering by such structures is suppressed in a strong magnetic field; therefore, the magnetoresistance is quite high at low temperatures, where the competition between the antiferromagnetic and ferromagnetic interactions is significant. As the temperature increases, such noncollinear structures disappear gradually due to the formation of the austenitic phase and the cubic γ phase [19], which leads to a decrease in the magnetoresistance.

In the general case, the Hall resistivity is described by the relation

$$\rho_H = R_0 B_z + 4\pi R_s M_z + \Delta\rho_H. \quad (1)$$

Here, the first term describes the normal Hall effect (NHE) appearing due to the action of the Lorentz force, R_0 is the NHE coefficient, and B_z is the z component of the magnetic induction. The second term in Eq. (1) characterizes anomalous Hall effect (AHE), which appears due to the spin–orbit interaction, and R_s is the AHE coefficient. The third term describes the possible topological and antiferromagnetic Hall effects (see, e.g., [16]). The field dependence of the Hall resistivity (Fig. 4) has a standard shape for ferromagnetic alloys and correlates well with the field dependence of the magnetization without any signs of the topological Hall effect or the antiferromagnetic Hall effect, and this contribution is not considered further. Then, approximating the data for the field dependence of the Hall resistivity (Fig. 5) by the field dependence of the magnetization (see Fig. 1b) and following the definition of the Hall effect resistivity (Eq. (1)) without the last term, we can determine the NHE (R_0) and AHE (R_s) coefficients. This technique of separating the contributions of these effects was described in [14]. The result of this separation of the NHE and AHE coefficients is shown in Fig. 5.

The NHE coefficient is relatively weakly dependent on temperature as compared to the earlier studied Heusler alloys undergoing MT [14, 16]. The AHE coefficient increases monotonically with the temperature, as that in low-resistance metals and alloys. For Ni, the AHE coefficient is negative; however, it changes its sign in alloys with other metals in the region of medium concentrations [24]. Therefore, the positive sign of this coefficient in the case under consideration with 49.7% Ni does not contradict this tendency. For the temperature dependence of R_s, the relations of the form $R_s \propto \rho^n$, where $n = 1$ for skew scattering and $n = 2$ for the intrinsic mechanism and the intrinsic mechanism and the side jump mechanism [25], are not applicable. For the current case, the approximation procedure leads to $n = 0.7$, which confirms the inapplicability of the relation $R_s \propto \rho^n$ to describe the temperature dependence of R_s. For the

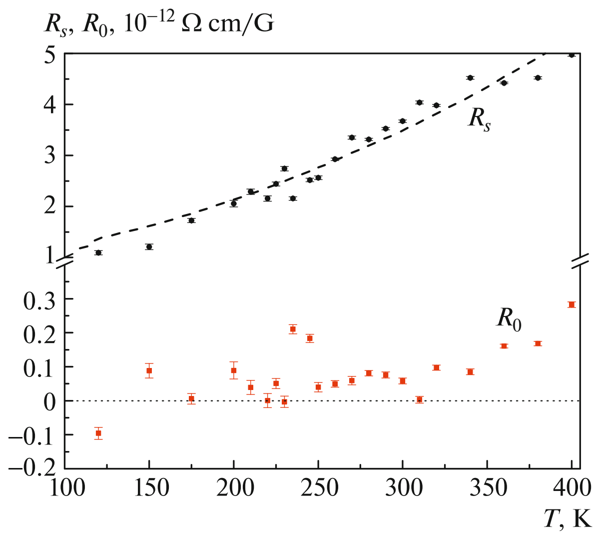

Fig. 5. Temperature dependences of the NHE (R_0) and AHE (R_s) coefficients of the $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ films. (dashed line) Approximation by the relation $R_s = \alpha\rho + \beta\rho^2$ at α [G^{-1}] = $4.92 \times 10^{-8} + 2.34 \times 10^{-22}$ and β [$(\Omega \text{ cm G})^{-1}$] = $4.48 \times 10^{-9} + 4.52 \times 10^{-25}$.

temperature dependence of R_s in ferromagnetic alloys, the authors of [26] proposed the dependence

$$R_s = \alpha\rho + \beta\rho^2. \quad (2)$$

Here the first term describes the skew scattering by impurities and the interference contribution of impurity–phonon scattering, and it is higher than the second term, which describes scattering by phonons and the contributions of the side jump mechanism and the intrinsic mechanism. Taking into account magnetic scattering, i.e., scattering by the temperature fluctuations of the magnetic moment, at elevated temperatures does not change the form of Eq. (2). Figure 5 shows that Eq. (2) describes the experimental data well and the second term is really lower than the first one. Thus, we conclude that the AHE in low-resistance Heusler alloys undergoing a hidden MT behaves similarly to that in homogeneous ferromagnetic alloys and that the temperature dependence of R_s is determined by skew scattering and interference scattering by phonons and impurities.

4. CONCLUSIONS

The MT in $Ni_{49.7}Fe_{17.4}Co_{4.2}Ga_{28.7}$ films is hidden, since it occurs over a wide temperature range and is not accompanied by a magnetic phase transition and the magnetizations of the martensitic and austenitic phases differ slightly. Nevertheless, the MT is clearly pronounced in the behavior of the magnetoresistance, which decreases in the range 100–250 K with increasing temperature in the martensitic phase and then increases in the austenitic phase. This behavior is associated with the suppression of antiferromagnetic correlations at low temperatures and scattering by magnetic moment fluctuations. The AHE coefficient is well described by the relation $R_s = \alpha\rho + \beta\rho^2$, which is typical of homogeneous ferromagnetic alloys. The second term in this relation corresponds to scattering by phonon, the intrinsic mechanism, and the side jump mechanism and is substantially lower than the first term. This fact indicates the dominant role of skew scattering and interference impurity–phonon scattering for the low-resistance composition under study. The experimental data point to insignificant changes in both the total DOS at the Fermi level and the distribution of *d* spin-up and spin-down states during the MT.

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