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Features of the electroresistivity, magnetic and galvanomagnetic characteristics in Co_2MeSi Heusler alloys

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The electro- and magnetotransport as well as magnetic properties of Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) Heusler alloys were studied. The electroresistivity was measured from 4.2 to 300 K, the galvanomagnetic properties (magnetoresistivity and Hall effect) were measured at T = 4.2 K in magnetic fields of up to 100 kOe, and the magnetization at T = 4.2 and 300 K in fields of up to 70 kOe. The normal and anomalous Hall coefficients, saturation magnetization, residual resistivity, current carrier concentration, coefficients at linear contributions into the electroresistivity and magnetoresistivity were obtained. It was shown that on the one hand, there is quite clear correlation between the electronic and magnetic characteristics of Heusler alloys studied, and the spin polarization coefficients of current carriers, taken from well know literature data, on the other hand. The obtained results can be used for creation of new materials for spintronics.

Keywords: Heusler alloys, galvanomagnetic properties, spin polarization, spintronics.

1. Introduction

Heusler alloys [1–3] can be attributed to unique functional materials, since they exhibit a wide variety of practically important properties, for example, a giant magnetocaloric effect [2, 4, 5], shape memory effect [2, 6], unusual thermoelectric, thermal and semiconducting properties [7–10], the properties of topological insulator and semimetal [2, 11], etc. A special place in this series is occupied by Heusler alloys, which are in the state of a half-metal ferromagnet (HMF) [12–15] and a spin gapless semiconductor (SGS) [16–19]. In the HMF state, an interesting feature is observed in the electronic energy spectrum near the Fermi level E_F: for one spin direction (usually for the spin oriented against the direction of magnetization, i.e., spin “down”), there is a wide energy gap (~ 1 eV) at E_F, while for the opposite direction of spin (spin “up”) the band gap is absent at E_F. In SGS materials, there is a wide gap at E_F for spin-down electrons and a zero gap for spin-up electrons (the valence and conduction bands are in a contact). Thus, it is possible to obtain the 100 % spin polarization of charge carriers. Consequently, these compounds can find application in spintronics, which determines the progress in modern technologies for recording, processing and reading information.

Heusler alloys based on cobalt are promising HMF and SGS materials, since many of them have high values of the Curie temperature, magnetic moment, and in some cases, large values of the spin polarization of charge carriers at room temperature [20–22]. Therefore, such compounds can be used for spintronic devices [23–25].

In order to develop and synthesize new Heusler compounds, including those based on cobalt, new information about the features of their electronic structure and magnetic state is needed. Such data can be obtained by studying their electrical resistivity, magnetic and galvanomagnetic properties. Therefore, the purpose of this work is to study experimentally the features of low-temperature electronic transport and magnetic properties of Heusler alloys based on Co, namely, Co_2MeSi, with the change in the Me-component, where Me = Ti, V, Cr, Mn, Fe, Co, Ni.

2. Experimental

Polycrystalline alloys were prepared in an induction furnace in a purified argon atmosphere. Then the obtained Co_2VSi, Co_2CrSi, Co_2FeSi, and Co_2CoSi ingots were annealed at 1100 °C for 3 days and quenched. The Co_2TiSi, Co_2MnSi, and Co_2NiSi alloys were annealed at 800 °C for 9 days with subsequent cooling to room temperature, according to [26–28].

An elemental analysis of the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) alloy system was performed using an Inspect F scanning electron microscope (FEI Company, USA) equipped with a field-emission cathode and an EDAX spectrometer. An elemental composition was determined in at least three regions selected at three different points of the sample. As a rule, two opposite sides of the sample and its middle are selected. Table 1 shows the analysis results.

Table 1. The chemical composition of the Co_2MeSi, where Me = Ti, V, Cr, Mn, Fe, Co, Ni

Alloy	Composition	Co, %	Me, %	Si, %
Co_2TiSi	$Co_{1.98}Ti_{0.95}Si_{1.07}$	49.5	23.75	26.75
Co_2VSi	$Co_{1.98}V_{0.93}Si_{1.09}$	49.5	23.25	27.25
Co_2CrSi	$Co_{1.95}Cr_{0.99}Si_{1.06}$	48.75	24.75	26.5
Co_2MnSi	$Co_{1.9}Mn_{1.01}Si_{1.09}$	47.5	25.25	27.25
Co_2FeSi	$Co_{2.19}Fe_{0.65}Si_{1.16}$	54.75	16.25	29
Co_3Si	$Co_{2.86}Si_{1.14}$	71.5	–	28.5
Co_2NiSi	$Co_{1.87}Ni_{1.03}Si_{1.1}$	46.75	25.75	27.5

According to the Table 1, the deviation from the stoichiometric composition is seen to be less than 5 %, excluding Co_2FeSi.

According to the data of the x-ray structural analysis, all alloys are found to be ordered in the $L2_1$ structure (face-centered cubic lattice), but almost all samples contain an insignificant amount of the second phase, with the exception of Co_3Si.

The electrical resistivity and magnetoresistivity, as well as the Hall effect were measured using conventional techniques, for example, described in [29, 30]. The galvanomagnetic properties were measured using a PPMS setup, and the magnetization on the SQUID magnetometer (Quantum Design) at the Collaborative Access Center "Testing Center of Nanotechnology and Advanced Materials" of the Institute of Metal Physics, UB RAS.

3. Results and discussion

Figure 1 shows the results of measurements of the electrical resistivity temperature dependences $\rho(T)$ of the Co_2MeSi alloy system.

All samples are found to have a metallic type of conductivity. In addition, the dependences for Co_2MeSi alloys (Me = V, Cr) tend to saturation, while the dependence for Co_2MeSi alloys (Me = Ti, Fe) is superlinear; at Me = Ni, Co, and Mn — linear at temperatures above 100 K.

Table 2 shows the values of the residual resistivity ρ_0 of the studied alloys, which is defined as the value of the electrical resistivity at a liquid helium temperature of 4.2 K (Fig. 1). The values of the residual resistivity of the investigated alloys are found to differ significantly from low values for the Co_2MnSi, Co_2FeSi, and Co_3Si alloys (from 16 to 69 μΩ·cm) to high values of ρ_0 for the Co_2VSi and Co_2CrSi alloys (294 and 318 μΩ·cm). In this case, the values of ρ_0 differ by almost an order of magnitude. Besides, it should be noted that it is for these "high-resistivity" Co_2VSi and Co_2CrSi alloys with large values of ρ_0 the $\rho(T)$ dependence is observed with saturation at high temperatures.

It is known [31] that at low temperatures in ferromagnets, the temperature dependence of the electrical resistivity of a metal can be expressed using formula (1):

$$\rho(T) = \rho_0 + AT + BT^2, \quad (1)$$

where ρ_0 is the residual resistivity, A and B are constants.

Table 2. Residual resistivity ρ_0, coefficients A and B at linear and quadratic terms of $\rho(T)$, saturation magnetization M_s, Curie temperature T_C, normal R_0 and anomalous R_s Hall coefficients, main type of charge carriers, their concentration n and mobility μ, coefficient k at linear term of magnetoresistivity of the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) Heusler alloys

Alloy	Co_2TiSi	Co_2VSi	Co_2CrSi	Co_2MnSi	Co_2FeSi	Co_3Si	Co_2NiSi
ρ_0, μΩ·cm (T = 4.2 K)	155	294	318	16	27	69	131
A, 10^{-2} μΩ·cm·K^{-1}	–2.89	11.52	44.59	–1.62	0.18	17.41	1.4
B, 10^{-3} μΩ·cm·K^{-2}	1.71	0.06	–1.97	0.44	0.23	–0.21	0.35
M_s, emu/g	48.8	5.9	1.0	114.2	96.7	67.9	45.9
T_C, K	385[32]	566 [32]	747 [32]	985 [32]	1100 [32]	622 [33]	589 [33]
R_0, 10^{-4} cm^3/C	7.23	–1.21	–5.01	–1.50	1.83	1.26	–0.77
R_s, 10^{-2} cm^3/C	4	29	46	0.04	0.05	2	2
Main type of charge carriers	holes	electrons	electrons	electrons	holes	holes	electrons
n, 10^{22} cm^{-3}	0.9	5	1	4	3	5	8
μ, cm^2/(V·s)	4.7	0.4	1.6	9.7	6.7	1.8	0.6
k, 10^{-3} kOe^{-1}	–2.2	20.8	22.7	–1.9	1.5	–15.1	–5.1

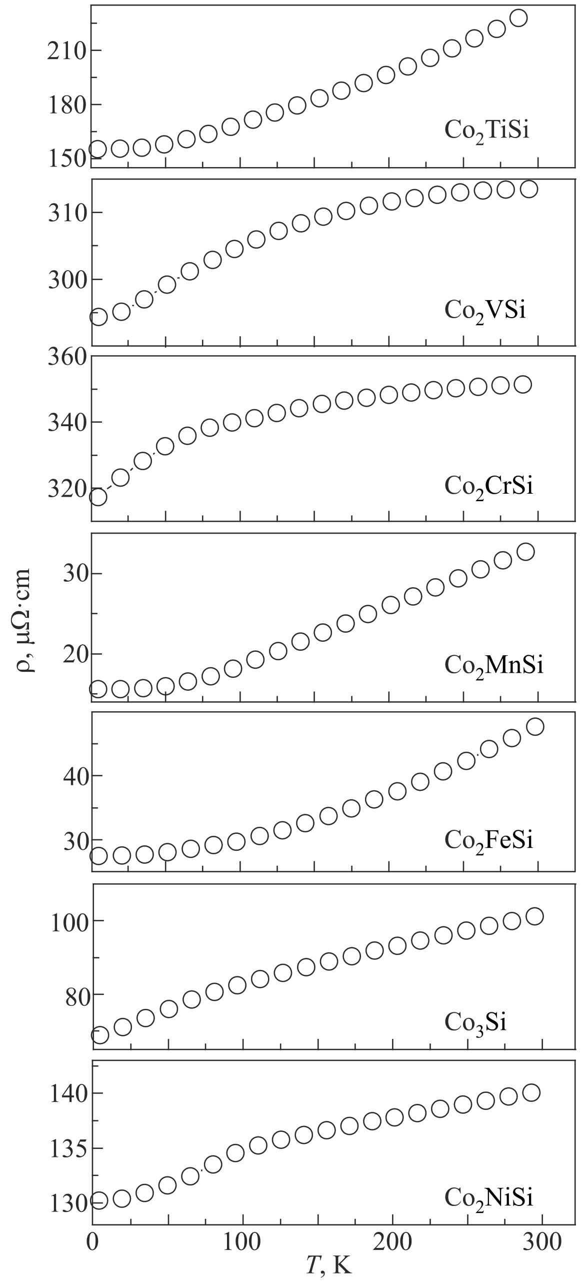

Fig. 1. Temperature dependences of the electrical resistivity of the Co_2MeSi alloys.

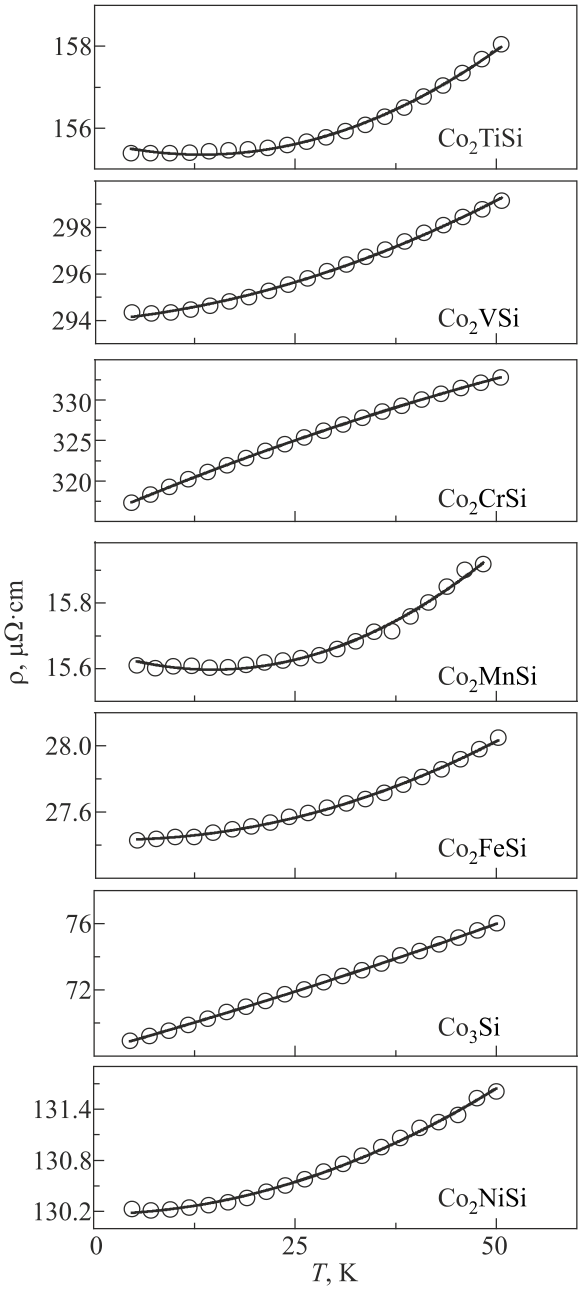

Fig. 2. Temperature dependence of electrical resistivity ρ(T) at 4.2 K ≤ T ≤ 50 K. Here, the dots denote the experimental data, and the solid lines denote the data calculated by the formula (1).

This type of low-temperature dependence of the electrical resistivity is characteristic of ferromagnetic alloys because the processes of conduction electrons scattering by magnetic inhomogeneities become significant in the low-temperature region. The interaction of current carriers with the spin magnetic subsystem due to the *s*-*d* exchange coupling or due to the spin-orbit interaction leads to additional linear and quadratic contributions. Figure 2 demonstrates that the dependence of the form (1) is observed for all investigated alloys. The coefficients *A* and *B* determined in this way are presented in Table 2.

Apparently, the observed differences in the values of the residual resistivity, the form of the temperature dependences of the electrical resistivity, as well as the coefficients *A* and *B* can be associated both with the features of the electronic structure (density of states at the Fermi level E_F) and with the processes of scattering of charge carriers in a particular alloy, depending on the magnetic state of a particular compound, among other things.

Additional information on the electronic and magnetic characteristics of the studied alloys can be obtained from measurements of their magnetic and galvanomagnetic properties.

Figure 3 shows that the $M(H)$ dependences of most of the investigated alloys (except for the alloy with V, Cr) saturate in fields exceeding 10 kOe. Using the obtained data (Fig. 3), the values of the saturation magnetization M_s

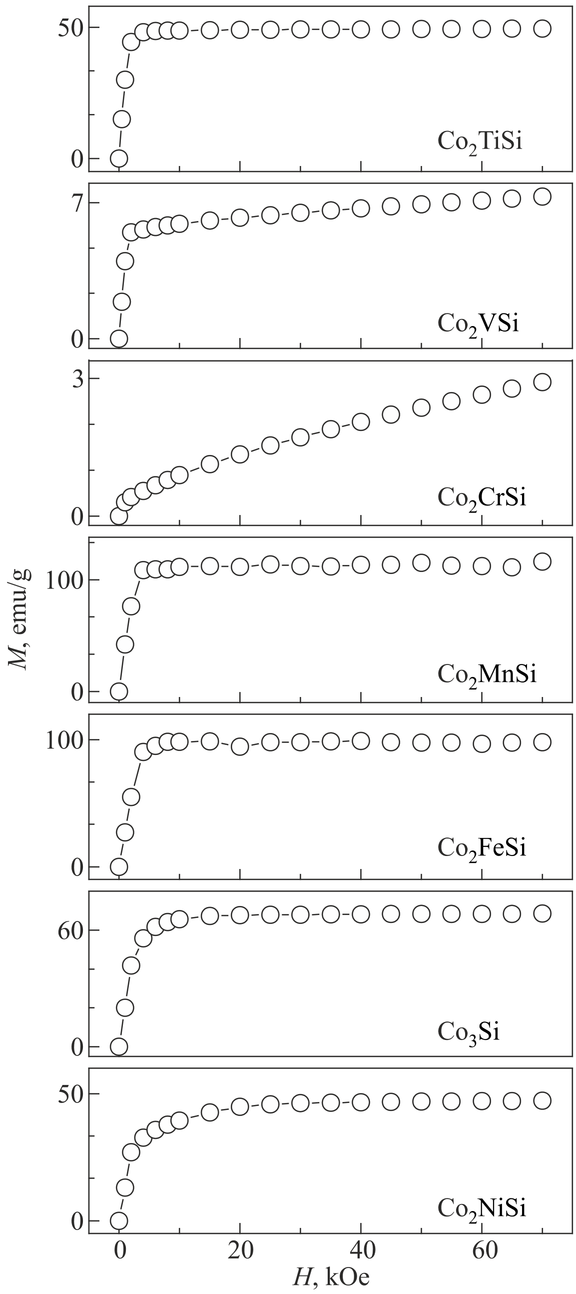

Fig. 3. Field dependences of the magnetization $M(H)$ for the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) alloy system at $T = 4.2$ K.

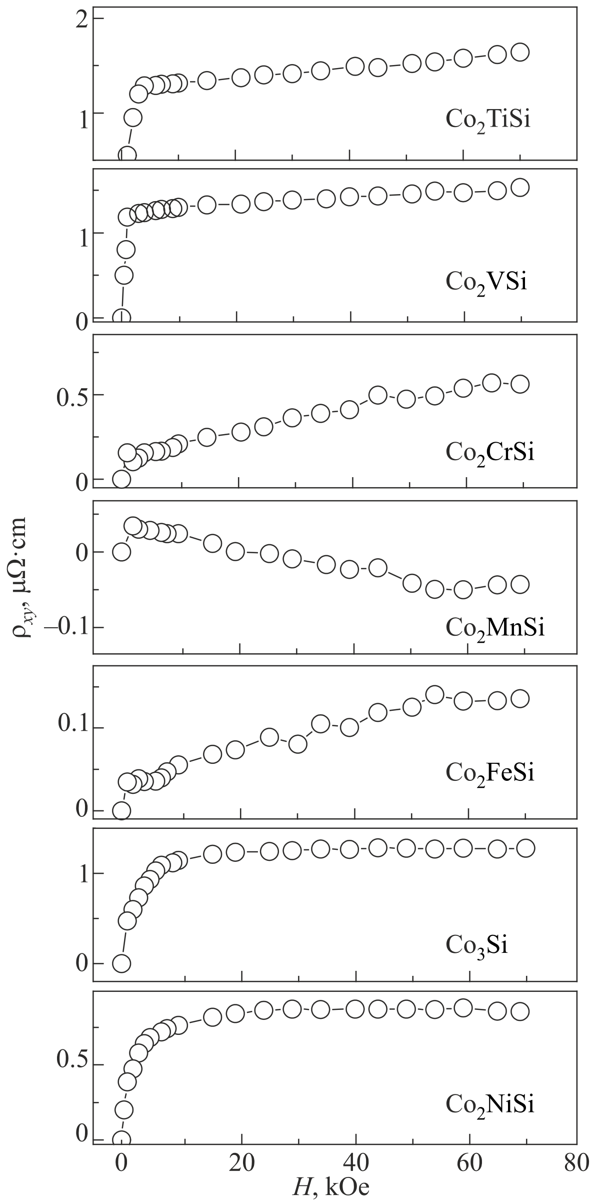

Fig. 4. Field dependences of the Hall resistivity ρ_{xy} (H) for the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) alloy system at $T = 4.2$ K.

of all alloys were determined (Table 2). The values of M_s are found to differ significantly as well. According to [20], at temperatures below the Curie point, all alloys of the Co_2MeSi system are in a ferromagnetic state. The values of the Curie temperature T_C for all investigated alloys [32, 33] are presented in Table 2.

Figure 4 shows the field dependences of the Hall resistivity. Their form is seen to coincide largely with the field dependences of the magnetization (Fig. 3).

At a liquid helium temperature $T = 4.2$ K, all alloys of the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) system are in a ferromagnetic state. Therefore, both normal and anomalous Hall effects should be observed in them.

It is known that in ferromagnets the Hall coefficient R contains both normal R_0 and anomalous R_s components. Formula (2) was used to separate them. Provided that the demagnetizing factor N of the samples is close to 1, we obtain formula (2):

$$\rho_{xy}/H = R_0 + 4\pi R_s M/H. \qquad (2)$$

According to the formula (2), a dependence of the form $\rho_{xy}/H = f(M/H)$ should be observed, from which it is possible to determine the values of both the normal Hall coefficient (NHE) R_0 and the anomalous Hall coefficient (AHE) R_s.

Figure 5 shows the dependence $\rho_{xy}/H = f(M/H)$. In the limit of strong fields ($H > 10$ kOe), this formula (2) is seen to be indeed valid for all investigated alloys. Using

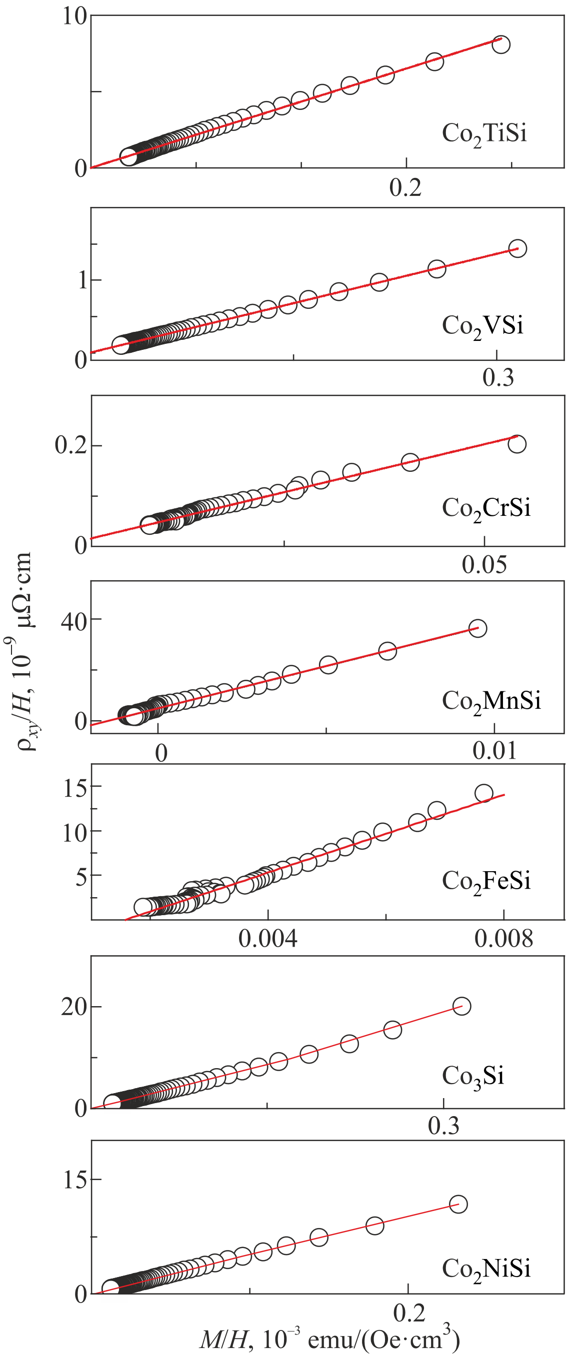

Fig. 5. The dependence ρ_{xy}/H from (M/H) for the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) alloy system.

the obtained data (Fig. 5), the values of the coefficients of the normal R_0 and anomalous R_s of the Hall effect were determined (Table 2).

Table 2 shows that the R_s coefficient exceeds R_0 by two to three orders of magnitude, which is typical for ferromagnetic alloys.

Knowing the sign and value of the NHE coefficient, as well as the value of the electrical resistivity of a particular compound, it is possible to estimate the type, concentration and mobility of charge carriers using formulas (3) and (4).

$$n = \frac{1}{qR_0}, \tag{3}$$

where q — electron charge equal to $1{,}6\cdot 10^{-19}$C.

$$\mu = \frac{R_0}{\rho_0}, \tag{4}$$

where ρ_0 — alloy resistivity without magnetic field at $T = 4.2$ K (residual resistivity).

The values presented in Table 2 are observed to be typical for metals. It should be noted that the measurements were carried out on polycrystalline samples; therefore, the estimates of n and μ are only qualitative. In addition, formula (3) was used for the estimation, i.e., the one-band approximation was used, although in fact the investigated alloys have a much more complex electronic structure. In this case, the Fermi surface of each of the investigated compounds consists of several sheets, both the electronic and hole types. Consequently, to accurately determine the sign, concentration and mobility of current carriers, their partial contributions to the galvanomagnetic properties, it is necessary to have data on the details of their Fermi surfaces. As far as we know, such data are currently lacking.

It is interesting to study the behavior of the magnetoresistivity (MR) with a change in the Me component in the studied system of Co_2MeSi alloys. Figure 6 shows the field dependences of the magnetoresistivity.

Figure 6 shows that in alloys of the Co_2MeSi system, where Me = Ti, Co, and Ni, negative magnetoresistivity is observed, while there are positive magnetoresistivity in alloys with Me = Fe, V, and Cr. The Co_2MnSi alloy has a positive magnetoresistivity in low fields with a change of sign to negative in fields above 90 kOe. In this case, for all compounds, a magnetoresistivity linear in the magnetic field is observed: either in the entire range of fields (over 15 kOe), or in individual high-field regions (for example, for alloys with Mn and Co).

For the magnetoresistivity of all studied alloys in the range of magnetic fields from 50 to 100 kOe, a contribution that is linear in the magnetic field can be distinguished (Table 2). Then the field dependence of the magnetoresistivity can be represented in the form (5):

$$\frac{\Delta\rho}{\rho_0} = k_0 + kH, \tag{5}$$

where k_0 — field independent, k — linear in magnetic field contributions.

According to theoretical calculations [34], under certain conditions, the so-called two-magnon scattering processes can arise in HMFs, leading to a specific dependence of the electrical resistivity $\rho \sim T^n$, where $3.5 \le n \le 4.5$, as well as a negative and linear contribution to the MR. It is possible that the experimentally observed (Fig. 6) linear

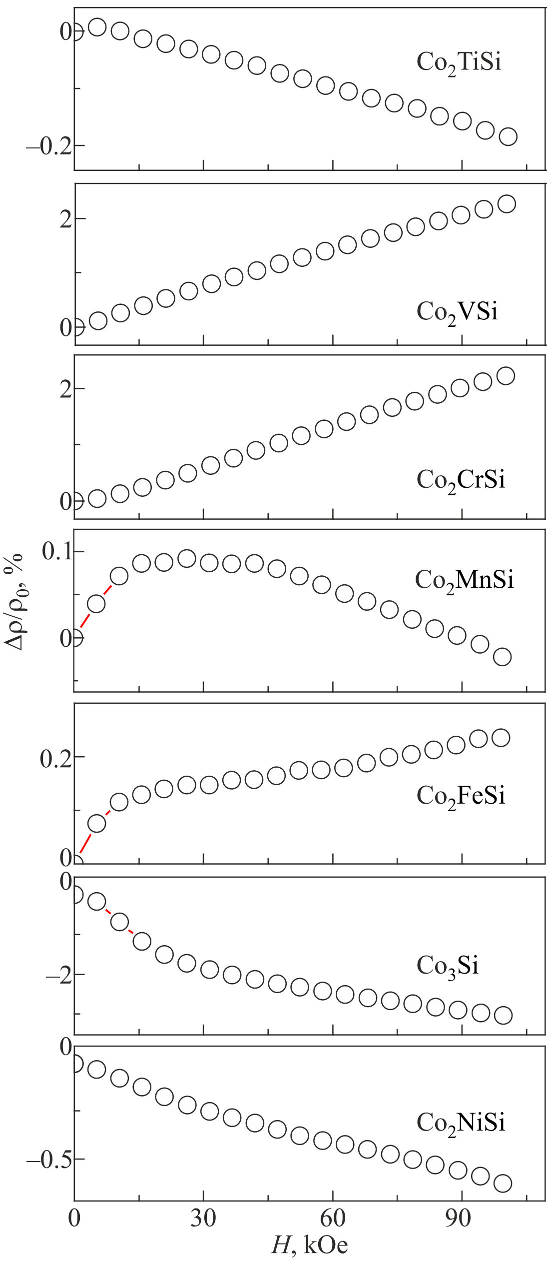

Fig. 6. Transverse magnetoresistivity of the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) alloy system.

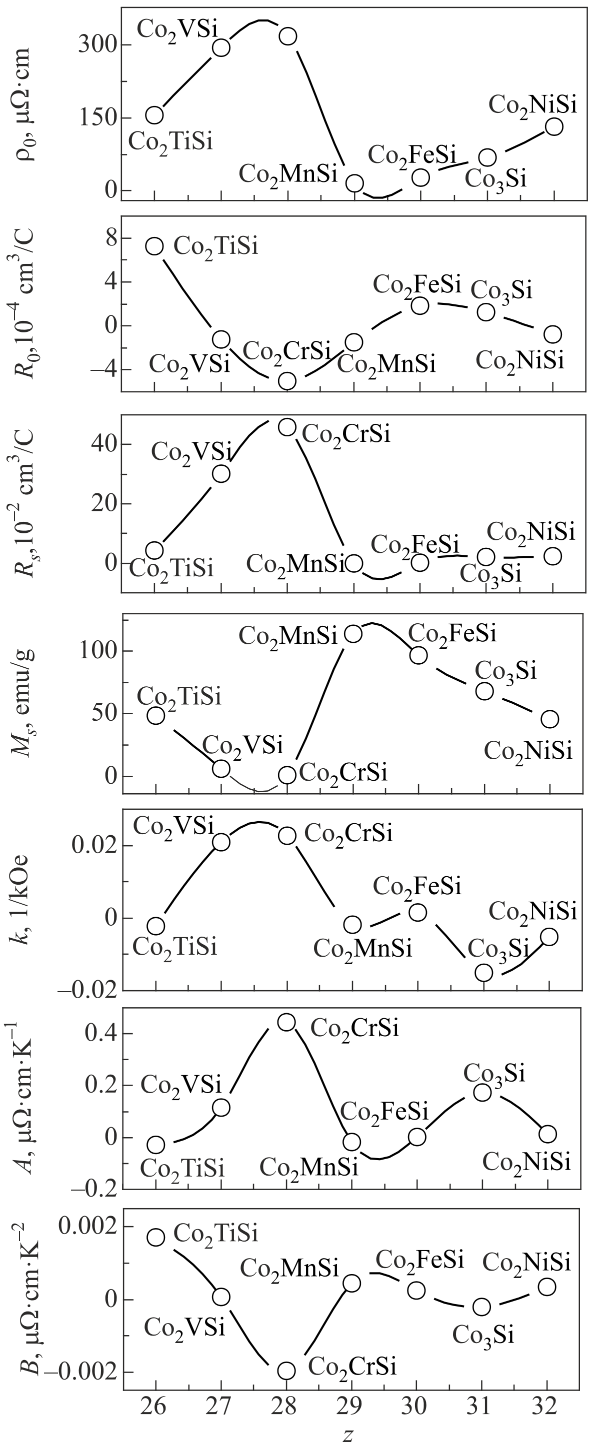

Fig. 7. The dependence of residual resistivity ρ_0, coefficients of normal R_0 and anomalous R_s Hall effect, saturation magnetization M_s. A and B constants, which determine the contribution to the electrical resistivity at low temperatures, and k determining the contribution to the linear magnetoresistivity from the number of valence electrons z in the Co_2MeSi alloy system, where Me = Ti, V, Cr, Mn, Fe, Co, Ni.

and negative magnetic field dependences of the MR are a manifestation of the mechanism of two-magnon scattering of charge carriers [34].

Besides, it should be noted that a similar field dependence of the MR for the Co_2FeSi single crystal alloy was observed in [22]. In this case, the contribution that is linear and positive in the magnetic field at low temperatures, as in our case at $T = 4.2$ K, was replaced by negative and also linear as well in the field at temperatures above 120 K [22].

Earlier, in [15, 35], a strong change and correlation of macroscopic properties, electronic parameters, and magnetic characteristics on the number of valence electrons was observed with a change in the Y and/or Z components in the Heusler alloys Fe_2YAl, Co_2YAl, and Co_2FeZ. It can be assumed that a similar correlation can be observed in the case of the Co_2MeSi system.

Figure 7 shows a summary graph of the dependences of the residual resistivity ρ_0, the coefficients of the normal R_0, the anomalous R_s of the Hall effect, the saturation magnetization M_s, as well as the constants A, B, which determine the contribution to the electrical resistivity at low temperatures, and k, which determine the contribution to the magnetoresistivity in fields from 50 to 100 kOe of the number of valence electrons z in alloys of the Co_2MeSi system when z changes from 26 (for Ti) to 32 (for Ni).

Figure 7 shows clear correlation between the presented values of ρ_0, R_0, R_s, M_s, A, B, and k. Thus, the anomalous Hall coefficient R_s and the residual resistivity ρ_0, the coefficients A and k increase at $z < 28$, while the normal Hall coefficient R_0, the magnetization M_s and the coefficient B decrease. These correlations are possibly related to the HMF- and SGS-states. For example, the Co_2MnSi and Co_2FeSi alloys were demonstrated to be half-metallic ferromagnets [21, 36]. Figure 7 shows that these compounds reveal the maximum values of magnetization and minimum values of residual resistivity. Apparently, these alloys predominantly contain charge carriers providing the “metallic” type of conductivity, i.e., with spin “up”, as well as a large magnetic moment, which should lead to high spin polarization of carriers. It would be interesting to experimentally determine the spin polarization coefficient in these alloys, as well as to study its behavior with a change in the number of valence electrons.

The spin polarization coefficient of current carriers is determined by the expression (6):

$$P(x) = \frac{n_\uparrow(x) - n_\downarrow(x)}{n_\uparrow(x) + n_\downarrow(x)}, \tag{6}$$

where $n_\uparrow(x)$ and $n_\downarrow(x)$ are the concentration of electrons with spin-up and spin-down orientations, respectively.

Figure 8 shows the calculated and experimental values of the coefficients of spin polarization of current carriers taken from [21, 32, 33, 36] on the number of valence electrons. Methods for determining spin polarization: [32, 33] VASP + GGA functional, [36] Point Contact Andreev Reflection (PCAR) spectroscopy, [21] Spin resolved ultraviolet-photoemission spectroscopy (SRUPS).

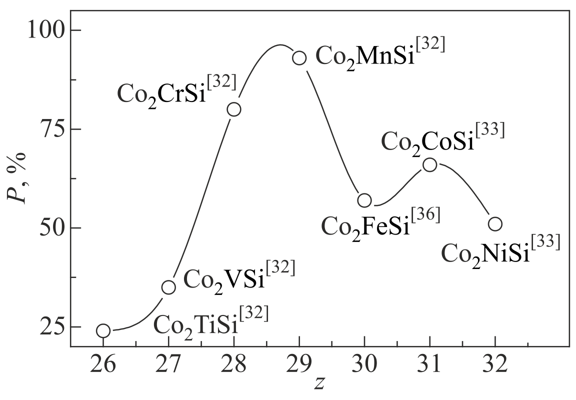

Fig. 8. Spin polarization versus the number of valence electrons *z*.

Figure 8 shows that when the number of valence electrons changes from Ti to Mn, the spin polarization of electrons increases (maximum 93 %). The spin polarization values shown in Fig. 8 are consistent with the previously stated assumption that the Co_2MnSi alloy has a high spin polarization. It seems very interesting to analyze the behavior of the spin polarization with a change in the number of valence electrons and compare it with the data in [20].

4. Conclusions

Thus, according to the results of an experimental study of the low-temperature electronic transport and magnetic properties of the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) Heusler alloys, the following main conclusions can be drawn.

The temperature dependences of the electrical resistivity of Co_2MeSi alloys (Me = V, Cr) are established to tend to saturation, while the $\rho(T)$ dependences for Co_2MeSi alloys (Me = Ti, Fe) are superlinear, and for Co_2MeSi alloys (Me = Ni, Co, and Mn), the $\rho(T)$ dependences are linear at temperatures above 100 K.

The residual resistivity ρ_0 of the investigated alloys is shown to differ significantly from low values for the Co_2MnSi, Co_2FeSi, and Co_3Si alloys (from 16 to 69 μΩ·cm) to high values of ρ_0 for the Co_2VSi and Co_2CrSi alloys (294 and 318 μΩ·cm). It should be noted that it is precisely for these “high-resistivity” alloys (Co_2VSi and Co_2CrSi) with large values of ρ_0 that the $\rho(T)$ dependence is observed with saturation at high temperatures.

It is found that negative magnetoresistivity is observed in Co_2MeSi alloys (Me = Ti, Co, and Ni), while positive magnetoresistivity is observed in alloys with Me = Fe, V, and Cr. In addition, the Co_2MnSi alloy has a positive magnetoresistivity in low fields with a change of sign to negative in fields above 90 kOe. In this case, for all compounds, a linear magnetoresistivity is observed in the magnetic field: either in the entire range of fields (over 15 kOe), or in strong fields above 50 kOe (for example, for the Co_2MnSi and Co_2CoSi alloys). It is assumed that the experimentally observed linear and negative in the magnetic field dependences of the magnetoresistivity can be a manifestation of two-magnon scattering processes of charge carriers, which are characteristic of half-metallic ferromagnets.

It was found that between the values of the residual electrical resistivity ρ_0, the saturation magnetization M_s, the coefficients of the normal R_0 and anomalous R_s of the Hall effects, the coefficients A, B for a linear and quadratic in temperature contributions to the low-temperature electrical resistivity, and the coefficient k as well for a contribution to the magnetoresistivity that is linear in the magnetic field above 50 kOe, a clear correlation is observed depending on the number z of valence electrons.

A comparison of the literature data on the coefficient of current carriers spin polarization *P* and the experimental data obtained in this work indicates a good correlation between them as well.

Thus, new information has been obtained on the features of the electronic and magnetic characteristics of the Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni) Heusler alloys, which may be useful in choosing the optimal materials for spintronic devices.

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Особливості електроопору, магнітних та гальваномагнітних властивостей сплавів Гейслера Co_2MeSi

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Вивчено електро- та магнітотранспортні, а також магнітні властивості сплавів Гейслера Co_2MeSi (Me = Ti, V, Cr, Mn, Fe, Co, Ni). Електроопір виміряно у температурному інтервалі від 4,2 до 300 К, гальваномагнітні властивості (магнітоопір та ефект Холла) — при $T = 4{,}2$ К у магнітних полях до 100 кЕ, намагніченість — при $T = 4{,}2$ і 300 К у полях до 70 кЕ. З експерименту отримано значення нормального та аномального коефіцієнта Холла, намагніченості насичення, залишкового опору, концентрації носіїв струму, коефіцієнтів при лінійних внесках в електроопір та магнітоопір. Показано, що існує досить чітка кореляція між електронними й магнітними характеристиками досліджуваних сплавів Гейслера, з одного боку, та коефіцієнтами спінової поляризації носіїв струму, взятими з добре відомих літературних даних, з іншого боку. Отримані результати можуть бути використані для створення нових матеріалів для спінтроніки.

Ключові слова: сплави Гейслера, гальваномагнітні властивості, спінова поляризація, спінтроніка.