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Magnetic and thermal properties of alloys close in composition to the spin gapless semiconductor Mn_2CoAl

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ABSTRACT

The field dependence of magnetization at $T = 4.2$ K and in magnetic fields of up to 70 kOe, temperature dependences of magnetization (2 K < T < 300 K), heat capacity (2 K < T < 30 K) and magnetic susceptibility (2 K < T < 1000 K) for $Mn_{1.99}Co_{0.96}Al_{1.05}$ and $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloys, closed in composition to the Mn_2CoAl spin gapless semiconductor, were studied. Alloys studied were demonstrated to be the band ferromagnets. Their high-field (H > 11 kOe) magnetization is described in the Stoner models with the Rhodes-Wohlfarth parameter $p_{RW} = 1.3$ for $Mn_{1.99}Co_{0.96}Al_{1.05}$ and $p_{RW} = 2.3$ for $Mn_{1.79}Co_{1.25}Al_{0.96}$. When the composition deviates from the stoichiometric Mn_2CoAl, the spontaneous moment decreases slightly, the effective moment, on the contrary, increases. In this case, a negative sign of the temperature-independent component of the paramagnetic susceptibility is observed. The density of states $n(E_F)$ at Fermi level and the Debye temperature Θ_D of studied alloys have the usual values for 3d-metal alloys.

1. INTRODUCTION

The huge family of Heusler alloys, which can be considered as intermetallic compounds, is very attractive for practical applications due to their interesting and diverse physical properties (see, for example, reviews Refs. 1 and 2 and references therein). The Heusler compounds family has more than 1500 alloys which exhibit various functional properties, i.e., magnetocaloric effect,[3,4] unusual thermal,[5,6] thermoelectric[7] and semiconductor[8–10] characteristics, properties of half-metallic ferromagnets[11,12,18] and spin gapless semiconductors.[13,14,19] The so-called full and inverse Heusler alloys are studied the most. These alloys have the same general formula X_2YZ, where X and Y are 3d metals, and Z are s, p element, but a different crystal structure: $L2_1$ of the Cu_2MnAl-type (full) and X_a of the Hg_2CuTi-type (inverse). According to many *ab initio* calculations, alloys with the $L2_1$ structure are of interest as materials such as half-metallic ferromagnets,[15–18] and inverse alloys are interesting as gapless semiconductors with ferrimagnetic properties or spin gapless semiconductors (SGS).[14,19] Calculations show that the X_a structure is characteristic of the Mn_2CoAl alloy.[20]

According to Ref. 19, SGS materials have many unique properties associated with their unusual zone structure. This is the presence of a wide ($\Delta E \sim 1$ eV) gap at the Fermi level for current carriers with spin-down and zero energy gap for carriers with spin-up. In such SGS materials, strong ferromagnetism is expected, namely, a high Curie temperature and 100% charge carrier polarization at room temperature. Therefore, they can be promising objects for practical applications in spintronic instruments and devices. Zone calculations[20–23] and experimental studies of transport,[21,24] magnetic,[20,21,23] and optical[25] properties showed that this class of materials includes the Mn_2CoAl alloy ordered in a structure of the $F\bar{4}3m$ (Hg_2CuTi) type. According to Ref. 23, this alloy has ferromagnetic ordering at temperatures below $T_C = 720$ K, and its magnetic moment in the ground state is $2\mu_B$/f.u.

However, it was shown in Refs. 21 and 22 that with decreasing c/a lattice constant ratio in Mn_2CoAl, the energy gap in the electron spectrum completely closes. Moreover, even when a cla ratio corresponds to normal conditions, any type of disorder (mutual substitutions of Mn, Co, or Al atoms) leads to an increase in the

density of states for electrons with a spin-up projection, whereas for electrons with a spin-down projection, the energy gap decreases or even closes in some cases. Despite a large number of calculations, currently, from an experimental point of view, inverse alloys are not sufficiently studied. Therefore, it is of interest to synthesize and study the behavior of various physical properties of Mn_2CoAl-based alloys both near the stoichiometric composition and when deviating from stoichiometry.

In this work, the $Mn_{1.99}Co_{0.96}Al_{1.05}$ with an almost stoichiometric composition and the $Mn_{1.79}Co_{1.25}Al_{0.96}$ with a deviation from the stoichiometry alloys were prepared. Their magnetic properties were studied in a wide range of magnetic fields ($H \leq 70$ kOe) and temperatures ($2\ K < T < 1000\ K$), as well as low-temperature ($2\ K < T < 30\ K$) heat capacity for the $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloy.

2. EXPERIMENTAL

The alloys were melted in an induction furnace in a purified argon atmosphere. Then, the alloys were annealed for 48 h at 800 K in an argon atmosphere, followed by cooling to room temperature at a rate of about 100 K/h.

Samples for measurements of the magnetic susceptibility, magnetization, and heat capacity were cut out from the obtained ingots using the electro spark method. The surface layer damaged as a result of electric spark cutting was removed by grinding, chemical etching and electro polishing, as described in Refs. 26 and 27.

The atomic content of elements in the alloys was monitored using a FEI Company Quanta 200 scanning electron microscope equipped with an EDAX x-ray microanalysis attachment. Studies have shown that the resulting alloys have the $Mn_{1.99}Co_{0.96}Al_{1.05}$ and $Mn_{1.79}Co_{1.25}Al_{0.96}$ compositions. Structural certification of the samples was performed at the Collaborative Access Center «Testing Center of Nano-technology and Advanced Materials» of the Institute of Metal Physics, UB RAS.

The field and temperature dependences of the magnetization $M(H, T)$ were measured using the SQUID magnetometer MPMS XL7 (Quantum Design) and the VSM 7407 vibromagnetometer (LakeShore), the heat capacity was measured using a Quantum Design RPMS-9 system at the Collaborative Access Center «Testing Center of Nanotechnology and Advanced Materials» of the Institute of Metal Physics, UB RAS.

3. RESULTS AND DISCUSSION

3.1. Magnetic properties

The results of measurements of the magnetization curves $M(H)$ at $T = 4.2$ K [Figs. 1(a) and 2(a)] show that the process of technical magnetization in the $Mn_{1.99}Co_{0.96}Al_{1.05}$ alloy ends at $H \geq 4$ kOe, and in the alloy with a large deviation from stoichiometry $Mn_{1.79}Co_{1.25}Al_{0.96}$ at $H \geq 11$ kOe. In stronger magnetic fields, the processes of technical magnetization are practically absent and both compounds are in a single-domain state. In the case of the band nature of magnetism, the magnetization here should be determined mainly by the rearrangement of the electronic band structure of the alloys in a magnetic field. Based on the Stoner model,[28] the high-field magnetization in the considered zone ferromagnets

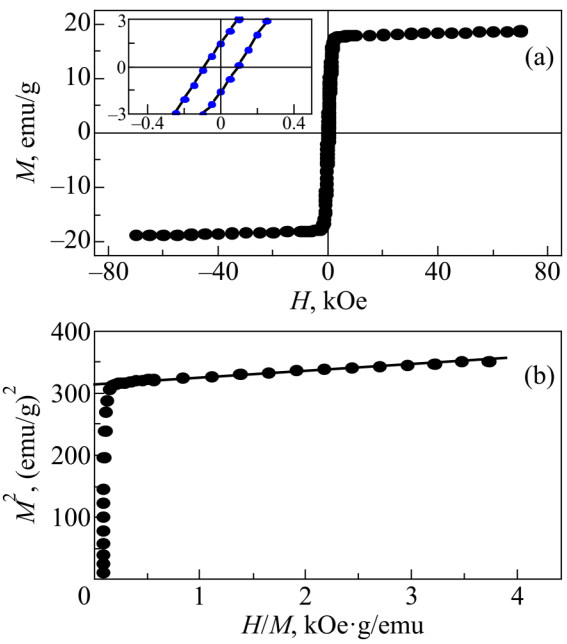

FIG. 1. (a) Field dependences of the magnetization $M(H)$ at $T = 4.2$ K for the $Mn_{1.99}Co_{0.96}Al_{1.05}$ alloy. (b) Dependence of M^2 on H/M for the $Mn_{1.99}Co_{0.96}Al_{1.05}$ alloy.

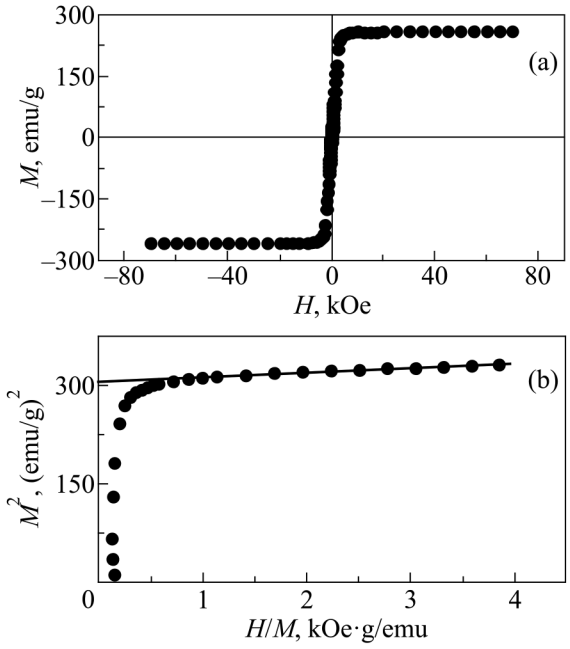

FIG. 2. (a) Field dependences of the magnetization $M(H)$ at $T = 4.2$ K for the $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloy. (b) Dependence of M^2 on H/M for the $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloy.

should be described by the relation[29]

$$M^2 = M_S^2 + 2\chi_0 M_0^2\, H/M, \quad (1)$$

where M_S is the spontaneous magnetization, M_0 is the saturation magnetization, and χ_0 is the susceptibility of the paraprocess. In turn,

$$M_S^2 = M_0^2[1 - (T/T_C)^2]. \quad (2)$$

Figures 1(b) and 2(b) show the dependences of M^2 on H/M for the studied $Mn_{1.99}Co_{0.96}Al_{1.05}$ and $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloys, measured at $T = 4.2$ K. It can be seen that relation (1) is valid in the region of the single domain for both compounds. In this case, the values of spontaneous magnetization M_S in the studied alloys have rather close values: $M_S = 17.8$ emu/g and $\mu_S = 0.62\,\mu_B$/f.u. for the $Mn_{1.99}Co_{0.96}Al_{1.05}$ alloy, and $M_S = 17.5$ emu/g and $\mu_S = 0.56\,\mu_B$/f.u. for $Mn_{1.79}Co_{1.25}Al_{0.96}$.

The validity of relation (2) for the considered alloys can be checked by taking into account the values of the paramagnetic Curie temperatures θ determined from measurements of the paramagnetic susceptibility (Fig. 3). The $M(T)$ dependences in the field $H = 50$ kOe are shown in Fig. 4. Due to the absence of the M_S values of the studied alloys in a wide temperature range, the procedure described earlier in Ref. 30 was used to verify the validity of relation (2). Considering that in the region of strong magnetic fields ($H \geq 11$ kOe) the magnetization is $M \approx M_S$, the square of the saturation magnetization M_S^2 in expression (2) can be replaced by M^2.

Figure 5 shows the dependences of the square of magnetizations M^2 obtained at $H = 50$ kOe on $(T/\theta)^2$. It can be seen that for the $Mn_{1.99}Co_{0.96}Al_{1.05}$ alloy there is a fairly wide temperature range where the value of M^2 is proportional to $(T/\theta)^2$ indeed. This indicates the validity of relation (2) for the $Mn_{1.99}Co_{0.96}Al_{1.05}$ band ferromagnet and allows us to determine for it the values of the magnetic moment per formula unit $\mu_0 = 0.65\,\mu_B$/f.u. and paraprocess susceptibility $\chi_0 = 1.5\cdot10^{-5}$ cm^3/g. However, band calculations for the spin gapless semiconductor Mn_2CoAl ordered in the $F\bar{4}3m$ -type structure give the value $\mu = 2\,\mu_B$/f.u.[20–22] As noted above, any deviation from stoichiometry or disordering removes this alloy from the state of a spin gapless semiconductor. This may be one of the explanations for the deviation of the experimentally determined value of μ_0 for a given alloy from μ_0 defined in Refs. 20–22.

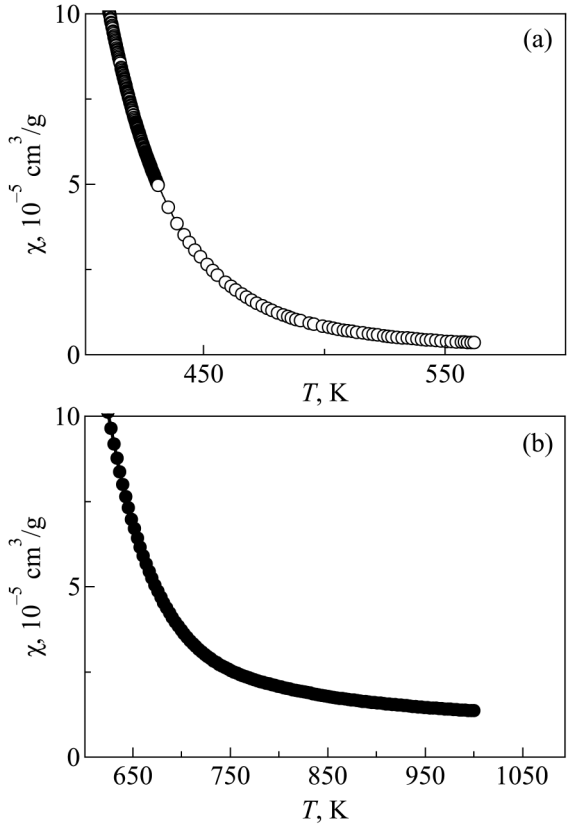

FIG. 3. Temperature dependences of paramagnetics susceptibility: (a) $Mn_{1.99}Co_{0.96}Al_{1.05}$; (b) $Mn_{1.79}Co_{1.25}Al_{0.96}$. Solid lines show the calculation results using formula (3).

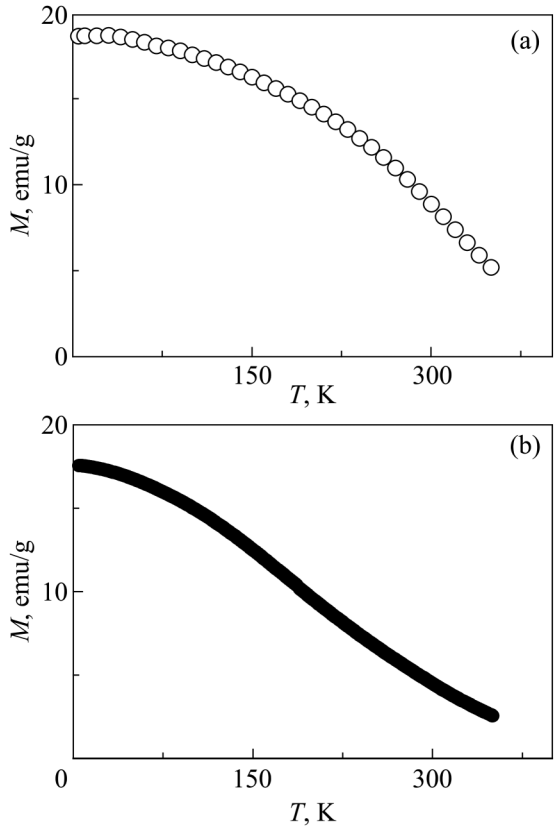

FIG. 4. Temperature dependences of magnetization in a magnetic field $H = 50$ kOe: (a) $Mn_{1\ 99}Co_{0\ 96}Al_{1.05}$; (b)$Mn_{1.79}Co_{1\ 25}Al_{0\ 96}$.

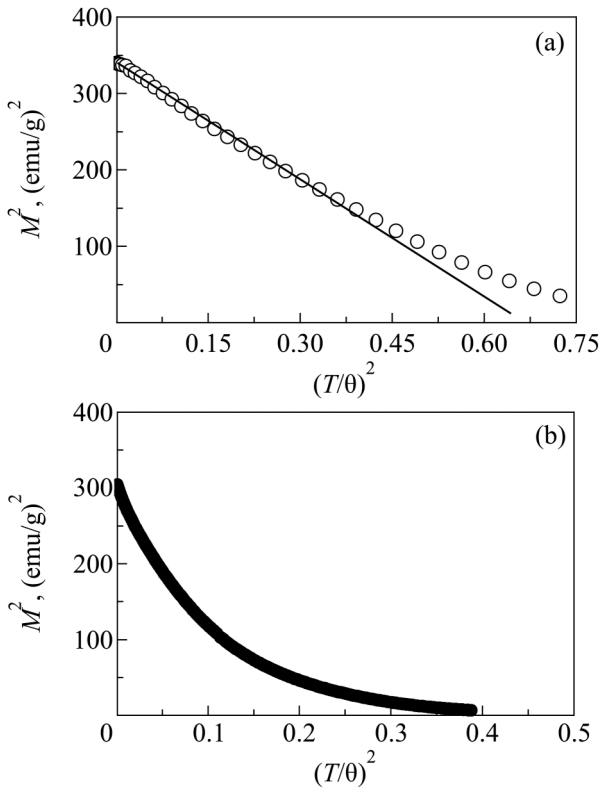

FIG. 5. Dependences of the square of the magnetization measured at $H = 50$ kOe on $(T/\theta)^2$: (a) $Mn_{1.99}Co_{0.96}Al_{1.05}$; (b)$Mn_{1.79}Co_{1.25}Al_{0.96}$.

On the contrary, in the $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloy, with a composition farther from the stoichiometric, the linear section is practically absent in the dependence $M^2 = f\,[(T\,/\,\theta)^2]$. Therefore, relation (2) is not satisfied with this alloy. Most likely, this is since the high-field magnetization values in this case significantly differ from the M_S in the entire temperature range studied.

In the considered alloys, at temperatures above the Curie point, the paramagnetic susceptibility $\chi(T)$ was measured. It can be seen from Fig. 3 that at $T > \theta$ in a fairly wide temperature range susceptibility $\chi(T)$ of the studied alloys is described by the modified Curie–Weiss law

$$\chi(T) = \chi_0 + C/(T - \theta). \tag{3}$$

Here Curie constant $C = \mu_{eff}{}^2/8M$, where μ_{eff} is the effective magnetic moment per atom, M is the molecular weight calculated per atom. This allows us to determine the numerical values of μ_{eff} and θ in the band ferromagnets under consideration. For the $Mn_{1.99}Co_{0.96}Al_{1.05}$ alloy, $\theta = 413$ K, and for $Mn_{1.79}Co_{1.25}Al_{0.96}$ $\theta = 620$ K. Accordingly, the squares of the effective magnetic moment are $\mu_{eff}{}^2 = 1.18(\mu_B/\text{at.})^2$ for $Mn_{1.99}Co_{0.96}Al_{1.05}$ and $\mu_{eff}{}^2 = 3.43(\mu_B/\text{at.})^2$ for $Mn_{1.79}Co_{1.25}Al_{0.96}$. From the results of measurements of low-temperature magnetization, we can obtain the Rhodes- Wohlfarth parameter $p_{RW} = \mu_C/\mu_S$, where μ_S is the spontaneous magnetic moment per atom, and μ_C is the number of unpaired electrons, which is determined from the relation $\mu_{eff}{}^2 = \mu_C(\mu_C + 2)$.[31] For the $Mn_{1.99}Co_{0.96}Al_{1.05}$ alloy, we obtain $p_{RW} = 1.3$, and for $Mn_{1.79}Co_{1.25}Al_{0.96}$, we get $p_{RW} = 2.3$.

When describing the results of measurements of $\chi(T)$ by expression (3), we as well obtained the values of temperature- independent magnetic susceptibility: $\chi_0 = -4.8\cdot10^{-6}$ cm^3/g for the $Mn_{1.99}Co_{0.96}Al_{1.05}$ alloy and $\chi_0 = -7.4\cdot10^{-6}$ cm^3/g for the $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloy. It is known[32] that, in transition metal alloys, the value of χ_0 is mainly determined by the weakly temperature-dependent Pauli paramagnetic susceptibility, which can be written in the form $\chi_p = \mu_B^2 n(E_F)$, where $n(E_F)$ is the density of states at the Fermi level E_F. The contribution χ_p has a positive sign. However, it follows from the experiment that $\chi_0 < 0$ in the studied alloys. The presence of a large diamagnetic contribution to the magnetic susceptibility may be because the effective mass m^* of the conduction electrons of the compounds under study is much smaller than the mass of free electrons m_0.[32] In this case, the paramagnetic Pauli component, which is proportional to the density of states at the Fermi level, is suppressed by the Landau diamagnetism of the conduction electrons and the ion core diamagnetism. This indicates a significant rearrangement of the electronic band structure near E_F when the composition of the alloy under study deviates from stoichiometric and in its disordering.

3.2. Heat capacity

To estimate the density of states $n(E_F)$ in the non-stoichiometric $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloy, we measured its specific heat C_P in the temperature range ($2\text{ K} < T < 30$ K), the results are shown in Fig. 6. Traditionally, the dependences of $C_P(T)$ of ferromagnetic alloys at low temperatures $T < 0.1\ \Theta_D$ (where Θ_D is the Debye temperature) are described taking into account the electron components $\sim \gamma T$ and the lattice, in the Debye approximation

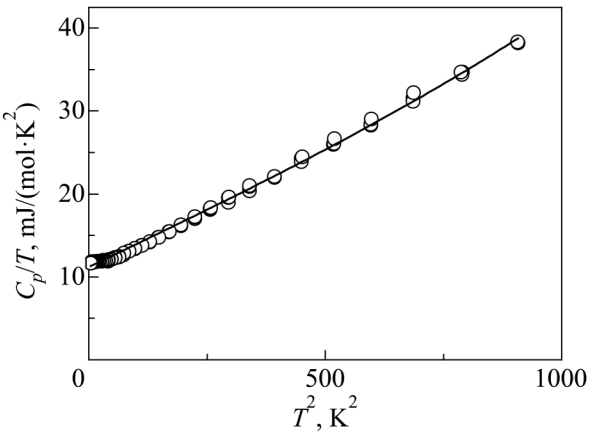

FIG. 6. Low-temperature specific heat of the $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloy. The solid line is approximation of the experimental data of $C_P(T)$ according by expression (4).

$\sim\beta T^3$. In this case, the spin-wave contribution $\sim\alpha T^{3/2}$, as a rule, is insignificant. However, in the studied band magnets, such a simple approximation does not provide a satisfactory description of the obtained heat capacity results. As can be seen in Fig. 6, the dependence of $C_P(T)$ in this case is quite satisfactorily described by the expression

$$C_P = C_0 + \gamma T + \beta T^3, \quad (4)$$

where the constants $\gamma = (\pi^2/3)k_B^2 n(E_F)$ is the Sommerfeld electron coefficient and $\beta = (12 / 5)\pi^4 N_A k_B/\Theta_D$ characterizes the phonon contribution to the specific heat in the Debye model, $n(E_F)$ is the density of electronic states at the Fermi level, N_A is the Avogadro constant. The temperature- independent term in expression (4) is usually associated with the presence of magnetic inhomogeneities (clusters) in the sample. In this case, the presence of C_0 in (4) can be explained by analogy with[33] as follows. The magnetic moments of weakly interacting ferromagnetic clusters in the studied alloy oscillate in the field of crystalline anisotropy. This leads to additional absorption of the thermal energy of the sample. In the analysis of heat capacity, such a process can be considered as a system of oscillators with a low Einstein temperature ($\leq$ 1 K), above which the corresponding excitations, and, consequently, the contribution of C_0 are independent of temperature.

The coefficients values obtained as a result of fitting the experimental data using expression (4) are: $C_0 = 5.2$ mJ/(mol·K), $\gamma = 9.83$ mJ/(mol·K^2) and $\beta = 0.0312$ mJ/(mol·K^4). It follows that the density of electronic states at E_F and the Debye temperature of the sample are typical for band ferromagnets: $n(E_F) = 4.2$ states/(eV·at.) and $\Theta_D = 396$ K. At the same time, according to band calculations,[20] the electron density of states at E_F in the gapless Mn_2CoAl semiconductor should be close to zero. The presence of a sufficiently high value of $n(E_F)$ in the studied alloy can be explained according to Ref. 22 by the absence of a gapless state on E_F due to the deviation of the studied sample in composition from stoichiometric Mn_2CoAl and, accordingly, having a significant atomic disorder. This is in agreement with the presence of a temperature-independent term C_0 in the specific heat.

4. CONCLUSIONS

Thus, the performed studies show that the $Mn_{1.99}Co_{0.96}Al_{1.05}$ and $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloys, which are close in composition to the Mn_2CoAl spin gapless semiconductor, are band ferromagnets. It was shown that the high-field magnetization for both compositions is described in the Stoner models with the following Rhodes-Wohlfarth parameters: $p_{RW} = 1.3$ for $Mn_{1.99}Co_{0.96}Al_{1.05}$ and $p_{RW} = 2.3$ for $Mn_{1.79}Co_{1.25}Al_{0.96}$ alloys. It was observed that the spontaneous moments decrease slightly and the effective moments increase at deviation from the stoichiometric composition of Mn_2CoAl that leads to the appearance of a negative sign of the temperature-independent component of the paramagnetic susceptibility. It was demonstrated that the values of density of states at Fermi level and the Debye temperature of studied alloys are usual ones for 3*d* metals.

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