

Spin-Polarized Electric Current in $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ Nanocomposite

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Abstract—For the first time, the temperature dependences of the electrical resistivity and magnetization of the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite were measured in the temperature range of 10–350 K. It is shown that the electrical properties of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ are associated with spin polarization of intrinsic electrons in the Cd_3As_2 matrix by spin-polarized electrons injected into it from ferromagnetic MnAs nanoclusters. With an increase in the magnetization of the entire sample, the angle between the magnetization directions of individual nanoclusters decreases and the spin-polarized current increases. Furthermore, an increase in the concentration of intrinsic carriers in the matrix leads to an increase in the spin-polarized current. This concept is also confirmed by measurements of current–voltage characteristics (CVCs) at voltages of up to 5 V at temperatures both below critical temperature $T_{\text{cg}} = 241$ K of cluster glass formation (at 77 and 172 K) and above it (at 273.15 and 373.15 K), which exhibit an increasing deviation from the ohmic behavior with an increase in the stress. This means that the greater the spin polarization of intrinsic electrons in Cd_3As_2 due to an increase in the injection of spin-polarized electrons from MnAs with an increase in the voltage, the higher the current.

Keywords: spin, nanocomposites, current–voltage characteristics, resistivity, spin polarization, semiconductors, spin injection, magnetization

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1. INTRODUCTION

Composites with high magnetoresistance values, in which semiconducting compounds of cadmium arsenide were used as a matrix and compounds of MnAs were used as ferromagnetic nanoclusters, were synthesized in [1]. As was shown in [2–6], the electrical and magnetic properties of the $(\text{Cd}_x\text{Mn}_{1-x})_3\text{As}_2$ nanocomposite are determined by MnAs nanoclusters. The pressure dependences of the electrical resistivity, the Hall coefficient, the carrier mobility, the carrier concentration, and the magnetoresistance in the range of 3–4 GPa exhibit features associated with phase transitions. The structural transition from the tetragonal to monoclinic structure in Cd_3As_2 occurs in the range of 2.6–4.67 GPa [7]. The spin-reorientation transition in MnAs at about 3.8 GPa and 110 K [8] has a substantial effect on the transport of charge carriers and the magnetoresistance of this composite. The magnetoresistance peaks in the pressure dependences increase with an increase in the magnetic field strength. A study shows that the magnetoresistance of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$

undergoes a sign reversal at 7.8 GPa, and the negative magnetoresistance (NMR) reaches 0.36%. The behavior of the electrical resistivity of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ at high pressures (up to 50 GPa) is in good agreement with the behavior of doped ternary oxides of transition elements and film heterostructures based on them [9–11].

The main objective of this article is to explain the nature of the observed deviation from the ohmic behavior of the current–voltage dependence in the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite.

2. EXPERIMENTAL

To study the temperature dependences of the current–voltage characteristics (CVCs), a sample with contacts was placed in a hermetically sealed chamber filled with gaseous helium. First, the chamber was placed into a vessel with liquid nitrogen for measurements in the temperature range of 77–300 K and then placed in a thermostat for measurements in the range

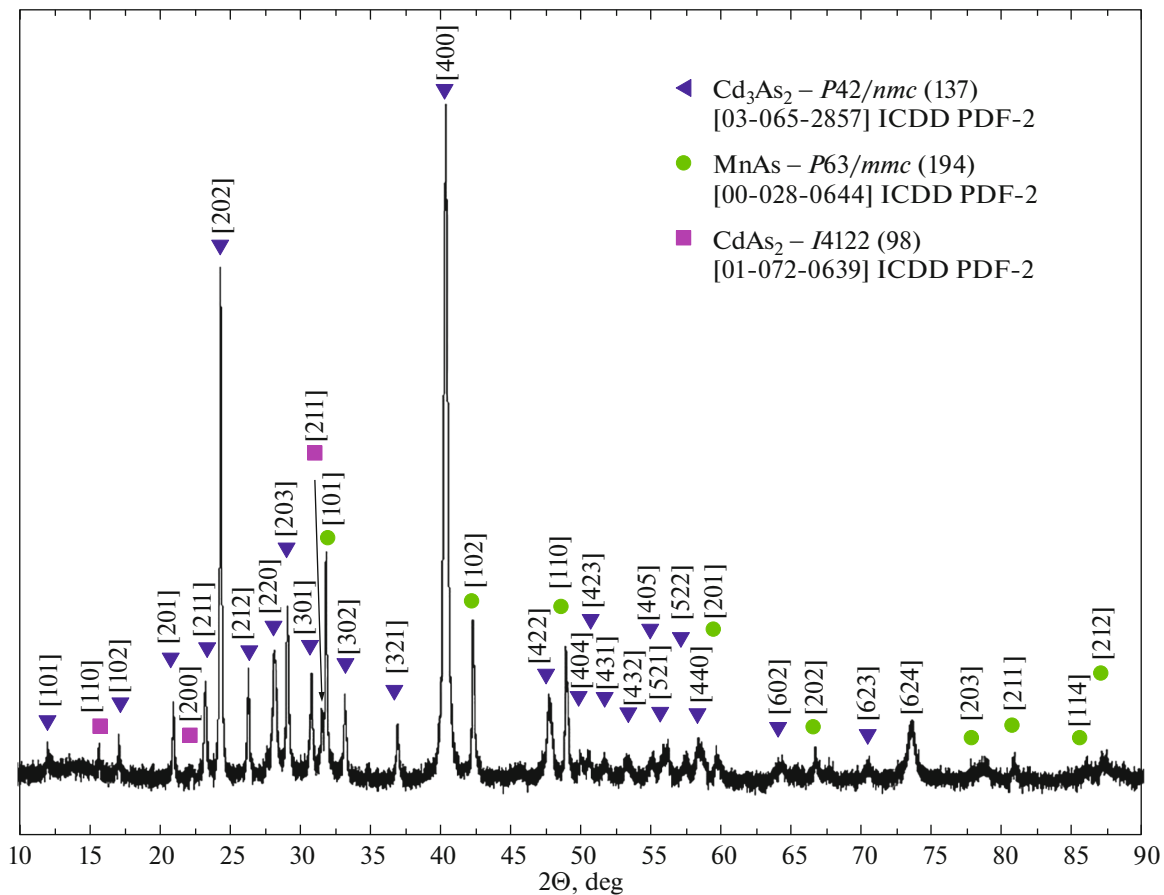


Fig. 1. X-ray diffraction pattern of the sample of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$.

of 300–372 K. The electromotive force on the sample and the readings of the copper–constantan thermocouples were measured with an Sh-300 potentiometer. To determine the temperature dependence of the CVC, the voltage and current were measured using a Keithley 2000 instrument.

The magnetization of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ was studied using a SQUID magnetometer (Magnetic Property Measurement System-XL-7EC) with a sensitivity of $1 \times 10^{-8} \text{ G cm}^3$ when measuring the magnetic moment. The sample weight was 67.09 mg. The sample was cooled to a temperature of 10 K in the absence of a field and then was heated to 350 K to measure the magnetization in a field of 100 Oe.

3. RESULTS AND DISCUSSION

Compound $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ is a complex system comprised of ferromagnetic granules of MnAs randomly arranged in a Cd_3As_2 semiconductor matrix. This morphology leads to an uneven distribution of the electric field in the sample volume. An electron microscopic study of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ made it possible to establish that the size of MnAs nanoclusters varies in the range of 10–50 nm [12]. Typically, the charac-

teristic diameter of MnAs nanoclusters is in the range of 20–30 nm. In our opinion, individual specimens with particle sizes greater than 50 nm can be located only in the near-surface regions, rather than in the bulk of the composite, since the latter case would lead to a substantial increase in the deformation energies of both the matrix and its inclusions. Consequently, it can be reasonably assumed that the bulk physical properties of the composite material under study are determined by MnAs nanoclusters with particle diameters from 20 to 30 nm.

The X-ray diffraction pattern of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ (Fig. 1) confirms that it consists of the following two phases: Cd_3As_2 and MnAs (some X-ray diffraction patterns show minor peaks attributed to CdAs_2).

The microstructure of the sample indicates the eutectic segregation of the melt upon solidification into a system of the following two substances: Cd_3As_2 and MnAs. Moreover, the latter is present in the form of spherical granules, which may indicate that such a segregation took place even in the melt [12].

As can be seen from the temperature dependence of the electrical resistivity of the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ composite (Fig. 2), it is characterized by a metallic type of conductivity at temperatures below critical tempera-

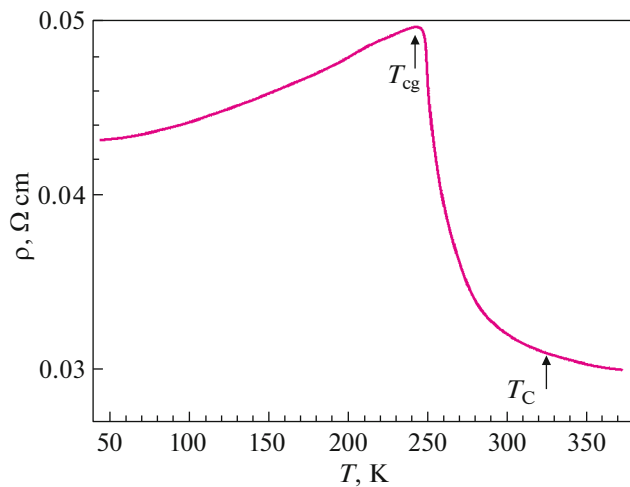


Fig. 2. Temperature dependence of the electrical resistivity of the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite in the temperature range of 40–380 K; the critical temperature of the cluster glass is $T_{\text{cg}} = 241$ K, and the Curie point is $T_{\text{C}} = 325$ K.

ture $T_{\text{cg}} = 241$ K of cluster glass formation (the maximum point on the curve) and by a semiconducting (thermoactivated) type of conductivity above this temperature. This kind of behavior of the electrical resistivity is consistent with the SEM image of a cleaved surface of the studied sample [12], which shows that the sample is a eutectic alloy of MnAs nanoclusters that are hardly in contact with one another and embedded in a Cd_3As_2 semiconductor matrix. If there were any significant percolation conductivity of the Cd_3As_2 phase, then the metallic type of conductivity would be observed for MnAs up to the Curie temperature at 325 K. As seen from Fig. 2, the metallic conductivity is replaced by the semiconductor conductivity above $T_{\text{cg}} \approx 241$ K. Since the two-phase system comprised of ferromagnetic MnAs and a nonmagnetic Cd_3As_2 matrix is observed both above and below T_{cg} , this behavior is difficult to explain. It would seem that there should be a complex behavior corresponding to the sum of metallic and semiconducting types of electrical conductivity, and a certain monotonic dependence should be observed up to Curie temperature $T_{\text{C}} \approx 325$ K as a result of their competition. However, the real pattern shown in Fig. 2 suggests nanoclusters have an effect on the electrical resistance properties of the matrix.

The Zener double exchange model is a key to unraveling this anomalous behavior of electrical conductivity in a mesoscopic nanocomposite system, since the dependences of the electrical resistivity are qualitatively the same in manganites and in the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite. However, manganites represent microscopic atomic systems, in which an electron is transferred from manganese cation Mn^{3+} to another Mn^{3+} cation through diamagnetic

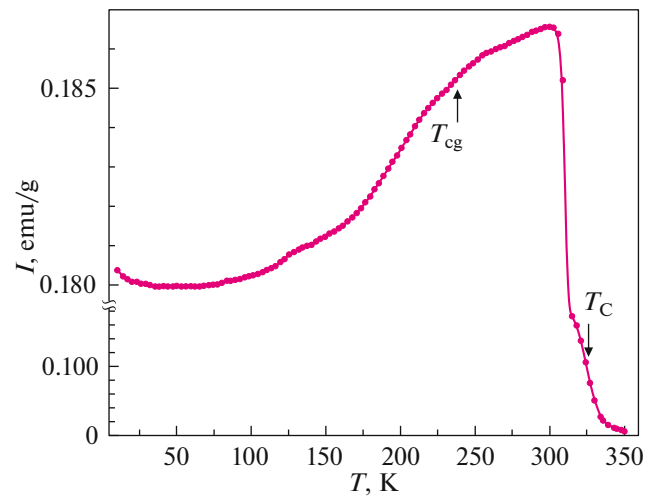


Fig. 3. Temperature dependence of the specific magnetization of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$; Curie point $T_{\text{C}} = 325$ K corresponds to the inflection point of the curve ($\partial^2 I(T)/\partial T^2 = 0$).

oxygen anion O^{2-} . Moreover, the probability of exchange increases substantially when the spins of the electron and Mn^{3+} cations are coaligned, i.e., the exchange is ferromagnetic. In this case, the electrical resistivity at $T < T_{\text{C}}$ behaves in accordance with the metallic type of conductivity. If the spins of neighboring cations are not parallel due to temperature disorientation, i.e., they are in the paramagnetic state at $T > T_{\text{C}}$, the electrical resistance is of the semiconductor type.

Thus, Curie point T_{C} of a microscopic system with double exchange (for example, manganite) is similar to cluster glass point T_{cg} of the mesoscopic $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ system. Continuing the analogy, this suggests that not only the dipole–dipole magnetic interaction between MnAs nanoclusters, but also the spin polarization of the current caused by their magnetization promotes ferromagnetic ordering.

The plausibility of such a scenario is indicated by an anomalous decrease in the magnetization with a decrease in the temperature after the transition to the ferromagnetic state (Fig. 3). In such a case, the magnetization usually increases as a result of reducing the disordering effect of temperature. Indeed, the concentration of intrinsic carriers in the Cd_3As_2 matrix decreases with a decrease in the temperature, which leads to a decrease in the spin-polarized current between the MnAs nanoclusters. In turn, this leads to a decrease in the system magnetization caused by the ordering effect of the spin-polarized current. The decrease in the magnetization upon cooling the sample from T_{cg} to 10 K is about 3.3%. Hence, even though the effect is small, but still reliable against the background of the high accuracy of magnetization measurements. This suggests that we are dealing with

a mesoscopic version of double exchange in this case, i.e., not only the collapse of angles between the magnetization vectors of ferromagnetic nanoclusters leads to a noticeable increase in the spin-polarized current between them, but this current itself favors the ferromagnetic ordering of nanocluster magnetization vectors in due course and leads to an increase in the magnetization of the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite. Therefore, it is possible to change the magnetization of the sample by changing the spin-polarized current by means of an electric field applied to the sample. For example, this takes place in manganites [9] and iron-based superconductors [11], as well as in multilayer magnetic sandwich structures based on them [10], but with a very important advantage that the test-tube melting of nanocomposite is much cheaper than the preparation of a semiconductor heterostructure by molecular beam epitaxy. In the macroscopic analogue of double exchange, the mesoscopic MnAs cluster plays a role of magnetoactive cations, and the Dirac semimetal Cd_3As_2 with the n -type of conductivity acts as a medium that transfers the charge and electron spin instead of the oxygen anion O^{2-} . It should be noted that structural and magnetic phase transitions are inherent to the studied nanostructured eutectic alloy of Cd_3As_2 with mesoscopic ferromagnetic inclusions of MnAs, as well as to the systems with double exchange, as can be seen from the above-mentioned NMR [5].

As can be seen from Fig. 3, a sharp decrease in the magnetic susceptibility of the material is observed with an increase in the temperature after reaching $T \approx 310$ K, which is explained by the transition from the ferromagnetic to paramagnetic state. Curie temperature T_C of the composite, which was estimated from the inflection point of the $\chi(T)$ curve (where $\partial^2\chi/\partial T^2 = 0$) obtained at a constant magnetic field strength of 100 Oe, is 325 K (see Fig. 3). This agrees with the data obtained earlier for the composites with a different content of MnAs clusters [13] and is close to present Curie point $T_C = 318$ K of true ferromagnet MnAs. As is seen from Fig. 4, the studied composite is characterized by a small value of the coercive force (<10 Oe), which is consistent with the small, i.e., superparamagnetic, sizes of MnAs particles included into the Cd_3As_2 matrix. Being a complex of MnAs nanoclusters in dielectric filler Cd_3As_2 , the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ composite behaves as a ferromagnet only in some external manifestations. In particular, it only imitates the Curie point. That is why the Curie temperature for it is conditional, and the corresponding index is denoted by a lowercase letter indicating the first letter of the word “critical” rather than Curie’s surname, which begins with an uppercase letter. By not calling the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ composite “a true ferromagnet,” we imply that the magnetic interactions in its volume are inhomogeneous, i.e., a ferromagnetic exchange interaction takes place only inside

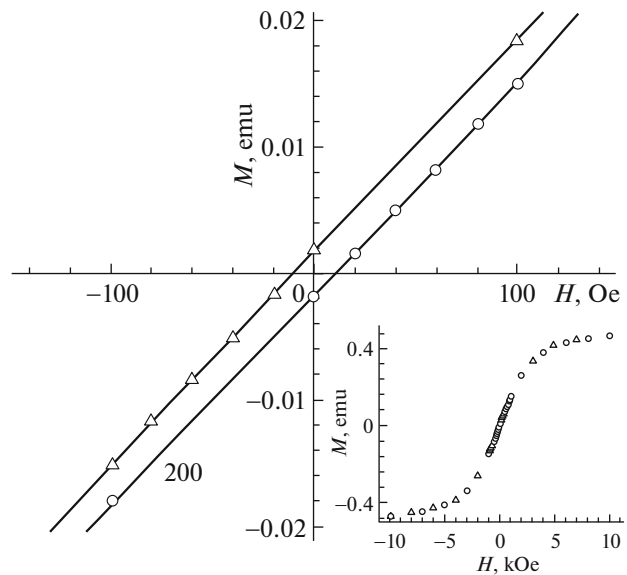


Fig. 4. Section of the isothermal magnetization hysteresis loop of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ at 300 K, which demonstrates a small value of the coercive force (10 Oe). The inset shows a magnetization loop; the hysteresis is indistinguishable because of its smallness. The circle dots are recorded in the course of increasing the magnetic field, and the triangular dots are recorded in the course of decreasing the magnetic field.

the clusters, while only the magnetic dipole–dipole interaction takes place between them, which is capable to order superparamagnetic MnAs into ferromagnetically ordered clusters at the so-called “Curie point” equal to $T_C = 325$ K. In many studies, no terminological distinction is made between the true ferromagnetic and superparamagnetic states. In our opinion, both parties are correct. If we consider the magnetic interactions in the entire volume of the sample, then these states should be distinguished. However, if only the external ordered state with all the magnetic moment arrows pointing in the same direction is of interest, then the absence of a strict terminological distinction between ferromagnetism and superparamagnetism is acceptable. Moreover, superparamagnetism has nothing to do with paramagnetism in reality, since a single-domain ferromagnetic particle is called “superparamagnet” only in order to emphasize the spontaneous instability of its magnetization relative to the direction of the external magnetic field, similar to the behavior of the magnetic moment of a paramagnetic atom or molecule or an elementary particle with a nonzero magnetic moment (electron, proton, etc.).

To reveal the possible contribution of the spin polarization of current carriers to the electrical conductivity, we measured the isothermal dependences of the electrical resistivity of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ on the applied voltage both at temperatures below $T_{cg} = 241$ K (namely, at 77.8 and 172 K), and above it at 273.15 and 373.15 K (Fig. 5).

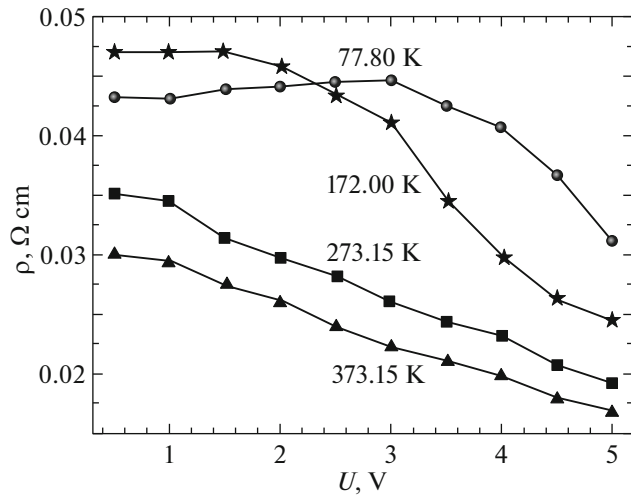


Fig. 5. Isothermal dependences of the electrical resistivity of the granular $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite in the ferromagnetic (77 and 172 K) and paramagnetic (273.15 and 373.15 K) states on the voltage at various temperatures.

In the absence of spin polarization of carriers in the sample under study, the current is proportional to the applied voltage and Ohm's law is fulfilled, i.e., the electrical resistivity is constant. However, the electrical resistivity depends on the voltage (Fig. 5). In the state of cluster glass at $T < T_{\text{cg}}$, the resistivity drops nonlinearly and sharply. Moreover, its relative change at 172 K is much higher than at the temperature of liquid nitrogen; namely, the relevant resistivity values reduce by factors of 2.2 and 1.3, respectively. The fact that the onset of the dependence at 172 K is higher than the onset of the dependence at 77 K is explained by the metallic behavior of the conductivity in the state of cluster glass at $T < T_{\text{cg}}$.

At temperatures of $T > T_{\text{cg}} = 241$ K, i.e., at 273.15 and 373.15 K, the resistivity decreases linearly and with almost the same line slopes. At temperatures above T_{cg} , temperature disordering of the magnetization directions of MnAs nanoclusters begins to manifest itself. Therefore, spin polarization is less pronounced at $T > T_{\text{cg}}$. At $T > T_{\text{cg}}$, the temperature dependence of the electrical resistivity is replaced by semiconductor behavior, at which the resistivity decreases with an increase in the temperature. That is why the dependence at 373.15 K in Fig. 6 is located lower than the dependence at 273.15 K.

A decrease in the electrical resistivity with an increase in the applied voltage is explained by the enhancement of spin polarization with an increase in the current, since the larger the number of spin-polarized electrons penetrating from the MnAs nanoclusters into the host Cd_3As_2 matrix, the larger the extent to which they polarize the intrinsic electrons of this matrix. As a result, the current increases, since there is no need for electrons to spend energy for spin flip, as

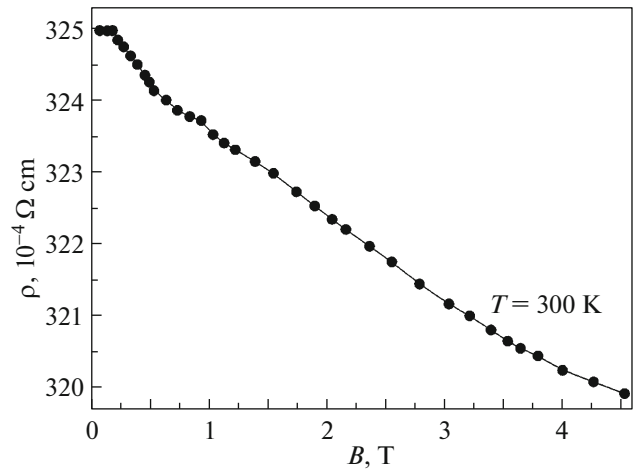


Fig. 6. Isothermal dependence of the electrical resistivity of $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ on the magnetic field strength at 300 K.

it happens when its direction does not coincide with the magnetization of neighboring MnAs nanoclusters. This is confirmed by the dependences of the electrical resistivity on the magnetic field (Fig. 6). As can be seen from Fig. 6, the electrical resistivity at a temperature of 300 K, i.e., near T_{C} , decreases almost linearly with an increase in the magnetic field. This indicates that the ordering of magnetization vectors of MnAs nanoclusters by a magnetic field favors the flow of an electric current of spin-polarized electrons between them.

The isothermal dependence of the electrical resistivity on the voltage at 373.15 K requires separate consideration, since it is observed when a significant fraction of MnAs nanoclusters are already in the paramagnetic state (the Curie point of MnAs nanoclusters in the Cd_3As_2 matrix is $T_{\text{C}} = 325$ K). Nevertheless, the influence of spin polarization takes place in this case as well. It is possible that this behavior comes from the fact that a certain concentration of ferromagnetic MnAs nanoclusters is also present above the Curie point, as was observed in manganites [14]. Indeed, there is some nonzero magnetization even at a temperature of 350 K, as can be seen from Fig. 3, which increases because of the spin-polarized current induced by an external electric field and reaches a temperature that is even higher than 373.15 K. In addition, mesoscopic ferromagnetic states can exist substantially above the Curie point in the paramagnetic state because of thermodynamic fluctuations and mesoscopic structural inhomogeneities (for example, different sizes of nanoclusters and, consequently, different compressive elastic forces from the side of the Cd_3As_2 matrix). This behavior is inherent in complexes of transition elements, such as ternary alloys, including $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$, and ternary oxides, i.e., manganites, nickelates, cobaltites, etc. As regards the

effect of the mother rock on the ferromagnetic inclusions contained in it, it should be noted that the Curie point for MnAs inside Cd_3As_2 is 325 K, while the Curie point for MnAs in free form is 318 K [15, 16]. That is, the Cd_3As_2 matrix increases the Curie point of the MnAs nanoclusters by as much as 7 K by squeezing them from all directions. This gives an additional technological advantage to nanocomposites, which consists in that the magnetic characteristics of inclusions can be controlled by selecting the appropriate matrix. It is also possible that the Cd_3As_2 matrix itself exhibits magnetic properties. From this point of view, our results agree with the fact that the presence of double exchange and the corresponding ferromagnetic behavior in some composites based on Cd_3As_2 has been reliably established [13, 17]. The spin-polarized current was observed in magnetic sandwich structures comprised of layers of magnetic and nonmagnetic material with approximately the same thickness as the distance between the MnAs clusters [9–11]. These studies support our explanation of the behavior of the electrical resistivity of the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite by the spin polarization of delocalized electrons from MnAs nanoclusters by their own current carriers in the conduction band of Cd_3As_2 .

A low concentration of electrons in the Cd_3As_2 matrix, especially at low temperatures of $T < T_{\text{cg}}$, does not allow one to use the Ruderman–Kittel–Kasuya–Yoshida mechanism (RKKY mechanism) of exchange interaction for explaining the observed effect of spin polarization of the electric current in the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite by the initial RKKY polarization of charge carriers in the Cd_3As_2 matrix by the magnetic moments of MnAs clusters. Thus, our interpretation of an anomalous decrease in the electrical resistivity of the investigated composite with an increase in the applied voltage as a consequence of the spin-polarized current in bulk granular nanocomposite structures, which leads to a linear decrease in the electrical resistivity of the medium, seems to be quite relevant.

4. CONCLUSIONS

The temperature dependences of the electrical resistivity and specific magnetization of the $\text{Cd}_{48.6}\text{Mn}_{11.4}\text{As}_{40}$ nanocomposite in the temperature range of 10–350 K are studied for the first time. The electrical resistivity isotherms in the ferro- and paramagnetic states are calculated. An explanation is given for an anomalous decrease in the electrical resistivity of this composite as a function of the applied voltage, which is interpreted as a consequence of the appearance of a spin-polarized current, i.e., spin polarization of delocalized electrons from MnAs nanoclusters by intrinsic charge carriers in the conduction band of Cd_3As_2 . Thus, our interpretation of an anomalous decrease in the electrical

resistivity of the investigated composite as a function of the applied voltage seems to be quite relevant.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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