

Electrical Resistance and Magnetoresistance of Cd_3As_2 –30 mol % MnAs under High Pressures

L. A. Saypulaeva^{a,*}, K. Sh. Khizriev^a, N. V. Melnikova^b, A. V. Tebenkov^b,
A. N. Babushkin^b, V. S. Zakhvalinskii^c, A. I. Ril^d, S. F. Marenkin^d,
M. M. Gadjaliev^a, and Z. Sh. Pirmagomedov^a

^a Amirkhanov Institute of Physics of the Daghestan Federal Research Center of the Russian Academy of Sciences, Makhachkala, 367003 Dagestan, Russia

^b Institute of Natural Sciences and Mathematics, Ural Federal University, Yekaterinburg, 620002 Russia

^c Belgorod National Research University, Belgorod, 308007 Russia

^d Kurnakov Institute of General and Inorganic Chemistry, Russian Academy of Sciences, Moscow, 119991 Russia

*e-mail: l.saypulaeva@gmail.com

Received January 24, 2021; revised March 30, 2021; accepted March 30, 2021

Abstract—The paper presents the results of a study on the Electrical resistance and magneto-resistance (MR) of the composite, consisting of a Dirac semimetal Cd_3As_2 and 30 mol % ferromagnetic dopant MnAs at pressure values up to 50 GPa. In the pressure range of 16–50 GPa, a hysteresis behavior of the transport properties was observed during a cycle of application and release of a pressure. Measurements on MR in the pressure rise and release mode revealed the features in the form of maxima of negative and positive MR, when the relative magneto-resistance ($\Delta R/R_0$) reached ~20% and ~5.3% for negative and positive values respectively. An instability of the monoclinic structure of Cd_3As_2 as a result of its partial decomposition during decompression has been established.

Keywords: high pressure, composite, electrical resistivity, negative magnetoresistance, structural phase transition

DOI: 10.1134/S1063783421080266

1. INTRODUCTION

Previously we established the existence of a pressure-induced negative magnetoresistance (NMR) in the Cd_3As_2 + MnAs structure with contents of 20, 30, and 44.7 mol % MnAs [1–4]. The studies of the magnetoresistance of Cd_3As_2 + MnAs under pressures of up to 9 GPa enabled one to find the pressure ranges in which NMR is observed. So, in the Cd_3As_2 + MnAs(44.7 mol % MnAs) sample, the maximum NMR was ~0.36% under pressures up to 7.8 GPa. The pressure dependences of the resistivity, the Hall coefficient, and the magnetoresistance of Cd_3As_2 + MnAs composites in the vicinity of pressures of 3–4 GPa are found to demonstrate the features in the behavior of the transport properties [1–4] related to a phase transition. This phase transition can be considered as a result of combined phase transition, namely, the structural phase transition in the Cd_3As_2 matrix and the spin-reorientational magnetic phase transition in MnAs nanoclusters which influence the carrier transport and MR in the composite.

The behavior of the transport properties of the composites under pressures higher than 9 GPa has

been studied inadequately. The studies of the transport properties of Cd_3As_2 + MnAs composites are also of interest due to the observation of possible phase transitions. For example, the studies of the thermopower of the Cd_3As_2 + 44.7 mol % MnAs composites as a parameter, which is one of most sensitive to phase transformations, detected the feature of the Seebeck coefficient near $P \sim 33$ GPa that was interpreted as the second phase transition [5]. Because of this, of great interest is the study of MR in the pressure range close to the second phase transformation region [5].

In this work, we consider the influence of pressures up to 50 GPa on the electrical and magnetotransport properties of the Cd_3As_2 + 30 mol % MnAs composites.

The results demonstrate the maxima and minima in the field dependence of MR, the nature of which can be interpreted from the point of view of the composite instability during pressure cycling.

2. EXPERIMENTAL

The Cd_3As_2 + 30 mol % MnAs crystals were synthesized by the vacuum-ampoule method from

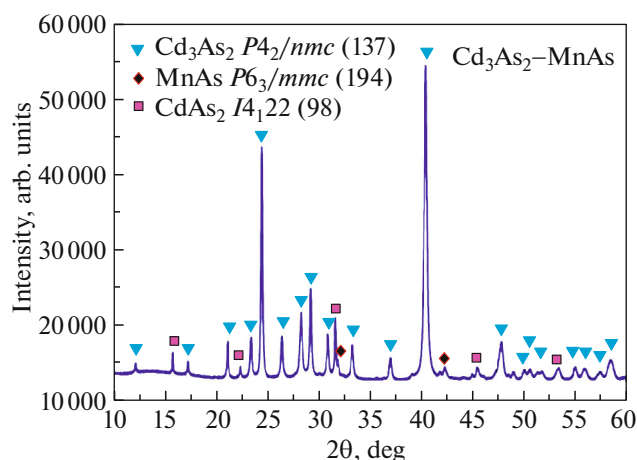


Fig. 1. X-ray diffraction pattern of the Cd_3As_2 + 30 mol % MnAs sample.

Cd_3As_2 and MnAs compounds at the melting temperature of manganese arsenide [6]. The samples were a composite consisting of nanometer-size MnAs ferromagnetic granules arranged chaotically in the volume of a semiconductor Cd_3As_2 matrix. The obtained samples were characterized by X-ray diffraction method and scanning electron microscopy (SEM). The elemental analysis of the Cd_3As_2 + 30 mol % MnAs composite showed that a large part of the volume is the Cd_3As_2 phase. The MnAs phase inclusion fraction is less than 5%. The interpretation of the X-ray diffraction data shows the peaks corresponding to two main phases of the material under study: tetragonal (Cd_3As_2) and magnetic hexagonal (MnAs) (Fig. 1). In addition, there is insignificant content of the CdAs_2 . The microstructures contain the Cd_3As_2 phase and the Cd_3As_2 eutectic. The feature of the Cd_3As_2 + MnAs composite is the existence of melts of Cd_3As_2 and MnAs phases. The electron-microscopy studies could not visualize granules at the content 30 mol % MnAs (Fig. 2), likely, because of their characteristic small sizes.

The influence of a high pressure on the electrophysical properties of the composites were studied in a high-pressure chamber (HPC) with diamond anvils of the “rounded cone–plane” type. The principle of generating pressures up to 50 GPa, the technical characteristics and the calibration of the HPC are described in detail in [7–9]. The equipment used in this work enables one to measure electrical characteristics of a material immediately during its deformation by applying high pressures. The peculiarities of this HPC make it possible to measure electrophysical characteristics of compressed samples under a minimum pressure no lower than 15 GPa. It is known that during a change (rise or relief) in the pressure, some time is needed for the electrical resistance to take a value unchanged in time. The thickness of the samples

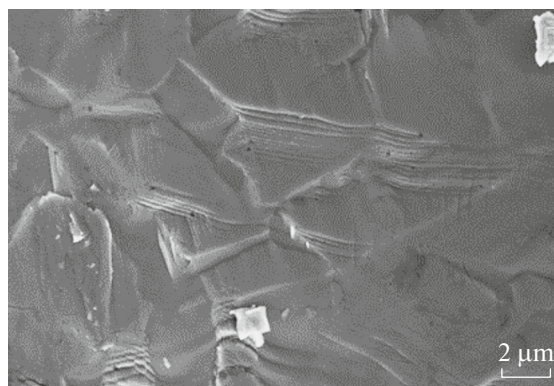


Fig. 2. SEM image of a Cd_3As_2 + 30 mol % MnAs sample cleavage.

during compression was $\sim 15 \mu\text{m}$, and the sample diameter in HPC was $\sim 200 \mu\text{m}$. When measuring MR, the high-pressure chamber was placed into a shell magnet to induce a transverse magnetic field ($0 \leq B \leq 1 \text{ T}$); in this case, the change and the control of pressure and magnetic field were performed immediately during the experiment. At each fixed pressure, the relative magnetoresistance $\Delta R/R_0$ (in percent) was estimated by relationship

$$\frac{\Delta R}{R_0} = 100 \frac{R(B) - R(0)}{R(0)}, \quad (1)$$

where $R(B)$ is the electrical resistance in the transverse magnetic field with induction B , and $R(0)$ is the electrical resistance in the absence of magnetic field.

3. RESULTS AND DISCUSSION

Figure 3 shows the dependence of the electrical resistance R of the Cd_3As_2 + 30 mol % MnAs composite on pressure P during two subsequent measurement cycles (pressure rises and reliefs) near room temperatures. In the pressure range 15–25 GPa of the first cycle, R significantly decreases as P rises, which agrees well, as a whole, with the results obtained for Cd_3As_2 [10]. In this pressure range, the resistance is changed almost linearly at a mean rate $dR/dP = -24.9 \Omega/\text{GPa}$ (inset in Fig. 3). At $P > 34 \text{ GPa}$, the pressure coefficient is $-1.1 \Omega/\text{GPa}$, which indicates a weak dependence of R on P to pressures of 50 GPa. During the pressure relief from 50 GPa, the $R(P)$ dependence demonstrated a hysteresis behavior in both measurement cycles. The predominance of the hysteresis of R in Cd_3As_2 + 30 mol % MnAs in the pressure range 15–50 GPa seems very interesting, since this fact can favor the structural transition which takes place in Cd_3As_2 , whose structure at pressures higher than 4.67 GPa is already monoclinic ($P2_1/C$) [10]. However, according to the X-ray diffraction data presented in [10], the monoclinic phase was registered up to pressures of

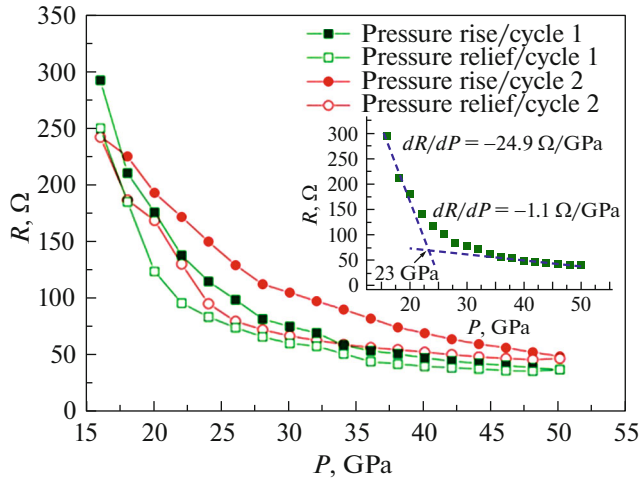


Fig. 3. Pressure dependences of the electrical resistance of $\text{Cd}_3\text{As}_2 + 30 \text{ mol } \%$ MnAs measured during two subsequent measurement cycles. The inset shows dependence $R(P)$ of the first cycle during pressure rise. The dashed lines correspond to the determination of the dR/dP slopes.

17.8 GPa, while its predominance up to 50 GPa has only a hypothetical character. Because, in this work, in situ X-ray diffraction studies at a high pressure for $\text{Cd}_3\text{As}_2 + 30 \text{ mol } \%$ MnAs were not performed, we can suggest that the existence of an insignificant CdAs_2 phase in the composite can also play a certain part in the appearance of the hysteresis of the $R(P)$ dependence. Now it is known that the CdAs_2 compound has been studied up to pressures of 9 GPa and, it was reported on an inverse structural transformation near 5.5 GPa, according to the data on the resistivity and the Hall coefficient [11]. On the other hand, we note that the appearance of the hysteresis behavior of the electrokinetic properties can most likely be associated with a partial decomposition of the $\text{Cd}_3\text{As}_2 + \text{MnAs}$ composite (pressure decomposition), rather than with the appearance of a structural transition at pressures higher than 17.8 GPa. This assumption also can be confirmed by the fact that the value of R is not regained to its initial value after the first cycle, and the hysteresis width increases during the second cycle as shown in Fig. 3.

Note that the magnetoresistive effect can be used as a structure-sensitive parameter to the structural transformation and the pressure decomposition in the composites [12, 13]. Figures 4 and 5 show the field dependences of MR measured at various pressures up to 50 GPa in magnetic fields up to 1 T at room temperature. The measurements were performed during the rise and the relief of pressure, which enables us to qualitatively establish the correlation between the hysteresis observed in the $R(P)$ dependence and the behavior of MR. The transverse MR was calculated by Eq. (1). As follows from Fig. 4, at $P = 16 \text{ GPa}$, a positive MR is observed in the composite. Similar positive

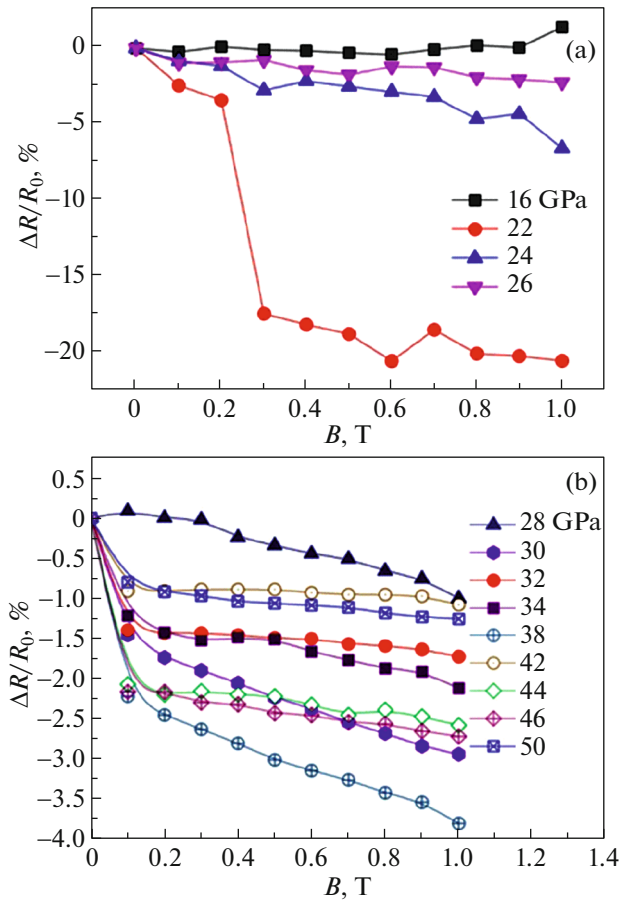


Fig. 4. Dependences of the magnetoresistance of $\text{Cd}_3\text{As}_2 + 30 \text{ mol } \%$ MnAs on the magnetic field induction in pressure ranges (a) 16–26 and (b) 28–50 GPa measured during pressure rise.

MR in $\text{Cd}_3\text{As}_2 + 30 \text{ mol } \%$ MnAs was observed before at comparatively low pressure 7.7 GPa and it was explained by the competition between the influence of the Lorentz force and spin-dependent scattering of charge carriers on MnAs clusters [2]. The positive MR is likely to exist up to pressure of 16 GPa; however, its value gradually decreases. The further rise in P leads to the change in the MR sign, and the maximum negative MR $\sim 20\%$ is observed at 22 GPa in field of 1 T. It should be noted that, in this pressure range, the rate dR/dP in the $R(P)$ dependence is changed sharply, and the intersection of the approximation lines of the ranges of low and high pressures corresponds to the value $\sim 23 \text{ GPa}$ (the inset in Fig. 1).

In this case, the maximum pressure-induced negative MR is realized in comparatively narrow pressure range 22–26 GPa, as clearly shown in Fig. 6; further, at $P > 26 \text{ GPa}$, the negative MR is no higher than 4% at 38 GPa. The dynamics of changing this negative MR with the pressure rise seems to be unambiguous, since there are several local minima in the $\Delta R/R_0(P)$ dependence. As pressure reliefs, the field dependence

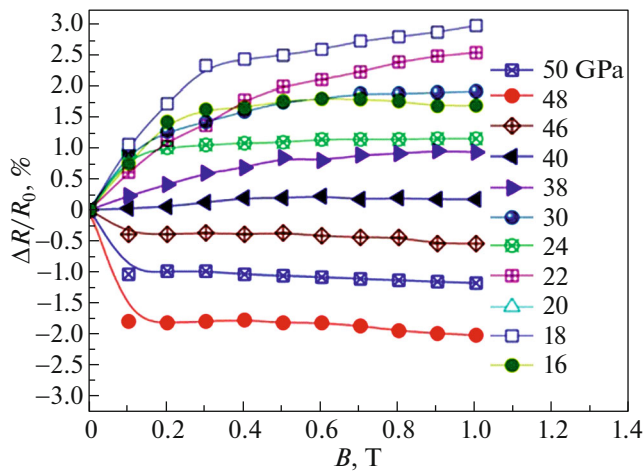


Fig. 5. Dependences of the magnetoresistance of $\text{Cd}_3\text{As}_2 + 30 \text{ mol } \% \text{ MnAs}$ on the magnetic field induction at some fixed values of pressure measured during pressure relief.

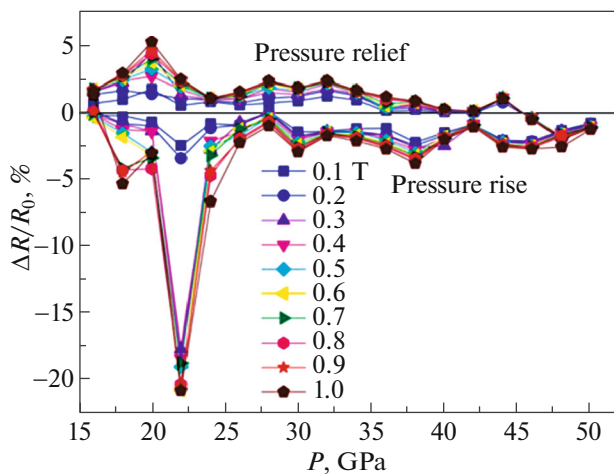


Fig. 6. Pressure dependences of the magnetoresistance of $\text{Cd}_3\text{As}_2 + 30 \text{ mol } \% \text{ MnAs}$ measured at various inductions of the transverse magnetic field to $B = 1 \text{ T}$.

of MR demonstrates the sign inversion at $P > 40 \text{ GPa}$ (Fig. 5), and the maximum positive MR $\sim 5.3\%$ is observed near 20 GPa (Fig. 6). Thus, the change in the MR sign during the pressure rise and relief can demonstrate the irreversibility of structural properties in $\text{Cd}_3\text{As}_2 + 30 \text{ mol } \% \text{ MnAs}$, which is due to a partial decomposition of the composite after its decompression. At the same time, the existence of the maxima of the negative and positive MR is supposedly related to the nature of the phase transformations in the electron subsystem of the composite [5].

4. CONCLUSIONS

The electrical resistance and the peculiarity of MR in the $\text{Cd}_3\text{As}_2 + 30 \text{ mol } \% \text{ MnAs}$ composite have been studied under high pressures up to 50 GPa. The measurement R during cycling P shows the hysteresis char-

acteristic in a wide pressure range, which can be due to an instability of the monoclinic structure of Cd_3As_2 at $P > 16 \text{ GPa}$ with its partial decomposition after the pressure relief. This behavior is confirmed by the results of measuring MR during pressure rise and pressure relief, which demonstrate the change in the MR sign from negative to positive. The $\Delta R/R_0(P)$ dependence has pronounced maxima of MR, in particular, the maximum of the negative MR is $\sim 20\%$ as pressure increases and the maximum of positive MR is $\sim 5.3\%$ during the pressure relief, the origin of which is caused most likely by the influence of magnetic impurity Mn as a result of a topological phase transformation.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

REFERENCES

1. L. A. Saipulaeva, M. M. Gadzhialiev, A. G. Alibekov, N. V. Melnikova, V. S. Zakhvalinskii, A. I. Ril', and A. Yu. Mollaev, *Inorg. Mater.* **55**, 873 (2019).
2. A. G. Alibekov, A. Yu. Mollaev, L. A. Saipulaeva, S. F. Marenkin, I. V. Fedorchenko, and A. I. Ril', *Russ. J. Inorg. Chem.* **62**, 90 (2017).
3. A. G. Alibekov, A. Yu. Mollaev, L. A. Saipulaeva, S. F. Marenkin, and I. V. Fedorchenko, *Inorg. Mater.* **52**, 357 (2016).
4. L. A. Saipulaeva, M. M. Gadzhialiev, A. G. Alibekov, N. V. Melnikova, V. S. Zakhvalinskii, A. I. Ril', S. F. Marenkin, and A. N. Babushkin, *Phys. Solid State* **62**, 942 (2020).
5. N. V. Melnikova, A. V. Tebenkov, G. V. Sukhanova, A. N. Babushkin, L. A. Saipulaeva, V. S. Zakhvalinskii, S. F. Gabibov, A. G. Alibekov, and A. Yu. Mollaev, *Phys. Solid State* **60**, 494 (2018).
6. A. I. Ril', A. V. Kochura, S. F. Marenkin, A. E. Kuz'ko, and B. A. Aronzon, *Izv. Yu.-Zap. Univ., Ser.: Tekh. Tekhnol.* **7** (2), 120 (2017).
7. L. F. Vereshchagin, E. N. Yakovlev, B. V. Vinogradov, G. N. Stepanov, K. Kh. Bibaev, T. I. Alaeva, and V. P. Sakun, *High Temp. – High Press.* **6**, 499 (1974).
8. A. N. Babushkin, G. I. Pilipenko, and F. F. Gavrilov, *J. Phys.: Condens. Matter* **5**, 8659 (1993).
9. A. N. Babushkin, *High Press. Res.* **6**, 349 (1992).
10. L. He, Y. Jia, S. Zhang, X. Hong, Ch. Jin, and S. Li, *Quantum Mater.* **1**, 16014 (2016).
11. A. Y. Mollaev, L. A. Saypulaeva, R. K. Arslanov, S. F. Gabibov, and S. F. Marenkin, *High Press. Res.* **22**, 181 (2002).
12. T. R. Arslanov, L. Kilanski, S. López-Moreno, A. Yu. Mollaev, R. K. Arslanov, I. V. Fedorchenko, T. Chatterji, S. F. Marenkin, and R. M. Emirov, *J. Phys. D* **49**, 125007 (2016).
13. T. R. Arslanov, U. Z. Zalibekov, L. Kilanski, I. V. Fedorchenko, T. Chatterji, and R. Ahuja, *J. Appl. Phys.* **128**, 213903 (2020).

Translated by Yu. Ryzhkov