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Magnetization of Cd_3As_2–30 mol % MnAs Composite at High Pressure

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Abstract—In this article, we report research data for the isothermal magnetization of a composite consisting of Cd_3As_2 Dirac semimetal and MnAs ferromagnet (30 mol %) at pressures up to 5 GPa. A magnetic transformation in MnAs inclusions, the size of which presumably varies from a micro- to nanoscale, has been found to be similar to that in bulk MnAs compound, except for a hysteresis in the range of magnetostructural transformation. Under hydrostatic pressure, a magnetization maximum has been revealed at pressures $P \geq 0.77$ GPa, which is indicative of ferromagnetic ordering improvement. The conclusion has been drawn that that the above features are due to the superparamagnetic nature of nanometer-sized MnAs inclusions in the Cd_3As_2 matrix.

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INTRODUCTION

Manganese dissolves in Cd_3As_2 with the formation of a variety of $(Cd_{1-x}Mn_x)_3As_2$ ternary solid solutions in a certain interval of concentrations. An excess of concentration over a limiting value results in the formation of an eutectic alloy containing, apart from $(Cd_{1-x}Mn_x)_3As_2$ solid solution, nanodimensional ferromagnetic MnAs inclusions with a unit cell having $P6_3/mmc$ space-group symmetry and parameters a = 3.72 Å and c = 5.71 Å [1]. At room temperature and a pressure of 0.45 GPa, MnAs experiences a structural transition: the hexagonal structure of the NiAs type with $P6_3/mmc$ symmetry changes to an orthorhombic structure of the MnP type with $Pnma$ symmetry [2]. MnAs compound is a ferromagnet with a Curie point near 318 K. This makes it promising for spintronic devices operating in the terahertz range [3–5].

From studies of thermal emf—a parameter that is very sensitive to phase transformation—it was found that Cd_3As_2 + 44.7 mol % MnAs exhibits a singularity near $P \approx 33$ GPa, which was identified as a sign of the second phase transition [6]. It may be assumed that Cd_3As_2 + MnAs composites subjected to high pressure show the result of concurrent phase transitions: a structural phase transition in the Cd_3As_2 matrix and a spin-reorientation magnetic phase transition in MnAs nanoclusters, which influences the carrier transport and magnetic structure of the composite. That is why it is of interest to examine the magnetic structure in a pressure range near the second phase transition [6]. At the same time, magnetic transformations in MnAs clusters, which are usually predicted at moderate pressures near the domain of the first phase transition (to 5 GPa), can be studied by measuring the magnetization under pressure.

In this article, the influence of pressure up to 5 GPa on the behavior of the isothermal magnetization of Cd_3As_2 + 30 mol % MnAs composite is considered.

1. EXPERIMENTAL

Cd_3As_2 + 30 mol % MnAs crystals were synthesized by the vacuum-sealed ampoule method from Cd_3As_2 and MnAs precursors at the manganese arsenide melting point [7]. Grown samples were examined by the methods of X-ray diffraction and scanning electron microscopy (SEM). The elemental analysis of Cd_3As_2 + 30 mol % MnAs composites showed that

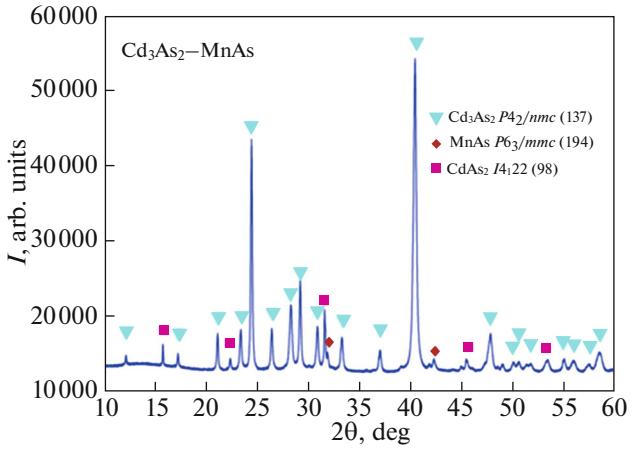

Fig. 1. X-ray diffraction pattern of the Cd_3As_2 + 30 mol % MnAs sample.

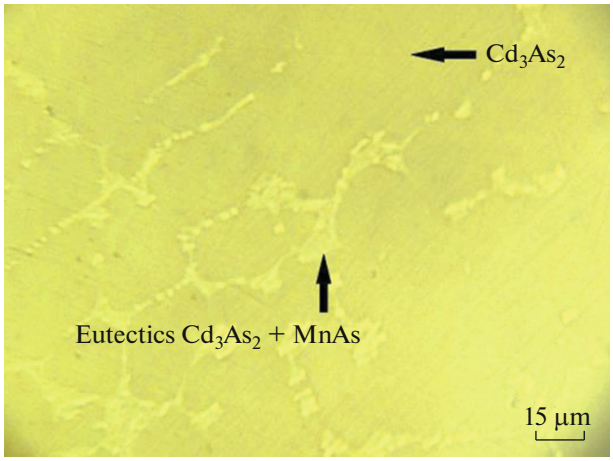

Fig. 2. SEM image of the cleavage plane of the Cd_3As_2 + 30 mol % MnAs sample.

most of the volume is occupied by the Cd_3As_2 phase and the fraction of MnAs inclusions is less than 5%. SEM studies did not reveal granules possibly because of small charactesristic sizes (Fig. 1).

It was established that two main phases in diffraction patterns are tetragonal α-CdAs and hexagonal MnAs (Fig. 2). In addition, there exists a minor amount of the $CdAs_2$ phase.

High-pressure (up to 6 GPa) magnetic measurements were made in a Toroid setup [8] both at room temperature and at atmospheric pressure in the temperature interval 180–350 K. The isothermal magnetization was measured under pressure by the induction method. A cylindrical sample ≈3.5 mm long and ≈1.2 mm in diameter was placed in two inductively coupled coils with a maximal number of turns $n = 10$. The absolute measurement error was no greater than 5%. An external magnetic field was generated by a multiturn solenoid with field intensity $H \le 5$ kOe. A teflon working cell ≈80 mm^3 in volume had eight electrical leads. A nonconducting ethanol : methanol = 4 : 1 mixture was used as a pressure-transmitting medium. The pressure inside the cell was controlled with a manganin sensor calibrated from phase transitions in bismuth.

2. RESULTS AND DISCUSSION

Figure 3 shows the temperature dependences of magnetization $M(T)$ measured in magnetic fields H up to 3.6 kOe under atmospheric pressure. Qualitatively, the run of the curves is the same: under cooling, M sharply grows near $T_C \approx 318$ K due to the nonmagnetic–ferromagnetic transition [9]. As the field rises to 3.6 kOe, the Curie point slightly shifts toward T_C = 321 K (T_C was found from maxima in the dependences dM/dT calculated for $M(T)$ under heating). Such behavior substantiates the supposition that the magnetic properties of MnAs nanoinclusions in the Cd_3As_2 matrix are similar to those of the bulk material. At the same time, a remarkable feature of the ferromagnetic transition is the absence of temperature hysteresis, which is characteristic of magnetization M and indicates a first-order phase transition in bulk MnAs. This magnetic transition is associated with the β–α transformation (where α is the orthorhombic nonmagnetic phase and β is the hexagonal ferromagnetic phase) и exhibits a hysteresis of about 8–12 K. However, the absence of magnetization hysteresis may be akin to the situation observed in MnAs(001)/GaAs(111) epitaxial layers, where the region of α and β phase coexistence is absent [10].

The isothermal ($T = 293$ K) pressure dependences of magnetization, $M(P)$, for two values of magnetic field strength are shown in Fig. 4. Under atmospheric pressure ($P = 0$ GPa), M equals 0.05 and 7.69 (A m^2)/kg in magnetic fields of 500 Oe and 5 kOe, respectively. The initial value of M in weak magnetic fields is consistent with earlier data [11]. The dependences $M(P)$ have maxima, which indicate a magnetic transformation taking place in MnAs clusters. The rise of M near the maxima reaches 80% and 23% of the initial value for $H = 500$ Oe and 5 kOe, respectively. In addition, with increasing H, the region of phase transformation shifts toward higher pressures from 0.77 to 0.90 GPa.

Thus, the following scenario may be suggested to explain the magnetization behavior: the rise of magnetization at low pressures ($P < 1$ GPa) is associated with a higher degree of ordering of the ferromagnetic state in MnAs inclusions, which are most likely in the superparamagnetic state under atmospheric pressure. That is, the application of pressure and magnetic field stabilizes ferromagnetic ordering owing to MnAs magnetic moment alignment, as can be judged from the sharp growth of M. This, in turn, may indicate that MnAs inclusions have nanometer sizes. At pressures $P > 1$ GPa, magnetization M monotonically decreases.

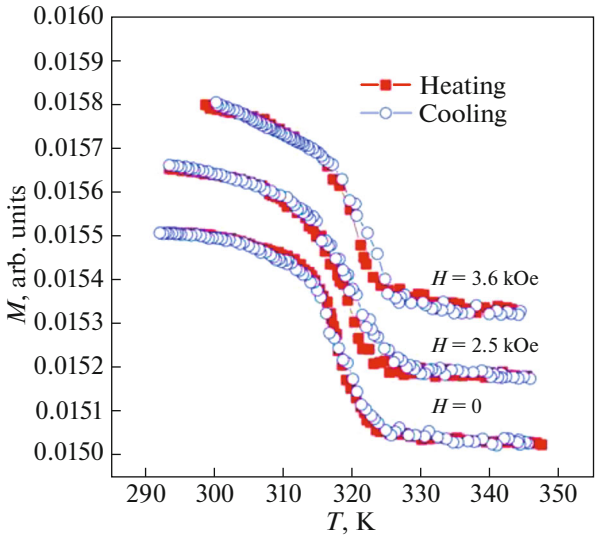

Fig. 3. Temperature dependences of magnetization near the region of the paramagnetic–ferromagnetic transition taken in magnetic fields H up to 3.6 kOe. Heating/cooling curves are shifted relative to each other.

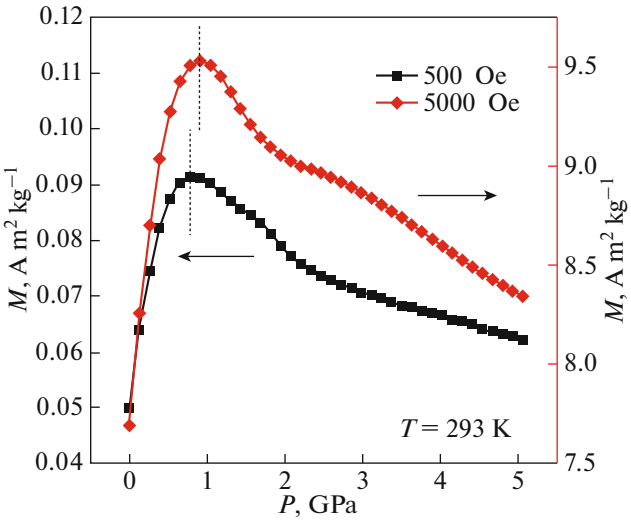

Fig. 4. Pressure dependences of magnetization taken at 293 K in magnetic fields of 500 Oe and 5 kOe.

This can be attributed to the formation of an antiferromagnetic state, since the maximum of M declines with magnetic field increasing to 5 kOe.

It should be noted that the same situation was observed for MnP clusters in a $CdGeP_2$ semiconductor matrix because of pressure-induced changes in magnetic interactions in clusters causing the formation of an antiferromagnetic phase [12].

Since $M(P)$ gradually drops with pressure increasing to 5 GPa (Fig. 4), one may conclude that MnAs clusters most likely become nonmagnetic at still higher pressures.

CONCLUSIONS

The temperature behavior of magnetization $M(T)$ in Cd_3As_2 + 30 mol % MnAs composite was studied in the temperature interval 290–350 K under atmospheric pressure and at pressures $M(P)$ up to 5 GPa. From the dependences $M(T)$ and $M(P)$ it was concluded that the observed features of magnetic transformations substantiate the nanoscale dimensionality of MnAs clusters (this conclusion is difficult to draw from SEM data). As follows from the dependence $M(P)$, clusters experience magnetic transformation into an antiferromagnetic state at pressures below 1 GPa and magnetization M monotonically declines with increasing pressure (up to 5 GPa).

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