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Changes in the magnetization hysteresis direction and structure-driven magnetoresistance of a chalcopyrite-based magnetic semiconductor

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

1 Amirkhanov Institute of Physics, Dagestan Scientific Center, RAS, 367003 Makhachkala, Russia
2 Institute of Physics, Polish Academy of Sciences, Al. Lotnikow 32/46, 02-668 Warsaw, Poland
3 CONACYT Research Fellow, Centre for Corrosion Research, Autonomous University of Campeche, Av. Héroé de Nacozari 480, Campeche, 24070 México
4 Kurnakov Institute of General and Inorganic Chemistry RAS, 119991 Moscow, Russia
5 National University of Science and Technology MISiS, 119049, Moscow, Russia
6 Institute Laue-Langevin, Boîte Postale 156, 38042 Grenoble Cedex 9, France
7 Dagestan State University, Faculty of Physics, 367025 Makhachkala, Russia

E-mail: arslanovt@gmail.com

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Abstract
An unusual change in the hysteresis direction is believed as rare phenomenon associated with perovskite-type structure. Such ‘anomalous’ magnetization hysteresis could possess a direct impact on the giant magnetoresistance (MR). Here we demonstrate that the room-temperature magnetization versus pressure for chalcopyrite semiconductor Zn_{1-x}Mn_{x}GeAs_{2} with x = 0.01 follows a usual direction of hysteresis, while the direction turns into anomalous for x = 0.07. Both these phenomena are results of a pressure-induced structural transition occurring in the host material, as is evident from volumetric measurements and \textit{ab initio} calculations. This structural transition gives rise to the pressure-enhanced large MR and changes it drastically. Unlike the case of x = 0.01 where MR can be well reproduced within a theoretical approach, the presence of magnetic inhomogeneities for x = 0.07 induces an unexpected crossover from large positive to non-saturating negative MR (~92% at H = 5 kOe) in the new high-pressure phase. These results suggest that Zn_{1-x}Mn_{x}GeAs_{2} provides an example of a chalcopyrite-based material whose functional possibilities could be expanded through a new type of ‘structure-driven’ MR.

Keywords: anomalous hysteresis, large magnetoresistance, high pressure, spintronics, magnetic semiconductor

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(Some figures may appear in colour only in the online journal)
accompanied with anomalous hysteresis. This phenomenon occurs when the rotation direction of the conventional hysteresis is inverted. Dho et al [2] reported an observation of the anomalous ‘clockwise’ behavior of a thermal hysteresis in magnetization and resistivity of half-doped manganites near the Néel temperature upon cooling and warming. Noteworthy is that the anomalous hysteresis characteristic in resistivity gives rise to a very large magnetoresistance (MR) effect [3, 4]. Since the origin of such unusual behavior remains unclear, available reports explain it by the geometrical characteristics of the magnetic ordered interactions in the lattice [2] or enhanced spin disorder at grain boundaries [3, 4]. Thus far, an anomalous change in hysteresis has been achieved by doping or applying a magnetic field. In this work, we present an observation of similar anomalous hysteresis in magnetization as a function of pressure discovered in a chalcopyrite-based magnetic semiconductor. As a guide, the usual hysteresis of the physical properties under pressure corresponds to the direction when the reverse run (decompression) goes back below with respect to the forward run (compression); otherwise, it will occur by anomalous or upstream hysteresis.

The family of Mn-doped II-IV-V 2 semiconductors have attracted considerable attention both experimentally [5–7] and theoretically [8, 9] due to room-temperature (RT) ferromagnetism which holds potential in the modern concept of spin devices. Although the scientific interest devoted to these materials is smaller than that of III-V diluted magnetic semiconductors (DMS) [10], interest in these compounds has been resurrected due to the unusual properties of their magnetic inhomogeneities such as recent demonstrations of metamagnetic-like behavior under pressure, as well as the path to pressure controlled response of magnetic clusters at RT [11, 12]. Mn-doped ZnGeAs2 is the most representative due to its high Curie temperature $T_C \approx 367\, K$ [13]. The strong dependence of the critical temperature from Mn-doping $x$ opens a unique picture for this compound, as shown in figure 1. In the limit when $x < 0.07$, the low-dilution regime is realized, as characterized by long-range carrier-mediated ferromagnetism, where only a small fraction of all the Mn$^{2+}$ ions are magnetically active [14, 15]. A more intriguing situation occurs when the chemical composition is between $0.07 \leq x \leq 0.182$ which leads to the existence of two types of phase separations: MnAs clusters with hexagonal or orthorhombic structure [16]. Very recently, the presence of MnAs clusters was also confirmed by Raman spectroscopy experiments [17]. In contrast, the low-temperature area shows a complex character with non-interacting MnAs clusters [18].

From figure 1, we have a special interest in two compositions of Zn$_{1-x}$Mn$_x$GeAs$_2$ with $x = 0.01$ and $x = 0.07$ since they show different magnetic nature. We find that applying a high pressure to these compositions reveals two type of hysteresis in isothermal magnetization. Both usual and anomalous hysteresis are closely linked with the structural transition occurring in the chalcopyrite lattice, as is supported by volumetric measurements and ab initio calculations. In addition, the structural transition gives rise to the pressure-enhanced large MR and changes it drastically. Our results highlight the role of pressure-induced structural transitions on the behavior of magnetization hysteresis and their contribution to MR that has remained beyond understanding. Thus, based on Zn$_{1-x}$Mn$_x$GeAs$_2$ we are able to identify a peculiar type of MR, called the ‘structure-driven MR effect’, which is favorably realized at RT.

2. Experimental and computational details

2.1. Samples
Zn$_{1-x}$Mn$_x$GeAs$_2$ crystals were grown using a direct fusion method from a stoichiometric ratio of high purity powders of ZnAs$_2$, Ge and Mn, as described previously in [13–16]. The growth was performed at a temperature of about 1200 K. Mn-doped crystals were cooled from the growth temperature down to 300 K with a relatively high speed (about 5–10 K s$^{-1}$). The chemical composition of the samples was controlled using the energy dispersive x-ray fluorescence method (EDXRF). EDXRF analysis shows that our samples have $x$ changing in the range from 0 to 0.07. Within our measurement accuracy, all the studied crystals have the correct stoichiometry of Zn$_{1-x}$Mn$_x$Ge:As equal to a ratio 1:1:2. For chemical composition with $x = 0.07$ we found that the addition of Mn to the alloy significantly lowered their crystal quality. For this sample the powder x-ray diffraction (XRD) pattern confirmed the existence of two more phases—hexagonal and orthorhombic MnAs phases, similar to [16].

2.2. High-pressure measurements
High-pressure measurements were conducted in the Toroid type device [19] under ambient temperature conditions. The pressure transmitting medium used is a mixture of ethanol-methanol 4:1, which remains liquid up to 10 GPa at RT and ensures highly hydrostatic pressure generation. The studied...
samples had a cylindrical shape for magnetization as well as Hall geometry (3 × 1 × 1 mm³) for transport and MR measurements. High-pressure zero-field magnetization measurements were carried out using inductively coupled small coils in which a sample was inserted. The measured magnetic field produced by the coils was about 20 Oe. Volumetric measurements were performed by using an accurate strain-gauge technique [20]. We also conducted an XRD analysis for the two selected compositions: \( x = 0.01 \) and \( x = 0.07 \). The quality of the diffraction profile remains equal for samples before and after the pressure cycle, confirming the recovery of the structure as well as reversibility of the observed phase transitions (see supporting information, figure S1, stacks.iop.org/JPhysD/49/125007/mmedia).

2.3. Ab initio calculations

Calculations of the total energy were performed within the framework of density functional theory and the projector-augmented wave (PAW) [21, 22] method as implemented in the Vienna ab initio simulation package (VASP) [23–26]. We use a plane-wave energy cutoff of 360 eV. For the exchange correlation energy, we have used the generalized gradient approximation (GGA) in the AM05 prescription [27–29]. The Monkhorst–Pack scheme was employed to discretize the Brillouin zone integrations [30] with meshes \( 4 \times 4 \times 2 \) and gamma point for chalcopyrite ZnGeAs² and for Zn₀.₉₈₁₅Mn₀.₀₁₈₅GeAs₂, respectively. In the relaxed equilibrium configuration, the forces are less than 2 meV Å⁻¹ per atom in each of the Cartesian directions.

3. Results and discussion

3.1. High-pressure magnetization and transport

In order to elucidate the hysteresis behavior in Zn₁₋ₓMnₓGeAs₂, we compared the high-pressure behavior upon compression and decompression between the magnetization for two compositions with \( x = 0.01 \) and \( x = 0.07 \) (figures 2(a) and (c)). First, after application of high pressure we observed unexpected upturn in magnetization by 6% at \( P \sim 2 \) GPa, because the composition with \( x = 0.01 \) is paramagnetic (PM) at RT (\( T_C \sim 30 \) K) [14]. As we will argue further, this event of pressure-tuned ferromagnetic ordering is consistent with MR evolution (section 3.4). Csontos et al. [31] reported such an observation caused by enhancement in the exchange interaction between uncorrelated Mn²⁺ ions in the PM state at very low temperatures. In the same way, the pressure dependence of ac susceptibility studies performed for the sample with \( x = 0.01 \) and nearest low-dilute composition (\( x \leq 0.04 \)) show distinct peaks, confirming a transition from PM to the magnetically ordered state (inset to figure 2(a)). Upon decompression, a pressure hysteresis in magnetization was developed, which corresponds to the usual direction (figure 2(a)).
The pressure-magnetization characteristic for $x = 0.07$ under compression follows without significant changes with initial value $2.7 \times 10^{-4}$ emu g$^{-1}$ and dropped around ~7 GPa. In contrast to the composition with $x = 0.01$, anomalous hysteresis arises in magnetization for $x = 0.07$, at which the direction of decompression is upstream (figure 2(c)). We want to stress that such a finding has not been previously observed. Nevertheless, a main reason of the changes in the magnetization hysteresis should be closely related to MnAs clusters within the host structure. Even for the bulk material of MnAs, whose magnetic properties are well studied [32], the behavior of isothermal magnetization versus pressure demonstrates the usual hysteresis direction.

Here we point out a different origin for such unusual magnetization hysteresis, in contrast to the situation observed in perovskite manganites. For instance, as discussed in [2], the anomalous thermal hysteresis of the magnetization in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ has been associated with the field-induced transition from the antiferromagnetic to the ferromagnetic phase. This anomalous hysteresis is explained by the competing interaction between the 3D ferromagnetic and 2D $A$-type antiferromagnetic orderings [2]. However, as displayed in figure 2(c), the behavior of the isothermal magnetization near 7 GPa indicates that the transition from the ferromagnetic to the paramagnetic state is caused by MnAs clusters at compression. A possible reason for the realization of the unusual hysteresis in $\text{Zn}_{1-x}\text{Mn}_x\text{GeAs}_2$ with $x = 0.07$ could be the following: the application of pressure leads to a non-uniform response of the chalcopyrite structure (matrix) and MnAs clusters at decompression. In other words, an ‘outrunning’ of the reverse direction of magnetization is probably due to different rates of cluster and matrix relaxation, because of the difference of their bulk properties. As a result, a reversible transition from the paramagnetic state to the ferromagnetic state occurs earlier, showing the change in the hysteresis direction. Furthermore, the adjustment of decompression rates (artificial acceleration) has no effect on the hysteresis process, indicating that the occurrence of unusual hysteresis is not an experimental artifact. Another explanation of the unusual hysteresis could be based on the competing interaction between the hexagonal and orthorhombic MnAs clusters under pressure, similar to that in $\text{Cd}_{1-x}\text{Mn}_x\text{GeAs}_2$ [11]. Therefore, high-pressure studies of transport and magnetotransport properties will be helpful to elucidate this complex issue.

Figures 2(b) and (d) show the changes in resistivity $\rho(P)$ for both samples. A sharp drop by at least two orders of magnitude of $\rho$ in the high-pressure area ($P \sim 6.2$ and ~7 GPa for $x = 0.01$ and $x = 0.07$, respectively) has taken place. During the decompression process, we observed that both samples follow the usual hysteresis that commonly indicates a polymorphic phase transition [33]. As can be seen from figures 3(a) and (b), the Hall effect measurements also point to such behavior, showing a dramatic increase in effective hole concentration as well as decrease of the Hall mobility. Thus, we expect the pressure-induced structural transformation occurring in a chalcopyrite lattice underpins both usual and the anomalous hysteresis.

For the visual analysis of hysteresis behaviour of magnetization and resistivity we considered the hysteresis width, which is defined as a difference between the points of discrepancy and convergence of the curves. Data were collected on the series of crystals of $\text{Zn}_{1-x}\text{Mn}_x\text{GeAs}_2$ with 0.01 $\leq x \leq$ 0.07 (see supporting information, figure S2). Remarkably, in the inhomogeneous regime ($x = 0.07$) the magnetization hysteresis width becomes significantly smaller while the hysteresis width reproduced from $\rho(P)$ increases (figures 2(c) and (d)). This circumstance allows one to conclude that the MnAs clusters contribute not only to magnetic behavior, but also to the structural transition of the host structure. However, the allocation of a distinct cluster contribution to the structural transitions is a difficult challenge.

3.2. Volume changes

As a next step we examined the possibility of expected structural transitions in our samples. It is known that the nearest polymorphic modification for pure ZnGeAs$_2$ is the disordered zinc blend structure with cubic ZnS type [34]. We carried out volumetric measurements on the $\text{Zn}_{1-x}\text{Mn}_x\text{GeAs}_2$ with $x = 0.01$, in order to evaluate the effect of Mn substitution in the lattice. Note that such a substitution appears in the whole range of Mn doping, since a very small fraction of Mn sites lie on a Zn vacancy. Figure 4 shows the experimentally obtained volume ratio $V/V_0$ for $x = 0.01$. We include the fit of the
data by using the third-order Birch–Murnaghan equation of state (EOS) [35]. A volume collapse of ~5% was observed at \( P \approx 6.5 \text{ GPa} \) which is consistent with the drop in \( \rho(P) \) (figure 2(b)), suggesting a first-order structural phase transition. EOS fitting to chalcopyrite (phase I) gives the value of bulk modulus \( B_0 = 83 \text{ GPa} \) and \( B'_0 = 4 \), which is in agreement with results obtained for pure ZnGeAs\(_2\) [36]. For the new high-pressure phase II the values of \( B_0 = 87.1 \text{ GPa} \) and \( B'_0 = 4 \) are quite different than those expected for ZnS type structure [37].

### 3.3. Ab initio calculations for the structural transition

To provide a better understanding at the atomic level of the structural changes, we performed ab initio calculations by using the VASP package [23–26]. Our calculations start with the optimization of ZnGeAs\(_2\) in the chalcopyrite structure, for which we found the structural parameters \( V_0 = 361.7 \text{ Å}^3 \), \( a = 5.6753 \text{ Å}, \text{ and } c = 11.2294 \text{ Å} \), in good agreement with experimental values [38]. To study the crystal of the low-dilution Zn\(_{1-x}\)Mn\(_x\)GeAs\(_2\) we use a supercell of 216 atoms \((3 \times 3 \times 3 \text{ times the primitive cell})\), which corresponds to \( x = 0.0185 \). For the high-pressure phase we consider several structures proposed in the literature for other tetragonal chalcopyrites including the distorted cubic zinc blend phase. The calculated total energy-volume curves for the most representative structures are displayed in figure 5(a). According to our results, the chalcopyrite structure undergoes a first-order phase transition to a distorted fcc structure at ~5 GPa (figure 5(b) and also see supporting information, figure S3). This high-pressure phase is similar to distorted ZnS and NaCl-type structures. We found that Mn has a coordination of 4 in the chalcopyrite, while in the high-pressure phase the coordination is 6, where the change in coordination is related with the packing efficiency criteria experimented at high pressures. To correlate the experimental and theoretical results, we calculate the bulk modulus of both phases. For chalcopyrite, the values are \( B_0 = 67 \text{ GPa} \) with \( B'_0 = 5.8 \), while for the second phase the values are \( B_0 = 84.4 \text{ GPa} \) with \( B'_0 = 4.0 \). According to an ab initio study performed with a local density approximation (LDA) [38], the bulk modulus of ZnGeAs\(_2\) was 72.1 GPa. It is well known that the generalized gradient approximation (GGA) underestimates the bulk modulus values of crystals. Hence, this could be the reason for obtaining a lower value of \( B_0 \) for the low-pressure phase in comparison with experimental results.

### 3.4. High-pressure magnetotransport

To shed light on the effect of the structural transition in the emergence of both usual and anomalous magnetization hysteresis, we made magnetotransport measurements over a wide pressure range. Figures 6 and 7 display the appearance of the pressure-enhanced MR effect in Zn\(_{1-x}\)Mn\(_x\)GeAs\(_2\) with \( x = 0.01 \) and \( x = 0.07 \), respectively. At ambient pressure the MR effect is negligible.

#### 3.4.1. Low-diluted case (\( x = 0.01 \))

Now we can see that the observable increase in magnetization for \( x = 0.01 \), as can be seen in figure 2(a), correlates well with the appearance of negative MR \( \approx 4 \% \) at \( P = 2.6 \text{ GPa} \) (figure 6). This negative MR in addition to magnetization upturn suggests a magnetic transition to the ferromagnetic state as discussed above. A very similar negative MR has been reported for most of the Mn-doped III-V DMS, as a sign of carrier-mediated ferromagnetism [39–41]. Hence, we adopted the \( p-d \) model of magnetic scattering that was successfully applied to the description of MR in (In,Mn)Sb DMS [39, 42]. In the ferromagnetically ordered state, the linear part of negative MR can be expressed as:

\[
\text{MR} = -4 \mathcal{P} \frac{J_{pd}}{g \mu_B} M(H),
\]

where \( \mathcal{P} \) is the spin polarization of the carriers, \( J_{pd}/\mathcal{P} \) is the strength of the scattering characterized by a constant of exchange interaction \( J_{pd} \) and the spin-independent part of the potential \( \mathcal{V} \), \( M = g \mu_B \langle S_z \rangle \) is magnetization, while \( \langle S_z \rangle \) is the average field-direction component given by \( B_{S2}(c) \), the Brillouin function with an argument of \( \alpha = g \mu_B H/ka \text{Teff} \). In the ferromagnetic state, where \( \mathcal{P} \propto M \) as typically for DMS [40], we used the experimental value for \( \mathcal{P} = 6\% \) at \( P = 2.6 \text{ GPa} \).
The value of $\xi$ in the scattering process, as is predicted by equation (1).

part of negative MR at $H \geq 1$ kOe is due to spin-polarized carrier arrangements in the scattering process, as is predicted by equation (1). The value of $J_{\text{pd}}/\sigma$ shows an increase of $\sim 494\%$ from 2.6 GPa to 6.24 GPa, evidencing a noticeable enhancement in the exchange coupling between isolated Mn$^{2+}$ ions and, hence, the magnitude of the negative MR with pressure.

Moreover, the full curves of negative MR at the limit of $H = 5$ kOe can be well fitted using a semiempirical expression [43]:

$$\text{MR} = -a^2 \ln(1 + b^2 H^2) + \frac{c^2 H^2}{(1 + d H^2)}, \tag{2}$$

where $a$ and $b$ are fitting parameters including the physical characteristics of the $p$–d exchange interaction, which has been demonstrated to increase with pressure.

Surprisingly, the structural transition entails a positive MR, with a record value of 110% at $P = 6.7$ GPa and $H = 5$ kOe (figure 6). At this pressure region of phase II, the effective hole concentration runs up dramatically and becomes metallic-like with reduced Hall mobility ($\mu_H \sim 0.25 \text{cm}^2/(V\cdot\text{s})^{-1}$, as can be seen from figure 3(a)). Thus, the observable positive MR is quite a lot larger than it should be, except when considering ordinary metals with spin orbital contribution. A two-band model [43, 44], which is included in the second term of equation (2) (where fitting parameters $c$ and $d$ mean the conductivity and mobility of carriers in the two spin-split bands) gives acceptable reproduction for the positive MR (see curve at 9.1 GPa in figure 6). In general, this analysis emphasizes that positive MR is likely related to $p$–d spin-split bands and strongly governed by pressure [45], but is a result of structural changes.

3.4.2. Inhomogeneous case ($x = 0.07$). A qualitatively different situation is presented in figure 7 for the sample with $x = 0.07$. Although MR curves at pressures up to $P \sim 1$ GPa can be fitted using equation (2) (inset to figure 7), the magnitude of the positive MR does not exceed $\sim 1.5\%$, so this effect is rather due to the action of the Lorentz contribution on the mobile carriers. A crossover from positive to negative MR is inhibited by the application of pressures with gradual tendency for saturation and finally becomes negative with an amplitude of about $\sim 54\%$ at $P = 7.2$ GPa. At a pressure $P \geq 2$ GPa, the MR curves could not be fitted by equation (2). This strongly suggests that negative MR dominates over carrier scattering on the localized MnAs clusters. The clusters become better correlated and rather ferromagnetically aligned with increasing pressure. When the transition occurs to phase II, the behavior of MR for $x = 0.07$ taken at $P = 8.9$ GPa is very unexpected. It shows a very large crossover from positive to negative MR that has a complicated nature. The possible causes of this crossover might be connected with intercluster interaction between two types of hexagonal and orthorhombic MnAs clusters [11, 16] within the host. Considering that the pressure reduces the distance between randomly spatial arrangements of clusters, eventually it may induce a tunneling channel with saturated negative MR, as commonly happens for granular materials [46]. During the structural transition,

The expression for coefficients $a$ and $b$ are given in [43]. The values of $a = 0.08$ and $b = 2.18$ at $P = 2.6$ GPa increase with $a = 0.17$ and $b = 3.46$ at $P = 6.24$ GPa, indicating an enhancement of the strength of the $p$–d coupling between Mn$^{2+}$ spin population by increasing pressure.

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**Figure 6.** The pressure-enhanced magnetoresistance (MR) in Zn$_{1-x}$Mn$_x$GeAs$_2$ with $x = 0.01$. Dashed lines in the inset to (a) are results of fitting with equations (1) and (2), denoted as fit 1 and fit 2, respectively.

**Figure 7.** The pressure-enhanced magnetoresistance (MR) in Zn$_{1-x}$Mn$_x$GeAs$_2$ with $x = 0.07$. The inset in (b) shows the applicability of equation (2) to fitting of positive and negative MR in the pressure range $P \geq 1$ GPa, where starting from pressures $2$ GPa it is not relevant.

and $T_{\text{eff}} = 297$ K. The fitted lines of the experimental results are presented in the inset of figure 6 showing that the linear part of negative MR at $H \geq 1$ kOe is due to spin-polarized carriers in the scattering process, as is predicted by equation (1). The value of $J_{\text{pd}}/\sigma$ shows an increase of $\sim 494\%$ from 2.6 GPa to 6.24 GPa, evidencing a noticeable enhancement in the

$^7$The effective temperature $T_{\text{eff}} = T + T_{\text{ef}}$ contains an empirical antiferromagnetic coupling parameter describing the interaction between Mn ions. We assumed this term is negligible, since it is only a fitting parameter.
there is a possible change in the transport from tunneling MR to the metallic regime [47]. Interestingly, the result of such behavior is accompanied by an unusual competition between structurally driven large positive and nearly-linear large negative MR at \( H > 4 \) kOe with no saturation trend (figure 7). The latter may be evidence for high-polarized transport of carriers by MnAs interfaces, similar to the case of self-organized Mn nanocolums in the Ge matrix [48].

Figure 8 shows the Hall resistance against magnetic fields at various pressures of the sample with \( x = 0.07 \). We note that the pressure-enhanced MR evolution is well correlated with the development of the anomalous Hall effect (AHE) for both samples. The characteristic is similar for the sample with \( x = 0.01 \) and not shown. In addition, AHE for the sample with \( x = 0.07 \) becomes very pronounced in the new high-pressure phase II, as seen in figure 8. Furthermore, this AHE confirms the transport mechanism due to high-polarized carriers in the transformed high-pressure phase where large crossover from positive to negative MR as well as anomalous magnetization hysteresis observed.

4. Conclusion

Summarizing, we presented the pressure-induced changes in magnetization hysteresis in connection with the pressure-enhanced large MR on two representative compositions of Zn\(_{1-x}\)Mn\(_x\)GeAs\(_2\) (\( x = 0.01 \) and \( x = 0.07 \), respectively) with different magnetic origin. For \( x = 0.07 \), we found that the direction of the isotothermal magnetization hysteresis turns into anomalous and therefore provides an initial observation of this phenomenon under pressure. An unexpected upturn in magnetization for \( x = 0.01 \) is verified by the continuously tuned parameter \( J_{zcl} \), which followed the usual hysteresis characteristics at decomposition. It is established that structural transformation in a chalcopyrite lattice is responsible for both types of magnetization hysteresis and causes a remarkable structure-driven MR effect where magnetotransport features depend on the magnetic nature of the compositions. Trying to explain an unusual behavior of hysteresis, we believe that this effect can possibly be attributed to strong interacting clusters and their volume properties [12] in a new high-pressure phase. A deeper understanding of the interplay between the host lattice distortion and MnAs clusters remains to be elucidated in future.

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