

Giant Magnetoresistance of $\text{Me}_x\text{Mn}_{1-x}\text{S}$ (Me = Fe, Cr) Sulfides

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The structural, electrical, and magnetic properties, as well as the magnetoresistance of polycrystalline $\text{Me}_x\text{Mn}_{1-x}\text{S}$ (Me = Fe and Cr) sulfides were investigated in longitudinal magnetic fields of up to 50 kOe over the temperature range 4.2–300 K. The ferromagnetic compound $\text{Fe}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.29$) exhibits the giant magnetoresistance (GMR) effect with magnitude $\delta_H = -450\%$ in a field of 30 kOe at 50 K. Antiferromagnetic $\text{Cr}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.5$) sulfide undergoes a transition to the GMR state ($\delta_H \sim -25\%$ in a field of 30 kOe at 4.2 K) in the region of antiferromagnet–ferromagnet transition ($T_c \sim 66$ K). A mechanism of the GMR in these compounds is discussed. © 2000 MAIK “Nauka/Interperiodica”.

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In spite of a great body of experimental data on the giant magnetoresistance (GMR) phenomenon in manganese lanthanide oxides with the perovskite structure, the mechanism of the GMR effect still remains to be clarified [1]. That is why a search for, and the study of, new compounds exhibiting the GMR effect and having a non-perovskite structure is a topical problem. Earlier [2], we revealed the GMR effect in $\text{Fe}_x\text{Mn}_{1-x}\text{S}$ sulfides derived from manganese monosulfide. The greatest effect ($\delta_H \sim -83\%$) in a transverse magnetic field of 10 kOe was observed for $x = 0.29$. This work reports the results of studying the structural, electrical, magnetic, and magnetoelectric properties of $\text{Fe}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.29$) and $\text{Cr}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.5$) sulfides at temperatures of 4.2–300 K in longitudinal magnetic fields of up to 50 kOe.

Polycrystalline samples of $\text{Me}_x\text{Mn}_{1-x}\text{S}$ (Me = Fe and Cr) were synthesized from pure elemental chromium, iron, manganese, and sulfur in evacuated quartz tubes by high-temperature annealing for a week [2]. X-ray structural analysis was performed on a DRON-2.0 diffractometer with monochromatic $\text{Cu } K_\alpha$ radiation in the temperature range 100–300 K. Electrical resistance was measured potentiometrically on a direct current over the temperature range of 4.2–300 K in longitudinal fields $H = 0, 2, 10, 30,$ and 50 kOe. The magnetic properties were measured on a vibrating-coil magnetometer with a superconducting solenoid in magnetic fields of up to 30 kOe in the range 77–300 K.

According to the X-ray data, the synthesized samples of $\text{Fe}_{0.29}\text{Mn}_{0.71}\text{S}$ and $\text{Cr}_{0.5}\text{Mn}_{0.5}\text{S}$ are solid solutions with the NaCl fcc lattice typical of manganese monosulfide [2, 3]. The X-ray patterns of the samples show

three extra lines with 5% relative intensity, indicating the presence of a possible impurity phase. The fcc lattice parameter of $\text{Fe}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.29$) is $a = 5.186$ Å at room temperature. The compound undergoes a structural transition at $T_s \sim 147$ K, similar to that observed

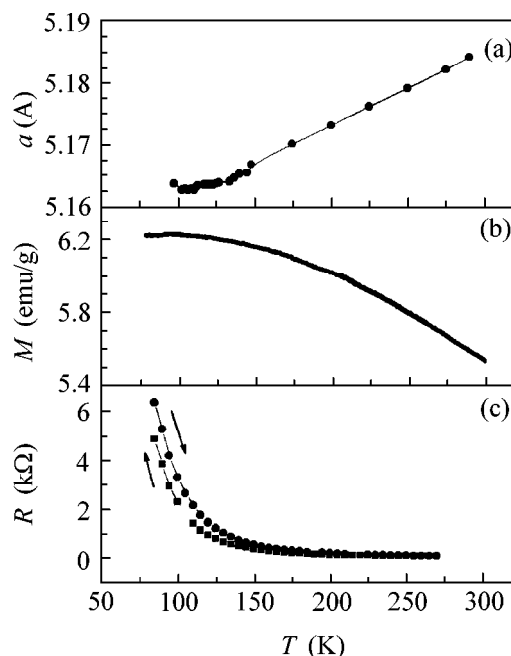


Fig. 1. Temperature-dependent (a) lattice parameter, (b) magnetization, and (c) resistivity of $\text{Fe}_{0.29}\text{Mn}_{0.71}\text{S}$.

for manganese monosulfide at 150 K [3]. As the temperature decreases, the lattice parameter first decreases, next is almost independent of temperature at 120–140 K, and then shows a tendency to increase below 120 K (Fig. 1a). An analogous structural transition also occurs in the $\text{Cr}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.5$) sulfide near 160 K.

Magnetic measurements showed that the samples are ferromagnets at temperatures $T < 300$ K (Me = Fe) and $T < T_c = 66$ K (Me = Cr). The saturation magnetization ($H \sim 7\text{--}10$ kOe) for $\text{Fe}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.29$) is 6.24 emu/g at 300 K. The temperature curves for the magnetization of the sulfides under study are shown in Figs. 1b and 3a. The value of $\rho_{77\text{ K}} \sim 10^2 \Omega \text{ cm}$ found for the resistivity of the samples at 77 K is five orders of magnitude lower than for $\alpha\text{-MnS}$. The conduction in the samples is of the semiconductor type with a thermal hysteresis in zero magnetic field at temperatures below 250 K (Fig. 1c).

Figure 2 demonstrates the temperature curves for the magnetoresistance δ_H of $\text{Fe}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.29$) in longitudinal magnetic fields of 10, 30, and 50 kOe. It was pointed out in [2] that the negative magnetoresistance δ_H for this sulfide increases on lowering the temperature below 250 K in a field of 10 kOe and reaches its maximum value at ~ 160 K. Below 120 K, where the NaCl structure is distorted, the magnetoresistance changes sign. It was established that the change in sign of magnetoresistance disappears upon repeated measurements with the same sample at 120 K. This is likely caused by the hysteresis effects and the magnetic-induced changes in the state of the samples. After field removal, the magnetoresistance is lower than its initial (prior to the experiment) value at $T = 170$ K ($T > T_s$) and higher at 110 K ($T < T_s$). The resulting δ_H value calculated for fields of 0 and 10 kOe at 170 K is equal to -70% for the field buildup and to -50% for the field removal; at 110 K, the corresponding values are $+15\%$ and -127% .

As the magnetic field increases to 50 kOe, the negative magnetoresistance peak shifts to lower temperatures (Fig. 2). At 30 kOe, the negative magnetoresistance δ_H reaches a value of -450% for $\text{Fe}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.29$) at ~ 50 K. At 50 kOe, this value is -87% .

Figure 3 shows the temperature curves for the magnetization (Fig. 3a) and magnetoresistance (Fig. 3b) of $\text{Cr}_x\text{Mn}_{1-x}\text{S}$ ($x = 0.5$). At 66 K, this sulfide undergoes the antiferromagnet–ferromagnet transition, whose nature is caused by the orbital degeneracy of the chromium ions and by the cooperative Jahn–Teller effect. One can see from Fig. 3 that the transition to the negative magnetoresistance state occurs in the range of magnetic transition. The δ_H value increases with lowering temperature and reaches a value of $\sim -25\%$ at 4.2 K in a field of 30 kOe.

An analysis of the experimental data obtained earlier in [2, 5] indicates that the concentration dependences of the electrical and magnetic properties of the

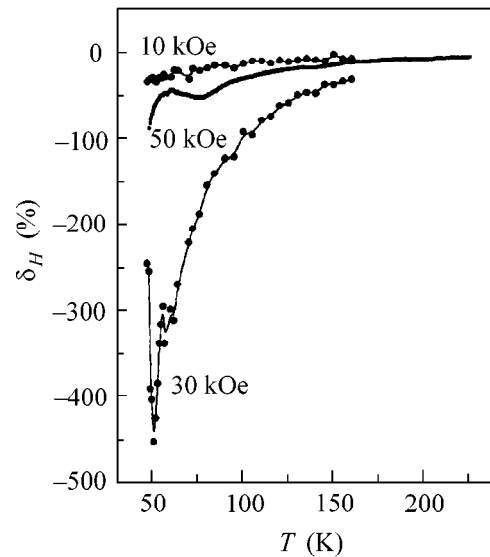


Fig. 2. Temperature-dependent magnetoresistances in a longitudinal magnetic field.

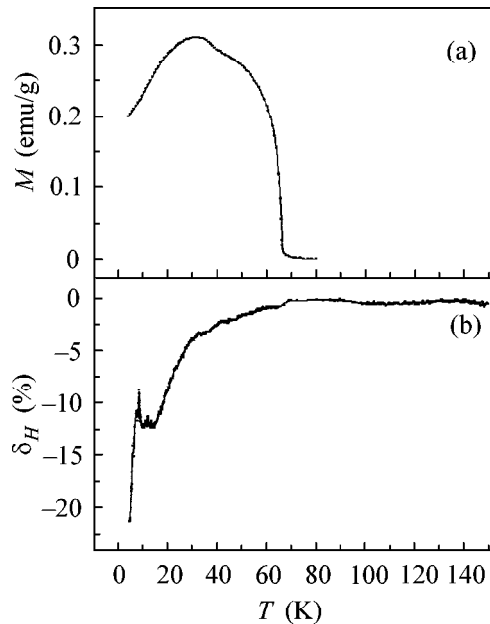


Fig. 3. Temperature-dependent (a) magnetization and (b) magnetoresistance of $\text{Cr}_{0.5}\text{Mn}_{0.5}\text{S}$ in a longitudinal magnetic field of 30 kOe.

solid solutions $\text{Me}_x\text{Mn}_{1-x}\text{S}$ are in many respects similar to those observed for manganese lanthanides. Namely, the ferromagnetic properties emerge simultaneously with the metallization of the samples; that is, the $\text{Me}_x\text{Mn}_{1-x}\text{S}$ samples with low concentration $x < 0.2$ are antiferromagnetic semiconductors, whereas the samples with $x \sim 0.4\text{--}0.6$ are ferromagnetic metals. The GMR effect is most pronounced in the intermediate compositions preceding the concentration transition to

the metallic state. It was assumed in [2] that among the possible GMR mechanisms in iron–manganese sulfides, there is a separation of magnetic and electronic phases, namely, the formation of a system in which the regions of antiferromagnetic semiconductor and ferromagnetic metal coexist. It is established in this work that the behavior of resistivity and magnetoresistance of the samples exhibits not only the temperature hysteresis but also the magnetic hysteresis. It is well known that the probability for the hysteretic effects to occur in the polycrystalline samples increases because of the presence of grain boundaries. However, the polycrystalline version of the hysteretic effects cannot explain the strong temperature shift of the magnetoresistance peak from 160 to 50 K in $\text{Fe}_x\text{Mn}_{1-x}\text{S}$. Since the GMR effect in $\text{Me}_x\text{Mn}_{1-x}\text{S}$ sulfides is observed in the region where the cubic lattice is distorted, a more thorough study of the crystal structure and the nature of the structural transition is necessary for the elucidation of the GMR mechanism. It is not improbable that the crystal structure of sulfides alters under the action of a magnetic field and thermocycling, as, e.g., it occurs in the $\text{RbDy}(\text{WO}_4)_2$ compound exhibiting the Jahn–Teller structural transition [6]. It should also be pointed out that the nature of the structural transition in manganese monosulfide remains to be explored.

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