

# Ferromagnetism and the metal-insulator transition in the magnetic semiconductor system $\text{Fe}_x\text{Mn}_{1-x}\text{S}$

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The results of investigations of the structure, electrical, and magnetic properties in the system of antiferromagnetic semiconductors  $\text{Fe}_x\text{Mn}_{1-x}\text{S}$  ( $0 < x \leq 0.5$ ) are described. It is established that metal-insulator transitions with respect to concentration and temperature are connected with changes in the magnetic properties of the system. © 1998 American Institute of Physics. [S1063-7834(98)02502-7]

It has been observed previously<sup>1,2</sup> that the electric and magnetic state of the system of antiferromagnetic semiconductors  $\text{Fe}_x\text{Mn}_{1-x}\text{S}$  changes as the concentration of iron  $x$  increases until a concentration-induced metal-insulator transition occurs. Studies of systems of magnetic semiconductors based on two monosulfides from the  $3d$  series containing magnetoactive cations such as  $\text{MnS-CrS}$ ,  $\text{MnS-FeS}$ ,  $\text{FeS-CrS}$ ,  $\text{CoS-CrS}$  have shown<sup>3</sup> that cation replacement in the solid solutions is one effective method that can be used not only to regulate the parameters of the metal-insulator transition in these materials but also to significantly change their magnetic characteristics, and also create new magnetic materials with metal-insulator transitions.

In this paper we report the results of studies of the structure, electrical, and magnetic properties of the system of antiferromagnetic semiconductors  $\text{Fe}_x\text{Mn}_{1-x}\text{S}$  undergoing a metal-insulator transition and the interrelation between the metal-insulator transition with respect to concentration and temperature and changes in the magnetic properties.

Samples of the  $\text{Fe}_x\text{Mn}_{1-x}\text{S}$  system ( $0 < x \leq 0.5$ ) were obtained by heating vacuum quartz ampoules containing electrolytic Mn, reduced Fe, and sulphur with a purity of 99.999%, at 1233 K for a week to induce cation replacement. A compound with  $x \sim 0.3$  having special electrical and magnetic properties<sup>2</sup> was synthesized several times using the same fabrication technology.

According to  $x$ -ray structure analysis, all the compounds investigated with  $0 < x \leq 0.5$  in the temperature range 80–300 K were solid solutions with the FCC lattice of  $\alpha$  MnS.

The electrical resistivity  $\rho$  measured by a four-probe potentiometer at constant current showed that, as  $x$  increases, a metal-insulator transition takes place with respect to concentration when  $x_c \sim 0.4$ .<sup>1</sup> It is clear from Fig. 1 that, as the temperature increases, the composition with  $x \sim 0.3$  exhibits a smooth change in the type of conductivity from semiconducting to semimetallic at  $T > 700$  K (a metal-insulator transition with respect to temperature) with a change in activation energy from 0.18 to 0.03 eV.

As the iron concentration  $x$  increases, ferromagnetism appears in the collinear antiferromagnetic host  $\alpha$  MnS (type II) when  $x_{cr} \sim 0.3$ . The following experimental effects confirm the appearance of ferromagnetism and coexistence of antiferromagnetism and ferromagnetism in these compounds:

the appearance of a hysteresis loop for the spontaneous moment, the presence of a Curie temperature, and a deviation from linear behavior for the field dependence of the magnetization at 300 K. The existence of ferromagnetic exchange in this system is confirmed by constructing the magnetic phase diagram based on magnetic measurements and Monte Carlo calculations.<sup>2</sup>

Magnetic measurements show that as  $x$  increases from 0 to 0.2 the temperature behavior of the magnetic susceptibility (the curve  $1/\chi(t)$ ) corresponds to antiferromagnetism in the temperature range 80–700 K. However, the low-temperature measurements of magnetization indicate nucleation of ferromagnetism at sufficiently low concentrations of  $x$ , for example  $x \sim 0.05$ . Figure 2 shows the temperature dependence of the magnetization for the composition  $x \sim 0.3$ . Measurements were made using an automated magnetometer with a superconducting solenoid in the temperature range 4.2–300 K in a field of 700 Oe. The curve  $\sigma(T)$  has a low-temperature anomaly in the range  $35 < T < 40$  K. A similar high-temperature anomaly was observed at  $\sim 45$  K in the system of antiferromagnetic semiconductors  $\text{Li}_x\text{Mn}_{1-x}\text{Se}$ , which is isostructural with the  $\text{Fe}_x\text{Mn}_{1-x}\text{S}$  system.<sup>4</sup> Measurements of the magnetic properties and neutron-scattering investigations of the system  $\text{Li}_x\text{Mn}_{1-x}\text{Se}$  lead these authors to conclude that this low-temperature anomaly is connected with the formation of a canted antiferromagnetic structure and the appearance of a magnetic moment. However, whereas increasing  $x$  and  $T$  in the system  $\text{Li}_x\text{Mn}_{1-x}\text{Se}$  leads to a change from antiferromagnetic ordering to ferromag-

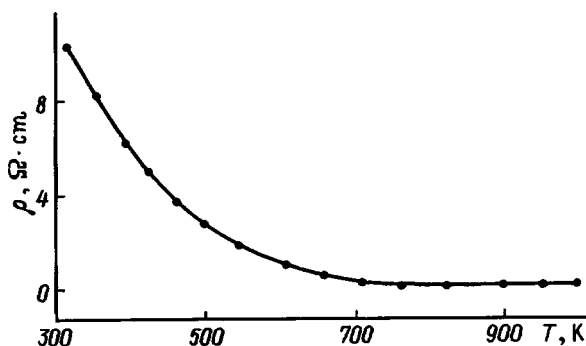


FIG. 1. Temperature dependence of the resistivity of  $\text{Fe}_{0.3}\text{Mn}_{0.7}\text{S}$ .

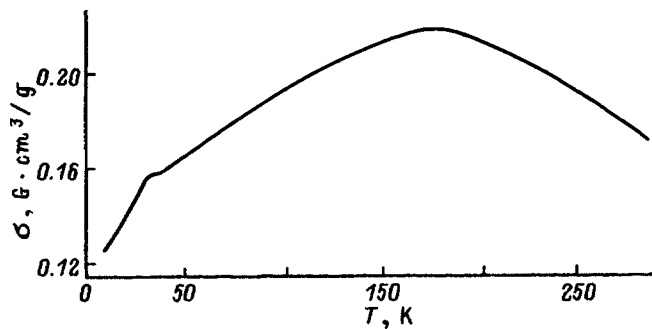


FIG. 2. Temperature dependence of the magnetization of  $\text{Fe}_{0.3}\text{Mn}_{0.7}\text{S}$  in a magnetic field of 700 Oe.

netic, with simultaneous replacement of semiconductor conductivity by metallic conductivity, in the system  $\text{Fe}_x\text{Mn}_{1-x}\text{S}$  the change in electrical and magnetic properties has its own peculiarities: 1) compositions with  $0 < x \leq 0.3$  are impurity semiconductors with nucleated ferromagnetism; 2) compositions with  $0.3 \leq x \leq 0.4$  are ferromagnetic semiconductors in which a smooth high-temperature transition takes place with increasing temperatures from semiconductor to semimetal in the temperature range  $\sim 700\text{--}1000$  K; 3) compositions with  $0.4 \leq x \leq 0.5$  are ferromagnetic semimetals with a Curie temperature  $T_c \sim 780$  K which become metallic at  $T \sim 1000$  K, i.e., they enter the paramagnetic phase.

Thus, the system  $\text{Fe}_x\text{Mn}_{1-x}\text{S}$  for  $0 < x \leq 0.5$  exhibits a metal-insulator transition with respect to concentration  $x$  and temperature  $T$ , which are connected with changes in the magnetic properties. In this system the concentration-induced metal-insulator transition occurs at  $x_c \sim 0.4$ , so that the magnetic AFM  $\rightarrow$  FM transition at  $x_{cr} \sim 0.3$  precedes the metal-insulator transition. As for the temperature transition, with increasing  $x$  and  $T$  the conductivity type is smoothly replaced in the system, accompanied by the magnetic transformation AFM  $\rightarrow$  FM  $\rightarrow$  PM and coexistence of antiferromagnetism and ferromagnetism.

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