Transition to a spin glass and anomalous temperature dependence of the Hall coefficient for a gap-free semiconductor

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A transition to a spin-glass state has been observed at $T \lesssim 1$ K in semimagnetic gap-free semiconductors with the composition $Hg_{1-X}Mn_XTe$ ($0 < X < 0.075$) at Mn concentrations well below the percolation threshold ($X_c = 0.17$). Under these conditions the direct exchange interaction between Mn ions, which gives rise to the spin glass, is negligibly small. The transition to the spin-glass state indicates the existence of a new low-temperature region on the $X-T$ phase diagram.

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The semimagnetic semiconductors $Hg_{1-X}Mn_XTe$ are gap-free semiconductors in the composition range $0<X<0.075$. Bastard and Lewiner[1] have shown that in this case virtural interband transitions across the zero gap can give rise to a long-range exchange interaction. Accordingly, a spin glass may form in the interval $0<X<0.075$ in the model of Ref. 1, even though oscillations of the spin density cannot be established because of the low concentration of free charge carriers. We wish to emphasize that in this situation the appearance of the spin-glass phase is expected at low Mn concentrations—well below the percolation threshold $X_c = 0.17$ determined from the model for direct exchange in a lattice of the HgTe type. According to the direct-exchange model, a spin glass cannot form[2] at $X<X_c$, so that we would expect the semimagnetic semiconductor $Hg_{1-X}Mn_XTe$ with $X<0.075$ to remain a paramagnet down to the very lowest temperatures. To the best of our knowledge, a transition to a spin glass has not previously been observed below the percolation threshold, $X<X_c = 0.17$, in $Hg_{1-X}Mn_XTe$ or in other semimagnetic semiconductors.

In the present experiments we measured the temperature dependence of the Hall coefficient, $R_H(T)$, and of the magnetic susceptibility, $\chi(T)$, over the temperature range $0.05<T<10$ K for *p*-type $Hg_{1-X}Mn_XTe$ ($0<X<0.075$) single crystals with acceptor and donor concentrations $N_A \sim 10^{16}$ cm^{-3} and $N_D \sim 10^{15}$ cm^{-3} (at $T = 4.2$ K). Since structural features resulting from the transition of $Hg_{1-X}Mn_X$ to a spin-glass phase might be observed in these experiments, we attempted to determine $\chi(T)$ in an external magnetic field as weak as possible. For this purpose we measured the magnetic susceptibility with a SQUID in a residual magnetic field $H \sim 0.05$ Oe.

As the temperature is lowered in the region $T<1$ K, the Hall coefficient varies in a quite monotonic manner (Fig. 1); specifically, for $0.2\ \mathrm{K}<T<0.8$ K the $R_H(T)$ curves exhibit a local maximum. The height of this maximum increases sharply with decreasing

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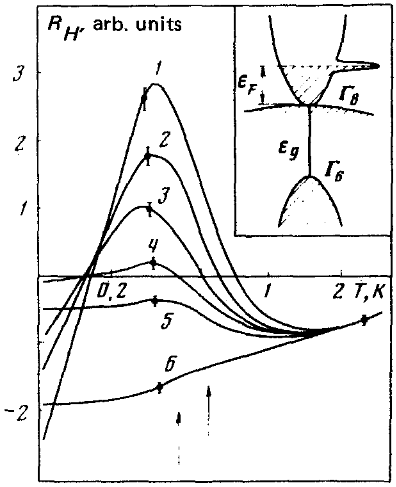

FIG. 1. Dependence of the Hall coefficient R_H on the temperature T for an $Hg_{0.98}Mn_{0.02}Te$ single crystal in various magnetic fields: 1–20 Oe; 2–50; 3–100; 4–150; 5–200; 6–500 Oe. The arrows show the maximum on the $\chi(T)$ curve for this sample. The inset shows the band structure of p-type $Hg_{1-X}Mn_XTe$ semimagnetic semiconductors.

value of the magnetic field in which the Hall voltage is measured. To the left of the peak, $R_H(T)$ does reach saturation even at the very lowest temperatures.

In a magnetic field $H > 200$ Oe the $R_H(T)$ curves become monotonic. It should be noted that the shape of the $R_H(T)$ curves in weak magnetic fields depends strongly on the temperature at which the magnetic field is switched. If it is switched below a certain characteristic temperature on the order of $T_{\max}$, then the field dependence of the Hall voltage, $U_H(H)$, for $T \lesssim T_{\max}$ tends toward a certain finite value rather than zero in the limit $H \to 0$. If **H** is switched at $T \approx 1$ K ($T > T_{\max}$), we find $U_H \to 0$ in the limit $H \to 0$. When this measurement procedure is used, the curves of $U_H(T)$ in a weak field in the range $T < T_{\max}$ change more slowly with the temperature and show a tendency toward saturation (Fig. 2).

The temperature dependence of the susceptibility, $\chi(T)$, is quite different from the law $\chi(T) \sim (T-\theta)^{-1}$ (Fig. 3). As the temperature is lowered, $\chi(T)$ goes through a maxi-

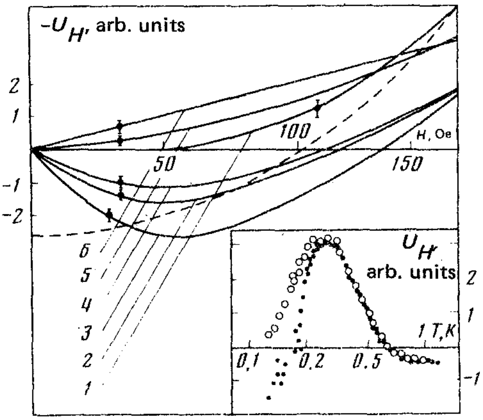

FIG. 2. Dependence of the Hall voltage U_H on the magnetic field H for an $Hg_{0.98}Mn_{0.02}Te$ sample for various temperatures: 1–150 mK; 2–200; 3–250; 4–350; 5–500; 6–700 mK. Solid curves and open circles in the inset: The field was switched at $T > T_{\max}$. Dashed curve and filled circles: At $T < T_{\max}$.

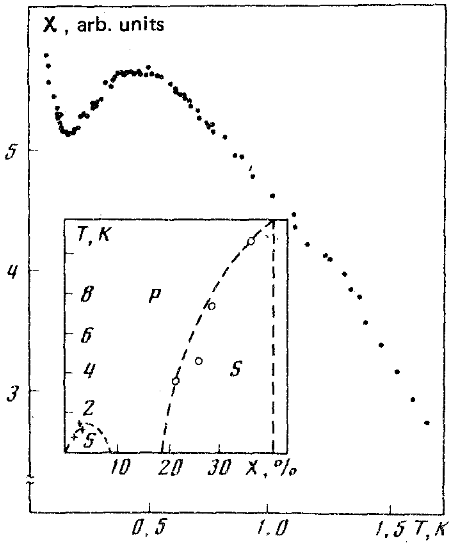

FIG. 3. Temperature dependence of the magnetic susceptibility $\chi(T)$ for an $Hg_{0.98}Mn_{0.02}Te$ sample. The inset shows the T-phase diagram. P—Paramagnetic phase; S—spin glass; +—data of the present experiments; ●—data of Ref. 2; ○—data of Ref. 4 for the semimagnetic semiconductor $Hg_{1-X}Mn_XSe$.

mum and a minimum ($T = T_{min}$). In the region $T < T_{min}$, $\chi(T)$ begins to increase again with decreasing temperature, and this increase continues to $T \approx 0.05$ K.

Anomalous features in the Hall coefficient (Figs. 1 and 2) and in the susceptibility (Fig. 3) are characteristic of a transition to a spin glass. The effects associated with such a transition are known to be extremely sensitive to the strength of the external magnetic field.[3] In a weak field (in the present case at $H < 200$ Oe) the energy of the exchange interaction of the magnetic moments is much higher than the energy of the interaction of these moments with the external magnetic field. This circumstance evidently explains the decrease in the height of the local maximum in $R_H(T)$ with increasing H (Fig. 1). The difference ΔU_H between the temperature dependences of the Hall coefficient in a weak field measured upon switching of the magnetic field either at $T > T_{max}$ or $T < T_{max}$ (Fig. 2) appears when a local freezing of the spins begins. The magnitude of ΔU_H increases with decreasing temperature.

The increase in $\chi(T)$ at $T < T_{min}$ (Fig. 3) may be due to a paramagnetic contribution to $\chi(T)$, either from isolated Mn^{2+} ions which remain in the "pores" of an infinite cluster which results from a transition of $Hg_{1-X}Mn_XTe$ to a spin-glass state or from localized magnetic moments associated with carriers which fill the impurity acceptor band (see the inset in Fig. 1). The acceptor concentration in these samples was $\sim 10^{16}$–10^{17} cm^{-3}, or much lower than the critical Mott concentration ($\sim 10^{18}$ cm^{-3}). Therefore, the presence of localized magnetic moments in the impurity band appears possible.

The slope change of the $\chi(T)$ curve, the local maximum on the $R_H(T)$ curve, and its dependence on the magnetic field and on the temperature at which the magnetic field was switched all imply a transition of the gap-free semimagnetic semiconductor $Hg_{1-X}Mn_XTe$ to a spin-glass state. The T-χ phase diagram (the inset in Fig. 3) thus turns out to be more complex than was suggested previously.[2,3] The spin-glass phase forms not only at $0.17 < X < 0.4$ but also in the interval $0 < X < 0.075$. The right-hand boundary of this interval is set by the opening of a direct gap: At $X > 0.075$, the semimagnetic semiconductors $Hg_{1-x}Mg_xTe$ are not gap-free, and the mechanism of Bastard and Lewiner[1] becomes impossible.

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