Magnetotransport Properties of p-Type (In,Mn)As Diluted Magnetic III-V Semiconductors

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Magnetotransport properties of p-type (In,Mn)As, a new diluted magnetic semiconductor based on a III-V semiconductor, are studied. The interaction between the holes and the Mn $3d$ spins is manifested in the anomalous Hall effect, which dominates the Hall resistivity from low temperature (0.4 K) to nearly room temperature, and in the formation of partial ferromagnetic order below 7.5 K, which is a cooperative phenomenon related to carrier localization. The coexistence of remanent magnetization and unsaturated spins as well as the large negative magnetoresistance at low temperatures is explained by the formation of large bound magnetic polarons.

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Recent progress in molecular beam epitaxy (MBE) has enabled the successful preparation of a new class of diluted magnetic semiconductors (DMS's) based on III-V semiconductors [1,2]. DMS's are alloys of semiconductors and magnetic ions (transition metals or rare earths) which exhibit a variety of cooperative effects via spin-exchange interactions [3] not present in nonmagnetic semiconductors. The exchange interaction between the conduction carriers and the localized moments of the magnetic ions can alter drastically the transport, optical, and magnetic properties of the host semiconductor. In the extreme case, the interaction may induce a ferromagnetic phase transition as observed in PbSnMnTe [4], where the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction between Mn spins via delocalized carriers is responsible for the transition.

Here, magnetotransport properties of a p-type (In,Mn)As film are studied. (In,Mn)As is especially suitable for studying carrier-spin interaction, since carrier type and concentration can be controlled either by doping impurities and/or by changing the MBE growth conditions. The exchange interaction between the conduction holes and the Mn spins in the p-type (In,Mn)As manifests itself in the anomalous Hall effect over nearly the entire temperature range studied, and especially in partial ferromagnetic order at low temperatures. This magnetic order is a cooperative phenomenon accompanied by carrier localization resulting in a large negative magnetoresistance, which is in sharp contrast to the conventional RKKY-driven magnetic order such as in PbSnMnTe, where high carrier concentration (3×10^{20} cm^{-3}) is required for the ferromagnetic phase transition [4].

The (In,Mn)As film studied by magnetotransport measurements consists of a top 1.3-μm-thick $In_{0.987}Mn_{0.013}As$ DMS layer, a 20-nm InAs buffer layer, and a 300-nm GaAs buffer layer grown by MBE on a (001) semi-insulating GaAs substrate. The substrate temperature T_s during MBE growth of the DMS was 275 °C. The relationship between T_s and the resulting (In,Mn)As properties is described elsewhere [5]. Hall resistivity $\rho_{\rm Hall}$ and resistivity ρ were measured in the van der Pauw configuration from room temperature to 0.4 K and in magnetic fields up to 9 T.

The resistivity and the Hall coefficient R_H ($=\rho_{\rm Hall}/B$, where B is the magnetic induction) of the sample from 10 K to room temperature are shown in Fig. 1. The resistivity stays constant from room temperature to 100 K and then starts to increase with decreasing temperature T. On the other hand, R_H increases monotonically from room temperature with lowering T. This temperature dependence of R_H can be understood by a dominating contribution from the anomalous Hall effect [6]. The Hall resistivity is expressed (in mksa units) as

$$\rho_{\rm Hall}=R_0B+R_sM\ , \tag{1}$$

where R_0 is the ordinary Hall coefficient, R_s the anomalous Hall coefficient, and M the magnetization of the film. Since $M=\chi H$, where H is the magnetic field, in the paramagnetic region and R_s is often proportional [6] to ρ,

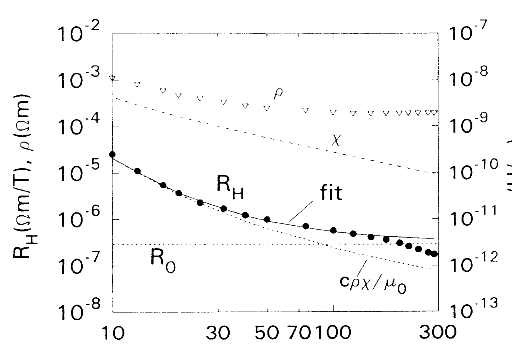

FIG. 1. Temperature dependence of the Hall coefficient R_H and resistivity ρ. R_H can be modeled over a wide range of temperature as $R_0+c\rho\chi/\mu_0$, which is shown by the solid line. $c=5.6$ is used. Susceptibility χ (shown by the dashed line) is calculated assuming the Curie-Weiss law with $N_{\rm Mn}=2.3\times10^{20}$ cm^{-3}, $S=\frac{5}{2}$, and $\theta_P=3.8$ K.

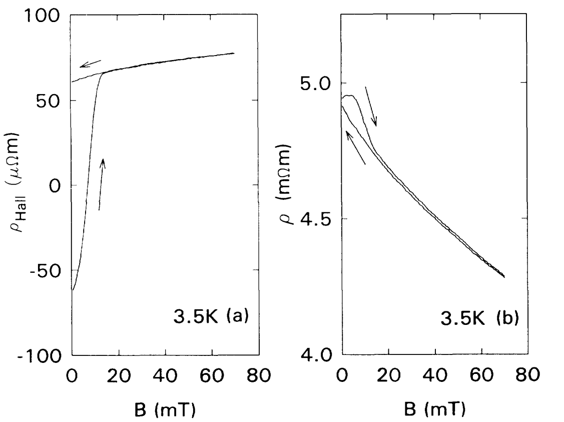

FIG. 2. Hysteresis observed (a) in the Hall resistivity ρ_{Hall} and (b) in the resistivity ρ.

Eq. (1) becomes

$$R_H = R_0 + c\rho\chi/\mu_0 , \tag{2}$$

where c is constant and μ_0 is the magnetic permeability of vacuum. Here, $B = \mu_0 H$ because of the thin-film demagnetization factor. The susceptibility χ may be approximated by $C/(T-\theta_P)$ (Curie-Weiss law) at high temperatures, where C is the Curie constant which can be calculated from the Mn concentration ($N_{Mn} = 2.3\times10^{20}$ cm^{-3} for $x = 0.013$) and the Mn spin $S_{Mn} = \frac{5}{2}$ at low Mn concentration [7]. A very good fit of Eq. (2) to the experimental R_H from 10 to 200 K is obtained using $\theta_P = 3.8$ K and $c = 5.6$ as shown in Fig. 1 by a solid line, together with dotted lines for $c\rho\chi/\mu_0$ and R_0. The good fit over a wide range of temperature indicates that R_H is indeed dominated by the anomalous Hall effect. It also shows that the underlying mechanism for the anomalous Hall effect is skew scattering [6], since R_s is proportional to ρ. The carrier type and concentration obtained from the fit are p type with $p = 2.2\times10^{19}$ cm^{-3}. Thermoelectric power also indicated p-type conduction.

At low temperatures below 8 K, hysteresis appears in $\rho_{Hall}(B)$ and $\rho(B)$ in the low-field region (< 20 mT). Figures 2(a) and 2(b) show examples of hysteresis observed in $\rho_{Hall}(B)$ and $\rho(B)$, respectively, at 3.5 K. The fact that $\rho(B)$ is symmetric with respect to the polarity of B, whereas $\rho_{Hall}(B)$ is antisymmetric, is used to separate the contribution of negative magnetoresistance from the raw Hall voltage data in the van der Pauw configuration. Distinct hysteretic behavior in $\rho_{Hall}(B)$ with a clear remanence, which reflects hysteresis and remanence in M as a result of the anomalous Hall effect, indicates that ferromagnetic interaction is present in the p-type (In,Mn)As film.

Since R_s is proportional to ρ, the remanent part of ρ_{Hall}, $\rho_{Hall}(0)$, divided by $\rho(0)$ (ρ at $B=0$) must be proportional to the remanent part of M, M_0. The temperature dependence of $\rho_{Hall}(0)/\rho(0)$ is shown in Fig. 3(a), where a critical temperature T_c of about 7.5 K is clearly

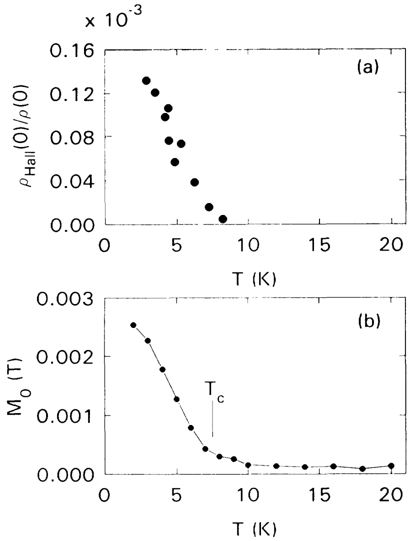

FIG. 3. (a) Temperature dependence of the remanent part of ρ_{Hall}, $\rho_{Hall}(0)$, divided by zero-field resistivity, $\rho(0)$. (b) Temperature dependence of remanent magnetization of the film.

seen, above which hysteresis in both ρ_{Hall} and ρ disappears. Figure 3(b) depicts the temperature dependence of the actual M_0 of the same (In,Mn)As film measured from 2 to 20 K using a SQUID magnetometer. Comparison of Figs. 3(a) and 3(b) shows that $\rho_{Hall}(0)/\rho(0)$ is indeed proportional to M_0, from which the constant c in $R_s = c\rho$ is determined as 6.3, in good agreement with $c = 5.6$ obtained from the paramagnetic region.

Figures 4(a) and 4(b) are high-field (up to 9 T) ρ_{Hall}/R_s and ρ data at 2.8 K, where $R_s = c\rho$ with $c = 6.3$.

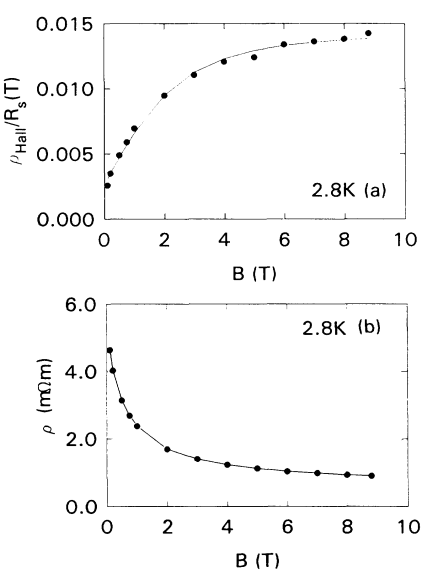

FIG. 4. (a) Magnetic field dependence of ρ_{Hall}/R_s, where $R_s = 6.3\rho$. The solid line is a fit by the empirical Brillouin function $B_J(x)$, where $J = \frac{5}{2}$ and $x = Jg\mu_B H/(T+1.5)$. (b) Magnetic field dependence of ρ.

The negative transverse magnetoresistance as well as the change in ρ_{Hall}/R_s extends to and almost saturates at 9 T, clearly indicating that the spins involved in the remanence are only partially saturated and the effects of unsaturated Mn spins become dominant at high magnetic fields. The magnetization measurements also revealed that both the remanent and paramagnetic contributions are present at low temperature. The magnetic field dependence of ρ_{Hall} again can be interpreted as solely determined by the anomalous Hall effect, i.e., $\rho_{Hall}/R_s = M$. The shape of the ρ_{Hall}/R_s vs B curve above the remanent magnetization part [above the offset in Fig. 4(a)] is Brillouin-function-like and can be fitted by a modified Brillouin function [8] as shown by the solid line in Fig. 4(a). The saturation magnetization M_{sat} obtained from the saturated ρ_{Hall}/R_s value in Fig. 4(a) is 0.014 T, in excellent agreement with the expected value of 0.014 T obtained from $M_{sat} = N_{Mn} g \mu_B S_{Mn}$, where $g=2$ is the g factor of Mn and μ_B is the Bohr magneton [9]. The negligible contribution of $R_0 B$, even at 9 T, is consistent with R_0 obtained from the paramagnetic temperature region.

The magnetic ordering occurring at T_c has a profound effect on the conductivity σ of the sample. The temperature dependence of σ with and without magnetic fields starts to deviate from each other below 30 K as shown in Fig. 5, displaying a large negative magnetoresistance at low temperatures. The temperature dependence of σ at $B=0$ shows a decrease of about a factor of 10 in the neighborhood of T_c, indicating that most of the holes have been localized by the onset of magnetic order. The remaining delocalized holes are either just above or just below the mobility edge. At 8 T, σ below 5 K can be expressed as $\sigma = \sigma_0 + AT^{1/3}$ over at least one order of temperature, where $A = 9.8$ with σ_0 being small ($\sigma_0 = 20$ S/m). The $T^{1/3}$ dependence is expected and has been observed in the weak localization regime near the metal-insulator (M-I) phase transition [10,11].

The origin of the coexistence of remanent magnetization and the paramagnetic component, the rapid decrease of σ around T_c, and the negative magnetoresistance extending over 9 T can be understood by the formation of large bound magnetic polarons (BMP) with partially aligned canted spins. Magnetically, the net aligned moments in BMP provide M_0, which reaches almost 20% of M_{sat} at $T=0$ K, and the subsequent alignment of the canted spins in BMP with field gives rise to the paramagnetic response. The canting of spins could result from the spin-orbit anisotropy involved in the hole-Mn-ion exchange [12], where the carrier-induced Mn-Mn interaction is ferromagnetic but with a texture reflecting anisotropy of the interaction. Or, the canting could arise from the competing coupling of the Mn spins; the carrier-induced ferromagnetic coupling and the direct antiferromagnetic coupling of the Mn spins. Note that in the low carrier concentration n-type (In,Mn)As ($n \cong 10^{16}$ cm^{-3}), the temperature dependence of susceptibility followed a Curie-Weiss law with no trace of ferromagnetic interaction, and the nearest-neighbor exchange constant was $J_{nn}/k = -1.9$ K (antiferromagnetic) [7]. The presence of two completely different phases, i.e., ferromagnetic and paramagnetic phases, is remote since no sign of divergence was observed in the χ vs T curve at T_c. The magnetic order can be classified as asperomagnetic order where a net magnetization develops over a correlation length much larger than the interatomic spacing [13].

The lower limit of the size of the magnetic domains responsible for the hysteresis may be estimated in the following way. At 3.5 K, hysteresis is completed at $B=0.015$ T [see Fig. 2(a)]. Assuming that there is no magnetic anisotropy, the magnetization M_c due to domain rotation can be expressed in terms of the Curie law as $M_c = \chi_c H = N_c g^2 \mu_B^2 S_c(S_c+1)H/3kT$, where N_c is the number of clusters and S_c the total spin in the domain. When all domains are rotated, the magnetization $M_c(\text{sat}) = N_c g \mu_B S_c$. Since $M_c/M_c(\text{sat}) \cong 1$ at $B = 0.015$ T at 3.5 K, S_c becomes 500 and, using $M_c(\text{sat}) \cong 0.002$ T [from Fig. 2(a)], $N_c = 2\times 10^{17}$ cm^{-3}. The magnetic correlation length is then > 100 Å. Here, $S_c = 500$ is the lower limit for S_c, since any magnetic anisotropy will increase the field required to rotate the domains.

The rotation of partially saturated domains is responsible for the hysteresis observed in the B dependence of M and ρ_{Hall} [see Fig. 2(a)]. On the other hand, only a small change in $\Delta\rho/\rho$ [about -3% at 3.5 K as in Fig. 2(b)] is observed in the hysteresis region, whereas $\Delta\rho/\rho$ reaches nearly -80% at $B=9$ T as in Fig. 4(b). This may be understood because when the direction of the domains is reversed, there is only a little change in the local magnetization responsible for determining transport [14].

The negative magnetoresistance below 30 K can be explained qualitatively by the magnetic contribution to the activation energy from the localized BMP state to the delocalized states above the mobility edge [15,16]. The delocalized states experience an average Mn magnetiza-

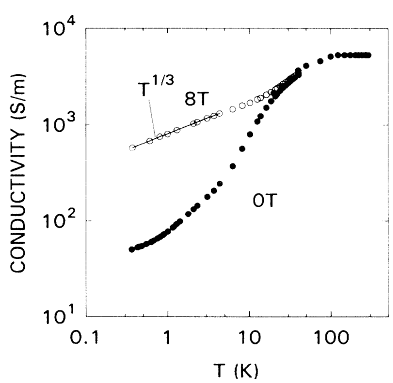

FIG. 5. Temperature dependence of σ with and without magnetic fields. Note the kink at about 7.5 K in the $B=0$ curve. When $B=8$ T, the low-temperature curve can be fitted by $\sigma_0 + AT^{1/3}$.

tion which is zero in zero magnetic field, while the Mn in a local BMP are more closely aligned with the spin of the hole and have a finite value of about 20% of the saturation magnetization. The effect of the magnetic field is to orient all of the Mn so that there is less local inhomogeneity in the Mn spin system and a lower activation energy between the local magnetic polaron state and the delocalized state.

The magnetic coupling of the Mn spins S is most probably caused by a local exchange J of the form $JS\cdot s$, where s is the spin of the localized hole. This is similar in origin to the RKKY interaction responsible for the carrier-induced magnetism observed in PbSnMnTe [4] and rare-earth metals, except that in the present case the hole is localized, while in RKKY it is delocalized with a well-defined Fermi k. Holes seem to provide considerably larger binding energy than electrons since no remanent magnetization was observed in n-type samples [17] with comparable doping and x.

In summary, the interaction between the conduction holes and Mn spins in p-type (In,Mn)As, a new diluted magnetic semiconductor based on a III-V semiconductor, is manifested in the anomalous Hall effect, the formation of remanent magnetization due to partially aligned spins, and the temperature dependence of conductivity with and without magnetic field. The anomalous Hall effect dominates the Hall resistivity from low temperature (0.4 K) to nearly room temperature. The formation of partial ferromagnetic order (asperomagnetism) reveals itself in the hysteresis of the Hall resistivity and resistivity below 7.5 K. These phenomena as well as the temperature dependence of the conductivity with large negative magnetoresistance are explained by the presence of large bound magnetic polarons.

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