Influence of the resonant acceptor state on the magnetotransport properties of zero-band-gap $Hg_{1-x}Mn_xTe$

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(Received 2 October 1987; revised manuscript received 25 February 1988)

A comparative study of magnetotransport in a series of annealed and unannealed zero-band-gap $Hg_{1-x}Mn_x$ Te samples reveals the influence of a resonant acceptor state. In this system, the lowtemperature transport properties result from the resonant nature of the acceptor level. We show that low-temperature negative magnetoresistance originates from an increase in the hole density within the resonant acceptor state rather than an increase in the hole mobility as proposed by others. Also, we have observed an unusual increase in the low-temperature electron mobility with decreasing temperature which we attribute to spatial ordering of the ionized acceptors within the resonant acceptor state.

INTRODUCTION

The disordered ternary alloy $Hg_{1-x}Mn_xTe$ (HMT) combines the electrical properties of a narrow-band-gap semiconductor such as $Hg_{1-x}Cd_{x}Te$ (HCT) with the magnetic properties of a system of interacting spins. This confluence of magnetism and semiconductor physics yields new phenomena such as magnetic-field-dependent band gaps, ¹ field-induced conduction-band-valence-ban overlap,² giant negative magnetoresistance,³ and nonmonotonic variations in the Shubnikov —de Haas oscillation (ShdH) amplitudes.

The HMT system is based on the semimetal HgTe which possesses a band structure that is inverted with respect to InSb. Random replacement of the Hg atoms in the HgTe system with Mn, as in HMT, or Cd, as in HCT the right system with Mil, as in Fiwh, or Cd, as in Fig. 1.
tends to increase the band gap E_0 , $E_0 = E_{\Gamma_6} - E_{\Gamma_8}$. The band gap as a function of the x value increases approximately twice as fast in the HMT system as in the HCT system. Thus, the semimetal to semiconductor transition occurs at $x = 0.07$ for HMT (Ref. 5) and $x = 0.165$ for HCT (Ref. 6) at $T = 4.2$ K. Generally, it is assumed that the nonmagnetic properties of these two systems are the same for equal band gaps, however even the HCT system is not well understood for x values other than $x \approx 0.2$.

Although the extrinsic properties of a normal semiconductor are determined by impurity levels that lie in the band gap, the lack of a gap in zero-band-gap or semimetallic systems forces these levels to be resonant with the conduction or valence bands according to the theory of Gelmont and Dyakonov.⁷ Therefore, semimetallic systems show unusual characteristics such as lack of compensation and anomalous temperature dependence of the mobility and the Hall coefficient compared to ordinary semiconductors.

Experimental work on zero-band-gap HCT has suggested the presence of up to three resonant acceptor levels denoted A_0 , A_1 , and A_2 .⁸ It has been shown, however, that experimental observations attributed to the presence of the A_2 level can also be explained by an unrelated intrinsic mechanism, interband scattering by longitudinal-optic (LO) phonons.⁹ Thus, the presence of the A_2 level is still controversial in the HCT system. The study of acceptor levels has also been complicated by experimental observation of a gap-dependent ionization energy in some cases⁸ while other studies show that the ionization energy does not depend on the gap.¹⁰ These observations have been reconciled by suggesting that resonant acceptor states due to substitutional impurities tend to be tied to the Γ_8 valence band so that their ionization energy is gap independent while the ionization energy of resonant states which originate from Hg vacancies would resonant states which originate from Hg vacancies would
tend to vary with the gap.¹¹ The magnetooptical result of Bastard et al.¹² have provided evidence for possibl existence of a resonant acceptor state even in the semimagnetic HMT system.

Magnetotransport measurements in zero-band-gap HCT have also indicated multiband conduction with electrons dominating at low fields and p-type behavior at high fields. As the overlap between the conduction and valence bands is negligible at zero field and the application of a field induces a gap in this non-magnetic system, the observation of hole transport at the lowest temperatures has been attributed to conduction within the resonant acceptor state.¹³

Our experimental investigation of the semimetallic HMT system through ShdH and magnetotransport measurements also reveals the presence of a resonant acceptor state at low temperatures. We show that the lowtemperature transport properties result from the resonant nature of the acceptor level. Specifically, the negative magnetoresistance that is observed in our zero-band-gap samples for $T < 60$ K originates from an increase in the hole concentration in the resonant acceptor state rather than an increase in the hole mobility as suggested by others. $14-16$ Also, our observation of an increase in the elec-

tron mobility with decreasing temperature is due to spatial ordering of the ionized acceptors within the partially filled resonant acceptor state.

EXPERIMENT

The HMT crystals used in our study were grown by the Bridgman technique. Parallelepiped samples were cut from 1-cm-diam wafers and etched prior to application of contacts. The sample x values were determined by correlating density, microprobe, magnetization, and x-ray fluorescence measurements.

ShdH and magnetotransport measurements were made in a superconducting solenoid to 40 kOe at temperatures down to 1.5 K. Standard field-modulation techniques were used to enhance the ShdH oscillations. The data were collected for both field directions and combined with the aid of a computer to eliminate any mixing of the Hall and magnetoresistance signals.

DATA AND DISCUSSION

In this discussion, we shall present the transport results of two samples which illustrate the transport behavior observed in a series of six p-type zero-band-gap HMT samples. The samples, labeled A (unannealed) and B (annealed in Hg vapor) had an x value, $x = 0.06$, which corresponds to $E_0 = E_{\Gamma_6} E_{\Gamma_8} \cong 50$ meV at 4.2 K.

In order to point out the general features of transport in this diluted magnetic zero-band-gap semiconductor, we have shown in Fig. ¹ the resistance and the Hall voltage as a function of the magnetic field over the temperature range $4.2 < T < 180$ K for the annealed sample B. Several details can be observed in this figure: First, negative magnetoresistance is seen at high fields for $T < 60$ K. Second, at low fields the resistance increases, sharply at low temperatures and more gradually as the temperature is raised. Third, Shubnikov —de Haas oscillations are observable in the 4.2-K plots. Finally, the Hall voltage changes sign from negative to positive with increasing field up to 40 kOe for temperatures less than 60 K and the sign change in the Hall voltage correlates with the onset of negative magnetoresistance. We associate the negative magnetoresistance with the exchange interaction between the magnetic moments of the Mn^{2+} ions and the carriers since this effect is not observed in the nonmagnetic HCT system.¹³

An issue of interest in this sample is the observed sign change in the Hall voltage for temperatures $T < 60$ K. The sign change indicates multiband conduction and raises the question about which bands are responsible for this behavior. For zero-band-gap HCT, theory⁷ (see Fig. 2) indicates that donor levels are ionized while the acceptor levels are superimposed upon the conduction band with an ionization energy E_A with respect to the heavyhold bapd. Thus, for large enough temperatures $(E_F/k_B T < 1)$, and a conduction-band electron concentration n_e such that the Fermi energy lies in the acceptor band, it is possible to have conduction take place simultaneously within the acceptor band, the conduction band, and the heavy-hole valence band. The n-type behavior is

FIG. 1. The magnetoresistance V_R and the Hall voltage, V_H are shown as a function of the magnetic field over the temperature range $4.2 < T < 180$ K for the annealed $Hg_{1-x}Mn_xTe$ $(x = 0.06)$ sample B. In the inset, we have plotted the fielddependent activation energy over the temperature range $20 < T < 60$ K.

due to conduction-band electrons while the p-type behavior could be due to holes in the valence band or the acceptor band. ptor band.
The magnetooptical results of Bastard *et al*. ^{12, 17} would

suggest the exchange contribution to the Hamiltonian causes the heavy-hole band to cross the conduction band

FIG. 2. Figure illustrates possible band structure for zeroband-gap $Hg_{1-x}Mn_xTe$. We suggest that the Fermi energy is pinned by the resonant acceptor band at low temperatures in our samples.

at a certain value of the magnetic field and the hole conductivity occurs because the Fermi energy is pinned by the high density of states in the heavy-hole valence band. However, this model fails to explain similar multiband conduction observed at the lowest temperatures in the nonmagnetic zero-band-gap HCT system 13 where it is well known that the magnetic field opens a gap between the conduction and valence bands. Thus, we assume the p-type behavior is due to conduction within a resonant acceptor band and show that it produces a self-consistent description of our experimental results. We point out that band overlaps proposed by Bastard et al. to explain their magnetooptical results for low-x $(x < 0.02)$ HMT are obtained only for certain values of the exchange parameters.

In the regime where conduction is dominated by holes in the acceptor band, the negative magnetoresistance could be due to either an increase in the mobility of the holes or an increase in the hole density. The temperature dependence of the negative magnetoresistance suggests a magnetic-field-dependent activation energy; we have measured this quantity for $20 < T < 60$ K assuming temperature-independent parameters and plotted the results in the inset of Fig. 1. The figure indicates that the activation energy decreases with the magnetic field. This result, when correlated with low-temperature data, suggests that the hole density increases with the field and is the cause of negative magnetoresistance. Although the measured acceptor activation energy is reasonable for

.6Ŕ

Magneto resistance

6

 $E_A - E_{\Gamma_s}$, we would not expect the zero-field activation energy to equal the true acceptor ionization energy at a fixed temperature, which we have estimated for $T < 6$ K from the ShdH frequency, since the gap and related parameters vary with the temperature.

Further evidence for attributing hole conduction to conduction within a resonant acceptor band is found by comparing the low-temperature $(T < 4.2 K)$ magnetotransport data of our samples A and B which originated from the same wafer and thus had the same residual donor density but different annealing histories. The annealing reduces the number of Hg vacancies that serve as both acceptor sites and scattering centers which reduce the conduction-electron mobility. By comparing samples with different annealing histories, we emphasize in each sample the contribution of a different band to the conductivity. At the lowest magnetic fields, the annealed sample B emphasizes the high-mobility electrons of the conduction band, while A emphasizes hole conduction within the acceptor band because of the reduced conductionelectron mobility in samples with a large defect (vacancy) concentration. This will be denoted as $\sigma_c \gg \sigma_i$ for B and $\sigma_i > \sigma_c$ for A where σ_c is the conductivity of the conduc tion band and σ_i is the conductivity of the acceptor band.

Shown in Figs. 3 and 4 are the measurements on samples B and A , respectively. The illustrated quantities are V_R , V_H , σ_{xx} , and σ_{xy} . Here, V_R is proportional to the resistance, V_H is the Hall voltage, and σ_{xx} and σ_{xy} are the diagonal and the off-diagonal components of the con-

 σ_{xx}

FIG. 3. From the top left, counterclockwise, are the resistance (A) , V_R , the Hall voltage (B) , V_H , the conductivity tensor components σ_{xy} (C), and σ_{xx} (D) for the annealed sample B. The curves correspond to measurements made at the following temperatures: $T = 4.2, 3.0, 2.4, 2.1, 1.8,$ and 1.6 K.

FIG. 4. From the top left counterclockwise are (a) the resistance V_R ; (b) the Hall voltage V_H ; (c) and (d) the conductivity tensorcomponents σ_{xy} and σ_{xx} , respectively, for the unannealed sample A. The curves correspond to measurements made at the following temperatures: $T = 4.2, 3.0, 2.4, 2.1, 1.8,$ and 1.6 K.

ductivity tensor which are defined as follows:

$$
\sigma_{xx} = \rho / [\rho^2 + (R_H H)^2]
$$
 (1)

$$
= \sum_{i} \{ n_i q_i \mu_i / [1 + (\mu_i H)^2] \} , \qquad (2)
$$

$$
\sigma_{xy} = R_H H / [\rho^2 + (R_H H)^2]
$$
\n(3)

$$
= \sum \{ n_i q_i \mu_i^2 H / [1 + (\mu_i H)^2] \} . \tag{4}
$$

The measured quantities have been converted to σ_{xx} and σ_{xy} because the contributions of different bands are additive in these quantities. We point out that although the magnitudes of σ_{xx} and σ_{xy} are sensitive to the geometrical factors involved in the process of inverting ρ and R_H , the general shapes of these curves as a function of the magnetic field remains the same.

In examining the data of sample B $(\sigma_c > \sigma_i)$ for $T \leq 4.2$ K, we observe an initial positive magnetoresistance upon which ShdH oscillations are superimposed. At fields above about 5 kOe, the oscillations disappear before the onset of negative magnetoresistance. At $T = 1.6$ K, the resistance decreases by a factor of 2.5 over the field range $5 < H < 40$ kOe. The Hall voltage is linear in the magnetic field at extremely low fields and indicates electron conduction. For fields greater than 5 kOe, the sample becomes p type and the Hall voltage eventually saturates. Notice that with decreasing temperature the Hall voltage changes sign at lower fields and the slope of

the V_H -versus-H curve at this sign change increases. The ShdH oscillations apparent in these curves were observed down to 500 Oe (i.e., seven cycles) using field-modulation techniques and the ShdH frequency ($F=3.4$ kOe $\rightarrow n_e$) $=1.1 \times 10^{15}$ cm⁻³) is independent of temperature over the range $1.5 < T < 4.2$ K within the resolution of our measurement (\sim 2%). As some studies in zero-gap-band HCT have indicated a temperature-dependent ShdH frequency even over this temperature range¹⁸ due to the $T^{3/2}$ temperature dependence of the carrier density in zero-band-gap systems, we suggest that the temperature independence of the carrier density in this sample is also consistent with our assumption that the Fermi energy is pinned in the resonant acceptor band.

At low magnetic fields, the transport behavior is very different in sample A ($\sigma_i > \sigma_c$). The resistance does not show a pronounced rise and ShdH oscillations are missing. However, the decrease in the magnetoresistance at $T = 1.6$ K is still roughly a factor of 2.5 over the field range ⁵—40 kOe. Also, the Hall voltage indicates that the sample is p type over all fields and tends to approach a temperature-independent value at high fields.

As sample B shows multiband conduction, evidenced by the sign change in the Hall voltage, we associate the initial positive magnetoresistance with a shift in the conduction from the high-mobility electrons which are responsible for the ShdH oscillations to the low-mobility holes in the acceptor band as the magnetic field decreases the contribution of the high-mobility carriers. Although we have not been able to determine the electron mobility in the unannealed sample (A) because the holes dominate even at very low fields, we believe the absence of similar positive magnetoresistance in this sample is due to lower electron mobility resulting from scattering at vacancies.

The sharp rise in σ_{xx} subsequent to the last ShdH osci11ation in the annealed sample is an especially important clue to understanding transport behavior in zero-bandgap HMT since the classical multiband model with fieldindependent parameters predicts that σ_{xx} is a monotonically decreasing function of the magnetic field. The nonmonotonic behavior in σ_{xx} and the correlation of this effect with the last ShdH oscillation suggests that the carriers are being transferred out of the conduction band. Usually, in a simple semiconductor all the electrons are in the $N = 0$, spin-up Landau level after the last ShdH oscillation and the Fermi energy rises with the Landau level. However, if the Landau level crossed a highly degenerate resonant acceptor state at a certain value of the field, then it would be energetically favorable for the electrons to abandon the Landau level in favor of the resonant acceptor state. The sudden increase of the carrier density in the acceptor band would be manifested as an increase in σ_{xx} since the lower mobility of the carriers within this band makes it less likely to satisfy the freezeout condition, $\mu_a H > 1$, compared to the conduction electrons at the same magnetic field. We point out that although there is an increase in σ_{xx} , it still remains below the zero-field value. This sharply rising feature of the data also implies that it is incorrect to assume fieldindependent electron and hole densities as is usually done when modeling this system.¹⁵

At magnetic fields above 5 kOe, both samples show negative magnetoresistance for $T < 4.2$ K that is once again associated with p-type conduction as in the highertemperature regime discussed previously. In this highfield limit, we assume transport is due to carriers in the acceptor band, the high-mobility electrons having been emptied out of the conduction band by the magnetic field. Then, assuming $\mu_a H \ll 1$,

$$
n_a = \sigma_{xx}^2 H / \sigma_{xy} q \tag{5}
$$

and

$$
\mu_a = \sigma_{xy} / \sigma_{xx} H \tag{6}
$$

For samples B and A, $log_{10}n_a$ and μ_a are plotted as a function of the magnetic field in Fig. 5. The low-field part of these curves, i.e., where the curves turn over, should be neglected because the single-band approximation is invalid in this region. Also, the hole mobilities of the two samples are the same within experimental error since geometrical factors are known only to about 10% . We note that the hole density in the annealed sample at fields immediately after the sharply rising feature in σ is approximately 1×10^{15} cm⁻³ and equals the low-field electron density obtained from the ShdH effect thus suggesting that holes result from transfer of conduction-band carriers into the resonant acceptor band. It also supports our interpretation that the sharp rise in σ_{xx} is a signature of charge transfer from the conduction band to the resonant acceptor state which pins the Fermi energy.

The figure also shows that in the region of negative magnetoresistance, the carrier concentration within the acceptor band increases and the mobility decreases with increasing magnetic field. This behavior contradicts the model originally proposed by Davydov et al.¹⁴ for nega tive magnetoresistance in semimetallic HMT. They had suggested that negative magnetoresistance originates from an increase in the hole mobility due to reduced scattering from the Mn^{2+} spins as they are ordered by the magnetic field. Here we have shown that negative magnetoresistance originates from an increase in the hole density within the acceptor band. This behavior is also consistent with the measured reduction in the activation energy with field at higher temperatures.

The reduction in the activation energy and the corresponding increase in the hole density can be understood by contrasting the effect of the magnetic field on an ordinary zero-band-gap semiconductor such as HCT with that on a diluted-magnetic zero-band-gap semiconductor like HMT. In zero-band-gap HCT, Landau quantization due to application of a magnetic field induces a gap between the lowest conduction-band Landau level and the highest heavy-hole valence-band Landau level. The

FIG. 5. The field dependence of the acceptor hole density (top) and the hole mobility (bottom) for the annealed and unannealed samples are shown for $T=2.1$ K. Here, we have assumed one-band conduction to obtain this plot from the transport data of Figs. 2 and 3.

field-induced gap would vary monotonically though not linearly due to nonparabolic effects. The effect of the magnetic field in the HMT system is complicated by the exchange interaction, due to the presence of the magnetic Mn^{2+} ions, which is incorporated into the Pidgeon-Brown Hamiltonian in the mean-field, virtual-crystal approximation.⁴ It is believed to prove nonmonotonic variations in the field-induced gap and possibly even overlaps between the lowest conduction band and the highes heavy-hole Landau levels.^{2,17} Here, we suggest that the observed reduction in the activation energy is a consequence of the highest heavy-hole Landau level approaching the acceptor state as was shown for open gap HMT (Ref. 19) and the observed increase in the hole density with field may be due to the activation of carriers from the heavy-hole valence band into the acceptor state. This model explains the general trend observed in the temperature dependence of the magnetoresistance. The negative magnetoresistance becomes more pronounced at lower temperatures since the exchange interaction is proportional to the magnetization which increases with decreasing temperature.²⁰ We should point out that impurity band conduction is not well understood partly because the sign of the carriers cannot be related to band curvature as in standard effective-mass theory.

We have also observed an unusual temperature dependence in the zero-field resistance, see Fig. 6, at low temperatures, $T < 10$ K, in the annealed sample B which further supports our assumption that the Fermi energy is pinned in the resonant acceptor band. In the inset of Fig. 6, we have sketched the temperature dependence of the resistance for both the annealed and unannealed samples. From the figure, we observe qualitatively different behavior in the zero-field, low-temperature resistance of sample

FIG. 6. The temperature dependence of the resistance for the annealed sample B. Note the anomalous behavior for $T < 10$ K. The insets illustrate the different temperature dependence of the resistance observed for the annealed (bottom) and unannealed (top) samples. Here, σ_c is the conduction-band conductivity and σ_i is the conductivity for the resonant acceptor band

B ($\sigma_c > \sigma_i$) where the conduction electrons dominate the conductivity, in comparison with sample $A(\sigma_{i} > \sigma_{c})$, where conduction within the impurity band is dominant. That is, the resistance increases with temperature in B while in A the behavior is just the opposite.

As ShdH measurements indicate a temperatureindependent electron concentration at low temperatures and also because the transport properties at the lowest fields are controlled by the high-mobility carriers, the increase in resistance with increasing temperature in B implies a decrease in the electron mobility with increasing temperature. This behavior is confirmed by a plot of the temperature dependence of the mobility, see Fig. 7, for the annealed sample B for $T < 10$ K. For charged center scattering, we would expect the mobility to increase with increasing temperature at low temperatures in contradiction to these experimental results. Similar behavior has also been observed by others in the HMT system¹⁵ as well as in the nonmagnetic HCT system²¹ though it is not well understood.

The resonant nature of the partially filled acceptor state allows for virtual transitions between the conduction band and the resonant acceptor state which limits the lifetime of a particular ionized acceptor. Although the acceptors (vacancies) are randomly distributed at fixed sites throughout the lattice, only a fraction are ionized, $N_A \gg n_e + N_A^- = N_D^+$, since the Fermi energy is pinned in the resonant acceptor band (see Fig. 2). Here, N_A is the total acceptor density, N_A^- is the density of ionized acceptors, and N_D^+ is the donor density. Thus, there are a large number of possible occupational configurations for the ionized acceptors on the lattice. The resonant nature of the acceptor band and the pinned Fermi energy allow the ionized acceptors to continuously change their configuration on the lattice due to thermal fluctuations. As the temperature is lowered and the

FIG. 7. The temperature dependence of the electron mobility for the annealed sample B. Notice the increase in the mobility with decreasing temperature for $T < 10$ K.

FIG. 8. A plot of the low-temperature ($T < 10$ K) zero-field resistance for the annealed sample shows activated behavior indicating possible ordering of charged acceptors.

thermal fluctuations are reduced, the system of ionized acceptors tends to minimize the total Coulomb energy of the system by condensing into a partially ordered superlattice of ionized acceptors. The ordering would be more complete when the ionized acceptor density is only a fraction of the total acceptor density. The increased ordering of the ionized acceptors with decreasing temperature would enhance the electron mobility since orderin suppresses scattering. $22,23$ This idea was originally proposed to explain the decrease in scattering that is observed in the $Hg_{1-x}Fe_xSe$ system. In that semiconducting system, it is believed that the resonant donor level due to the Fe serves to pin the Fermi energy and aids in forming an ordered substructure of charged centers. This model, when applied to the semimetallic HMT system, would also suggest an activated temperature dependence of the mobility over the range of temperatures ($T < 10 K$) where this unusual behavior is observed. In Fig. 8, we have plotted $ln(R)$ versus $1/T$ to determine an activation energy. The plot indicates that the zero-field resistance does follow activated behavior with an activation energy of \sim 0.1 meV. We believe this value probably represents the mean Coulomb energy among ionized acceptors.

In conclusion, negative magnetoresistance and the associated p-type conduction in semimetallic $Hg_{1-x}Mn_xTe$ occur due to an increase in the carrier concentration, rather than an increase in the mobility, within a resonant acceptor band. The features in our data that can be explained with a model that includes a resonant acceptor state are the following. First, the temperature independence of the ShdH frequency in this zero-band-gap system which possibly indicates a pinned Fermi energy. Second, the sudden $n \rightarrow p$ transition observed in sample B, the corresponding sharp rise in the σ_{xx} curve and the correlation of the hole density after the sharp rise in σ_{xx} with the low-field electron density. We have attributed this behavior to charge transfer from the conduction band to the resonant acceptor state. Third, observation of multiband conduction at the lowest temperatures in both the annealed and unannealed sample and the similarity to the nonmagnetic HCT system. Fourth, the decrease in the hole mobility over the range of magnetic fields where negative magnetoresistance is observed which contradicts predictions of other models for negative magnetoresistance in zero-band-gap HMT. Fifth, the correlation between the contribution of hole conductivity to the total conductivity and presence of Hg vacancies obtained from a comparison of annealed and unannealed samples. Thus, the resonant acceptor state is due to the presence of Hg vacancies which acts as acceptors. Finally, the unusual low-temperature behavior of the electron mobility which occurs due to the spatial ordering of the charged centers in the resonant acceptor state and the resulting decrease in the scattering.

ACKNOWLEDGMENTS

The samples used in this study were grown by Mr. D. Kreft, Mr. E. Larsen, and Mr. K. Hankin. This research was supported in part by the U.S. Defense Advanced Research Projects Agency (DARPA) under Grant No. DAAG29-85-K-0052.

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