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Resonant-acceptor-bound magnetic polarons in the zero-band-gap semimagnetic semiconductor $Hg_{1-x}Mn_xTe$

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Far-infrared magneto-optical studies of zero-band-gap p-type $Hg_{1-x}Mn_xTe$ ($x=0.065$ and 0.070) for temperatures down to 1.6 K and magnetic fields up to 9 T are presented. Transitions of the resonant-acceptor state to the uppermost heavy-hole level are identified by the selection rules, temperature-dependent exchange effect, and $\mathbf{k}\cdot\mathbf{p}$ Landau-level calculations. A remarkable shift of the resonance field with increasing temperature is observed, which is attributed to the strong sp-d exchange interaction. The plot of the resonance energy as a function of calculated magnetization, followed by a linear extrapolation using a least-squares fit, yields the negative zero-field resonant-acceptor energy whose magnitude increases as temperature is lowered and saturated above $T \sim 16$ K. This feature indicates the possible formation of resonant-acceptor-bound magnetic polarons (RABMP's) in the $Hg_{1-x}Mn_xTe$ at low temperature and low external magnetic field. The zero-field binding energies of RABMP's are found to be $\simeq 3.5$ meV (3.8 meV), in the limit of zero temperature for $x=0.065$ ($x=0.070$).

I. INTRODUCTION

Bound magnetic polarons (BMP's)—the ferromagnetic complexes consisting of an electron (hole) bound at a donor (acceptor) and the polarized cloud of Mn^{2+} ions within the electron (hole) orbit—have been extensively investigated only in wide-band-gap semimagnets, $Cd_{1-x}Mn_xS$, $Cd_{1-x}Mn_xSe$, and $Cd_{1-x}Mn_xTe$, where sp-d exchange effects are relatively large because of the small size of the BMP's.[1-5] The net nonzero spin magnetization of BMP's originates from two contributions: mean field produced by the localized spins within the donor (acceptor) orbit at low temperature, and their momentary thermodynamic spin fluctuations at higher temperature. In the case of narrow-band-gap semimagnetic semiconductors, zero-field spin splitting (spin-flip energy) observed in $Pb_{1-x}Mn_xS$ and $Pb_{1-x}Mn_xTe$,[6-8] and magneto-optical transition observed in $Hg_{1-x}Mn_xTe$ (Ref. 9) have been controversially interpreted as the possible evidences for the formation of the free magnetic polarons (FMP's). On the basis of mean-field theory,[10] however, free magnetic polarons are unstable. The stability of FMP's can be improved by considering the possibility of self-trapping of free electrons on thermodynamic spin fluctuations.[9,11] However, we would expect these effects to be negligible since spin fluctuations would vanish over length scales comparable to that of the free-electron wave packet. In contrast to the FMP's, the influence of the spin fluctuation of Mn ions on the BMP's can be relatively large because of the finite size of the bound-carrier orbit.[12,13] The situation is particularly favorable in the acceptor-BMP case because of larger p-d exchange interaction and smaller orbit due to heavy-hole effective mass.

Here we report, for the first time, evidence for possible existence of a resonant-acceptor-bound magnetic polaron (RABMP) in zero-band-gap p-type semimagnetic $Hg_{1-x}Mn_xTe$ ($x=0.065$ and $x=0.070$). The temperature-dependent resonant-acceptor binding energy has been deduced from far-infrared magneto-optical measurement of the impurity transition between the resonant acceptor and the uppermost heavy-hole state. The resulting zero-field binding energy of the resonant acceptor is observed to increase as temperature is lowered to zero and saturates above $T \sim 16$ K. As Dietl and Spałek show,[12] this feature is characteristic of BMP's. The net nonzero spin fluctuations due to sizeable effect remove critical behavior and, instead, the system evolves continuously from mean-field- (cooperative) dominated low-temperature state to fluctuation-dominated high-temperature regime.

II. EXPERIMENTAL DETAILS AND SAMPLE CHARACTERISTICS

Far-infrared magnetotransmission and magnetotransport measurements have been carried out on bulk single-crystal samples of p-type zero-band-gap $Hg_{1-x}Mn_xTe$ (HMT) grown by the Bridgman method. Two samples (1×1 cm^2) were cut from the same wafer, mechanically polished, and then etched for 5 min in a 5% bromine-methanol solution. The Mn concentration x was determined by correlating transmission, density, and electron microprobe measurements. The results for the two samples are $x=0.065\pm 0.004$ and $x=0.070\pm 0.004$. Hall measurement at 4.2 K shows n-type behavior for $B \leq 3$ kG and p-type for $B \geq 3$ kG. Reversal of the sign of the

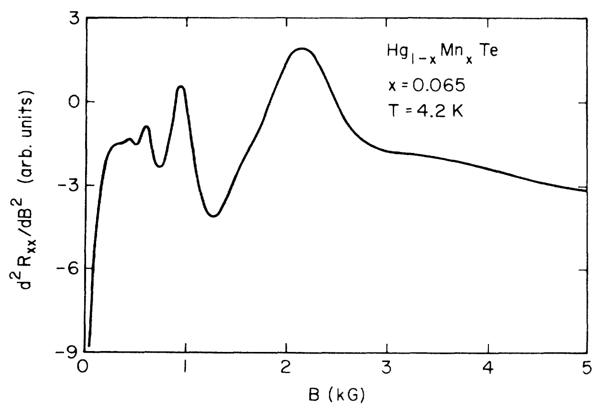

FIG. 1. Second derivative of transverse resistance, R_{xx}, respective to magnetic field. A series of Shubnikov–de Haas oscillations is observed at very low field ($B \leq 3$ kG).

Hall coefficient with increasing magnetic field indicates two types of carriers participating in low-field transport, i.e., high-mobility electrons in the conduction band and much lower mobility holes in the valence band. The carrier densities and mobilities deduced from low-field Hall measurement correspond to $n \sim 3\times10^{14}$ cm^{-3} and $\mu \sim 2\times10^{6}$ cm^2/V s at 4.2 K. A set of Shubnikov–de Haas (SdH) oscillations was also observed but only at very low fields, as shown in Fig. 1. Standard field-modulation techniques were used to enhance the weak SdH oscillations. The last SdH oscillation occurs at $B \sim 2$ kG. The electron density deduced from the SdH period agrees with Hall effect measurements within $\sim$20%. Two-band-carrier transport behavior observed in our studies confirms the x value and the zero-band-gap nature of these samples. We point out that these samples are below, yet very close to, the critical x value ($x \sim 0.075$) for the semimetal-semiconductor transition in HMT at $T = 4.2$ K.

For the far-infrared transmission measurement, the samples were mounted on an undoped Ge substrate chosen because of the close match of its dielectric constant and thermal expansion coefficient over a wide temperature range. This minimizes strain effects on that sample and interference fringes associated with the sample thickness. An optically pumped cw laser provided far-infrared photon energies ranging from $\lambda = 118.8$ to 888.9 μm. The radiation was circularly polarized using a polarizer consisting of a linear polarizer followed by quartz-crystal $\lambda/4$ plates. The detector was a composite bolometer operating at 4.2 K.

III. RESULTS

The temperature dependence of the magnetotransmission for the $x = 0.065$ sample for both σ^+ and σ^- polarizations in the Faraday configuration ($\mathbf{k}\|\mathbf{B}$, $\mathbf{E}\perp\mathbf{B}$) is shown in Fig. 2 for fixed photon energy of $\hbar\omega = 10.44$ meV. The spectra exhibit two major absorption features: (1) a sharp cyclotron resonance (CR) occurring at very low field in the σ^- polarization which was identified as $a_c(0)\rightarrow a_c(1)$ transitions and (2) a broad absorption in the

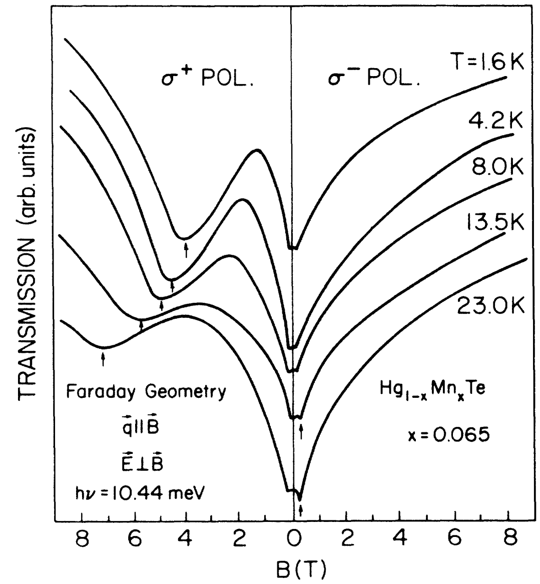

FIG. 2. Temperature dependence of magnetotransmission spectra of $x = 0.065$ sample for the σ^+ polarization in the Faraday geometry for a photon energy $\hbar\omega = 10.44$ meV. Two resonances indicated by arrows are identified as cyclotron resonance, $a_c(0)\rightarrow a_c(1)$, in the σ^- polarization, and the impurity transition, RA$\rightarrow b_v(-1)$, in the σ^+ polarization, respectively. We point out the remarkable shift of resonance position of the impurity transition with increasing temperature.

σ^+ polarization which was identified as impurity transitions from the resonant-acceptor state to the uppermost heavy-hole state, $b_v(-1)$. The CR shows a thermally activated behavior. It is nearly discernible at 4.2 K, but is seen to grow as temperature is raised. This thermally activated feature of the CR is explained in terms of the rapid increase of conduction-band Landau levels with magnetic field, which will be justified in Sec. IV. A remarkable shift of resonance position of the impurity transition with increasing temperature is observed, which is crucial in this study. It is attributed to the strong *sp-d* exchange effect in this semimagnetic material. Figure 3 shows absorption spectra in the σ^+ polarization for several photon energies at $T = 4.2$ K. The resonance energy versus magnetic field for the $x = 0.065$ sample is shown in Fig. 4 for temperatures ranging from 1.6 to 23 K. Unfortunately, the measurements of the impurity transition for $B \leq 1$ T were obscured by the strong absorption occurring at around zero field, which is attributed to Drude absorption, i.e., low-field cyclotron resonance. However, any straightforward extrapolation of the data is seen to yield a negative intercept for each temperatute, corresponding to an impurity level resonant with the conduction band at zero magnetic field. The zero-field binding energy of the resonant acceptor, which is seen to increase as temperature is lowered, was determined from an unbiased and more accurate extrapolation which will be presented in Sec. IV. The magnetic-field dependence of the cyclotron resonance observed in the σ^- polarization is plotted in Fig. 5. It is seen that the CR exhibits strong nonparabolicity, which indicates that the interacting gap,

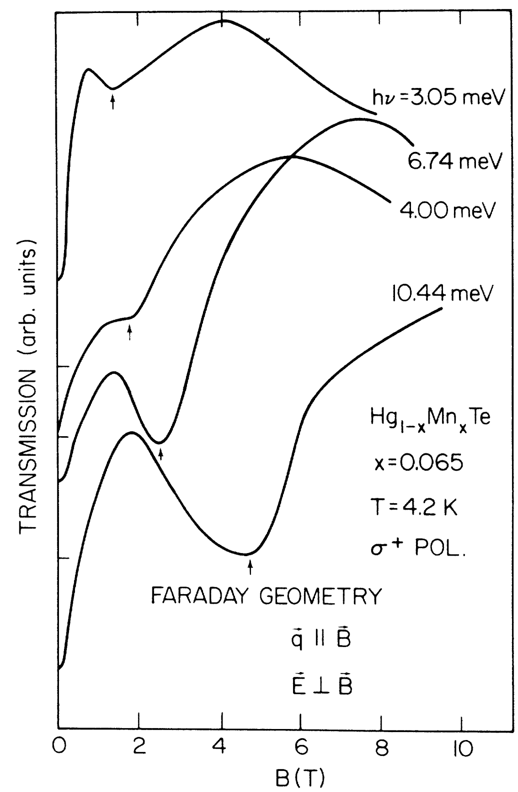

FIG. 3. Typical transmission spectra for sample $x = 0.065$ as a function of magnetic field for several photon energies in the Faraday σ^+ polarization at 4.2 K.

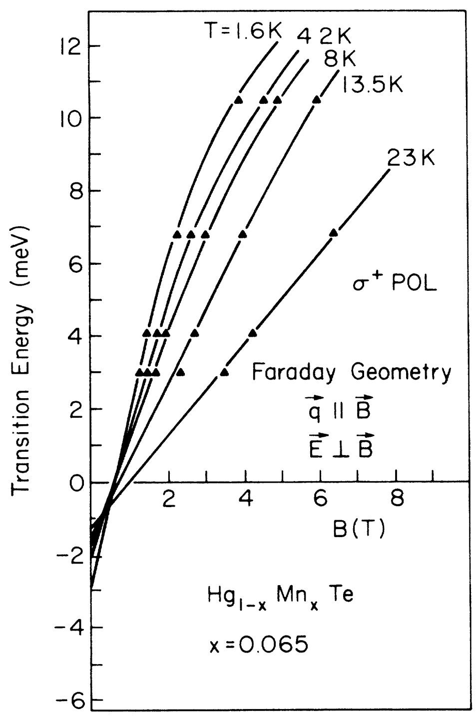

FIG. 4. Position of the transmission minima observed in the σ^+ polarization as a function of photon energy and magnetic field for temperatures ranging from 1.6 to 23 K. The zero-field extrapolated energy for each temperature is seen to be negative, and its actual value was determined as detailed in text (see Fig. 7).

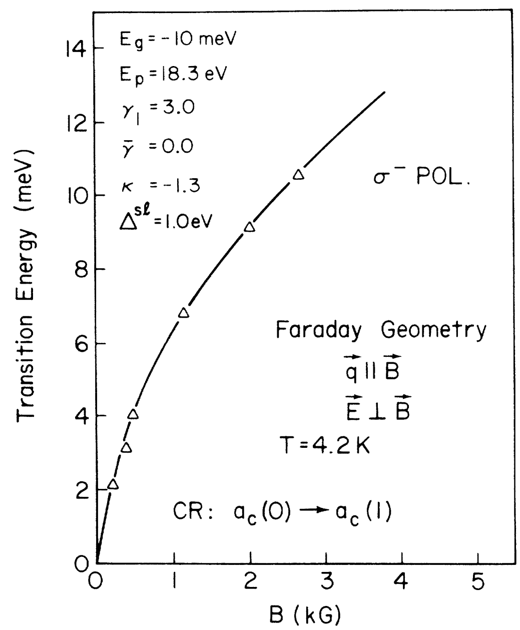

FIG. 5. Energy dispersion vs magnetic field for cyclotron resonance observed in the σ^- polarization. The solid line corresponds to the best fit to $\mathbf{k}\cdot\mathbf{p}$ Pidgeon-Brown Hamiltonian.

$E_g = E_{\Gamma_6} - E_{\Gamma_8}$, is very small. The resulting fit of the cyclotron resonance by the modified Pidgeon-Brown Hamiltonian is shown as a solid line in Fig. 5.

IV. ANALYSIS

A. Cyclotron resonance and k·p band parameters

In the analysis of CR data we used the modified Pidgeon-Brown Hamiltonian[14–17] which includes the exchange interaction between band electrons in the Γ_6, Γ_7, and Γ_8 levels and localized magnetic moments due to half-filled $3d$ electrons of the Mn ions. The optical transition energies in the Faraday geometry are almost insensitive to the exchange parameters since they involve the transitions within the same spin ladder solutions, i.e., $a(n) \rightarrow a(n+1)$ or $b(n) \rightarrow b(n+1)$. Calculations show that the exchange corrections to the initial and final states nearly cancel so that the $\mathbf{k}\cdot\mathbf{p}$ band parameters of a comparable nonmagnetic semiconductor are sufficient for fitting parameters. The observed CR was identified as the Γ_8 intraband $a_c(0) \rightarrow a_c(1)$ transition. The resulting $\mathbf{k}\cdot\mathbf{p}$ band parameters are $E_g = -10$ meV, $E_p = 18.3$ eV, $\gamma_1 = 3.0$, $\gamma = 0.0$, $\kappa = -1.3$, and the spin-orbit coupling constant $\Delta^{sl} = 1.0$ eV. The off-diagonal term G arising from the lack of inversion symmetry and the second-order conduction-band matrix element F both are set to zero.[18] The energy gap so determined is consistent with the measured Mn concentration of the sample. The interband Kane matrix energy E_p and the remote-band Luttinger parameters γ_1, γ, and κ are also in agreement with the values reported in the literature.[19–21] The rapid increase of conduction-band Landau levels with magnetic field, as shown in Fig. 6, explains the thermally activated behavior of the CR. The Fermi level is below the lowest conduction Landau level $a_c(0)$ at the resonance field, $B \simeq 3$ kG, and the CR is only observed for the electrons thermally excited to the $a_c(0)$ level. This observation is also consistent with the results of transport properties—Hall measurement which shows n-type behavior for $B \leq 3$ kG and low carrier density $n \sim 3\times10^{14}$ cm^{-3}, and the magnetoresistence measurements which show the last SdH oscillation occurring at $B \sim 2$ kG.

B. Resonant-acceptor transition

The observation of the remarkable temperature-induced shift of the broad resonance in the σ^+ polarization, which is central in this work, has been interpreted in terms of the sp-d exchange effects. using the $\mathbf{k}\cdot\mathbf{p}$ band parameters deduced from CR analysis, we have calculated Γ_8 Landau levels based on the modified Pidgeon-Brown Hamiltonian. The levels are sensitive to the values of the exchange parameters α,β and the magnetization of sample. In the analysis α and β are considered as adjustable parameters. We have used the result of Anderson and Gorska[22] for the values of the effective Mn concentration $\bar{x}$ and the antiferromagnetic temperature T_{AF} which are needed in the calculation of magnetization for each temperature. For samples of x ranging from 0.007 to 0.23, Anderson and Gorska fitted the magnetization data to a modified Brillouin function with two fitting parameters $\bar{x}$ and T_{AF}. The result of calculations of the Γ_8 Landau levels at 4.2 K are shown in Fig. 6, where we used $N_0\alpha=-0.4$ eV and $N_0\beta=0.6$ eV which are close to the most recent reported values for exchange.[20,21,23] The

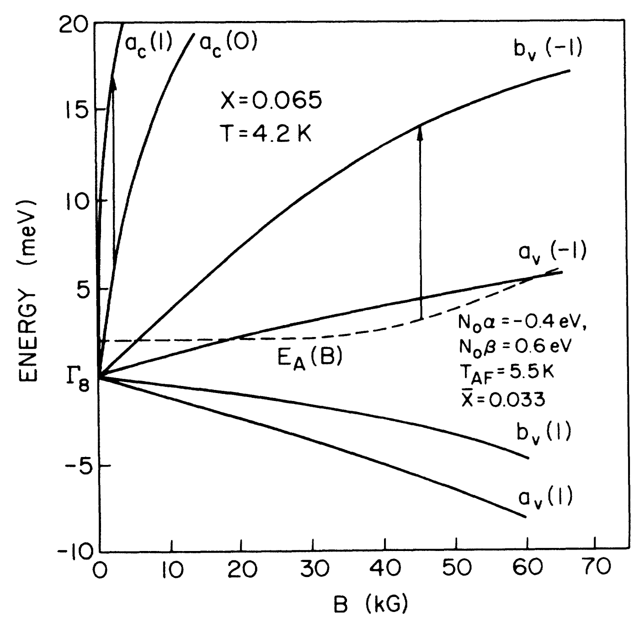

FIG. 6. Γ_8 Landau levels calculated at 4.2 K using a modified Pidgeon-Brown model. The dashed line corresponds to the resonant-acceptor level which was deduced from the transmission data and the calculated $b_v(-1)$.

only optical transition, consistent with energy conservation, selection rules, and temperature-dependent exchange effect, is either the hole spin resonance $a_v(-1)\rightarrow b_v(-1)$ or the impurity transition, resonant acceptor $\rightarrow b_v(-1)$.

The hole spin resonance may explain the broad absorption of the σ^+ polarization if we assume a splitting between $a_v(-1)$ and $b_v(-1)$ at zero field. The possible effects of a small amount of strain on the sample (due to the small difference in thermal expansion coefficients between the sample and substrate) may first appear to split the $a_v(-1)$ and $b_v(-1)$ valence levels at zero magnetic field. A negative extrapolation can be then obtained if the $a_v(-1)$ level is shifted upwards and crosses the $b_v(-1)$ level at a finite field. However, this strain-induced splitting is ruled out by noting that strain cannot remove the Kramers degeneracy. Since the strain Hamiltonian[24,25] affects only orbital angular momenta, the application of a uniaxial stress splits the $P_{3/2}$ multiplet into $|J=\frac{3}{2}, m_j=\pm\frac{3}{2}\rangle$ and $|J=\frac{3}{2}, m_j=\pm\frac{1}{2}\rangle$. It should be noted that the $a_v(-1)$ and $b_v(-1)$ states originate from the spin-up and spin-down state belonging to the Γ_8 heavy-hole valence band.

We point out further that any strain effects on our sample have been found to be too small to split even the $P_{3/2}$ valence multiplet. As mentioned in the experimental details, we have used undoped Ge as a substrate because of the close match of its thermal expansion coefficient over a wide temperature (note that $\alpha\sim5\times10^{-6}$ and 5.7×10^{-6} K^{-1} at 295 K for HgTe and Ge). We estimate the magnitude of the strain-induced splitting of the $P_{3/2}$ valence multiplet. Following the strain Hamiltonian by Bir and Picus,[24] the energy splitting at the Γ_8 valence-band extrema is $\sim b(S_{11}-S_{12})X$, where b is the shear deformation potential of the valence band, S_{11} and S_{12} are the elastic constants, and X is the in-plane stress experienced by the sample. The X can be written as

$$X=[1/(S_{11}+S_{12})][\delta a/a]_{H\text{-}G} ,$$

where $[\delta a/a]_{H\text{-}G}$ is the difference in the linear thermal contraction between the HMT and Ge when temperature is lowered from 295 to 1.5 K. The value of $[\delta a/a]_{H\text{-}G}$ is estimated to be $\sim10^{-4}$. Using $S_{11}=5.92$ N/m^2, $S_{12}=4.14$ N/m^2, and $b=-1.5$ eV for HgTe,[26,27] the energy splitting is found to be $\sim10^{-5}$ eV.

The source inducing the spin splitting between $a_v(-1)$ and $b_{-v}(-1)$ is rather the inversion asymmetry which is characteristic of the zinc-blende lattice. The lack of inversion symmetry yields a nonzero value of G in the Pidgeon-Brown Hamiltonian. It has been shown, however, that the removal of the twofold Kramers degeneracy due to nonzero G is at most $\sim10^{-4}$ eV for the Γ_8 valence band, which is nearly imperceptible.[28] The above considerations, therefore, lead us to conclude that the spin hole resonance is not able to explain the observed negative zero-field extrapolated energy.

The resonant-acceptor (RA) states due to Hg vacancies have been extensively studied in the nonmagnetic zero-band HgTe and $Hg_{1-x}Cd_xTe$.[29-31] In the case of semimagnetic $Hg_{1-x}Mn_xTe$ ($x \leq 0.02$), the magneto-optical results of Bastard *et al.*[32,33] have provided evidence for possible existence of a resonant-acceptor state whose binding energy at zero field is $E_A(0) \sim 2$ meV and nearly constant in the field ranging from 5 to 20 kG. Our data show very similar features to those of Bastard *et al.* except for the strong temperature-dependent exchange effects which are reported here for the first time. The negative zero-field extrapolation indicates that the resonant-acceptor energy crosses the uppermost heavy-hole state $b_v(-1)$ at a finite field and becomes lower than that level. This unusual level ordering indicates that the quasibound state of the resonant acceptor experiences a much weaker exchange contribution than the free valence level.[33]

The mechanism for inducing the impurity transition, $RA \rightarrow b_v(-1)$, can be described as the following. In the absence of a magnetic field, the low electron density $\sim 3 \times 10^{14}$ cm^{-3} results in the Fermi level that lies below or pinned by the resonant-acceptor level. Application of a magnetic field induces a gap between the $a_c(0)$ and $b_v(-1)$ levels as the $a_c(0)$ levels rapidly increases in energy, as shown in Fig. 6. As successive Landau levels cross the zero-field Fermi energy, SdH oscillations are observed and then conduction electrons are emptied on to acceptor. Following the magnetic depopulation of the conduction band, holes in the $b_v(-1)$ level begin to contribute to the conductivity leading to a p-type Hall effect and positive magnetoresistance. This qualitative picture satisfies charge neutrality of the crystal and allows for transition between the populated resonant-acceptor state and the partially filled $b_v(-1)$ level. The broad absorption linewidth is also connected to the high density of acceptor states which cannot be completely eliminated by annealing.

C. The temperature dependence of the resonant-acceptor binding energy: Resonant-acceptor-bound magnetic polarons

The temperatute-induced resonance shift of the impurity transition was completely explained in terms of the strong *sp-d* exchange between *band electron* and Mn ions. Here we extend our investigation on this strong temperature effect down to the extrapolated low-field region and present possible evidence for the exchange interaction between *a quasibound hole* and Mn ions. As mentioned in Sec. II, it is hard to doubt the fact that almost any straightforward extrapolation of the data of Fig. 4 would yield a negative intercept, which provides the crucial factor for the identification of the resonant-acceptor transition. However, the actual value of the intercept for each temperature will be very dependent on the extrapolation procedure used. For a given temperature, care must be taken during the extrapolation since the data are rather limited and the extrapolation rather long. We have done the extrapolation by plotting the transition energy as a function of calculated magnetization and a linear extrapolation using a least-squares fit. For the calculation of magnetization we have used the results of Anderson and Gorska, which were detailed in Sec. IV B. As seen in Fig.

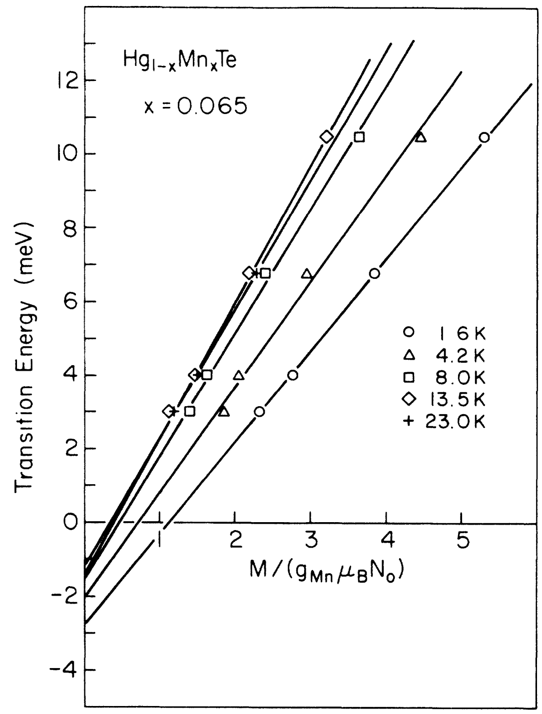

FIG. 7. Transition energy vs calculated magnetization. The solid line for each temperature corresponds to the resulting fit based on the linear-least-squares method.

7, the transition energy is nearly linear to the magnetization, indicating that the resonant-accpetor energy is nearly constant over the magnetic-field ranges studied. The remarkable result is the temperature dependence of the zero-field extrapolated energy, indicating that the zero-field binding energy of the resonant acceptor increases as temperature is lowered and saturates for $T \geq 16$ K. Figure 8 displays the temperature dependence of the zero-field binding energy of the resonant accpetor. This feature is characteristic of BMP's. It exhibits the full

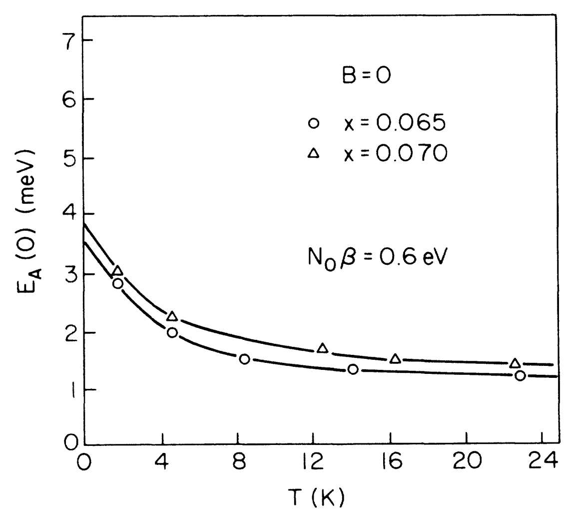

FIG. 8. The zero-field binding energy $E_A(0)$ of the resonant acceptor as a function of temperature for $N_0\beta = 0.6$ eV. The value of $E_A(0)$ in the limit of zero temperature extrapolates to 3.5 meV (3.8 meV) for $x = 0.065$ ($x = 0.07$).

range of BMP behaviors that is possible only for an acceptor BMP in this system; a low-temperature state dominated by cooperative effects and a fluctuation-dominated high-temperature regime. At low temperatures and low magnetic fields the cooperative exchange between a quasibound hole and its surrounding Mn ions leads to a deeper bound state than that caused by the Coulomb impurity potential alone. As the temperature increases, the correlated mean-field exchange interaction decreases, but, instead, the thermodynamic spin fluctuations start to contribute to the binding energy which results in temperature-independent enhanced acceptor binding energy relative to the nonmagnetic semiconductor. The zero-field binding energies of RABMP's extrapolate to ~ 3.5 and 3.8 meV for $x=0.065$ and $x=0.070$, in the limit of zero temperature for $N_0\beta=0.6$ eV.

V. CONCLUSIONS

In summary, we have observed the impurity transition $\mathrm{RA}\rightarrow b_v(-1)$, which was identified by the selection rules, temperature-dependent exchange effect, and $\mathbf{k}\cdot\mathbf{p}$ Landau-level calculations. The remarkable shift of the resonance field as temperature increases is attributed to the strong *sp-d* exchange interaction in this semimagnetic material. The Γ_8 Landau-level ordering leading to this transition is also consistent with the results of Hall and magnetotransport measurements. The alternative explanation for the observed transition by the spin hole resonance has been ruled out since there have been no evidences for an appreciable amount of the zero-field energy splitting between $a_v(-1)$ and $b_v(-1)$ levels. The plot of the transition energy as a function of calculated magnetization, followed by a linear extrapolation using a least-squares fit, yields the negative zero-field extrapolated energy whose magnitude increases rapidly as temperature is lowered. This feature was interpreted in terms of possible evidence for the formation of RABMP's at low temperature and low magnetic field in the zero-band-gap HMT. Unfortunatly, the measurements of the impurity transition at lower magnetic field ($B\leq 1$ T), which is crucial to confirm the conclusion, are obscured by the strong absorption occurring around at zero field (Drude absorption). Furthermore, the resonant-acceptor level was not resolved because of the large linewidth of the resonance due to the high density of acceptor states.

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