Magnetization of $Hg_{1-x}Mn_xTe$

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The magnetization of $Hg_{1-x}Mn_xTe$ for x values from 0.007 to 0.23 has been measured over a temperature range from about 2 to 25 K and in magnetic fields from 3 Oe to 40 kOe. The results for each x value can be well described by a modified Brillouin function with two fitting parameters $\bar{x}$ and T_0. The temperature dependence of the low-field susceptibility is proportional to $T^{\alpha(x)}$, where $-1 \leq \alpha(x) \leq 0$. Models based on random-exchange interactions predict such a power-law dependence.

I. INTRODUCTION

The first magnetic-susceptibility measurements in $Hg_{1-x}Mn_xTe$ by Savage *et al.*[1] showed the strong influence of the magnetic ion, Mn^{2+}, on this system. From a linear extrapolation of their measurements of inverse susceptibility (χ^{-1}) at high temperatures they found large negative paramagnetic Curie temperatures and concluded that clustering was responsible for this effect. Sonderman and Vogt,[2] also by linear extrapolation from high temperatures, found positive values of the Curie temperature for small concentrations of Mn ($x < 0.05$) and the expected negative temperatures for larger values of x. Davydov *et al.*,[3] however, determined that the Curie temperature was negative even for low concentrations of Mn. It is possible that the positive sign in Ref. 2 resulted from omission of the diamagnetic contribution of Te and Hg ions. In addition, Dobrowolski *et al.*[4] measured the magnetization at fields up to 150 kOe and fitted their data to modified Brillouin functions[5] for $x \geq 0.1$. Attempts by all these investigators to explain the results by cluster models based on a random distribution of Mn were unsuccessful even for small concentrations of Mn.

In contrast to the studies mentioned above, which were carried out at high fields (above 1 kOe), Nagata *et al.*[6] used a field of only 15 Oe to observe spin-glass effects in alloys with an x value of 0.35. These authors also concluded that clusters based upon a random distribution of Mn did not describe their experimental results. Brandt *et al.*,[7] carrying out measurements at temperatures below 1 K, found a spin-glass transition in the gapless regime ($0.02 \leq x \leq 0.075$) and speculated that the mechanism for the exchange interaction might be different in the gapless regime from that for large x. Recently, however, Shapira *et al.*[8] observed steps in the high-field magnetization of wide-gap, diluted magnetic semiconductors with small concentrations of Mn. These steps appear to provide evidence for a random distribution of Mn ions. It is not clear at the present time whether these results are applicable at higher concentrations or for narrow-gap materials such as $Hg_{1-x}Mn_xTe$. We will return to this point in the Results section.

Questions remain about the magnetization of $Hg_{1-x}Mn_xTe$ and its interpretation, and there are no measurements that extend from low magnetic fields to high fields on the same samples. Therefore in order to examine the effect of Mn on the magnetic properties of $Hg_{1-x}Mn_xTe$ and to obtain data useful in interpreting galvanomagnetic measurements, we have measured the magnetization for a range of x values from 0.007 to 0.23 from low fields, approximately 3 Oe, to 40 kOe. The magnetization data have been fitted to the modified Brillouin function.

Although this function has no theoretical basis, it has been used to fit the magnetization data in a number of diluted magnetic semiconductors with some success.[4,5,9,10] In $Hg_{1-x}Mn_xTe$ this function has not been used previously for small x values and a systematic study of the influence of temperature upon the fitting parameters has not been made.

The fitting expression for the magnetization in terms of a modified Brillouin function is given by

$$M = S_0 g_{\mathrm{Mn}} \mu_B \bar{x} N_0 B_{5/2}(\zeta) , \tag{1}$$

where

$$\zeta = S_0 g_{\mathrm{Mn}} \mu_B H / k_B (T + T_0) \tag{2}$$

and

$$B_S(\zeta) = \frac{2S+1}{2S} \coth\left[\frac{2S+1}{2S}\zeta\right] - \frac{1}{2S}\coth\left[\frac{\zeta}{2S}\right] \tag{3}$$

is a Brillouin function. Here the magnetization is given in emu/g, $S_0 (= \frac{5}{2})$ is the spin value of a Mn^{2+} ion, $g_{\mathrm{Mn}} (=2)$ is the g factor of the Mn^{2+} ion, μ_B is the Bohr magneton, N_0 is the number of cation sites per gram, k_B

is the Boltzmann constant, T is the temperature, and H is the magnetic field. There are two fitting parameters, T_0 and $\bar{x}$, which represent an exchange interaction and the effective occupation probability of a cation site by a Mn ion, respectively.

II. EXPERIMENTAL TECHNIQUE

A variable temperature susceptometer, with a superconducting quantum interference device (SQUID) detector,[11] was used to measure the susceptibility of $Hg_{1-x}Mn_xTe$ single crystals. Because this detection system is very sensitive, measurements could be made in fields as low as 3 Oe. Our measurements were made at selected fields up to 40 kOe and over a temperature range from about 2 to 25 K since this range covered most of the interesting magnetotransport results. In some cases measurements were carried out at temperatures up to 100 K.

For temperatures above 4 K the measured magnetization was the average of 16 measurements taken at fixed field and temperature. After a change in the magnetic field the superconducting magnet was switched into the persistent mode and the system was allowed to stabilize until the temperature was constant to within the experimental error of less than 1%. The entire switching and stabilizing process took about 15 min.

Since temperatures below 4 K could not be controlled automatically, measurements in this range were made for only a few of the samples and at a smaller number of magnetic fields than were used at higher temperatures. Under these conditions the magnetization was the average of four measurements and the error in temperature may have been as large as 5%.

In the beginning we did not know whether fields below 100 Oe could be obtained reproducibly using the SQUID system and its superconducting magnet. Our magnetization measurements, however, indicated that the uncertainty in field was less than 0.5 Oe for applied fields H less than 100 Oe. Above 1000 Oe the field error was less than 0.5%. The low-field measurements were made after the initial cooling of the magnet from room temperature. During this period the magnetic field was never increased above 100 Oe. After the high-field measurements to 40 kOe it was not possible to return to the low-field condition without warming up the system to allow the magnet to go normal.

The samples of $Hg_{1-x}Mn_xTe$ were grown by the modified Bridgman technique[1] from a mixture of the elements, Hg, Mn, and Te. After growth, slices were cut from each single-crystal boule and annealed in Hg vapor for about 30 days. The x values (see Table I) were determined by density, x-ray fluorescence, and x-ray microprobe measurements. From these analyses we estimate no more than a 5% variation in the value of x for each sample with $x > 0.08$. For smaller x values the variation was less than 15%. Each sample was suspended in the SQUID system by a cotton-polyester thread whose magnetization was negligible compared to that of the sample itself. The masses of the samples used in our magnetization studies were from 50 to 150 mg.

III. RESULTS

In Fig. 1 we show the magnetization M versus magnetic field H at different temperatures for three different values of x, representing our data for low, intermediate, and high concentrations of Mn. Each solid line was obtained from a nonlinear, least-squares fit of a modified Brillouin function, Eq. (1), to the data. At high temperatures M depends nearly linearly upon H, but at low temperatures there are appreciable deviations from a straight line. For all temperatures and x values it is clear that the data are fitted quite closely by Eq. (1), but the fitting parameters, $\bar{x}$ and T_0, depend upon temperature as shown in Table I. We have chosen to present the first parameter as

TABLE I. Fitting parameters at different temperatures for $Hg_{1-x}Mn_xTe$.

x	T (K)	4	6	8	10	15	25
0.007	$\bar{x}$	0.0067	0.0067	0.0067	0.0067	0.0067	0.0065
	T_0	1.6	1.6	1.6	1.6	1.7	1.8
0.05	$\bar{x}$	0.027	0.027	0.028	0.028	0.028	0.031
	T_0	4.9	4.9	5.2	5.2	5.2	6.3
0.06	$\bar{x}$	0.030	0.031	0.031	0.033	0.035	0.039
	T_0	5.5	5.7	5.7	6.6	7.8	9.7
0.07	$\bar{x}$	0.031	0.032	0.034	0.035	0.040	0.047
	T_0	5.8	6.1	6.7	7.3	9.6	14.4
0.10	$\bar{x}$	0.032	0.033	0.035	0.037	0.042	0.053
	T_0	5.7	6.1	7.0	7.7	9.9	16.7
0.12	$\bar{x}$	0.032	0.033	0.035	0.039	0.043	0.052
	T_0	5.8	6.0	6.6	7.6	9.3	12.9
0.16	$\bar{x}$	0.031	0.032	0.034	0.036	0.042	0.049
	T_0	5.7	5.5	5.8	6.2	7.7	8.6
0.23	$\bar{x}$	0.029	0.029	0.030	0.033	0.043	0.075
	T_0	7.7	6.4	5.7	5.5	8.3	24.7

an effective x rather than an effective spin since this seems to be a more consistent approach. [The spin occurs both in the coefficient and in the argument of the Brillouin function in Eq. (1), but only the spin quantity in the coefficient has been used as a fitting parameter by other authors, the spin appearing in the argument of the Brillouin function having been fixed at $\frac{5}{2}$.[4]] The number of Mn^{2+} ions contributing to the magnetization, represented by $\bar{x}$, may change with temperature due to antiferromagnetic interactions that occur among neighboring Mn^{2+} ions, but the spin of an individual Mn^{2+} ion would not be expected to vary.

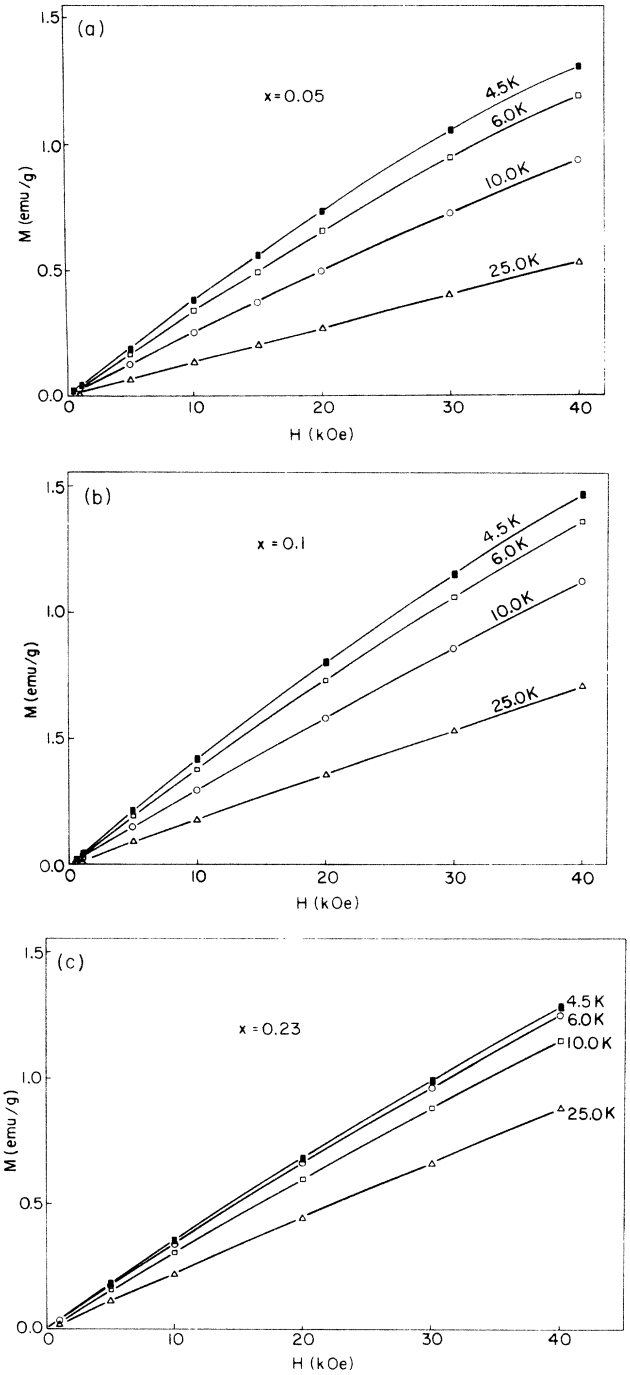

FIG. 1. (a) Magnetization versus magnetic field for $Hg_{0.95}Mn_{0.05}Te$. The lines are the fit to a modified Brillouin function with the parameters given in Table I. (b) Magnetization versus magnetic field for $Hg_{0.9}Mn_{0.1}Te$. The lines are the fit to a modified Brillouin function with the parameters given in Table I. (c) Magnetization versus magnetic field for $Hg_{0.77}Mn_{0.23}Te$. The lines are the fit to a modified Brillouin function with the parameters given in Table I.

The errors in the parameters shown in Table I may be estimated from the errors in the measurements of magnetization by using the standard error analysis for a nonlinear least-squares fit. We estimate for $T < 10$ K that the errors in $\bar{x}$ and T_0 are less than 3% and 5%, respectively. For $T > 10$ K, however, the maximum field of 40 kOe was too small to give an appreciable nonlinearity in M versus H. The standard error analysis gave errors of roughly 6% and 15% for $\bar{x}$ and T_0, respectively, but as one might expect showed a high correlation between the two parameters. Therefore we do not attach much significance to the values of $\bar{x}$ and T_0 for $T > 10$ K, but the numbers are given in Table I to show the trends and for completeness.

Ivanov-Omskii *et al.*[12] have measured the susceptibility χ_{HgTe} and found at low temperatures a nearly constant, negative χ_{HgTe} value of about -3.5×10^{-7} emu/g, which they attribute to a diamagnetic lattice contribution. This is about 1% of the susceptibility values we have measured for $Hg_{1-x}Mn_xTe$ with $x=0.007$ and about 0.1% for larger values of x. Thus we have included this constant term in our fits for the $x=0.007$ sample, but for higher x values this diamagnetic contribution has been omitted because it is so small.

We have also examined the effect of adding a constant fitting parameter C to the right-hand side of Eq. (1). This parameter represents approximately the uncertainty in our magnetic field determination as one can see by examining the Brillouin function for small arguments. When low-field data ($H < 100$ Oe) were included in our fit, the value of C corresponded to a field correction of about 0.2 Oe. When only high-field data ($H \geq 500$ Oe) were used, the value of the field correction corresponding to C was larger, roughly 20 Oe. The other parameters were changed by less than their uncertainty due to experimental error when C was included as a fitting parameter.

From Table I it can be seen that, except for $x=0.007$, $\bar{x}$ is much smaller than the nominal value of x and increases with temperature. This is not surprising if some of the Mn ions are located in clusters and the exchange interaction J among them is antiferromagnetic. Only at high temperatures where $k_BT > J$ should the parameter $\bar{x}$ approach the value of x, and Table I, in fact, shows that $\bar{x}$ does increase with T. The reduction in $\bar{x}$ should be smaller for smaller x, and for our lowest concentration we find that $\bar{x} \approx x$.

A partial understanding of the increase in T_0 with T (Table I) can be obtained from a plot of the low-field ($H \leq 100$ Oe), inverse susceptibility, χ^{-1} versus T as shown in Fig. 2 for samples with x values of 0.007, 0.05, 0.1, and 0.23. Except for the $x=0.007$ sample the curvatures of these plots are negative. Therefore, if a straight line, fitting the data over some temperature range, is extrapolated until it crosses the axis of the abscissa, a nega-

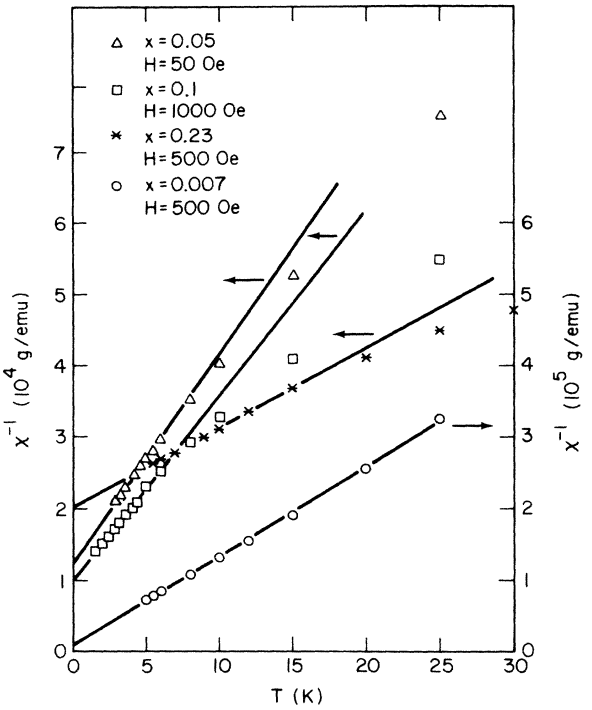

FIG. 2. Inverse susceptibility versus temperature for $Hg_{1-x}Mn_xTe$. The straight lines are fits to the low-temperature data.

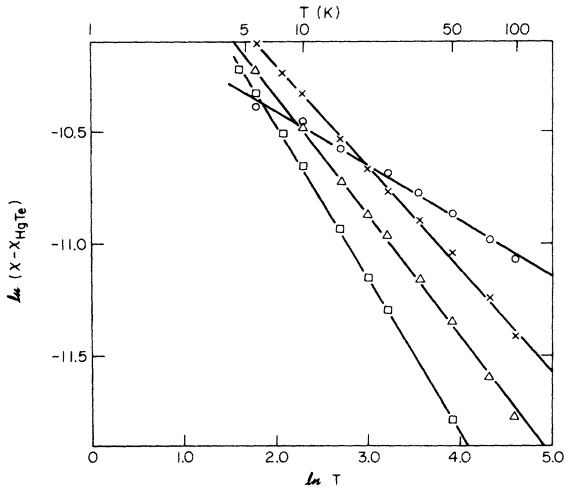

FIG. 3. $\ln(\chi-\chi_{HgTe})$ versus $\ln(T)$ for $Hg_{1-x}Mn_xTe$. ○, $x=0.23$; ×, $x=0.16$; △, $x=0.075$; □, $x=0.043$.

tive intercept is found which corresponds roughly to $T_0(T)$. This intercept moves to a more negative value if the linear fit is made at higher temperatures. In the case of the $x=0.007$ sample, however, χ^{-1} is almost linear with T and $T_0(T)$ is nearly constant.

A negative curvature similar to that in our χ^{-1} versus T plots was also found for phosphorus-doped silicon.[13–15] This behavior was explained by models which show that in a system with random-exchange interactions in any dimension, the low-temperature susceptibility follows a power law in temperature as T^{α} where $-1 \leq \alpha \leq 0$. Standard Curie-law behavior would correspond to $\alpha = -1$. Although it may seem surprising to compare phosphorus-doped silicon with $Hg_{1-x}Mn_xTe$, one may note that the extent of the wave function in phosphorus-doped silicon ($a_P \approx 17$ Å) compared with the separation of phosphorus atoms is similar to the ionic radius of Mn ($a_{Mn} \approx 0.8$ Å) compared to the separation of Mn ions. That is, the values of $n_P a_P^3$ studied by Andres *et al.*[13] range from 5.4×10^{-4} to 1.8×10^{-2}, while our values of $n_{Mn}a_{Mn}^3$ vary from 5×10^{-5} to 1.8×10^{-3}. Here n_P and n_{Mn} are the densities of phosphorus and manganese atoms, respectively. We have fitted the data for various concentrations x as shown in Fig. 3 and find that the random-exchange model fits the data very well except for the highest concentrations.

In Fig. 4 we show α, a measure of the random nature of the spin interactions, versus the Mn concentration. As expected, in the low-concentration limit one recovers a Curie contribution with a value of α approaching -1. Moreover, the fit to the susceptibility data at low concentrations allows no adjustable parameters since the concentration of Mn is known. Agreement is found between the measured susceptibility and the theoretical Curie contribution in this limit. Of course, in the very-low-temperature regime, below about 2 K, one would expect deviations from the pure Curie term since random spin interactions would affect the susceptibility. Generally speaking, one sees the power-law behavior in the susceptibility whenever the "characteristic" interaction energy between spins is of the order of k_BT.

For the highest concentrations it is reasonable to expect a significant influence on the susceptibility from antiferromagnetic interactions within clusters which can account for the poorer quality fits for these data. Nearest-

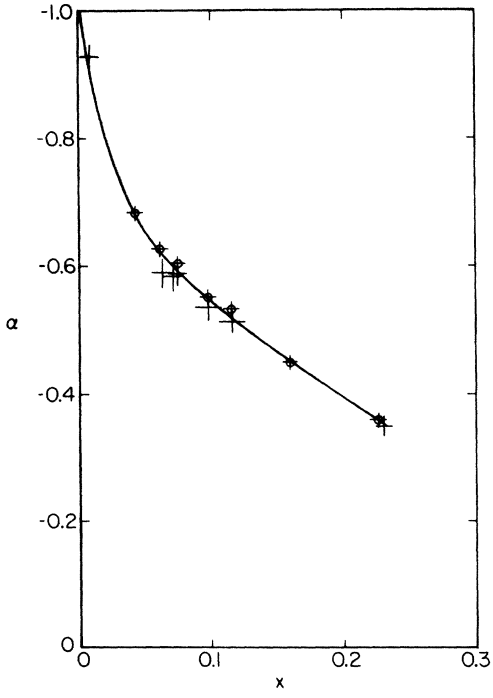

FIG. 4. $\ln\alpha$ versus x. ⊕ indicates the most accurate values.

neighbor cluster interactions should not have a random character to their interaction energies, and therefore the contribution to the susceptibility from these clusters would look Curie-like for k_BT much greater than the interaction energy and would go to zero exponentially for k_BT much less than the interaction energy. We have not attempted to fit the data from this point of view since inclusion of the nearest-neighbor clusters and random interactions introduces double the number of parameters and one cannot place much confidence in the uniqueness of the model. We merely wish to point out that our data are consistent with this point of view.

The model based on random-exchange interactions also provides another way to look at the temperature dependence of T_0. In the low-field regime the Brillouin function of Eq. (1) can be expanded for small argument ζ and the susceptibility equated to q_0T^{α}, where q_0 is independent of T, in order to determine $T_0(T)$. If this is done and if $\bar{x}$ is assumed to be constant, then one finds that $T_0(T)$ increases for low T to a rather flat maximum, whose position and width depend upon x, and then decreases. The maximum occurs at about 4 K for $x=0.23$ and at about 8 K for $x=0.12$. This maximum becomes sharper as x increases. The low-temperature data of Table I follow this behavior. For large x values the measurement temperature is not low enough to see the initial increase and $T_0(T)$ decreases. For x values in the vicinity of 0.1 there is an initial slow increase in $T_0(T)$. At low x values, after an initial increase, $T_0(T)$ is nearly constant. At higher temperature $\bar{x}$ increases as expected and this is accompanied by a rapid increase in $T_0(T)$. At temperatures above 10 K the fitting parameters of Eq. (1) are too highly correlated.

We also see in Fig. 2 that the curve for $x=0.23$ crosses the $x=0.1$ and 0.05 curves at low temperatures. In fact, at 5 K the low-field susceptibility has a maximum at $x\approx 0.14$. These results imply that at low temperatures the susceptibility for large x is actually less than that for small x, which, we believe, is due to the antiferromagnetic exchange interaction causing partial cancellation of the effects of Mn moments in clusters.

From Table I we observe that the samples with different Mn concentrations show two types of behavior based upon the change of T_0 with temperature. Because the two parameters, $\bar{x}$ and T_0, are correlated especially at high temperatures, it is likely that part of the increase in T_0 with temperature is related only to our fitting procedure. That is, an increase in $\bar{x}$ may produce an increase in T_0 as well. To account for this correlation we have plotted $T_0/\bar{x}$ versus temperature in Fig. 5 for four representative x values. For small x values $T_0/\bar{x}$ increases monotonically with T; the increase becomes stronger for larger x. For our largest x values ($x\geq 0.14$), as T increases, there is an initial decrease in $T_0/\bar{x}$ and then a rapid increase. This seems to be a real effect and not a result of our fitting procedure. The increase in $T_0/\bar{x}$ at higher temperatures reflects the behavior of χ^{-1} versus T shown in Fig. 2.

We have also compared our data with those of Dobrowolski *et al.*[4] by fitting their data to modified Brillouin functions for several different ranges of magnetic field. For $H\leq 70$ kOe the parameters obtained are nearly independent of the maximum field used in the fit and are the same as ours within experimental error. For higher maximum fields the fit is not nearly as good and both T_0 and and $\bar{x}$ are larger. (Dobrowolski also found larger values for his parameters for fits to 100 kOe in comparison with fits to 70 kOe.[16]) If we look carefully at the data of Dobrowolski *et al.*, we see that a smooth modified Brillouin function does not fit the data at high fields and, in fact, there may be steps in the data similar to those observed by Shapira *et al.*[8] in wide-gap diluted magnetic semiconductors. If these steps are real, then one would not expect a modified Brillouin function to fit the data at high magnetic fields, and this may account for the apparent difference between our parameters and those presented in Ref. 4, which included data to 150 kOe.

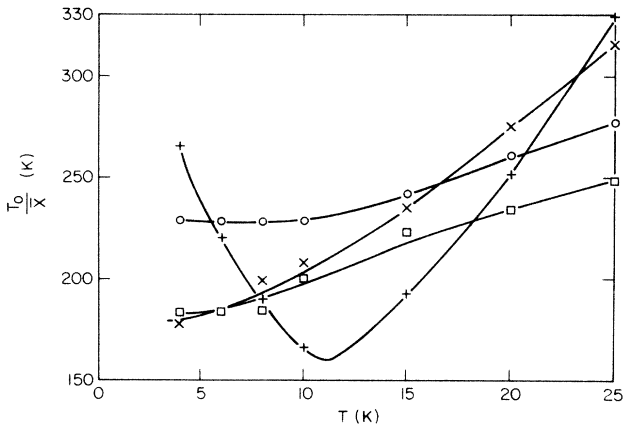

FIG. 5. $T_0/\bar{x}$ versus temperature for $Hg_{1-x}Mn_xTe$. ○, $x=0.007$; ×, $x=0.05$; □, $x=0.1$; +, $x=0.23$. The lines are drawn only as an aid in following the trend of the temperature dependence.

IV. CONCLUSIONS

The presence of Mn ions affects significantly the magnetic properties of $Hg_{1-x}Mn_xTe$. The magnetization for all x values at fields up to 40 kOe can be represented very well in terms of modified Brillouin functions with parameters that depend upon temperatures as shown in Fig. 5 and Table I. This functional form does not have a sound theoretical basis,[5] but attempts to interpret magnetization measurements theoretically in terms of different types of Mn clusters have not been successful even for concentrations as low as $x=0.01$. Several authors have suggested that the cluster models fail because the distribution of Mn ions is not random, but unfortunately a better model including a more realistic distribution of magnetic ions has not been proposed for $Hg_{1-x}Mn_xTe$. The fact that the low-field susceptibility is proportional to T^{α} over a temperature range from about 4 K to at least 100 K is consistent with a random distribution of exchange interactions and perhaps with a random distribution of Mn ions. Future studies should try to relate these experimental results and particularly the dependence of α upon x to better cluster models, but at the present time fitting to a modified Brillouin function seems appropriate.

Both $\bar{x}$ and T_0 are related to the antiferromagnetic exchange coupling among Mn ions and it should be possible to use our experimental fits to examine this exchange. An increase of $\bar{x}$ with temperature is expected because this implies that the interactions among Mn ions are less effective at high temperatures and the total magnetization approaches that of a system of isolated ions. For large values of x, there is an initial decrease in T_0 with temperature while $\bar{x}$ is increasing. Although we have no good explanation for the decrease in T_0 with T, if T_0 represents a characteristic temperature for clusters, T_0 would decrease as the Mn ions begin to act more independently at increasing temperatures.

We have also tried to fit all the data for one x value with one set of temperature-independent parameters. In this case the parameters obtained by our generalized least-squares fitting method were close to the high-temperature values and the low-temperature fit was poor. Thus we concluded that a temperature-independent set of parameters in Eq. (1) is unsatisfactory, unlike the situation in $Pb_{1-x}Mn_xTe$.[10] The modified Brillouin function expression has been useful, however, in obtaining the parameters of the exchange interaction between Mn^{2+} and the band carriers which are included in the field-dependent Hamiltonian for calculation of the dependence on magnetic field of the energy bands of $Hg_{1-x}Mn_xTe$.

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