Magneto-optical study of the spin freezing in $Hg_{1-x}Mn_xTe$ semimagnetic semiconductors

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The dynamic behavior of the magnetic susceptibility in the region of spin-glass formation is studied in $Hg_{1-x}Mn_xTe$ semimagnetic semiconductors. The Faraday rotation at low ac magnetic field is proportional to χ; this has allowed us to measure the ac susceptibility magneto-optically. The temperature dependence of the real component χ' (dispersion) as well as the imaginary component χ'' (absorption) of the complex susceptibility around the freezing temperature ($T_f \simeq 9.3$ K) is determined within the frequency range 70–3000 Hz, for the compound $Hg_{0.70}Mn_{0.30}Te$. Effects of the external static magnetic field ($H \leq 1000$ G) on the complex susceptibility are also investigated. The susceptibility behavior is consistent with a simple phenomenological model; in particular, the fundamental relation $\chi'' = -(\pi/2)\partial\chi'/\partial\ln\omega$ derived within the general assumption of a wide distribution of relaxation times, is quantitatively verified in the vicinity of T_f. The freezing temperature, obtained from low-field and low-frequency Faraday-rotation experiments, exhibits a linear composition dependence for $0.25 \leq x \leq 0.45$.

I. INTRODUCTION

The ternary compounds $Hg_{1-x}Mn_xTe$ ($x \leq 0.5$) are diluted magnetic semiconductors having the zinc-blende structure. Such systems have recently received considerable interest due to their important magneto-optical and magnetic properties.[1–5] The presence of localized magnetic moments ($S = \frac{5}{2}$) originating from the half-filled $3d$ shells of Mn results in many spectacular effects; e.g., giant Faraday rotation,[6] and anomalously large effective g factors of the electrons and holes[7] are the consequences of the strong s-d and p-d exchange interactions. The magnetic behavior of the Mn spin subsystem in $Hg_{1-x}Mn_xTe$ has been studied by magnetization,[8] magnetic susceptibility,[5,9,10] and specific-heat[5] measurements which have proven the existence of paramagnetic and spin-glass phases in these compounds.

Magnetic-susceptibility measurements lead, at high temperature, to a Curie-Weiss law ($\chi^{-1} \propto T + \Theta$), where the large and positive Curie-Weiss temperature Θ displays antiferromagnetic interactions between localized moments. The dominant nearest-neighbor exchange integral $J_1 = -7$ K is estimated from the Curie-Weiss temperature.[11]

One of the most striking properties of $Hg_{1-x}Mn_xTe$ compounds is the formation of a spin-glass phase at low temperature. The first experimental evidence of this was provided by the observation of a characteristic cusp in the temperature dependence of the dc magnetic susceptibility for an alloy of composition $x = 0.35$ at $T_g = 10.9$ K.[5] The occurrence of irreversible effects below T_g corroborates the existence of a spin-glass state in this compound. As in other diluted magnetic semiconductors,[12–14] the origin of the spin-glass phase in $Hg_{1-x}Mn_xTe$ is attributed to a frustration mechanism, inherent in the fcc lattice where magnetic ions interact only antiferromagnetically. As theoretically predicted by De Seze,[15] the topology of the lattice cannot accommodate all nearest-neighbor antiferromagnetic interactions, and the frustration leads to a frozen disordered state.

As a result of the strong exchange coupling between localized magnetic moments and mobile carriers, magneto-optical methods provide an access to the magnetic properties of diluted magnetic semiconductors. Faraday rotation experiments offer a sensitive method to study the evolution of magnetic properties from the paramagnetic to spin-glass state.[16–18] Since the dominant contribution to the valence- and conduction-band energy splittings in wide-gap semimagnetic semiconductors, in a magnetic field results essentially from the exchange interactions, the interband Faraday rotation[19] becomes proportional to the Mn spin magnetization, which leads to giant values of the Verdet constant (as large as 1000 deg/cm kG in $Cd_{1-x}Mn_xTe$).[20]

We previously reported on the paramagnetic-to-spin-glass transition observed in concentrated $Hg_{1-x}Mn_xTe$ compounds by simultaneous low-field Faraday rotation and ac magnetic susceptibility measurements.[16] However, no systematic study of the frequency dependence of the complex susceptibility has been so far performed. Measurements of the imaginary component are shown to be essential to obtain information on the freezing process in spin-glass systems.[21]

The purpose of this paper is to present a first experimental study of the spin freezing in $Hg_{1-x}Mn_xTe$ semiconducting compounds ($0.25 \leq x \leq 0.45$). The Faraday rotation method was applied to determine the real (dispersion) and imaginary (absorption) components of the magnetic susceptibility in the region of the spin-glass formation. Experimental results obtained in the frequency range $70 \leq \nu \leq 3000$ Hz are consistent with a simple phenomenological model based on a wide distribution of relaxation times.

II. EXPERIMENTAL DETAILS

$Hg_{1-x}Mn_xTe$ single crystals of high homogeneity were grown by a new technique combining the modified Bridgman method and recrystallization. Alloys of Mn content up to $x=0.50$ were synthesized with homogeneity better than 0.2%/cm along the growth axis. The samples were characterized by density measurements, electron microprobe analysis, and x-ray diffraction.

Single crystals of Mn content $0.25 \leq x \leq 0.45$ were investigated. These compounds display a semiconductor band structure (as opposed to semimetallic band ordering for $x<0.07$) with the energy gap, at 4.2 K, increasing from 0.6 to 1.1 eV in this range of composition.

We used a conventional method to measure the Faraday rotation induced by an alternating magnetic field of weak amplitude ($H_{ac}=10$–50 G). The sample is placed between two linear polarizers oriented at 45°. The transmitted light intensity at the frequency ω of the ac magnetic field is directly proportional to the Faraday rotation. The experiments are made at constant photon energy of the incident light ($E \sim 0.7E_g$–$0.9E_g$) where the transmission of the sample is found to be independent of temperature and magnetic field. At a fixed frequency, simultaneous measurements of the in-phase and out-of-phase components of the ac susceptibility were carried out as a function of temperature by slowly reducing the temperature from 150 K.

We have checked that the Faraday rotation measurements reproduce with excellent accuracy the temperature dependence of the ac susceptibility measured on the same sample by a mutual-inductance method. This comparison has allowed us to establish the absolute values for our optical measurements of the magnetic susceptibility.

At a given frequency, the relative accuracy of the measurements is 1% for χ', 10% for χ''. The sample temperature is determined, with an accuracy better than 0.05 K, by using a calibrated Allen-Bradley resistor located near the sample.

Measurements of the complex susceptibility were also carried out by the Faraday rotation method in the presence of a static magnetic field H_0 parallel to the ac driving field. These latter experiments were made on field-cooled samples.

III. RESULTS FOR $Hg_{0.70}Mn_{0.30}Te$

The temperature dependence of the in-phase (χ') and out-of-phase (χ'') components of the magnetic susceptibility is illustrated in Fig. 1, for the compound $Hg_{0.70}Mn_{0.30}Te$, at frequency $\nu=272$ Hz. χ' exhibits a characteristic maximum at $T_f=9.2$ K, which we identify as the freezing temperature. As one can see in Fig. 1, χ'' is not negligible for $T>T_f$, indicating that magnetization and field are not completely in phase at $T \sim T_f+0.5$ K. χ'' increases with decreasing temperature and an inflection point in $\chi''(T)$ occurs at a temperature very close to that corresponding to the χ' maximum. This behavior is very similar to that observed in many spin-glass systems.

The magnitude of the ac driving field was varied to test for possible alterations in the susceptibility. The $\chi''(T)$ curves obtained for $H_{ac}=26$ and 50 G are practically coincident.

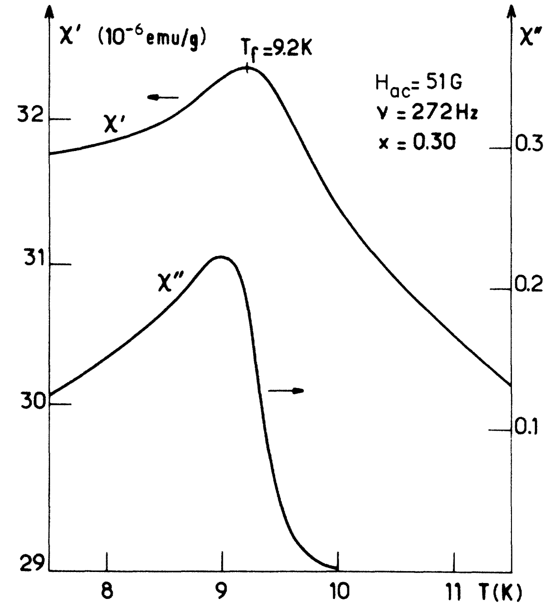

FIG. 1. Temperature dependence of the in-phase (χ') and out-of-phase (χ'') components of the ac susceptibility for $Hg_{0.70}Mn_{0.30}Te$ ($\nu=272$ Hz, $H_{ac}=51$ G).

The temperature dependence of the complex susceptibility was studied for different measuring frequencies between 70 and 3000 Hz. As for other insulating spin glasses,[17,21,22] the temperature dependence of χ', presented in Fig. 2, shows a noticeable frequency dependence with the maximum becoming progressively rounded and shifting to higher temperatures as the frequency is increased. The frequency dependence of the freezing temperature is

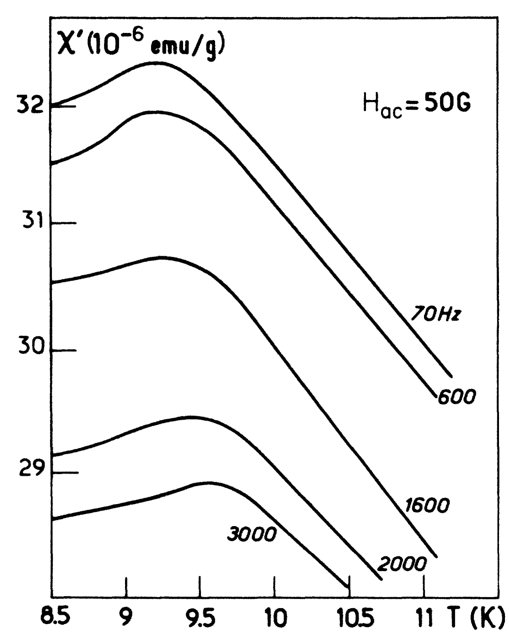

FIG. 2. Temperature dependence of χ' for $Hg_{0.70}Mn_{0.30}Te$ for several frequencies.

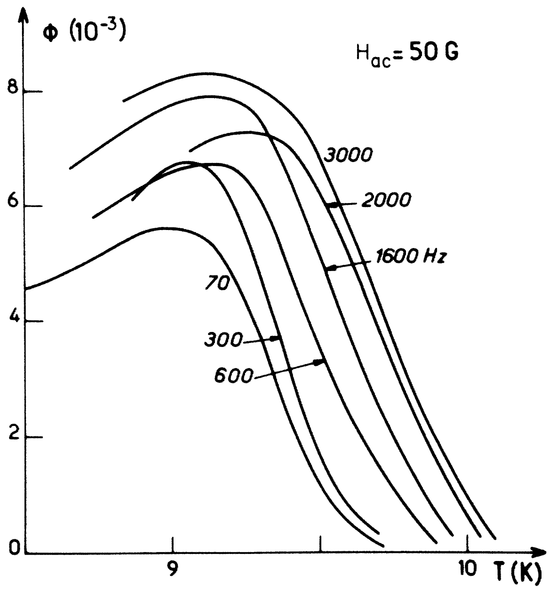

FIG. 3. Temperature dependence of $\Phi \simeq \chi''/\chi'$ for $Hg_{0.70}Mn_{0.30}Te$ for several frequencies.

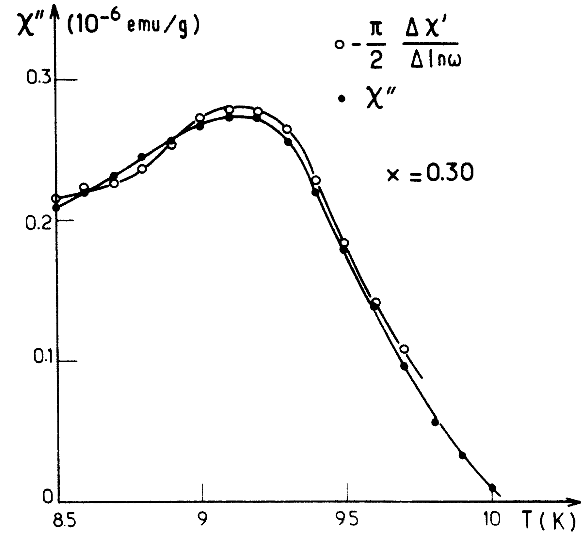

FIG. 4. Comparison between $\chi''(T)$ and $(-\pi/2)(\Delta\chi'/\Delta\ln\omega)$ for $Hg_{0.70}Mn_{0.30}Te$.

correlated with the shift of the χ'' curves between 70 and 3000 Hz. The dephasing factor $\Phi \simeq \chi''/\chi'$ measured at different frequencies is illustrated in Fig. 3. The dephasing factor remains always smaller than 10^{-2}. The relative shift of the freezing temperature $\Delta T_f/T_f \Delta\log\nu \simeq 3\times 10^{-2}$ per decade of frequency is comparable to the corresponding value observed for other insulating spin glasses.[17,21,22]

IV. ANALYSIS

A useful phenomenological model to discuss the complex susceptibility in spin-glass systems has been proposed by Lundgren *et al.*[23,24] This model assumes a random distribution of magnetic ions in the host material with short-range interactions leading to the formation of clusters of strongly coupled spins at temperatures well above the freezing temperature. This assumption seems to be well justified in the case of $Hg_{1-x}Mn_xTe$ compounds in which strong evidences for cluster formation are provided by magnetization and magnetic susceptibility measurements below 50 K. In this model, each cluster exhibits a local anisotropy energy E. The sum of the probabilities per unit time for the coupled spins to surmount the energy barrier E is expressed in terms of a relaxation time τ which is usually described by an Arrhenius law:

$$1/\tau = 1/\tau_0 \exp(-E/k_B T) . \quad (1)$$

Weak intercluster interactions[25] may modify the expression (1) for the relaxation time which is better described by a Vogel-Fulcher law of the form[26]

$$1/\tau = 1/\tau_0 \exp[-E/k_B(T-T_0)] . \quad (2)$$

A cluster appears to be frozen below a certain temperature when the time of observation is less than the relaxation time τ. In an alternating magnetic field of angular frequency ω, clusters of relaxation times $\tau > 1/\omega$ are blocked and do not contribute to the magnetic susceptibility while clusters with $\tau < 1/\omega$ give a paramagnetic temperature-dependent susceptibility.

In order to investigate the frequency dependence of χ, Lundgren *et al.* assign to each cluster a relaxation time τ and a magnetic moment m which at a constant applied magnetic field h_0 attains the value $m_0(\tau)$ at $t\rightarrow\infty$. In an oscillating magnetic field $h = h_0\sin(\omega t)$, the time variation of m is given by the simple differential equation[23]

$$\tau\frac{dm}{dt} + m = m_0(\tau)\sin(\omega t) ,$$

with the solutions

$$m = [m_0(\tau)/1+\omega^2\tau^2]\sin(\omega t) - [m_0(\tau)/1+\omega^2\tau^2]\omega\tau\cos(\omega t) .$$

If the spin glass is considered as a collection of magnetic

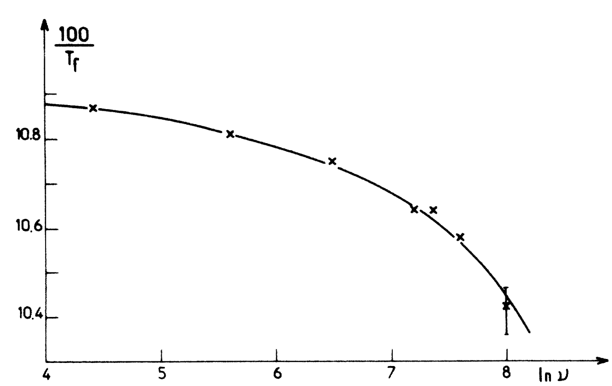

FIG. 5. Experimental variation of the inverse of the freezing temperature (corresponding to the maximum of χ') as a function of the logarithm of the frequency. Crosses are experiment data. The solid line is to guide the eyes.

entities with a broad spectrum of relaxation times $g(\tau)$, the in-phase and out-of-phase components of χ are

$$\chi'=(1/h_0)\int_{\tau_{\min}}^{\tau_{\max}}[m(\tau)/1+\omega^2\tau^2]g(\tau)d(\ln\tau)\ , \qquad (3)$$

$$\chi''=-(1/h_0)\int_{\tau_{\min}}^{\tau_{\max}}[m(\tau)\omega\tau/1+\omega^2\tau^2]g(\tau)d(\ln\tau)\ . \qquad (4)$$

$\tau_{\min}$ and $\tau_{\max}$ denote the minimum and maximum relaxation times in the system. Following Lundgren *et al.*,[23] we denote the system to be in a paramagnetic state when $\tau_{\max} \ll 1/\omega$ and in a spin-glass state when $\tau_{\min} \ll 1/\omega \ll \tau_{\max}$. We have analyzed our experimental data in terms of the above phenomenological approach.

(i) In the paramagnetic region ($\tau_{\max} \ll 1/\omega$), a characteristic average relaxation time $\bar{\tau}$ of the system may be extracted from the dephasing $\Phi \simeq \omega\bar{\tau}$ measured at sufficiently high temperature: The condition $\omega\tau \ll 1$ is satisfied where Φ remains small compared to its value at the inflection point of χ''.[27] In $Hg_{0.70}Mn_{0.30}Te$, at $T=10$ K ($\Phi \ll \Phi_{T_f}$), we obtain $\bar{\tau} \simeq 3\times10^{-7}$ sec for 3000 Hz and 2×10^{-7} sec for 1300 Hz.

(ii) In the spin-glass region ($\tau_{\min} \ll 1/\omega \ll \tau_{\max}$), if $g(\tau)$ is slowly varying in $\ln\tau$, χ' and χ'' are expected to obey the fundamental relation[23]

$$\chi''=-\frac{\pi}{2}\frac{\partial\chi'}{\partial\ln\omega}\ . \qquad (5)$$

This relationship was tested experimentally for the $Hg_{0.70}Mn_{0.30}Te$ compound in the vicinity of the spin-glass freezing temperature (8.5–9.7 K).

Figure 4 shows the comparison between $\chi''(T)$ measured at $\nu=1350$ Hz and $(-\pi/2)(\partial\chi'/\partial\ln\omega)$ calculated between 70 and 3000 Hz. Despite the large difference between the two frequencies, the relation (5) is quantitatively verified by the experimental data. This infers that the spin-glass behavior of $Hg_{1-x}Mn_xTe$ alloy may may be described in terms of a model involving a wide distribution of relaxation times.

(iii) The frequency dependence of the freezing temperature is compared with expressions (1) and (2). It is usually assumed[23] that the maximum of χ' occurs at the blocking temperature of the clusters with the largest energy barrier ($\tau_{\max} \simeq 1/\omega$).

We have compared our experimental data with the following expressions:

$$T_f(\nu)=(E_{\max}/k)[1/(\ln\nu_0-\ln\nu)] \qquad (6)$$

and

$$T_f(\nu)=T_0+(E_{\max}/k)[1/(\ln\nu_0-\ln\nu)]\ , \qquad (7)$$

which result from the Arrhenius (1) and Vogel-Fulcher (2) law's, respectively. Plots of T_f^{-1} versus $\ln\nu$ reported in Fig. 5 show that an Arrhenius law cannot account for our experimental data in the investigated frequency range.

Using various attempt frequencies ν_0, we find that the relation (7) is valid over a large range of ν_0, $T_f(\nu)$, increasing linearly with $1/[\ln(\nu_0/\nu)]$. A quantitative determination of the parameters $T_0,\nu_0,E_{\max}/k$ however, requires measurements over a wider frequency range.

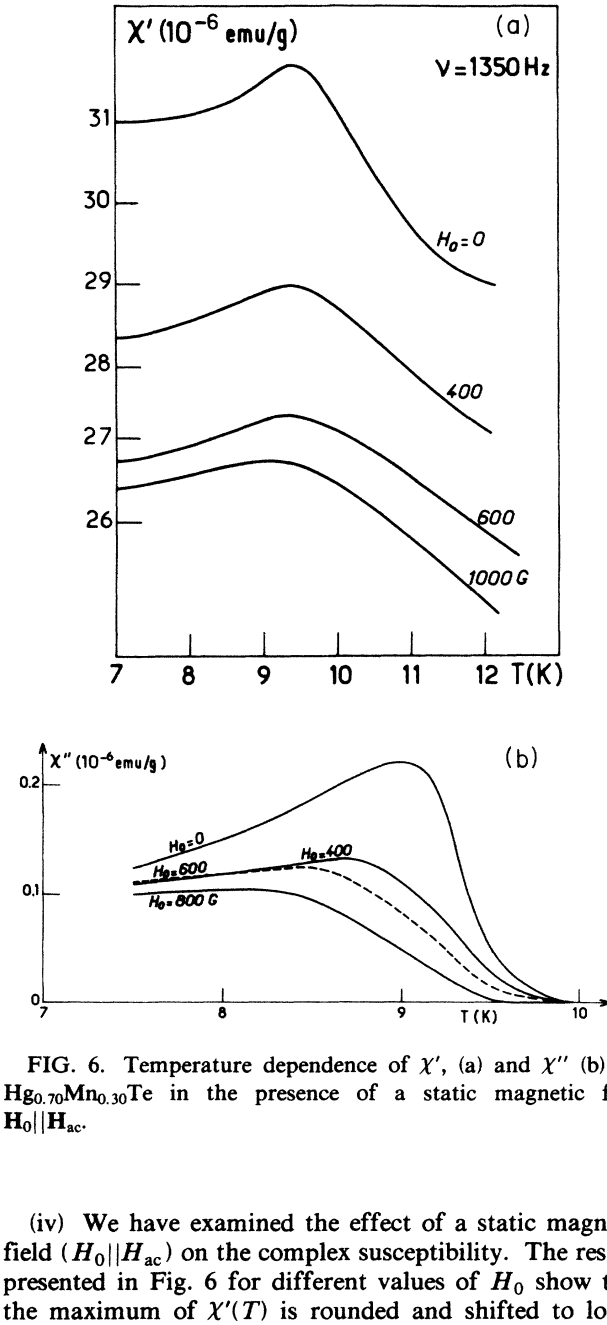

FIG. 6. Temperature dependence of χ', (a) and χ'' (b) for $Hg_{0.70}Mn_{0.30}Te$ in the presence of a static magnetic field $\mathbf{H}_0 || \mathbf{H}_{ac}$.

(iv) We have examined the effect of a static magnetic field ($H_0 || H_{ac}$) on the complex susceptibility. The results presented in Fig. 6 for different values of H_0 show that the maximum of $\chi'(T)$ is rounded and shifted to lower temperatures in the presence of the field. The magnitude of χ'' is strongly reduced in the region of the spin-glass formation with a shift of the threshold of χ'' towards lower temperatures. This behavior is indicative of a decrease of the relaxation times of the clusters in the presence of the field.

V. COMPOSITION DEPENDENCE OF THE FREEZING TEMPERATURE

Figure 7 shows the composition dependence of the freezing temperature in $Hg_{1-x}Mn_xTe$ semiconductors. In the range of composition $0.25 \le x \le 0.45$, T_f are obtained from Faraday rotation experiments performed in weak ac field ($H_{ac}=20$ G) of low frequency ($\nu=30$ Hz). We have also reported in Fig. 7 the results obtained from ac sus-

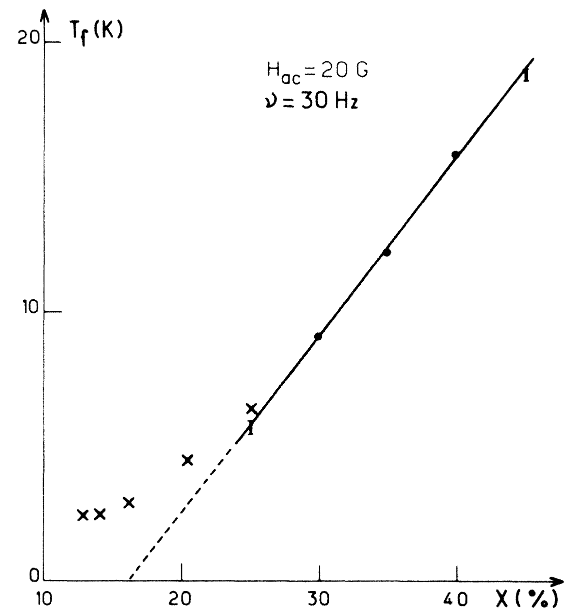

FIG. 7. Composition dependence of the freezing temperature for $Hg_{1-x}Mn_xTe$ semiconductor. ●, Present data; Faraday rotation at weak ac field (H_{ac}=20 G, ν=30 Hz)×, Otto *et al.* (Ref. 28) [ac susceptibility measurements (ν=76 Hz)].

ceptibility measurements[28] (ν=76 Hz) in semiconducting alloys of composition between 0.13 and 0.20.

Above $x \simeq 0.25$, the composition dependence of T_f displays a linear variation, the line $T_f(x)$ intersecting the composition axis at $x_0 \simeq 0.17$. The same behavior was also observed by Oseroff[14] in $Cd_{1-x}Mn_xTe$ compounds: Freezing temperatures obtained from low-field dc susceptibility (after zero-field cooling) are found to vary linearly with composition above $x = 0.20$ and the linear extrapolation of $T_f(H \to 0)$ leads to the same composition $x_0 = 0.17$ at $T \to 0$.

In the fcc lattice, the percolation threshold for the formation of an infinite spin cluster is predicted theoretically for $x_p^{(1)} = 0.195$ when only nearest-neighbor interactions are considered, and for $x_p^{(2)} = 0.136$ when next-nearest-neighbor interactions are taken into account.[29]

The spin-glass behavior[4,28] observed in $Hg_{1-x}Mn_xTe$ compounds of composition below the percolation thresholds $x_p^{(1)}$ and $x_p^{(2)}$ is indicative of the existence of long-range interactions which could result from indirect exchange mechanism or dipolar interactions.

In conclusion, we have reported here a first experimental study of the ac susceptibility in the vicinity of spin freezing in $Hg_{0.70}Mn_{0.30}Te$, a wide-gap semiconductor with short-range antiferromagnetic interactions. The dynamical behavior of the complex susceptibility in the region of the spin-freezing temperature could be described by the existence of relaxing magnetic entities with a broad spectrum of relaxation times, in particular the relation $\chi'' = (-\pi/2)(\partial\chi'/\partial \ln\omega)$ shows excellent agreement with the data.

Further experiments over a wider frequency range (especially for long measuring times) are needed to test more precisely the validity of the Fulcher law.

Static measurements would be of particular interest to characterize the behavior of the nonlinear magnetization when approaching T_f. Such experiments would permit the elucidation of whether the spin freezing observed in this system is just a manifestation of a strong relaxation process or whether it results from a true spin-glass transition.

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