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Dynamics of the spin-glass freezing in semimagnetic semiconductors

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Accurate measurements of the imaginary part of the complex susceptibility are used in order to compare the validity of different dynamic scaling models on the two related spin-glass compounds $Hg_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ ($x=0.3$). The conventional power-law scaling yields in both compounds a dynamic exponent $z\nu=9\pm1$, $T_c=8.4$ K for $Hg_{0.7}Mn_{0.3}Te$ and 6.45 K for $Cd_{0.7}Mn_{0.3}Te$. The generalized in-field scaling leads to an independent and consistent determination of T_c. Good scalings may also be achieved according to activated dynamics, but the P values appear to differ in various systems, a result at odds with the expected universality of the critical exponents.

Semimagnetic semiconductors (SMSC's), such as $Hg_{1-x}Mn_xTe$ or $Cd_{1-x}Mn_xTe$, are disordered Heisenberg antiferromagnets in which the conjunction of randomness and frustration gives rise to a spin glass (SG) at low temperature. Recent studies[1] of the spin dynamics carried out in $Cd_{0.6}Mn_{0.4}Te$ support the existence of a SG transition at finite temperature. The spin freezing has been analyzed in terms of a critical slowing down above the static freezing temperature, using a power-law scaling relation with a critical dynamic exponent $z\nu=9\pm1$. A different dynamic scaling model based on a thermally activated process was also proposed to analyze the departure from equilibrium of the in-phase component of the ac susceptibility in $Cd_{0.6}Mn_{0.4}Te$.[2]

In order to elucidate the spin freezing process in SMSC we have optically studied the dynamic magnetic properties of both systems $Hg_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ for the same composition $x=0.30$, in the region of the SG transition. We consider two different zero-field scaling models (power law as well as activated dynamics) to analyze the frequency and temperature dependences of the ac magnetic susceptibility. The most relevant test of the critical behavior is obtained by measuring and analyzing the imaginary component of $\chi''(\omega,T)$: This quantity is much more accurately determined than $\Delta\chi=\chi'(\omega,T)-\chi_{eq}$ which requires an extrapolation of the equilibrium susceptibility from the lowest available frequency down to zero frequency without the knowledge of the transition temperature T_c. We use generalized field scalings as a sensitive test to determine the values of T_c, which agree quite well with those deduced from the zero-field analysis. Therefore, while the available frequency range is smaller in the present work than in earlier investigations,[2,3] the use of two independent scaling plots may eventually be a more appropriate route to define T_c and, hence, the $z\nu$ exponent. Our conclusion is that the spin freezing in SMSC may be consistently interpreted in terms of a conventional critical slowing down above a finite transition temperature T_c, with reasonable values of the dynamic exponent 9 ± 1 for both compounds.

ac magnetic susceptibility (χ_{ac}) was obtained from Faraday rotation experiments carried out under weak fields ($\Delta H\approx10$–16 G), at a photon energy E slightly smaller than the energy gap ($E/E_g\approx0.9$). Accurate Faraday rotation measurements were made by using a sensitive modulation technique.[4] In-phase (χ') and out-of-phase (χ'') components of χ_{ac} were measured simultaneously in the vicinity of the SG transition, in the frequency range of the oscillating magnetic field (4–10^5 Hz). Nonlinear effects are negligible up to $\Delta H=32$ G. To explore spin dynamics at longer times (0.1–100 s) we have studied the relaxation of the thermoremanent magnetization (TRM) after switching off a small field, at fixed temperatures. The temperature is determined within an accuracy of 0.02 K.

Both types of experiments (χ_{ac} and TRM) were also performed in the presence of an additional static magnetic field H ($H\leq800$ G) applied perpendicularly to the driving field ΔH, to determine the constant relaxation time contours in the field-temperature plane. In $Eu_{0.4}Sr_{0.6}S$ similar results have been obtained for H applied parallel or perpendicular to the driving field.[5] An extensive study of these two field configurations is under way.

The temperature dependence of the static and ac magnetic susceptibility of $Hg_{0.70}Mn_{0.30}Te$ is illustrated in Fig. 1. In static measurements, FC susceptibility is obtained by cooling the sample in a constant field ($\Delta H\approx16$ G) at a rate of 1 mK per s.

In conventional slowing down, the divergence of τ as the SG transition is approached from above is usually assumed to follow the power law:[6]

$$\tau=\tau_0[T_f(\omega)/T_c-1]^{-z\nu}. \tag{1}$$

$T_f(\omega)$ is the frequency-dependent freezing temperature to be defined experimentally, ν is the critical exponent for the correlation length ξ, z is the dynamic exponent relating ξ and $\tau(\tau\propto\xi^z)$, and τ_0 is a microscopic relaxation time.

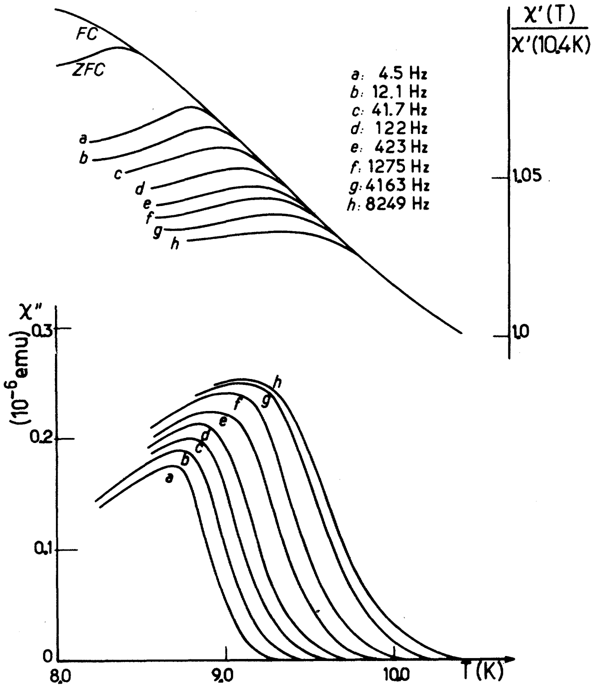

FIG. 1. Temperature dependence of χ' and χ'' at different frequencies for the compound $Hg_{0.7}Mn_{0.3}Te$ ($\Delta H = 16$ G). FC and ZFC curves refer to static measurements.

Dynamic scaling may be investigated by using the relation

$$\chi''(\omega,T) = \omega^{\beta/z\nu} G(\omega\tau) ,$$

or similarly

$$\chi''(\omega,T) = t^{\beta} H(\omega\tau) . \qquad (2)$$

$G(x)$ and $H(x)$ are universal functions of x, β is the exponent of the order parameter, and t is the zero-field reduced temperature $[t = (T - T_c)/T_c]$.

In the limit where $\omega\tau \ll 1$ one gets

$$\chi''(\omega,T) \approx \omega\tau_0 t^{-z\nu+\beta} . \qquad (3)$$

The internal consistency between (2) and (3) is worth being checked since these two scalings yielded contradictory results in $Cd_{0.6}Mn_{0.4}Te$.[1,2] One possible reason for this discrepancy has already been discussed.[7]

In order to verify (3), one defines for each frequency ω the temperatures $T_f(\omega)$ which set χ'' at the same small constant value. To ensure that $\omega\tau \ll 1$, we have checked as in Ref. 8 that the ratio $\chi''/\chi'\omega$ has reached a frequency-independent value at the selected $T_f(\omega)$.

Scaling (3) yields the values

$$\tau_0 \sim 10^{-12}\,\text{s}, \quad z\nu - \beta = 8.5 \pm 0.5 ,$$

$$T_c = 8.42 \pm 0.05\ \text{K} \quad (Hg_{0.7}Mn_{0.3}Te) ,$$

$$\tau_0 \sim 10^{-12}\,\text{s}; \quad z\nu - \beta = 8.85 \pm 0.45 ,$$

$$T_c = 6.45 \pm 0.05\ \text{K} \quad (Cd_{0.7}Mn_{0.3}Te) .$$

Second, we have tested the dynamic scaling law (2) using the full data $\chi''(T,\omega)$ at different frequencies. The best scalings are then achieved for

$$z\nu = 9.5 \pm 0.5; \quad \beta = 0.8 \pm 0.1 ,$$

$$T_c = 8.45 \pm 0.05\ \text{K} \quad (Hg_{0.7}Mn_{0.3}Te) ,$$

$$z\nu = 9.25 \pm 0.75; \quad \beta = 0.8 \pm 0.1 ,$$

$$T_c = 6.45 \pm 0.05\ \text{K} \quad (Cd_{0.7}Mn_{0.3}Te) .$$

Figure 2 shows in the same plot the scaling performed for both compounds when using relation (2). We obtain the same $H(x)$ function as expected from the universality. We consider a set of parameters $(T_c, z\nu, \beta)$ as acceptable when the scattering of experimental points is smaller or comparable to the error bars. As an example, for $T_c = 6.35$ K, the best scaling ($z\nu = 10$; $\beta = 0.8$) is not acceptable as the scattering of data exceeds the experimental accuracy. This criterion leads to the possible range of variation of the parameters T_c, $z\nu$, β.

However, extracting T_c from a best fit through the scaling relation (2) could lead to incorporate experimental points *below* T_c and results in meaningless conclusions. Thus an independent determination of T_c is necessary. We have used measurements in applied static fields to obtain it.

In an applied magnetic field H, in the limit $\omega\tau \ll 1$, the field dependence of ξ yields the generalized scaling relation[9]

$$\chi''(\omega,T,H) = \omega\tau_0 [t(H)]^{\beta - z\nu} F[h^{2/\phi}/t(H)] , \qquad (4)$$

where $t(H) = [T(H) - T_c]/T_c$, $\phi = \beta + \gamma$ is the crossover exponent and $h = g\mu_B H/kT(H)$. $T(H)$ is the freezing temperature which depends on the observation time: In the frequency range $4\text{–}10^3$ Hz, $T(H)$ corresponds to a small constant value of χ''.[5] In the time range $10^{-1}\text{–}10^2$ s, $T(H)$ is defined as the temperature where the decay of TRM after switching off ΔH, in the applied static field H, matches the decay observed in zero field at the temperature T. Relations (3) and (4) imply that for a small constant value of χ'', $t(0)/t(H)$ should be a unique function of $h^{2/\phi}/t(H)$. The scaling of the in-field data is very sensitive to the choice of T_c and can therefore be used as a critical test for its determination. The criterion for an acceptable scaling is, as previously stated, that the scattering of data remains smaller or comparable to the experimental errors. The best scalings correspond to $T_c = 6.42 \pm 0.07$ K; $\phi = 4 \pm 1$ ($Cd_{0.7}Mn_{0.3}Te$) and $T_c = 8.40 \pm 0.1$ K; $\phi = 3.5 \pm 1$ ($Hg_{0.7}Mn_{0.3}Te$). Figure 3 illustrates the scaling of the in-field data for $Cd_{0.7}Mn_{0.3}Te$.

Values of ϕ larger than 5 are unacceptable taking into account the values of β and γ. Our present data yield $\beta = 0.8 \pm 0.1$. Mauger, Ferré, and Beauvillain[10] obtain $\beta = 0.9 \pm 0.2$ and $\gamma = 3.3 \pm 0.3$ from nonlinear susceptibility measurements in $Cd_{0.6}Mn_{0.4}Te$. Our determination of T_c from in-field measurements is therefore quite consistent with the results of the zero-field analysis.

The two equivalent dynamic scaling procedures [relations (2) and (3)] lead to the same value of the dynamic exponent $z\nu$. Moreover, the fitting parameters obtained from the dynamic scaling model are quite comparable for

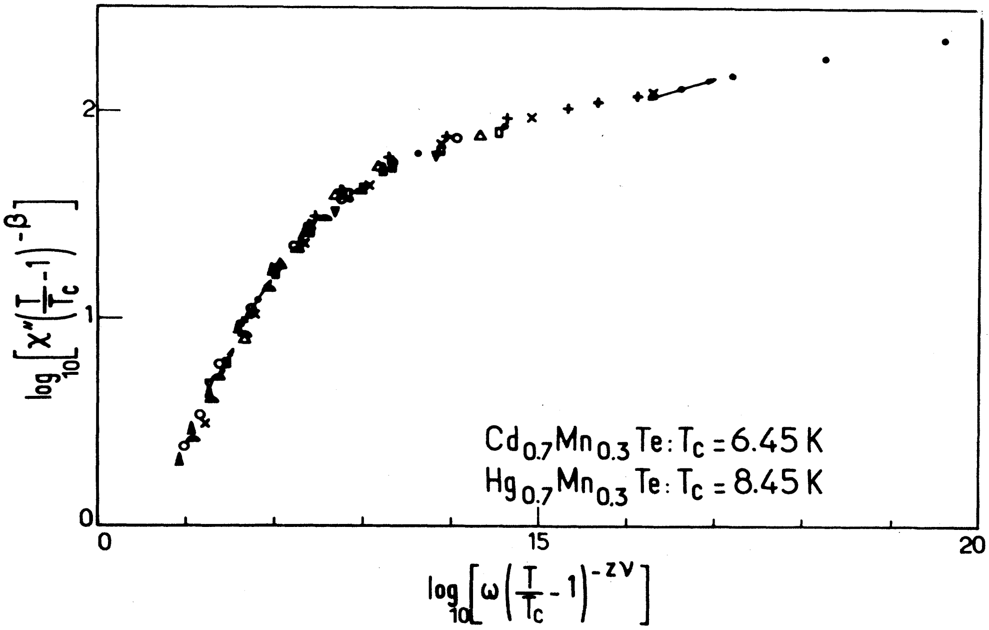

FIG. 2. Power-law scaling of $\chi''(\omega,T)$ data for $\mathrm{Cd_{0.7}Mn_{0.3}Te}$ and $\mathrm{Hg_{0.7}Mn_{0.3}Te}$ according to (2). Symbols refer to different frequencies: ●, +, △, □, ▽ from 7 to 7×10^4 Hz in decade steps for $\mathrm{Cd_{0.7}Mn_{0.3}Te}$; ×, ○, ▲, ◆ from 4 to 4×10^3 Hz for $\mathrm{Hg_{0.7}Mn_{0.3}Te}$.

$\mathrm{Cd_{0.7}Mn_{0.3}Te}$ and $\mathrm{Hg_{0.7}Mn_{0.3}Te}$: $z\nu = 9\pm1$; $\beta = 0.8 \pm 0.1$. For both compounds, the microscopic relaxation time compares well with $\hbar/kT_c$. The dynamic exponent $z\nu$ is in excellent agreement with the value obtained for $\mathrm{Cd_{1-x}Mn_xTe}$ of different compositions [$x = 0.40$ (Ref. 1); $x = 0.45$ and 0.55 (Ref. 11)] and also with simulation data in the case of three-dimensional (3D) SG with short-range interactions.[12] Our analyses do not confirm the large value of $z\nu = 14\pm1$ obtained by Geschwind *et al.*[2] We have checked that whatever the choice of T_c our $\chi''(\omega,T)$ data cannot be satisfactorily fitted for such large values of the dynamic exponent.

Malozemoff and Pytte[13] inspired by Fisher[14] have introduced the so-called activated dynamic scaling to describe the spin dynamics close to T_c. The ac susceptibility is written as

$$\chi''(T,\omega) = t^P G[-t^Q \ln(\omega\tau_0)]\,, \tag{5}$$

where P and Q are critical exponents and G is a scaling function. We have used the relation (5) to analyze the

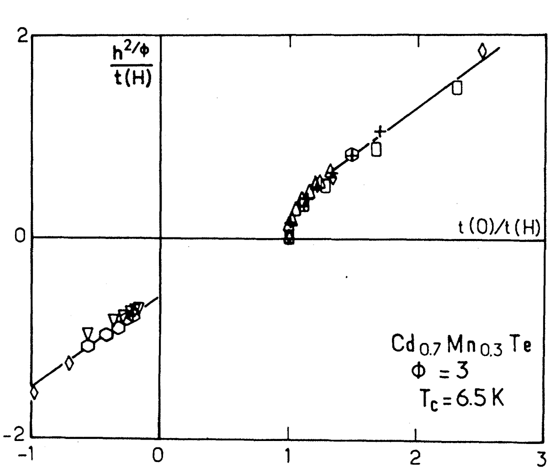

FIG. 3. Scaling of the in-field data for $\mathrm{Cd_{0.7}Mn_{0.3}Te}$: plots of $h^{2/\phi}/t(H)$ vs $t(0)/t(H)$ for $\phi = 3$ and $T_c = 6.50$ K. Symbols correspond to different observation times: △, 721 Hz; +, 70 Hz; □, 7 Hz; ◇, 0.2 s; ○, 2 s; ▽, 10 s.

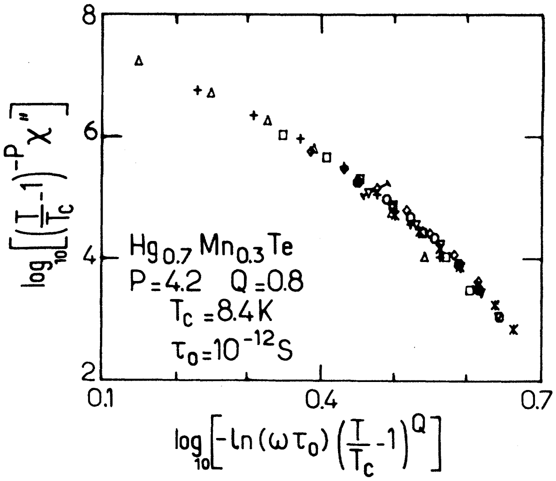

FIG. 4. Activated dynamic scaling of $\chi''(\omega,T)$ data for $\mathrm{Hg_{0.7}Mn_{0.3}Te}$ according to (5). Symbols refer to different frequencies: △, +, □, ◇, ○, ✱, ▽ denote frequencies from 4 to 4×10^3 Hz in half-decade steps. The temperature range is 8.60–10 K.

$\chi''(T,\omega)$ data: taking $\tau_0=10^{-12}$s, excellent scalings are achieved with $3.65 \leq P \leq 4.2$; $Q=0.8$; $T_c=8.40$ K for $\mathrm{Hg_{0.7}Mn_{0.3}Te}$ and $3.65 \leq P \leq 4.2$; $Q=1.2$; $T_c=6.48$ K for $\mathrm{Cd_{0.7}Mn_{0.3}Te}$. Figure 4 shows one of the best scalings according to activated dynamics in the case of $\mathrm{Hg_{0.7}Mn_{0.3}Te}$. However, in contrast with the power-law scaling, the χ'' data cannot be scaled simultaneously for both compounds for the same values of the critical exponents.

In order to compare our results with those of Ref. 2, it is important to come back to the definition of P and Q.[14] Fisher has argued and it has been experimentally demonstrated that the so-called "$\pi/2$ rule,"

$$\chi''(\omega)=(-\pi/2)d\chi'/d\ln(\omega)\,, \tag{6}$$

holds for systems obeying activated dynamics.[13,14] Whereas it can be easily shown that the scaling relation is formally the same for $\Delta\chi'$ and χ'' in the case of the power-law scaling[7] (which is consistent with the $\pi/2$ rule), the use of this rule yields a different P exponent for $\Delta\chi'$ and χ'' in the case of the activated dynamic scaling. If

$$\Delta\chi'(T,\omega)=t^P G[-t^Q \ln(\omega\tau_0)]\,,$$

then

$$\chi''(T,\omega)=t^{P+Q}G'[-t^Q \ln(\omega\tau_0)]\,. \tag{7}$$

Hence, $P(\chi'')=P(\Delta\chi')+Q$. We have checked that our $\chi''(T,\omega)$ data cannot be scaled for the values $P(\Delta\chi')+Q=1.3(Q=0.65)$ found by Geschwind whatever the choice of T_c. The exponents P and Q compare favorably with those obtained with χ'' data by Malozemoff and Pytte[13] for the $\mathrm{Eu_{0.4}Sr_{0.6}S}$ spin glass ($P=3\pm1$; $Q=0.65\pm0.15$). They are entirely different from those found by Nordblad, Lundgren, and Svedlingh[3] in $\mathrm{Fe_{0.5}Mn_{0.5}TiO_2}$ ($P=0.9\pm0.2$; $Q=0.55\pm0.1$). We stress that these exponents should be universal. It is therefore extremely irritating that one could find such scattered values.

The nonuniversality of the P exponents, even if properly taking into account the fact that $P(\Delta\chi')+Q=P(\chi'')$ added to the inconsistencies observed on other systems, can be taken as an indication that the activated dynamic scaling is inappropriate for our compounds. In contrast, power-law scaling yields values for the dynamic exponent which compare remarkably well with earlier determinations and the internal consistency of the analysis has been in this work successfully verified. We therefore conclude that power-law dynamic scaling is appropriate to the SMSC which appear to behave like conventional spin glasses.

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