PHYSICAL REVIEW B VOLUME 40, NUMBER 10 1 OCTOBER 1989

Equilibrium magnetic fluctuations of a short-range Ising spin glass

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(Received 5 April 1989)

Equilibrium magnetic fluctuations of the short-range Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$ have been investigated in a superconducting quantum interference device (SQUID) magnetometer. At temperatures above the spin-glass temperature the noise power spectra exhibit a $1/f$ dependence at high frequencies and a convergence to plateaus in the low-frequency limit. At temperatures below the spin-glass temperature only a $1/f$ dependence of the noise power spectra is observed. The equilibrium magnetic fluctuations and the out-of-phase component of the ac susceptibility show behavior in accordance with the predictions of the fluctuation-dissipation theorem. The functional form of the relaxation, $R(t) \propto t^{-\alpha} e^{-(t/\tau)^{\beta}}$, successfully used to describe results from Monte Carlo simulations on a three-dimensional Ising spin-glass model, is found to account very well for the observed behavior of the equilibrium magnetic noise.

I. INTRODUCTION

Spin-glass relaxation in the vicinity of the spin-glass temperature T_g extends over a large time interval. Experimentally, time-dependent magnetization and ac susceptibility measurements are frequently used to map the relaxation in spin glasses. Together, they offer the possibility of covering more than nine decades in time[1] of the spin-glass relaxation. Another important tool used to investigate the low-frequency behavior of the spin-glass dynamics is the measurements of the *equilibrium magnetic noise*. A complication with the ac susceptibility and time-dependent magnetization measurements is the nonlinear field dependence of the relaxation close to T_g. In contrast, measurements of the equilibrium magnetic noise are performed in zero field; i.e., the nonlinearities with fields are effectively avoided. Experiments of this kind have been reported by, e.g., Reim *et al.*,[2] Refregier *et al.*,[3] and Alba *et al.*[4] A general observation in these measurements was the $1/f$ character of the noise power spectra, both below and above the spin-glass temperature. In contrast, in a recent work by Svedlindh *et al.*,[5] a markedly different behavior of the noise power at temperatures above T_g was reported. In the low-frequency limit it was then shown that the noise power spectra converged towards constant values.

In this paper we report equilibrium magnetic noise measurements on the short-range Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$. This system has been shown to be a good model system for a three-dimensional (3D) Ising spin glass.[1,6] At temperatures above the spin-glass temperature the characteristics of the noise power spectra exhibit a high-frequency regime where the spectra display a $1/f$-like dependence and a low-frequency regime where the noise power levels off to constant noise values (plateaus) with decreasing frequency. The level of the plateaus increases rapidly as the temperature approaches T_g from above. At lower temperatures the noise power spectra only display a $1/f$-like dependence. Extensive measurements of the out-of-phase component of the ac susceptibility are shown to be in good agreement with the observed behavior of the equilibrium magnetic noise and unambiguously confirms the validity of the fluctuation-dissipation theorem in Ising spin-glass systems. Also, the observed behavior is consistent with the functional form used in Monte Carlo simulations[7] of a 3D Ising spin glass to describe the time dependence of the spin-correlation function.

II. EXPERIMENTAL

The measurements were performed on a single crystal of $Fe_{0.5}Mn_{0.5}TiO_3$ in the shape of a parallelepiped, $2\times2\times5$ mm^3, using a superconductor quantum interference device (SQUID) magnetometer. The crystal was centered in a first-order gradiometer pickup coil, connected to the signal coil of the SQUID. Layers of μ metal were used to screen the sample from the earth magnetic field. To further reduce the residual magnetic field, a compensating field was generated using a superconducting magnet operating in persistant mode, resulting in a residual magnetic field of less than 10^{-3} G. The temperature was measured using a copper resistance thermometer giving a long-time stability of better than 0.1 mK. At constant temperature the SQUID signal was sampled at a given rate. By varying the sampling frequency, 0.2 Hz $< f_s <$ 200 Hz, a frequency interval of (5×10^{-3}) Hz $< f <$ 10^2 Hz of the noise power spectra was covered. For each sampling frequency an antialiasing filter, with a cutoff frequency set equal to the Nyqvist frequency $(f_s/2)$, was used. The data were transformed by a standard fast Fourier transform (FFT) algorithm to get the noise power spectra.

III. RESULTS

A. ac susceptibility

In order to investigate the validity of the fluctuation dissipation theorem, ac susceptibility data are taken from a previous study.[1] These measurements were made in the frequency interval 10^{-3} Hz $<\omega<2\pi<5\times10^4$ Hz. The sample was stepwise cooled in a small sinusoidal field (≈ 0.1 G). At constant temperature, the in-phase $\chi'(\omega)$ and the out-of-phase $\chi''(\omega)$ components of the complex susceptibility $\chi(\omega)=\chi'(\omega)+i\chi''(\omega)$ were simultaneously detected.

Figure 1 shows $\log_{10}[\chi''(\omega)]$ versus $\log_{10}(\omega)$ at different temperatures in the vicinity of the spin-glass temperature, $T_g=20.9$ K (this value of T_g has been previously determined through dynamic scaling analyses[1]). In Fig. 1(a), the $\chi''(\omega)$ curves exhibit different characteristics at low and at high frequencies. At low frequencies,

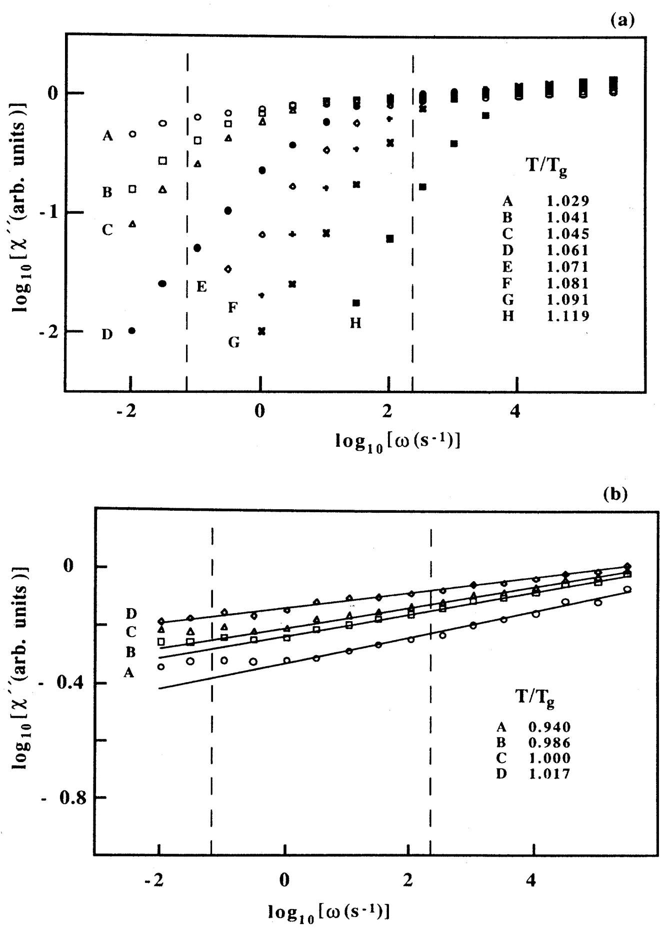

FIG. 1. $\log_{10}[\chi''(\omega)]$ from ac susceptibility measurements vs $\log_{10}(\omega)$ for $Fe_{0.5}Mn_{0.5}TiO_3$. The dashed lines mark the frequency interval covered by the magnetic noise measurements. (a) The different curves refer to temperatures above T_g ($T_g=20.9$ K). (b) The different curves refer to temperatures close to and below T_g. The slope of the curves α' are $T/T_g=0.940$, $\alpha'=0.045$; $T/T_g=0.986$, $\alpha'=0.04$; $T/T_g=1.000$, $\alpha'=0.04$; $T/T_g=1.017$, $\alpha'=0.025$.

where $\chi''(\omega)$ approaches zero, a near $\chi''(\omega)\propto\omega$ dependence is apparent. The width in frequency where such a frequency dependence can be resolved is, however, rather narrow (limited by the experimental resolution). The onset of this regime gradually shifts towards lower frequencies when the temperature approaches T_g from above. At high frequencies, $\chi''(\omega)$ is only weakly frequency dependent. Fitting the high-frequency part of these curves to a power law, $\omega^{\alpha'}$, gives an "effective" exponent $\alpha'\approx 0.02-0.03$. Close to and below T_g, the measured $\chi''(\omega)$ curves only display the weak algebraic dependence on frequency [Fig. 1(b)] and an approach towards zero is never attained at the experimental frequencies. Fitting these curves to a power law gives an increase of α' from 0.03 to 0.05 when the temperature is decreased through T_g. Below T_g, in the measured temperature range ($0.9<T/T_g<1.0$), α' is approximately constant. At the lowest frequencies of the $\chi''(\omega)$ curves shown in Fig. 1(b), a clear deviation from the algebraic behavior is noticed. This deviation is due to the aging phenomenon, i.e., the nonequilibrium character of the spin-glass phase which gives rise to a time dependence of the ac susceptibility.[8] At a given temperature $\chi''(\omega)$ decreases with time and the equilibrium value is *only* obtained after a very long wait time[8,9] at this temperature. At the lowest frequencies used in our ac susceptibility measurements, this requirement is never fulfilled and thus the measured $\chi''(\omega)$ is larger than the equilibrium value. We estimate that $\chi''(\omega)$ is about 20% larger than the equilibrium value at the lowest frequencies and temperatures shown in Fig. 1(b).

B. Magnetic noise

The fluctuation dissipation theorem[10] yields the following relation between the magnetic noise power, $S(\omega)$, and $\chi''(\omega)$:

$$S(\omega)=4kT\frac{\chi''(\omega)}{\omega},\quad \omega=2\pi f\ , \tag{1}$$

where k is the Boltzmann constant. If the fluctuation dissipation theorem holds, the behavior of $\chi''(\omega)$ implies certain characteristics of the noise power spectra. At temperatures well above T_g, a low-frequency regime with frequency independent noise levels [$S(\omega)=$const] and a high-frequency regime where the noise power follows a $1/f$-like dependence [$S(\omega)\propto\omega^{-1+\alpha'}$] are expected. Closer to and below T_g, only the $1/f$ character of the noise power spectra is expected. Complementary measurements of $\chi''(\omega)$ and $S(\omega)$ on the insulating spin-glass $CdIr_{0.3}Cr_{1.7}S_4$ by Alba *et al.*[4] have shown that the fluctuation dissipation theorem is well obeyed at temperatures below T_g.

Figure 2(a) shows the SQUID signal versus time. The different curves are recorded at three different temperatures. The figure illustrates the dramatic increase of the equilibrium magnetic fluctuations as T_g is approached from above. This behavior is consistent with the observed increase of $\chi''(\omega)$ in the frequency interval covered by the noise measurements (see Fig. 1). It is important to carefully check the noise data also in time domain since any small jump or drift occurring during sampling of the SQUID signal results in extraneous low-frequency contributions to the noise power spectra. Figure 2(b) shows Fourier transformed data at the corresponding temperatures. Each curve is an average of at least ten different noise power spectra at the same temperature. At the highest temperature shown ($T/T_g = 1.15$), the magnetic noise spectrum is indistinguishable from the background noise spectrum of the SQUID magnetometer, as measured in an empty coil experiment. This is an important and fundamental result that should be obtained in noise measurements on spin glasses since there are no low-frequency relaxation phenomena at temperatures well above T_g that can give any resolvable contribution to the noise power spectra. At $T/T_g = 1.061$ the spectrum converges to a constant value in the low-frequency limit which is consistent with the observed behavior of $\chi''(\omega)$ at the same temperature [see Fig. 1(a)]. At T_g, the noise power spectrum is $1/f$ like which accords with the weak algebraic frequency dependence of $\chi''(\omega)$ at this temperature [see Fig. 1(b)].

Figure 3(a) shows noise power spectra $S(\omega)$ (open symbols) at some temperatures above T_g. The empty coil noise power spectrum has been subtracted from these spectra. The figure illustrates the occurrence of plateaus

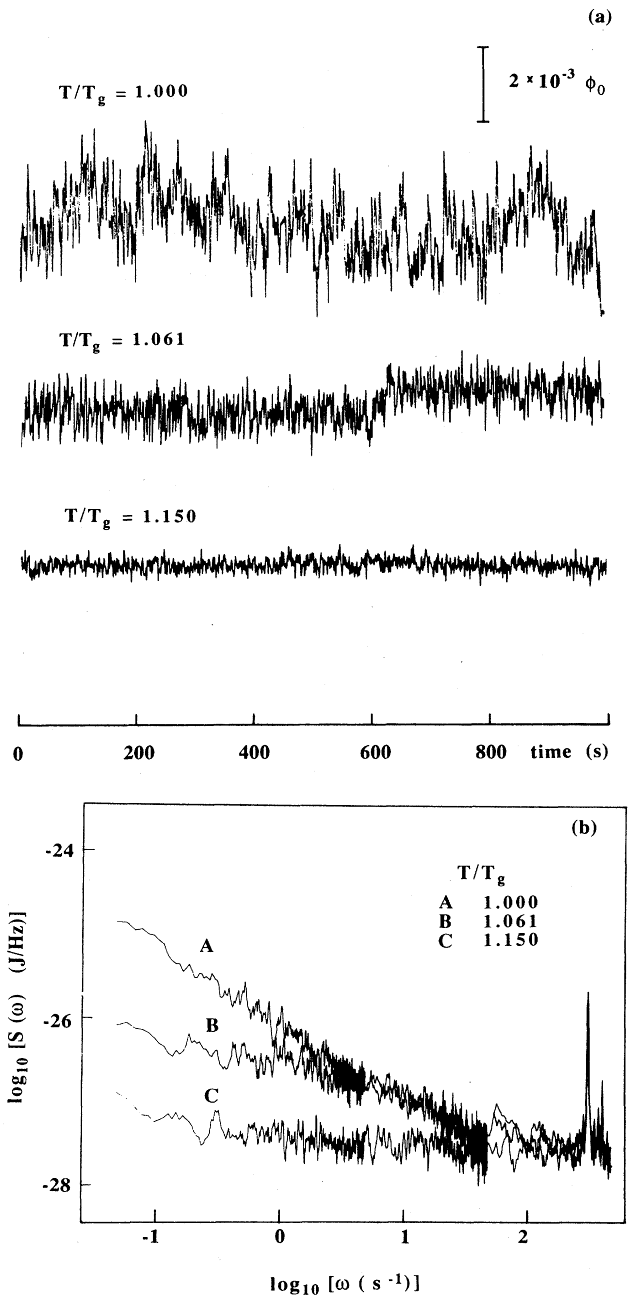

FIG. 2. (a) Magnetic-flux noise as a function of time for $Fe_{0.5}Mn_{0.5}TiO_3$. The different curves refer to different temperatures close to T_g. A relative flux change in the SQUID sensor of $2\times10^{-3}\phi_0$ ($\phi_0 = 2.07\times10^{-15}$ V s) is indicated. The sampling frequency is $f_s = 2$ Hz and an antialiasing filter of 1 Hz is used. (b) Magnetic noise power $S(\omega)$ as detected in the signal coil of the SQUID vs ω in a log-log diagram. The different curves refer to the same temperatures as in (a).

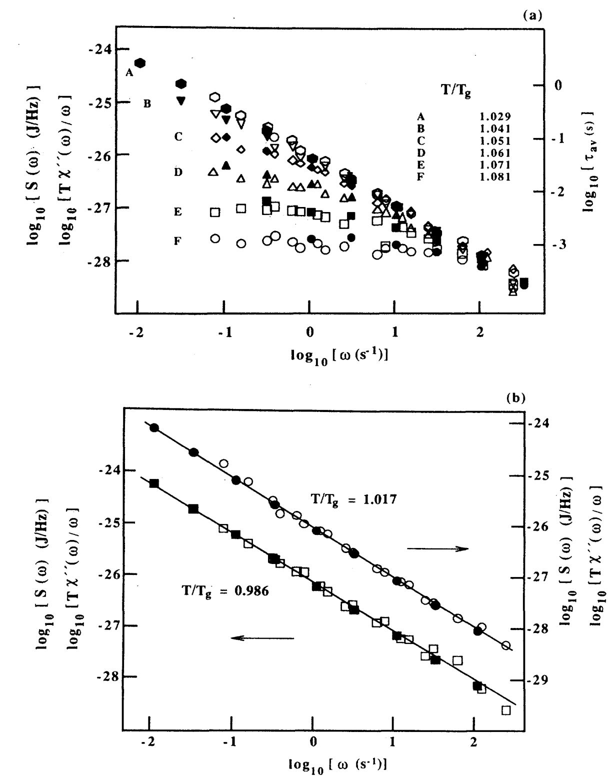

FIG. 3. $\log_{10}[S(\omega)]$ from magnetic noise measurements (open symbols) and $\log_{10}[T\chi''(\omega)/\omega]$ from ac susceptibility measurements (solid symbols) vs $\log_{10}(\omega)$ for $Fe_{0.5}Mn_{0.5}TiO_3$. The spectra have been obtained by averaging raw data over logarithmic frequency increments. (a) The different curves correspond to different temperatures above T_g. The right-hand scale indicates the average relaxation time τ_{av}, defined through Eq. (2). (b) The different curves correspond to temperatures close to and below T_g. The upper curve, $T/T_g = 1.017$, corresponds to the right-hand side scale and the lower curve, $T/T_g = 0.986$, to the left-hand side scale. The slope of the noise power spectrum, $-1+\alpha'$, is at both temperatures ≈ 0.96.

in the low-frequency limit and shows how the level of the plateaus sharply increases when the spin-glass temperature is approached from above. Also shown in Fig. 3(a) are $T\chi''(\omega)/\omega$ data from the ac susceptibility measurements (solid symbols). The proportionality constant, between the ac susceptibility and noise data, was determined by scaling the data at $T/T_g=1.029$. The excellent agreement between the two sets of data, at all temperatures shown in Fig. 3(a), confirms the validity of the fluctuation-dissipation theorem [Eq. (1)] for $Fe_{0.5}Mn_{0.5}TiO_3$. Since $S(\omega)/\chi''(\omega)\propto 1/\omega$, with decreasing frequency magnetic noise measurements become increasingly better than ac susceptibility measurements as a probe of the spin-glass dynamics. In addition, nonlinear field effects, which unavoidably affect the low-frequency $\chi''(\omega)$ data close to T_g,[11] can be effectively avoided in magnetic noise measurements. Using a SQUID sensor with a lower white noise level and by using a higher-order gradiometer, the quality of the present measurements can be improved considerably. Then, equilibrium magnetic noise measurements have the potential of becoming a nearly ideal tool to investigate the spin-glass dynamics in the zero-frequency and zero-field limits. The level of the plateaus in Fig. 3(a) can be related to the average relaxation time of the spin system, through[1,7]

$$\tau_{\mathrm{av}}=[1/\chi'(0)]\lim_{\omega\to 0}[\chi''(\omega)/\omega]\propto \lim_{\omega\to 0} S(\omega)\ , \tag{2}$$

where the second relation is obtained by assuming $\chi'(0)\propto 1/T$. The right-hand side scale in Fig. 3(a) indicates τ_{av}. The sharp increase of these levels is a manifestation of the rapid slowing down of the spin-glass dynamics when T_g is approached. Furthermore, at a given temperature the noise spectra start to deviate from a $1/f$-like dependence at $\omega\approx 1/\tau_{\mathrm{av}}$.

Figure 3(b) shows $\log_{10}[S(\omega)]$ and $\log_{10}[T\chi''(\omega)/\omega]$ versus $\log_{10}(\omega)$ at two temperatures close to T_g. Both of these spectra display a $1/f$-like dependence. Fitting the $S(\omega)$ curves to a $\omega^{-1+\alpha'}$ law the extracted effective exponent is at both temperatures $\alpha'\approx 0.04$, which is in good agreement with the exponents extracted using $\chi''(\omega)$ data at the same temperatures. One should expect an effect of aging on the noise experiments at $T<T_g$, appearing as nonstationary excess noise in the low-frequency part of the noise power spectra. However, the magnitude of the excess noise [estimated to about 0.05 decades on a logarithmic scale at $\omega/2\pi=10^{-2}$ Hz, cf. Fig. 1(b)] is too small to be resolved in these measurements.

IV. DISCUSSION

The relaxation function $R(t)$ of a magnetic system describes the relaxation of the magnetization towards equilibrium, i.e., $R(t)=M_{\mathrm{eq}}-M(t)$, where $M_{\mathrm{eq}}=M(t\to\infty)$. The response function, $f(t)$, is defined as the time derivative of the relaxation function $-dR(t)/dt$ and the complex susceptibility $\chi(\omega)$ is given by the Fourier transform of the response function.[12] The imaginary part, $\chi''(\omega)$, of the complex susceptibility is then given by the sine transform of $f(t)$, through

$$\chi''(\omega)=\frac{1}{H}\int_0^\infty \frac{dM(t)}{dt}\sin(\omega t)dt\ . \tag{3}$$

It has been found that the relaxation function of $Fe_{0.5}Mn_{0.5}TiO_3$ (Ref. 1) and the dynamic spin-correlation function, $q(t)$, of a 3D Ising spin-glass model,[7] both can be described by the following empirical formula:

$$R(t)=A\frac{\exp[-(t/\tau)^\beta]}{t^\alpha}\ , \tag{4}$$

where A, α, τ, and β are temperature-dependent parameters. The power-law term in Eq. (4) describes the relaxation at short times, while the stretched-exponential term governs the approach towards zero at long times. At a particular temperature, a crossover between the short- and long-time behaviors of the relaxation occurs at $t\approx\tau$ and thus τ can be interpreted as a relaxation time of the spin glass. The extracted values of τ, previously determined from time-dependent magnetization and ac susceptibility measurements,[1] sharply increase as the spin-glass temperature is approached from above and exhibit a very similar temperature dependence as the average relaxation time [Eq. (2)]. At temperatures below T_g and at dynamic equilibrium, both Monte Carlo (MC) simulations[7] and experiments on $Fe_{0.5}Mn_{0.5}TiO_3$ (Refs. 1, 6, and 13) indicate that a simple power law describes the spin-glass relaxation function, i.e., $R(t)\propto t^{-\alpha}$. It should be noted that if $\chi''(\omega)$ is fitted to a power law in a limited frequency range the effective "exponent" α' is not equal to α appearing in the expression for $R(t)$. It is only at temperatures below T_g, where $R(t)$ is described by a simple power law, that the two exponents are equal. At temperatures above T_g, the exponent α is found to be about three times larger than α' extracted from $\chi''(\omega)$ data. Assuming that Eq. (4) gives a correct description of $R(t)$ and by using Eq. (3) to calculate $\chi''(\omega)$, this discrepancy is due to the influence of the stretched exponential term in Eq. (4) which in the frequency region $\log_{10}(\omega)>\log_{10}(1/\tau)$ flattens the appearance of $\chi''(\omega)$, and thereby yields a value of the effective exponent α' that is lower than α.

Using the functional form of Eq. (4), it is possible to calculate $\chi''(\omega)$ from Eq. (3) by means of numerical integration. If Eq. (4) gives a correct description of $R(t)$, then the calculated $\chi''(\omega)$ curves should display a $\chi''(\omega)\propto\omega$ dependence in the low-frequency limit. It can be shown that this requires that the relaxation function fulfills the following condition:

$$\int_0^\infty t\frac{dR(t)}{dt}dt=Q\ , \tag{5}$$

where Q has a finite value. Applying the functional form of $R(t)$ shown in Eq. (4) and by using values of the parameters α and β found in measurements on $Fe_{0.5}Mn_{0.5}TiO_3$ (Ref. 1) and in MC simulations on a 3D Ising spin-glass model[7] ($0<\alpha<0.5$ and $0<\beta<1$), it can be shown that the condition in Eq. (5) is obeyed.

After transformation to a logarithmic time scale in Eq. (3), numerical integrations were performed; the values of the parameters in Eq. (4) are taken from Ref. 1. The integration was performed over a wide logarithmic time in-

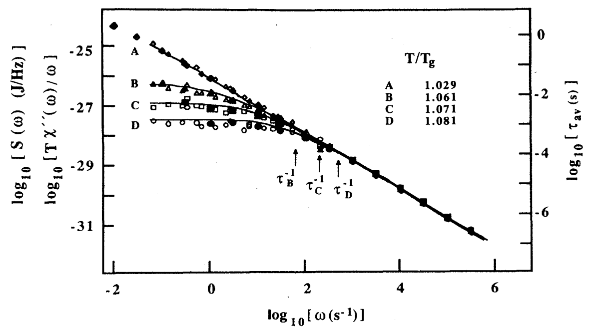

FIG. 4. $\log_{10}[S(\omega)]$ from magnetic noise measurements (open symbols) and $\log_{10}[T\chi''(\omega)/\omega]$ from ac susceptibility measurements (solid symbols) vs $\log_{10}(\omega)$ for $Fe_{0.5}Mn_{0.5}TiO_3$. The right-hand side scale shows τ_{av}, defined through Eq. (2). The solid lines indicate $\log_{10}[T\chi''(\omega)/\omega]$ obtained by numerical integration of Eq. (3), using the functional form of $R(t)$ shown in Eq. (4). The arrows indicate τ values for the three highest temperatures. The values of the different parameters used in the equation of $R(t)$ are $T/T_g=1.029$, $A=3.92$, $\alpha=0.073$, $\tau=7.9$, $\beta=0.25$; $T/T_g=1.061$, $A=2.93$, $\alpha=0.079$, $\tau=0.015$, $\beta=0.31$; $T/T_g=1.071$, $A=2.41$, $\alpha=0.088$, $\tau=0.0050$, $\beta=0.34$; $T/T_g=1.081$, $A=2.09$, $\alpha=0.094$, $\tau=0.0019$, $\beta=0.36$.

terval, the lower limit corresponding to the single spin-flip time τ_0 and the upper limit to a time that is ten decades larger than τ. The results of $T\chi''(\omega)/\omega$ are shown as solid lines in Fig. 4, together with data from ac susceptibility and magnetic noise measurements. The right-hand side scale indicates τ_{av} as defined in Eq. (2). To scale the calculated values of $T\chi''(\omega)/\omega$ to the noise spectra it was sufficient to use the same proportionality constant as was determined previously for the ac susceptibility measurements. The numerically integrated curves agree remarkably well with the experimental results and give further support to the use of the functional form in Eq. (4) to describe the relaxation function in Ising spin-glass systems. The breakaway point of the calculated $T\chi''(\omega)/\omega$ curves from a $1/f$ dependence occurs roughly at $1/\tau$ (indicated in the figure by arrows). The plateaus are obtained at a frequency 2–3 decades lower than this.

V. SUMMARY

Our measurements clearly verify the fluctuation-dissipation theorem of short-range Ising spin glasses. It is stressed that to investigate the spin-glass dynamics in the zero-field and zero-frequency limits the equilibrium magnetic noise is superior to other experimental methods. Further, we have outlined in behavior of $\chi''(\omega)$ versus ω in a wide frequency interval. Experimentally, in the high-frequency region, a weak algebraic dependence is found and in the zero-frequency limit, a $\chi''(\omega)\propto\omega$ dependence is observed. In the magnetic noise spectra, these features are the origin of the $1/f$ behavior in the high-frequency region and the convergence to plateaus in the low-frequency limit, respectively. The same qualitative features of the equilibrium magnetic noise spectra have been observed in an amorphous metallic spin glass.[5] Specifically, plateaus were observed in the same range of reduced temperatures as for $Fe_{0.5}Mn_{0.5}TiO_3$, $T/T_g>1.04$. Considering that dynamic scaling analyses on different spin-glass systems have yielded similar results when it concerns the temperature variation of the relaxation times in the vicinity of the spin-glass temperature, we anticipate that plateaus should be observed, in noise measurements on *any spin-glass system* covering a similar frequency interval of the noise power spectra as in this investigation, already at temperatures moderately higher than the spin-glass temperature.

ACKNOWLEDGMENTS

The authors wish to thank Gunnar Berg of the Department of Mathematics, Uppsala University for informative discussions. Financial support from the Swedish Natural Science Research Council (NFR) is acknowledged.

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