No Phase Transition in a Magnetic Field in the Ising Spin Glass $Fe_{0.5}Mn_{0.5}TiO_3$

J. Mattsson, T. Jonsson, and P. Nordblad
Department of Technology, Uppsala University, Box 534, S-751 21 Uppsala, Sweden

H. Aruga Katori
Institute for Solid State Physics, University of Tokyo, Roppongi, Minatu-ku, Tokyo 106, Japan

A. Ito
Ochanomizu University, Department of Physics, Faculty of Science, Bunkyo-ku, Tokyo 112, Japan
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The dynamic scaling behavior of the Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$ in superimposed dc magnetic fields has been studied. The dynamic freezing temperatures $T_f(\omega, H)$ were determined as a function of field $0 \leq H \leq 3.1$ T and frequency $0.017 \leq \omega/2\pi \leq 5100$ Hz. The in-field scaling behavior is strikingly different from the zero-field behavior and is well described by activated dynamics and a correlation length that depends on field as $\xi \propto H^{-\nu_H}$. The results are in agreement with the droplet model predictions and strongly suggest that there is *no* phase transition in a magnetic field in 3D Ising spin glasses.

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For a couple of decades there has been a continuous interest in trying to understand the physical properties of spin glasses [1,2]. The Sherrington-Kirkpatrick mean-field model of spin glasses has been successfully solved [3], but as yet virtually no exact results exist in the physical dimensions two (2D) and three (3D). In the mean-field model the low-temperature spin-glass phase has many thermodynamic states which are unrelated by symmetry. When a magnetic field is applied, the transition temperature decreases with increasing field following the so-called Almeida-Thouless line in the (H, T) plane [4].

An approach to describe low dimensional spin glasses is the droplet model based on renormalization group arguments [5–8]. In dimensions above the lower critical dimension $2 \leq d_l < 3$, the droplet model finds a low-temperature spin-glass phase in zero magnetic field. This low-temperature phase differs substantially from the spin-glass phase of the mean-field picture. In the droplet model there are only two thermodynamic states related to each other by a global spin flip, and there is no phase transition to a spin-glass phase in a magnetic field.

Whether the spin-glass phase of 3D Ising spin glasses really is destroyed by a magnetic field or if a mean-field picture [9–11] may be applicable is not yet settled. Numerical work is not conclusive [12–15] and there is, until now, no answer from experimental studies of spin glasses in a magnetic field.

In this Letter the question of the existence of a phase transition in a magnetic field is addressed by studies of the dynamic susceptibility of the 3D Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$. The main result of the Letter is that scaling analyses of the dynamic susceptibility in the (H, T) plane strongly indicate that there is no spin-glass phase in a finite magnetic field in the 3D Ising spin glass.

$Fe_{0.5}Mn_{0.5}TiO_3$ is regarded as a model system for a 3D Ising spin glass, and its physical properties have been extensively investigated [16–18]. Static and dynamic scaling analysis in the vicinity of the zero-field phase transition temperature $T_g(H = 0)$ have yielded good scaling behaviors with consistent values of the critical exponents [16,17]. The magnetic structure of $Fe_{0.5}Mn_{0.5}TiO_3$ is most conveniently described by a hexagonal unit cell with the spins aligned along the c axis [18]. The spin-glass behavior is due to a random mixture of ferromagnetic and antiferromagnetic nearest neighbor interactions within the hexagonal layers causing bond disorder.

ac susceptibility and dc magnetization measurements were performed on a single crystal of $Fe_{0.5}Mn_{0.5}TiO_3$ in the shape of a rectangular parallelepiped $2 \times 2 \times 4$ mm^3. The long axis of the crystal corresponds to the crystalline c axis, and in all measurements it was mounted parallel to the external dc field and the probing ac field. The amplitude of the ac field was low enough to ensure linear response [19] in the temperature and field regimes where the dynamics limits the susceptibility of $Fe_{0.5}Mn_{0.5}TiO_3$.

The temperature and frequency dependences of the ac susceptibility $\chi(\omega, T, H) = \chi'(\omega, T, H) + i\chi''(\omega, T, H)$ were measured in superimposed external dc magnetic fields $H \leq 5$ T in the frequency interval $15 \leq \omega/2\pi \leq 1000$ Hz, using a Lake-Shore 7225 ac susceptometer. The data were taken stepwise, increasing the temperature in increments of $\approx$0.3 K starting from $T = 5$ K. The ac susceptibility in zero external dc field in a wider frequency interval $0.017 \leq \omega/2\pi \leq 5100$ Hz was measured in a noncommercial low-field SQUID magnetometer. The field-cooled (FC) dc magnetization was measured in a Quantum Design MPMS SQUID magnetometer. The magnetic field $H \leq 5$ T was applied at a temperature well above $T_g(0)$ and the FC magnetization

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was recorded at different fields stepwise, decreasing the temperature at a cooling rate of ~0.1 K/min.

We first consider the dynamic susceptibility in zero magnetic field. In Figs. 1(a) and 1(b) the zero-field susceptibilities $\chi'(\omega, T, H = 0)$ and $\chi''(\omega, T, H = 0)$ are plotted vs T for $0.051 \leq \omega/2\pi \leq 5100$ Hz. Each susceptibility is measured down to the lowest temperature accessible in the noncommercial SQUID magnetometer $T \approx 9$ K. From these data the freezing temperatures $T_f(\omega)$ can be derived [$T_f(\omega)$ being the temperature at which the maximum relaxation time of the system $t_{\max} = 1/\omega$]. In zero field $T_f(\omega)$ is usually determined either as the maximum of $\chi'(\omega)$ or as the inflection point of $\chi''(\omega)$. In both cases the increase of the maximum relaxation time with decreasing temperature can be nicely fit by conventional critical slowing down at a finite transition temperature

$$\frac{t}{t_0} \propto \left(\frac{T_f(\omega) - T_g}{T_g}\right)^{-z\nu}, \qquad (1)$$

with $T_g(0) \approx 20.3$ K, $t_0 \approx 10^{-13}$ sec, and $z\nu = 11$. An extensive analysis of the zero-field dynamics of the same sample of $Fe_{0.5}Mn_{0.5}TiO_3$ has been reported earlier by Gunnarsson *et al.* [16]. In general, scaling analyses of the dynamical susceptibility in zero external field have proven very useful for observing and investigating both the finite temperature phase transition in 3D and the zero temperature phase transition in 2D spin glasses [20].

In Figs. 2(a) and 2(b) $\chi'(\omega, T, H)$ and $\chi''(\omega, T, H)$ are plotted vs T for different external fields H at the frequency $\omega/2\pi = 125$ Hz. In the insets of the figures, $\chi'(\omega, T, H)$ and $\chi''(\omega, T, H)$ are plotted vs T for $\omega/2\pi = 15$, 125, and 1000 Hz and $H = 0$ and 2 T. Both components of χ obey

$$\chi(\omega, T, H) = \begin{cases} \chi(\omega, T, H = 0), & T < T_f(\omega, H), \qquad (2a) \\ \chi(\omega \to 0, T, H), & T > T_f(\omega, H), \qquad (2b) \end{cases}$$

where the frequency and field dependent freezing temperature $T_f(\omega, H)$ is the temperature where the maximum relaxation time of the system $t_{\max} = 1/\omega$ at field H [21]. In other words, the dynamics of the spin glass is unaware of any external field on time scales shorter than $t_{\max}$, whereas on time scales longer than $t_{\max}$ the susceptibility is independent of frequency and governed by H. This is an important feature of spin-glass dynamics in

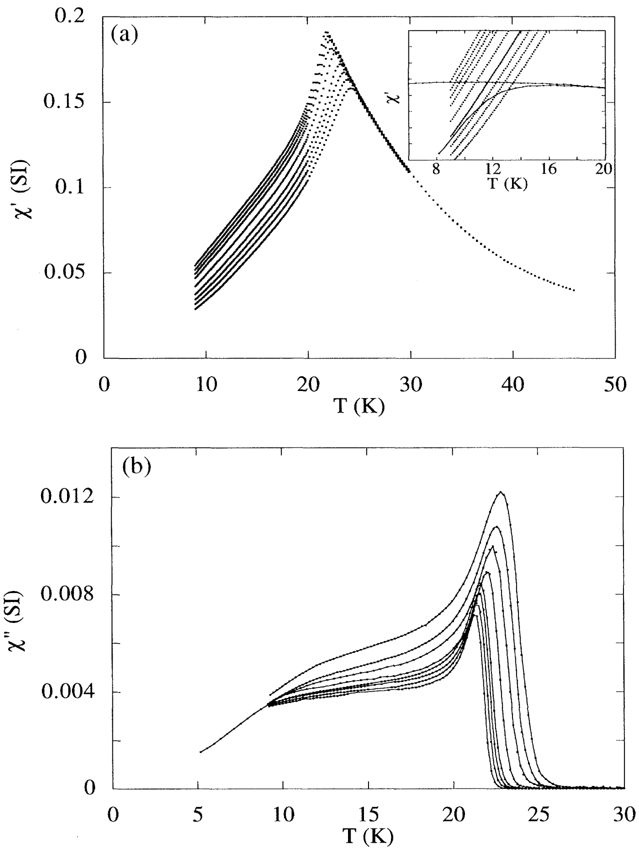

FIG. 1. The ac susceptibility of $Fe_{0.5}Mn_{0.5}TiO_3$ at different frequencies and zero dc field. (a) The in-phase component χ' at $\omega/2\pi =$ (bottom to top) 5100, 1700, 510, 125, 15, 1.7, 0.51, 0.17, and 0.051 Hz. The inset shows a low-temperature cut from the figure where also the 125 Hz curve and FC curve in an applied field of 2 T has been included. (b) The out-of-phase component χ'' at $\omega/2\pi =$ (top to bottom) 1700, 510, 125, 15, 1.7, 0.51, 0.17, and 0.051 Hz.

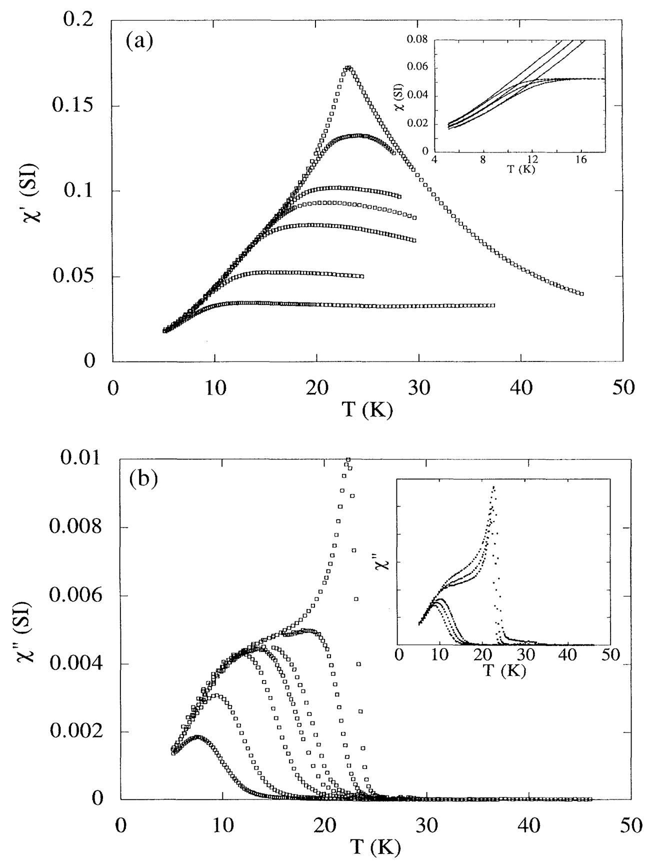

FIG. 2. The ac susceptibility of $Fe_{0.5}Mn_{0.5}TiO_3$ at frequency $\omega/2\pi = 125$ Hz and different superimposed dc fields: (top to bottom) 0, 0.1, 0.4, 0.6, 1, 2, and 3 T. The insets show the low-temperature part of $\chi(\omega, T, H)$ at three different frequencies, 15, 125, and 1000 Hz in external dc fields of 0 and 2 T. (a) χ' and (b) χ''.

a magnetic field [19]. One crucial consequence of this behavior is that $T_f(\omega, H)$ equals the temperature where $\chi'(\omega \to 0, T, H) = \chi'(\omega, T, H = 0)$ and hence can be directly determined from ac susceptibility measurements in zero dc field combined with the equilibrium susceptibility in the field H [see inset to Fig. 1(a)]. This also implies that there is no need for special ac susceptibility measurements in external magnetic dc fields to determine $T_f(\omega, H)$ in spin glasses [22].

To determine the equilibrium susceptibility at low temperatures we use the FC magnetization recorded in different magnetic fields. The FC magnetization eventually falls out of equilibrium when the temperature is lowered. This occurs at a temperature where the maximum relaxation time of the system is of the order of the characteristic time scale set by the cooling rate, in our experiments about 100 sec. The differential FC susceptibility was numerically calculated through $\chi_{FC} = \Delta M_{FC}/\Delta H$. It was checked that smaller ΔH and other differentiation methods did not alter χ_{FC} within the experimental accuracy. Futhermore, $\chi_{FC}(T, H)$ coincides with the ac susceptibility data $\chi(\omega, T, H)$ for $T > T_f(\omega, H)$. We conclude that in the range of frequencies and temperatures where we determine $T_f(\omega, H)$, $\chi_{FC}(T, H) \approx \chi'(\omega \to 0, T, H)$. In the inset of Fig. 1(a), part of the data are shown together with $\chi_{FC}(T, H = 2\ \text{T})$ and $\chi'(\omega/2\pi = 125\ \text{Hz}, T, H = 2\ \text{T})$.

In Fig. 3 $T_f(\omega, H)$, derived as the temperature at which $\chi_{FC}(T, H) = \chi'(\omega, T, H = 0)$, is plotted in the (H, T) plane for $0.017 \le \omega/2\pi \le 1700$ Hz. To compare adequately the mean-field and droplet predictions, the frequency dependence of the freezing temperatures was analyzed along different paths in the (H, T) plane. Here we present results along lines of constant H and lines of constant H/T (two such lines are indicated in the inset to Fig. 3). If there is a phase transition at a finite temperature in a magnetic field, the maximum relaxation times along a line of constant H/T are to diverge at a finite temperature. If not, the relaxation times will diverge at $T = H = 0$.

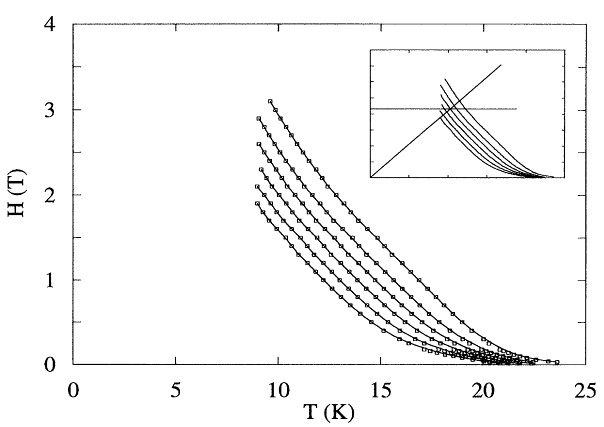

FIG. 3. $T_f(\omega, H)$ contours at $\omega/2\pi =$ (from right to left) 1700, 125, 15, 17, 0.17, and 0.017 Hz. The inset shows two scaling paths $H = 2.2$ T and $H/T = 0.21$ T/K.

At zero and low-enough fields the freezing temperatures at our measurement frequencies are adequately described by conventional critical slowing down [Eq. (2)]. However, at higher fields the dynamics becomes strikingly different and already at quite low fields $H = 0.2$ T fits by Eq. (2) yield unphysical parameters. For example, analyzing the freezing temperatures with $H/T = 0.21$ T/K gives a best fit with $z\nu = 19$, $t_0 \approx 10^{-5}$ sec, and $T_g = 6$ K. Generally, the values of $z\nu$ become unphysically large and the values of t_0 unphysically long, i.e., conventional critical slowing down at a finite temperature does not apply to the in-field dynamics.

In the droplet model the correlation length at low temperatures depends on the magnetic field as [7]

$$\xi_H \propto H^{-\nu_H}, \tag{3}$$

where $\nu_H = (d/2 - \theta)^{-1}$, d is the dimensionality of the sample (here $d = 3$), and θ is an exponent determining the free energy of a droplet excitation. The barrier height depends on length scale L as [7]

$$B = \Delta L^{\psi}. \tag{4}$$

Thermally activated dynamics $B = T \ln(t/t_0)$ then yields

$$\ln(t/t_0) \propto \Delta T_f^{-1}, \qquad \text{for } H = \text{const} \tag{5}$$

and

$$\ln(t/t_0) \propto \Delta T_f^{-(1+\nu_H\psi)}, \qquad \text{for } H/T = \text{const}. \tag{6}$$

The barrier height coefficient $\Delta = \Delta(T, t)$ depends in general on both time and temperature [23]. However, for $T < T_g/2$, Δ is approximately constant [7,23]. We therefore concentrate on this low-temperature region in the scaling analyses and assume $\Delta =$ const [24].

In Fig. 4, $\log_{10}(t_{max})$ is plotted both vs T_f^{-1} for $H = 2.2$ T and vs $T_f^{-1.65}$ for $H/T = 0.21$ T/K (the paths are indicated in Fig. 3). The slowing down along $H = 2.2$ T is nicely described by Eq. (5) giving $t_0 \approx 10^{-14}$ sec and implying that the correlation length is independent of temperature [Eq. (3)] at this constant field. For the slowing down along $H/T = 0.21$ T/K a best fit by Eq. (6) is plotted in Fig. 4 and obtained with $\nu_H\psi = 0.65$ and $t_0 \approx 10^{-14}$ sec.

In fact, all freezing temperatures at temperatures below and around $T_g/2$ are well described by the droplet model with a constant Δ and $\nu_H\psi = 0.65$. The exponent θ has a value ≈ 0.25 [2] which yields $\nu_H \approx 0.8$. The value of the barrier height exponent then becomes $\psi \approx 0.8$, which is well within its inferred limits $\theta \le \psi \le d - 1$ [7].

For the dynamics in a magnetic field, we also attempted activated dynamics, including a finite critical temperature which is likely to describe the critical dynamics of the random field Ising model

$$\ln(t/t_0) \propto T_f^{-1}(T_f - T_g)^{-x}. \tag{7}$$

Adopting this relation for the path $H/T = 0.21$ T/K gives a best scaling for $T_g \approx 0$ K, this reducing Eq. (7) to Eq. (6). Similar results are found for other paths $H/T =$ const.

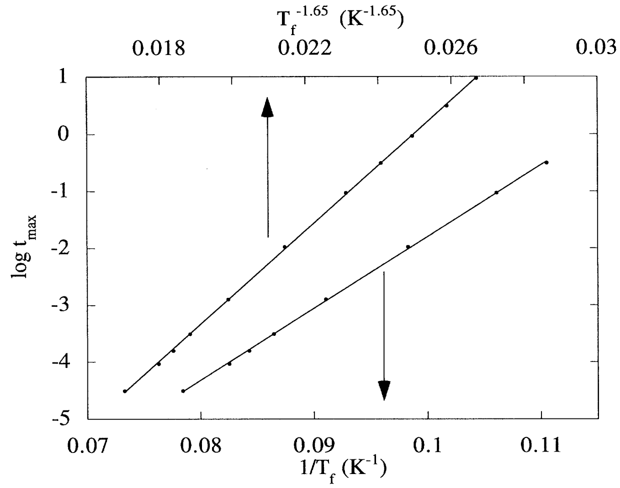

FIG. 4. $\log_{10}(t_{\max})$ vs $1/T_f$ along $H = 2.2$ T and vs $T_f^{-1.65}$ along $H/T = 0.21$ T/K.

The lowest frequency measured is slightly influenced by aging, i.e., the response is influenced by nonequilibrium dynamics. Furthermore, it is possible that $\chi_{FC}(T,H)$ differs somewhat from $\chi'(\omega \to 0, T, H)$ at the freezing temperature of the lowest frequencies. Both these effects are, however, weak and can only marginally affect the values of the exponents derived in our dynamic scaling analyses.

To summarize, we have introduced a method to determine accurately $T_f(\omega, H)$ in spin glasses in magnetic fields and investigated the dynamic scaling behavior of a 3D Ising system in the (H,T) plane. The main result of the Letter is that our dynamic scaling analyses strongly suggest that there is *no* phase transition in a magnetic field in 3D Ising spin glasses, the in-field scaling behavior being well described by the droplet model predictions with $\nu_H \psi = 0.65$.

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[1] K. Binder and A.P. Young, Rev. Mod. Phys. **58**, 801 (1986).

[2] K.H. Fischer and J. Hertz, *Spin Glasses* (Cambridge University Press, Cambridge, 1991).

[3] M. Mézard, G. Parisi, and M.A. Virasoro, *Spin Glass Theory and Beyond* (World Scientific, Singapore, 1987).

[4] J.R.L. de Almeida and D.J. Thouless, J. Phys. A **11**, 983 (1978).

[5] W.L. McMillan, J. Phys. C **17**, 3179 (1984).

[6] A.J. Bray and M.A. Moore, Phys. Rev. Lett. **58**, 57 (1987).

[7] D.S. Fisher and D.A. Huse, Phys. Rev. B **38**, 373 (1988); **38**, 386 (1988).

[8] M. Nifle and H. Hilhorst, Phys. Rev. Lett. **68**, 2992 (1992).

[9] J.E. Green, M.A. Moore, and A.J. Bray, J. Phys. C **16**, L815 (1983).

[10] T. Temesvári, I. Kondor, and C. De Dominicis, J. Phys. A **21**, 1145 (1988).

[11] C. De Dominicis and I. Kondor, J. Phys. A **22**, L743 (1989).

[12] S. Caracciolo, G. Parisi, S. Patarnello, and N. Sourlas, Europhys. Lett. **11**, 783 (1990); J. Phys. (Paris) **51**, 1877 (1990); J. Phys. I (France) **1**, 627 (1991); D. Badoni, J.C. Ciria, G. Parisi, F. Ritort, J. Pech, and J.J. Ruiz-Lorenzo, Europhys. Lett. **21**, 495 (1993).

[13] D.A. Huse and D.S. Fisher, J. Phys. I (France) **1**, 621 (1991).

[14] E.R. Grannan and R.E. Hetzel, Phys. Rev. Lett. **67**, 907 (1991).

[15] J.D. Reger, R.N. Bhatt, and A.P. Young, Phys. Rev. Lett. **64**, 1859 (1990).

[16] K. Gunnarsson, P. Svedlindh, P. Nordblad, L. Lundgren, H. Aruga, and A. Ito, Phys. Rev. Lett. **61**, 754 (1988). The same sample of $Fe_{0.5}Mn_{0.5}SiO_3$ was used in this and the present study. The different values of T_g (20.9 and 20.3 K, respectively) arise mainly from different thermometry in the two experiments.

[17] K. Gunnarsson, P. Svedlindh, P. Nordblad, L. Lundgren, H. Aruga, and A. Ito, Phys. Rev. B **43**, 8199 (1991).

[18] A. Ito, H. Aruga, E. Torikai, M. Kikuchi, Y. Syono, and H. Takei, Phys. Rev. Lett. **57**, 483 (1986); A. Ito, H. Aruga, M. Kikuchi, Y. Syono, and H. Takei, Solid State Commun. **66**, 475 (1988).

[19] C. Djurberg, J. Mattsson, and P. Nordblad, Europhys. Lett. **29**, 163 (1995).

[20] C. Dekker, A. Arts, H. De Wijn, A. Duyneveldt, and J. Myodosh, Phys. Rev. Lett. **61**, 1780 (1988); N. Bontemps, J. Rajchenbach, R.V. Chamberlin, and R. Orbach, Phys. Rev. B **30**, 6514 (1984); A.T. Ogielski, Phys. Rev. B **32**, 7384 (1985).

[21] As seen in the insets of Figs. 1 and 2, there is of course a crossover rounding between Eqs. (2a) and (2b) in the vicinity of $T_f(\omega, H)$.

[22] At very low fields the equilibrium susceptibility crosses $\chi'(\omega, T, H = 0)$ where $\chi'(\omega, T, H = 0)$ is rounded because of the vicinity to $T_f(\omega, H = 0)$. For these fields the usual methods to determine T_f, e.g., the inflection point of $\chi''(\omega, T, H)$, should be used. The here introduced definition of T_f in large fields also made four extra decades in frequency available for our in-field dynamic scaling analysis compared to what was achievable using a conventional determination of T_f from direct ac susceptometry date in superimposed fields. It is important to note that the specific definition of T_f employed, inflection point of χ'', the temperature where $\chi''(\omega)$ is first resolvable, or the position of the maximum in $\chi'(\omega)$ (when zero field dynamics are studied) does not significantly alter the values of critical exponents derived in a dynamic scaling analysis and does not add to the difficulty of the question whether there is a finite temperature phase transition or not.

[23] J. Mattsson, J.O. Andersson, and P. Svedlindh, Physica (Amsterdam) **194B–196B**, 305 (1994).

[24] To be able to analyze the dynamics at higher temperatures within the droplet model, predictions for $\Delta(T,t)$ are needed. In the absence of such, we mention only that the results are consistent with a slight decrease of Δ with increasing T and t, which agrees with earlier observations (Ref. [23]).