

Dynamic scaling in spin glasses

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We present neutron spin echo (NSE) results and a revisited analysis of historical data on spin glasses, which reveal a pure power-law time decay of the spin autocorrelation function $s(Q,t) = S(Q,t)/S(Q)$ at the glass temperature T_g . The power law exponent is in excellent agreement with that calculated from dynamic and static critical exponents deduced from macroscopic susceptibility measurements made on a quite different time scale. This scaling relation involving exponents of different physical quantities determined by completely independent experimental methods is stringently verified experimentally in a spin glass. As spin glasses are a subgroup of the vast family of glassy systems also comprising structural glasses and other noncrystalline systems the observed strict critical scaling behavior is important. Above the phase transition the strikingly nonexponential relaxation, best fitted by the Ogielski (power-law times stretched exponential) function, appears as an intrinsic, homogeneous feature of spin glasses.

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The glass transition, characterized by a dramatic slowing down of the dynamics without any noticeable change in the spatial order, is a generic phenomenon seen in systems as different as disordered magnets, polymers, and biological substances. In spite of its universality and of intense experimental and theoretical efforts it is still controversial whether the glass transition in structural glasses is a gradual freezing or a phase transition. The difficulty to identify the nature of the glass transition is due to the absence of an observable order parameter analogous to magnetization in the low-temperature phase, usually a key quantity in the study of phase transitions. This is due to the absence of any static spacial fingerprint; instead, the order parameter appears in the dynamics.^{1,2} In fact, the “snapshot” structure factor $S(Q) = S(Q, t=0)$, which reflects the short and medium range static correlations, shows no essential change when passing from the high-temperature liquid (or paramagnetic) to the low-temperature frozen glass phase. In this situation the observation of dynamic scaling relations, which are the direct consequence of the homogeneity hypothesis in the vicinity of a critical instability³ can reveal the crucial signature of a true phase transition.

In spin glasses, which are the simplest realizations of glassy systems from the experimental as well as from the theoretical point of view, a phase transition is well established.^{4,5} Low frequency dynamic susceptibility as well as the nonlinear part of the static susceptibility follow the scal-

ing relations and the analysis leads to accurate determination of the static critical exponents γ , β , δ , and of the dynamic exponent z .⁶⁻⁹ The verification of scaling relations between certain exponents, determined by completely independent experimental methods, however, was up to now impossible due to the absence of any obvious critical behavior on other physical quantities, such as the specific heat. Here we show that scaling relations can also be verified experimentally in spin glasses. New neutron spin echo (NSE) results and also a revisited analysis of historical data¹⁰⁻¹² show a pure power-law time decay of the spin autocorrelation function $s(Q,t) = S(Q,t)/S(Q, t=0)$ at the glass temperature T_g , with a power law decay exponent which is in excellent agreement with that calculated from the dynamic and static critical exponents deduced from zero and low frequency susceptibility measurements. The interplay between neutron scattering, macroscopic magnetic, hyperfine field measurements, and simulations has always been decisive in understanding spin glasses and these results constitute the strongest experimental evidence yet for a true phase transition with a nonconventional order parameter; they also imply that the prominently nonexponential relaxation is an intrinsic homogeneous feature of spin glasses.

Most of the information about glass transitions comes from the high-temperature unfrozen phase, where thermodynamic equilibrium is easily reached without any long-time drifts and aging phenomena. In spin glasses, where the mag-

netization is always zero, the fundamental parameter is the mean spin autocorrelation function $q(t-t') = \langle \mathbf{S}_i(t) \cdot \mathbf{S}_i(t') \rangle$ where \mathbf{S}_i is the spin at a site i and the average runs over all sites and configurations of the sample. Critical behavior in the paramagnetic phase is seen in the nonlinear susceptibility (or “spin glass susceptibility”).^{6–9} Below T_g the Edwards Anderson order parameter $q(t \rightarrow \infty) = \langle \mathbf{S}_i(t=0) \mathbf{S}_i(t \rightarrow \infty) \rangle$, becomes nonzero.^{1,13} Neutron spin echo spectroscopy measures the scattering function $S(Q, t)$ and after normalization by $S(Q, 0)$ delivers a direct determination of the autocorrelation function $q(t) = s(Q \rightarrow 0, t)$. NSE covers a time domain ranging from 10^{-12} to some 10^{-8} s, i.e., from characteristic microscopic times up to times, which already belong to the “long” time relaxation domain. The first NSE experiment ever performed on a glassy system was made on the reference spin glass CuMn 5% in 1979 (Ref. 10) and the results strongly influenced subsequent thinking on (spin)glass dynamics. It was shown that nonconventional dynamics is not limited to the spin glass phase but also extends into the paramagnetic phase well above T_g . Nonexponential and Q -independent relaxation occurs in a large temperature range up to $2-3 T_g$, which can arguably be identified with the Griffiths phase.¹⁴ For about $T > 1.2 T_g$ the relaxation can be described by a broad distribution of Arrhenius activation energies. Closer to T_g , however, a more dramatic slowing down sets in, which can be interpreted as the footprint of a phase transition with a critical region of usual extent. Here we report a detailed analysis of $s(Q, t)$ around T_g in spin glasses, based on enhanced quality data obtained by using new generation NSE spectrometers.

For an accurate determination of the NSE spectra we chose $\text{Au}_{0.86}\text{Fe}_{0.14}$. AuFe is a classical metallic Heisenberg spin glass with significant local anisotropy¹⁵ and with strong ferromagnetic correlations which amplify the magnetic scattering in the forward direction so improving the ratio between the magnetic signal and all nonmagnetic (structural) contributions, i.e., the signal to noise ratio. The sample was a polycrystalline disc 0.5 mm thick with a diameter of 37 mm prepared by arc melting of the constituents. It was subsequently cold worked, homogenized at 900 °C, annealed a 550 °C and then quenched and kept in liquid nitrogen.¹⁶ Given the vicinity to the percolation threshold x_c (~ 15.5 at. % Fe), above which ferromagnetism sets in,¹⁷ the annealing and quenching procedure was repeated before every series of measurements. The NSE data were collected at the high resolution spectrometer IN15 of ILL¹⁸ at an incoming wavelength of 0.8 nm for $Q=0.4$ and 0.8 nm^{-1} , respectively. These results were supplemented by measurements at the wide angle NSE spectrometer SPAN of BENS¹⁹ at an incoming wavelength of 0.45 nm for $0.6 \text{ nm}^{-1} \leq Q \leq 2.6 \text{ nm}^{-1}$. We used the paramagnetic NSE setup, which directly delivers the magnetic part of the NSE signal and for this reason no background correction was required. All NSE spectra were normalized against the resolution function of the spectrometers, determined with the sample well below T_g , at 2 K, where the spin dynamics is completely frozen. A small part of the sample was taken out for dc susceptibility measurements with a commercial SQUID magnetometer at the HMI and for ac susceptibility

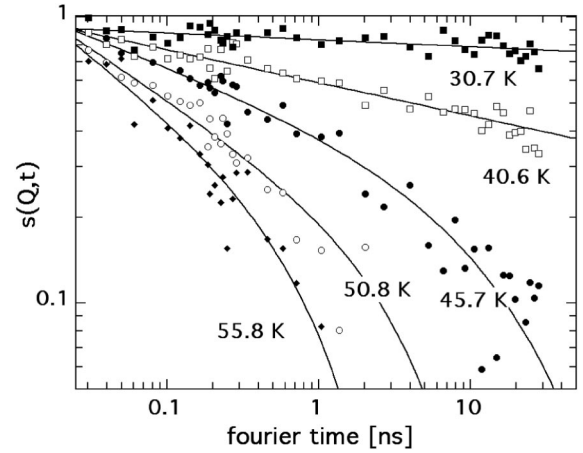


FIG. 1. Temperature dependence of the normalized intermediate scattering function $s(Q, t)$ of $\text{Au}_{0.86}\text{Fe}_{0.14}$. The spectra were collected at $Q=0.4 \text{ nm}^{-1}$ with the neutron spin echo spectrometer IN15 (ILL) for $T=30.7 \text{ K}$ (closed squares), 40.6 K ($\sim T_g$, open squares), 45.7 K (closed circles), 50.8 K (open circles), and 55.8 K (closed rhombs), respectively. The continuous lines are the best fits to the data of a simple power law decay below T_g ($\sim 41 \text{ K}$) and of the Ogielski function above T_g (see text).

measurements, which were made from 10 Hz up to 10 KHz with the MAGLAB setup at the Physics and Astronomy Department, University of Leeds. The spin glass temperature of $T_g = 41.0 \pm 0.3 \text{ K}$ was determined from the maximum of the static susceptibility.

The normalized intermediate scattering function $s(Q, t)$ of $\text{Au}_{0.86}\text{Fe}_{0.14}$ at $Q=0.4 \text{ nm}^{-1}$ is shown in Fig. 1 plotted in a log-log scale. The spectra span a dynamic range of three orders of magnitude and by combining spectra collected at two wavelengths on IN15 and SPAN the time domain of the observation is extended up to almost four decades (Fig. 2). The time dependence of the experimental $s(Q, t)$ is impressively similar to that of the numerical $q(t)$ found in large scale Ising spin glass simulations, which revealed the existence of a phase transition in three-dimensional Ising spin glasses.²⁰ From quite general scaling arguments,²¹ at a continuous phase transition relaxation must be of the form $t^{-x} f[t/\tau(T)]$ where $\tau(T)$ diverges as $(T-T_g)^{-z}$, and f is a nonuniversal function to be determined for each system. For a spin glass the power law exponent x is related to the standard critical exponents through $x = (d-2+\eta)/2z$.²⁰ Here η is the Fisher or “anomalous dimension” exponent and z is again the dynamical exponent. The Ising simulations showed that, as T_g is approached from above, $q(t)$ is strongly nonexponential. Ogielski chose to represent f by the stretched exponential or KWW function, familiar in fragile glass dynamics. Excellent fits were obtained with $q(t) \propto t^{-x} \times \exp\{-[t/\tau(T)]^\beta\}$ and T dependent τ and β . (The KWW β is not to be confused with the critical exponent β .) He and others found a temperature dependent β tending to near 1/3 at T_g and increasing with T .^{2,20,22,23} The most important point for our data analysis is that precisely at T_g dynamic scaling predicts a pure power law decay for the autocorrelation function $q(t) \propto t^{-x}$. This rule is quite general; its functional form does not depend on details such as the Ising character of the

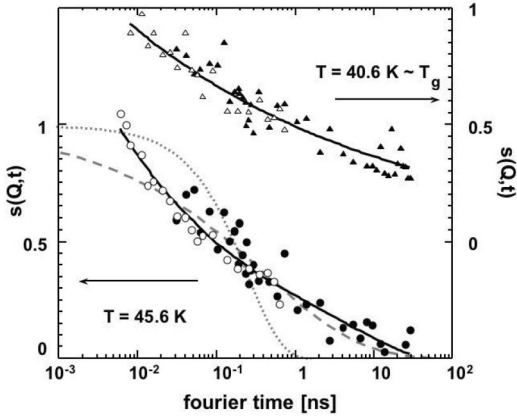


FIG. 2. NSE spectra of $\text{Au}_{0.86}\text{Fe}_{0.14}$ collected at $Q = 0.08 \text{ \AA}^{-1}$ at the ILL spectrometer IN15 (full symbols) and at the BENS spectrometer SPAN (open symbols) plotted in a lin-log scale. The circles are measured at 45.6 K and the triangles at 40.6 K, respectively. For the sake of clarity the data sets corresponding to each of the temperatures have been shifted with respect to each other in the vertical scale. The data at 40.6 K are fitted to a simple power law (continuous line). The continuous line through the 45.6 K data represents the fit of an Ogielski function (see text). The dashed and dotted curves correspond to a stretched exponential and a simple exponential decay, respectively.

spins but the value of x depends on the exponents for the particular system under study. Scaling therefore gives a unique opportunity for describing the NSE spectra at T_g in terms of exponents determined by completely independent dynamic and nonlinear macroscopic susceptibility measurements.

As seen on Fig. 1, for $T \leq T_g$ we found $s(Q,t) \propto t^{-x}$ and the lines represent the best fits to the data with $x = 0.116 \pm 0.007$ and 0.025 ± 0.005 at $T = 40.6 (\sim T_g)$ and 30.7 K, respectively. Above T_g the relaxation is also strongly nonexponential. As shown in Fig. 2, pure exponential as well as stretched exponential decay (without the power-law prefactor) can be definitively ruled out at all T in the range studied. In fact at 45.6 K, the fit with the Ogielski function leads to $\chi^2 = 0.28$, which is significantly lower than $\chi^2 = 0.55$, the value obtained for the stretched exponential and $\chi^2 = 1.67$ for the simple exponential, respectively. The power law part of the Ogielski function, which holds at short times, describes the main part of the relaxation above T_g and the spectra of Fig. 1 lead to an accurate determination of the effective power law exponent x as function of T . On the other hand, the stretched exponential only influences the tail of the relaxation so the parameters $\tau(T)$ and β are obtained here to low accuracy: $\tau(T) \approx 0.3 \text{ ns}$ and $\beta \approx 1$ at 55.8 and 50.8 K whereas $\tau(T) \approx 22 \text{ ns}$ and $\beta \approx 0.66$ at 45.7 K. In similar systems, muon spin depolarization measurements which are sensitive to the time range from 10 ns up to about 50 μs (i.e., in the range where the relaxation above T_g is mainly described by the stretched exponential) showed that in fact β approaches about $1/3$ at T_g ^{24,25} as expected by the simulations. Susceptibility measurements very close to T_g in an Ising spin glass show a similar limiting value of β .²⁷ Furthermore, recent magnetic-field-dependent muon spin depolar-

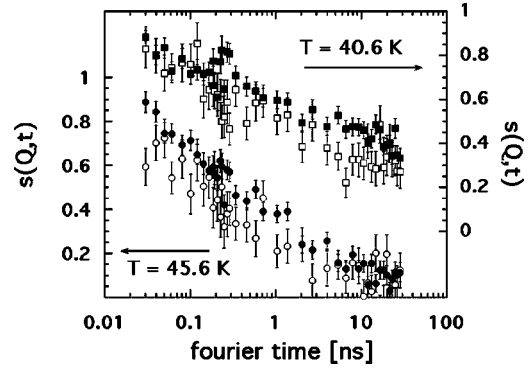


FIG. 3. $s(Q,t)$ of $\text{Au}_{0.86}\text{Fe}_{0.14}$ measured at 0.04 \AA^{-1} (close symbols) and 0.08 \AA^{-1} (open symbols) for 45.6 and 40.6 K, respectively. For the sake of clarity the data sets corresponding to each of the temperatures have been shifted with respect to each other in the vertical scale.

ization measurements on several AgMn spin glasses, analyzed assuming an Ogielski-like decay of the correlations, lead to values of x tending to ~ 0.15 at T_g in agreement with our results.²⁵

The large dynamic range covered by our data and their accuracy allow us to distinguish between a simple stretched exponential decay, the Ogielski function with $\beta = 1$ (power law times simple exponential), and the full Ogielski function; the full function is needed to give an acceptable fit of the data over the whole time and temperature range. In spin glasses the normalized $s(Q,t)$ does not vary with Q , in dramatic contrast to the strong Q dependence of the dynamics in ferromagnets. This remarkable Q independence of the relaxation in spin glasses¹⁰ implies that the NSE $s(Q,t)$ can be identified with $q(t)$ and analyzed in the frame of dynamic scaling. The NSE spectra of $\text{Au}_{0.86}\text{Fe}_{0.14}$ collected on IN15 for $Q = 0.4$ and 0.8 nm^{-1} and on SPAN for $0.6 \text{ nm}^{-1} \leq Q \leq 2.6 \text{ nm}^{-1}$ confirmed this behavior. Figure 3 compares, in a lin-log plot, spectra collected at 45.6 and 40.6 K for $Q = 0.04$ and 0.08 \AA^{-1} , respectively. The data almost overlap although the Q values differ by a factor of 2 and the magnetic intensity decreases by almost a factor of 3, which explains the larger error bars of the data set at 0.08 \AA^{-1} . The Q independence of $s(Q,t)$ was an important check of the high quality of our samples in a concentration range close to ferromagnetism. It also implies that $s(Q,t)$ can be directly related to the macroscopic ac susceptibility $\chi(Q=0, \omega) = S(Q=0)[1 - s(Q=0, t)]/kT$.¹¹ The meaning of this equation is quite simple: $s(Q,t)$ is the fraction of the total magnetic response $S(Q)$ which does not relax before the time t , in other words $s(Q,t)$ is the part of $S(Q)$ which cannot respond to a driving field of frequency $1/t$. Figure 4 shows the NSE spectra for $T \leq T_g$ combined with $s(Q,t)$ values deduced from macroscopic dynamic (ac) susceptibility measurements on the same sample. The data follow the power law decay over an impressively large range of at least nine orders of magnitude in time.

The impressive similarity between the experimental $s(Q,t)$ and the decay of $q(t)$ found in large scale simulations and more particularly the simple power law decay found at

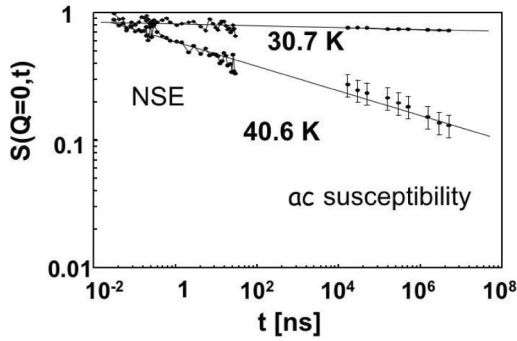


FIG. 4. Combined NSE spectra with $s(Q=0,t)$ values deduced from macroscopic ac susceptibility measurements below T_g . Also in the close vicinity of T_g , the pure power law decay of $s(Q,t)$ holds over an impressively large dynamic range of more than nine orders of magnitude in time, from the microscopic to the macroscopic times.

and below T_g incited us to revisit the historical CuMn 5% data. Below T_g , the spectra of CuMn 5% were combined with macroscopic dynamic (ac) susceptibility measurements and thus covered more than 9 orders of magnitude in time down to the microscopic time scale.¹¹ These data, plotted now on a log-log scale, also reveal a power law decay below T_g over an impressively large range of at least nine orders of magnitude in time as shown in Fig. 5. Above T_g , which is at 27.5 K, the power law holds only at short times and the decay can be described by the Ogielski function with $\beta \approx 1$ just as in AuFe 14%.

The values of the exponent x , plotted versus the reduced temperature T/T_g in Fig. 6, are similar for both metallic systems. This confirms the similarity of the dynamic behavior of AuFe, AgMn, and CuMn systems around T_g , pointed out by Uemura after comparing the NSE spectra of CuMn 5% with μ SR spectra of AgMn and AuFe.²⁶ As already mentioned, dynamic scaling relates the value of x at T_g to the static and dynamic critical exponents, which can be determined completely independently from nonlinear and ac macroscopic susceptibility measurements. Well established values of these

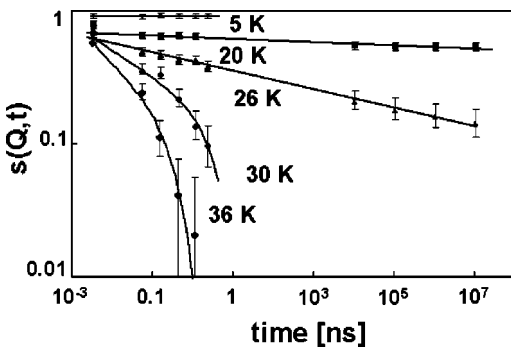


FIG. 5. Revisited analysis of the historical $s(Q,t)$ data on CuMn 5% [Eq. (5)]. Below T_g (~ 27.5 K) the NSE spectra were combined with values calculated from the dynamic susceptibility ($10^4 \geq t \geq 10^7$ ns). The data are plotted in a log-log scale and the continuous lines correspond to a simple power law decay below T_g and to the Ogielski function above T_g .

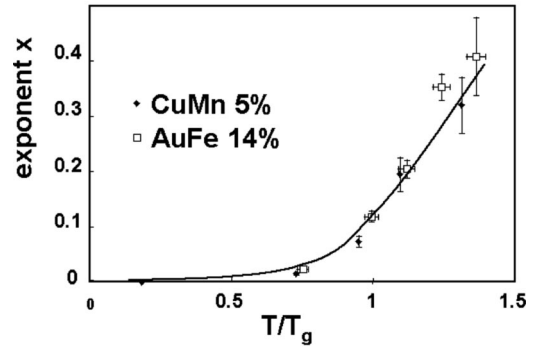


FIG. 6. Temperature dependence of the exponent x obtained by fitting the NSE spectra to a simple power law decay below T_g and to the Ogielski function above T_g . At T_g $x \sim 0.12$ as predicted by dynamic scaling.

exponents are available for a AgMn spin glass⁶ and for CuMn and AgMn spin glasses doped with Au,⁷ in agreement with more recent data on AuFe 14%. The static exponents lead to $\eta = 2 - \gamma/\nu = 0.23 \pm 0.3$ and the dynamic exponent $z = 5.3 \pm 0.8$ was determined by low frequency susceptibility measurements. From these values we calculate $x = 0.116 \pm 0.026$ in excellent agreement with $x = 0.116 \pm 0.007$, the value we have observed on $\text{Au}_{0.86}\text{Fe}_{0.14}$ at T_g . It is important to note that the value of x in these basically Heisenberg spin glasses is considerably larger than that seen numerically²⁰ or experimentally²⁷ in Ising spin glasses where $x \approx 0.07$.

Our results represent verification of scaling in spin glasses relating quantities of very different natures and measured by different methods on very different time scales, namely, the microscopic time dependence of the autocorrelation function at times between the microscopic time and 10^{-7} s on the one hand, and the macroscopic ac susceptibility at time scales greater than 10^{-3} s together with the static nonlinear susceptibility on the other. This agreement constitutes a most compelling evidence for a true phase transition in spin glasses at T_g . This demonstration of critical relaxation characteristic of a phase transition also implies that the nonexponential relaxation is an intrinsic, homogeneous feature of spin glasses. Similar conclusion was also drawn by Uemura after comparing μ SR, NSE, and dynamic susceptibility data.²⁶ This result is, however, in obvious contrast to the clear evidence for the heterogeneous origin of the nonexponential relaxation in some structural glasses, where also no clear signature of a phase transition could be found.²⁸ The clear evidence for a phase transition with a nonconventional order parameter and for an intrinsic, homogeneous nonexponential relaxation in spin glasses is of particular importance in view of the unsolved central question of the nature of the glass transition in general.

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