

Dynamic susceptibility of a reentrant ferromagnet

K. Jonason, J. Mattsson, and P. Nordblad

Department of Technology, Uppsala University, Box 534, S-751 21 Uppsala, Sweden

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The temperature, frequency, and magnetic-field dependence of the susceptibility of the standard reentrant ferromagnet $(\text{Fe}_{0.20}\text{Ni}_{0.80})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ has been investigated by ac-susceptibility measurements. In general the magnetic response is strongly nonlinear in the magnitude of the applied field h , but a distinct low-field region is found, $h < h_0$, where the dynamic susceptibility does not depend on the magnitude of the field. Thus, ac-susceptibility or zero-field-cooled magnetization measurements in low enough fields, $h < h_0$, allow studies of the inherent zero-field dynamics of reentrant ferromagnets. A dynamic scaling analysis of low-field dynamic susceptibility data near the ferromagnetic to reentrant spin-glass transition indicates a transition to a true spin-glass phase. Magnetic aging, another signature of spin-glass dynamics, is shown to exist in the reentrant spin-glass phase by low-field time-dependent zero-field-cooled magnetization measurements.

I. INTRODUCTION

When a piece of material is cooled down its constituent atoms or molecules become more and more ordered. Some systems, however, become seemingly disordered again when further lowering the temperature. Such a behavior is commonly referred to as reentrance (the system reenters a disordered phase when lowering the temperature). Reentrant behavior was first seen in binary liquid mixtures¹ and it has since then been observed and investigated in a variety of different physical systems, e.g., magnets,² superconductors,³ and liquid crystals.⁴ The usual mechanism for reentrance is the existence of interactions that are capable of lowering the entropy in some hidden way while at the same time reducing the energy of the system. Reentrant magnets differs from most other reentrant systems in that the interactions are quenched and, despite this or maybe because of this, no “hidden” interaction responsible for reentrance has been identified.⁵

Reentrant behavior has been found in a variety of disordered magnetic materials in which there is a competition between spin-glass order and long-range ferromagnetic order, i.e., in systems where there is a majority of ferromagnetic couplings between the individual spins but a sufficiently large number of antiferromagnetic couplings to create substantial frustration. When the temperature is lowered in such a material it exhibits a transition from a paramagnetic (PM) to a ferromagnetic (FM) phase and on further lowering the temperature typical spin glass, commonly called reentrant spin-glass (RSG), behavior appears.⁶

In theory of reentrant magnets, only mean-field models are well understood. In the Parisi solution⁷ of the Sherrington-Kirkpatrick mean-field model (SK model),⁸ ferromagnetic long-range order never disappears when lowering the temperature. However, below a certain temperature in the ferromagnetic phase, replica symmetry is broken, which means that there is a low-temperature “mixed phase” where spin-glass behavior coexists with long-range ferromagnetic order. Thus there is no true reentrance in the SK model (true reentrance implies that the ferromagnetic long-range order disappears at a low-temperature FM-RSG phase transition).

In low-dimensional samples the situation might well be different. Neutron-scattering results on several reentrant ferromagnets have shown that, within experimental resolution, the ferromagnetic Bragg scattering, present in the ferromagnetic phase, disappears at low temperatures.⁹ It is therefore quite generally concluded that true reentrance (the sequence of phases, PM→FM→RSG, when lowering the temperature) may occur in low-dimensional magnets. However, neutron depolarization experiments on reentrant ferromagnets suggest a randomly canted ferromagnet rather than a pure spin glass at low temperatures.¹⁰

II. OBJECTIVES AND METHODS

The distinguishing tool to recognize and characterize a reentrant ferromagnet is low-field susceptibility and magnetization measurements. It is however already difficult to conclusively interpret static and dynamic susceptibility or magnetization results on only a disordered ferromagnet or a pure spin glass and thus even more complicated to decipher the corresponding reentrant transitions, since both the FM and the RSG phase inherently exhibits strong nonlinearities and slow dynamics. It can be quite difficult even to reasonably accurately determine the ferromagnetic transition temperature, as the divergence of the susceptibility at the ferromagnetic transition often is severely smeared due to a strongly nonlinear field dependence. Moreover, the occurrence of reentrant behavior is often indistinguishable from only hysteresis effects within the ferromagnetic phase. To unambiguously establish the inherent static and dynamic properties of the two “ordered phases” and the true nature of a reentrant phase transition is even more involved. In spite of these obstacles, some presumed universal signatures of the temperature dependence of the susceptibility and magnetization of reentrant ferromagnets have been generally adopted,^{2,6,11} unfortunately however, these attributes suffer from being extracted using uncertain (or nonexistent) extrapolations to the zero-field behavior. The main objective of this study is to achieve a comprehensive empiricism on the inherent zero-field dynamic properties of reentrant ferromagnets through the reentrant spin-glass transition.

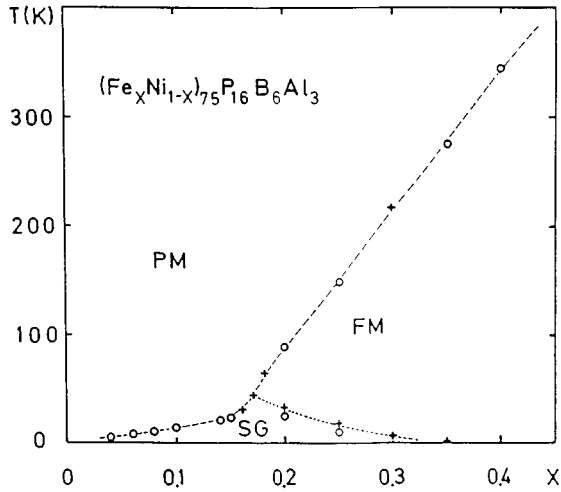


FIG. 1. Magnetic phase diagram of $(\text{Fe}_x\text{Ni}_{1-x})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$, from Ref. 14.

A standard diluted reentrant ferromagnet, $(\text{Fe}_{0.20}\text{Ni}_{0.80})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$, was investigated by susceptibility and magnetization methods. We first present the general behavior of the dynamic susceptibility measured at a specific frequency but using different magnitudes of the ac field and different superimposed dc fields. A remarkable and well-known sensitivity to the applied field strength is observed. However, for low enough fields and at low temperatures, the measured dynamic susceptibility does not depend on the magnitude of the ac field, i.e., the system exhibits linear response. This field regime allows direct studies of the inherent zero-field dynamics of reentrant ferromagnets. Utilizing this possibility, we demonstrate two distinguishing physical characteristics of reentrant ferromagnets: A dynamic scaling analysis of the ac-susceptibility data indicates a transition from a ferromagnetic to a true reentrant spin-glass phase and magnetic aging, a typical spin-glass feature, is found to exist throughout the reentrant spin-glass phase.

III. EXPERIMENTAL

The amorphous system $(\text{Fe}_x\text{Ni}_{1-x})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ is a metallic glass with Ruderman-Kittel-Kasuya-Yosida-type interactions between the magnetic ions. Its magnetic properties are mainly determined by the Fe atoms since the magnetic moment of Ni is quenched due to charge transfer from the metalloids.¹² The pure Ni alloy is nonmagnetic and with increasing Fe concentration long-range ferromagnetic order sets in at $x=0.17$. For lower concentrations, $x<0.17$, typical spin-glass behavior is found and for a range of concentrations with $x>0.17$, reentrant spin-glass behavior occurs at low temperatures. Both the spin-glass and the reentrant concentrations of this compound have been studied in a large number of investigations.¹³ In Fig. 1, the magnetic phase diagram of $(\text{Fe}_x\text{Ni}_{1-x})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ quoted from Beckman *et al.*¹⁴ is shown. The specific concentration $x=0.20$ shows a PM→FM transition at $T_c \approx 92$ K and reentrant behavior at low temperatures. Our sample of $(\text{Fe}_{0.20}\text{Ni}_{0.80})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ has the form of a ribbon (cross section 0.01×1.0 mm² and length 4 mm) yielding an effective demagnetization factor $N<0.002$ along the longest axis. The measurements were

performed with the applied magnetic field along this direction in two different susceptometer/magnetometers, a non-commercial superconducting quantum interference device magnetometer and a commercial Lake Shore 7225 ac susceptometer.

IV. RESULTS AND DISCUSSION

A. Field and temperature dependence of the ac susceptibility

We first focus on ac-susceptibility results measured at different amplitudes of the ac field. In Figs. 2(a) and 2(b), $\chi'(\omega)$ and $\chi''(\omega)$ are plotted vs temperature for $\omega/2\pi=125$ Hz. The different curves correspond to different amplitudes of the ac field, $0.13 \leq h \leq 20$ G. The susceptibility is strongly dependent upon the ac-field amplitude. The position of the high-temperature knee (cusp at the lowest fields) in the $\chi'(\omega)$ vs T curve signals the ferromagnetic phase, the rather abrupt decrease of $\chi'(\omega)$ at a lower temperature corresponds to a temperature where the coercive field, H_c , has increased to become of the order of the ac-field amplitude. This decrease is accompanied by a corresponding pronounced maximum in $\chi''(\omega)$ [see Fig. 2(b)]. The decreasing level of the plateau in $\chi'(\omega)$ with increasing amplitude of the ac field in-between these points is due to saturation of the induced magnetization on traversing the narrow hysteresis loop to reach the peak values of the ac field.¹⁵ A significant observation is also that the amplitude of the very largest part of the $\chi(T, \omega)$ curves is not limited by the demagnetizing field (the demagnetizing field puts an upper limit on χ at $1/N$, where N is the demagnetization factor). The highest values of the susceptibility for the fields $h=0.5$ and 1 G are however close to this limit (the measured $\chi'_{\text{max}}(\text{SI}) \approx 500$ and $1/N > 500$).

A most important observation for the interpretability of susceptibility data is that well below the maxima in $\chi'(\omega)$ and $\chi''(\omega)$, both components of $\chi(\omega)$ apparently become independent of h . Thus, for a specific temperature, $T < T_c$, there exists a field, $h_0(T, \omega)$, such that for $h < h_0(T, \omega)$ both components of $\chi(\omega)$ are independent of h within experimental accuracy. It can be postulated from Fig. 1, that $h_0(T, \omega)$ decreases as T increases. (Physically natural limits for h_0 are $h_0(T, \omega) \rightarrow 0$ as $T \rightarrow T_c$ or as $\omega \rightarrow 0$). In the insets to Fig. 2 the behavior at low temperatures is enlarged. From the curves at lower fields one observes a pronounced second downward deviation in $\chi'(\omega)$ and a corresponding maximum in $\chi''(\omega)$, these two features are the signatures of the reentrant transition.

Figures 3(a) and 3(b) show the ac susceptibility, $\chi'(T, \omega)$ and $\chi''(T, \omega)$ for $\omega/2\pi=125$ Hz, $h=0.125$ G in different superposed dc fields. Already at very modest background fields both $\chi'(\omega)$ and $\chi''(\omega)$ are suppressed drastically in the ferromagnetic phase (the ac loops are traversed at fields above the coercivity field in the higher superposed dc fields). The field dependence of the susceptibility at lower temperatures is governed by spin-glass dynamics and responds quite differently (see the inset of Fig. 3). The effect of a magnetic field on the susceptibility at the spin-glass transition is to shift the position of the maximum in $\chi''(\omega)$ to lower temperatures and also decrease its magnitude and to suppress the magnitude of $\chi'(\omega)$. At temperatures well below the position of the maximum in $\chi''(\omega)$, both components of the ac susceptibility are

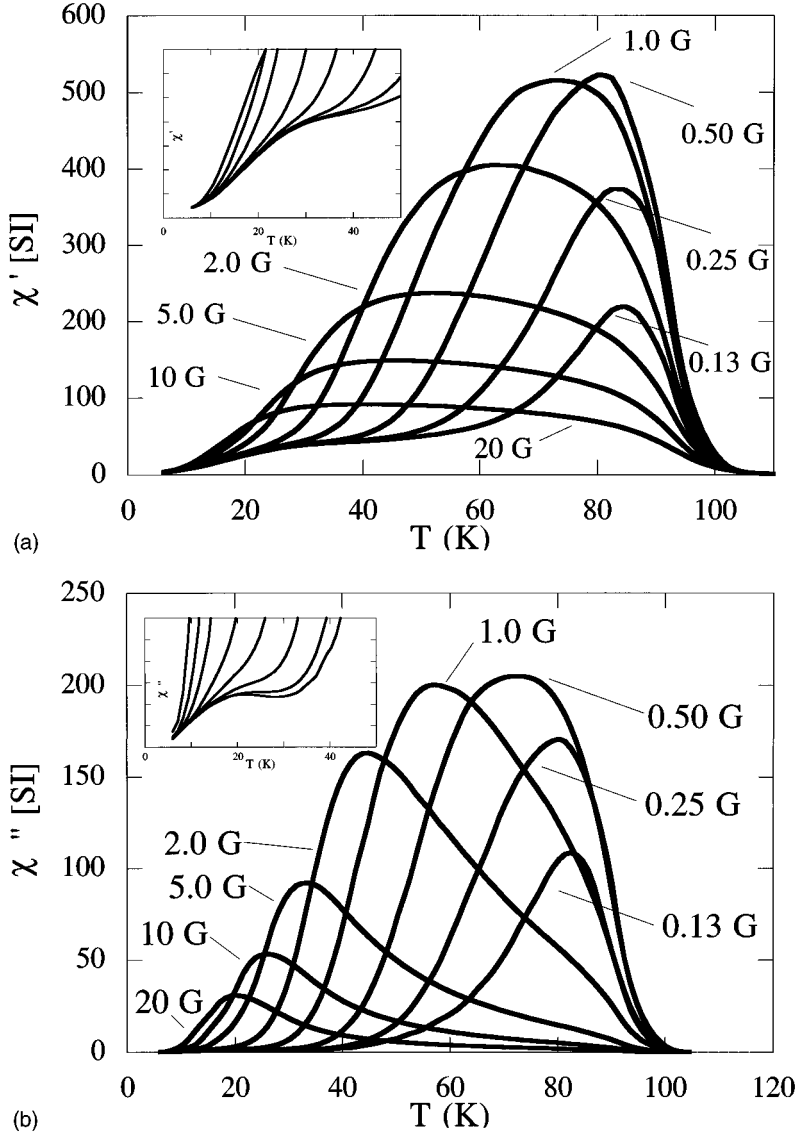


FIG. 2. Ac susceptibility for $\omega/2\pi=125$ Hz measured by different amplitudes of the ac field, $h=0.13, 0.25, 0.50, 1.0, 2.0, 5.0, 10,$ and 20 G. (a) $\chi'(\omega)$ vs T , (b) $\chi''(\omega)$ vs T . The insets show enlargements of the low-temperature region.

unaffected by the superimposed dc-magnetic field. This behavior is similar to what is found for ordinary spin glasses in a magnetic field near a PM-SG phase transition,¹⁶ and contrasts sharply to the remarkable dc-field dependence of the ferromagnetic phase. The magnitude of the maximum in $\chi''(\omega)$, signifying the reentrant behavior, is of almost diminishing size compared with the huge maximum associated with the coercivity.

A crucial new result for the interpretability of susceptibility and magnetization measurements is the existence of linear response at low enough fields, which in the low-temperature phase and at low temperatures in the FM phase is established within our experimental accuracy. The experimental consequence is that the inherent dynamics of the RSG phase (and of the FM phase) can be studied by susceptibility and magnetization measurements if a sufficiently low probing field $h < h_0(T, \omega)$ is used. In the remaining part of the paper, we discuss the two examples where we utilize this possibility. First we examine the dynamics around the FM-RSG transition and then we investigate a possible occurrence of magnetic aging.

B. Frequency dependence of the ac susceptibility and dynamic scaling

In Figs. 4(a) and 4(b), $\chi'(\omega)$ and $\chi''(\omega)$ measured in $h=2 \times 10^{-4}$ G are plotted for $\omega/2\pi=0.051, 0.51, 5.1, 51,$ and 510 Hz and $11 \leq T \leq 40$ K. In the insets of Figs. 3(a) and 3(b) where $\chi'(\omega)$ and $\chi''(\omega)$ are plotted at $\omega/2\pi=170$ Hz for two different probing fields, $h=2 \times 10^{-4}$ and 5×10^{-5} G. The results are identical within experimental accuracy showing that $h=2 \times 10^{-4}$ G $< h_0$ at these temperatures. In Fig. 4(b) there is a maximum in the $\chi''(\omega)$ vs T curves around 15 K which is frequency dependent and shifts towards lower temperatures with decreasing frequency. This is a characteristic feature of spin-glass dynamics (without spin-glass dynamics at low temperatures $\chi''(T, \omega)$ would decrease monotonously with decreasing temperature from the maximum at $h \approx H_c$). It is worth noting in this connection that if a too large magnitude of the ac field is used, the faint maximum associated with the reentrant spin-glass transition inevitably becomes dominated and hidden by the large maximum in $\chi''(T, \omega)$ governed by the coercivity, see inset of Fig. 2(b). Also the corresponding rapid decreases in $\chi'(T, \omega)$ coalesce at too high ac fields and

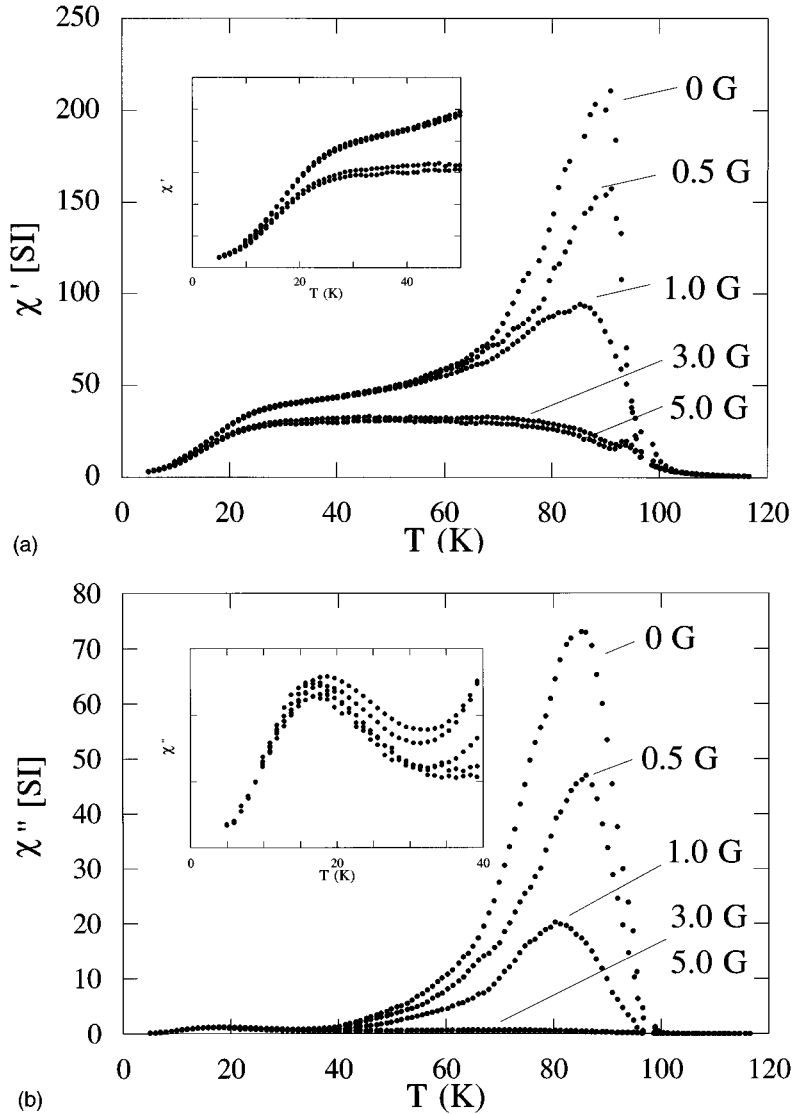


FIG. 3. $\chi'(\omega)$ (a) and $\chi''(\omega)$ (b) at $\omega/2\pi=125$ Hz and $h=0.13$ G in different applied dc-biasing fields, $H_{dc}=0, 0.5, 1.0, 3.0,$ and 5.0 G. The insets show enlargements of the low-temperature region.

yield a similar confusion. There are frequent examples in the literature where these features have mistakenly been associated only with the reentrant spin-glass transition).

The low-temperature behavior of $\chi'(\omega)$ and $\chi''(\omega)$ vs T appears indistinguishable from an ordinary low-temperature spin-glass phase below the spin-glass temperature, T_g . However, on exceeding the glass temperature, T_{RSG} , where the in-phase component of the susceptibility of an ordinary spin glass exhibits a frequency-dependent cusp and the out-of-phase component rapidly decreases to zero, the reentrant system on the other hand shows: (i) $\chi'(\omega)$ continuously increases on increasing temperature and (ii) $\chi''(\omega)$ exhibits a frequency-dependent maximum followed by a weakly temperature-dependent finite level in the ferromagnetic phase. These observations imply that some new high-frequency ($\omega > 1$ kHz) relaxation processes appear on increasing the temperature through T_{RSG} and give a contribution to $\chi'(\omega)$ in the ferromagnetic phase that continuously increases on increasing the temperature. Such a contribution is alien to a true spin-glass phase and apparently also disappears below the estimated T_{RSG} .

In studies of the ordinary PM-SG phase transition, a frequency-dependent freezing temperature, $T_f(\omega)$, can be de-

efined as the temperature where the maximum relaxation time, $t \approx 1/\omega$, of the system corresponds to the measurement frequency. The divergence of the maximum relaxation time, occurring at the spin-glass transition temperature, can thereafter be investigated using dynamic scaling. In the case of the possible FM-RSG transition, the maximum relaxation time of the system is very long also in the high-temperature FM phase as is seen by the frequency dependence of $\chi'(\omega)$ and the finite $\chi''(\omega)$ in Fig. 3. It is thus not correct to assign all the dynamics at our frequencies to an emerging spin-glass phase. Nevertheless, we can associate the appearance of the second maximum in $\chi''(\omega)$ to the slowing down due to spin-glass dynamics and analyze the data using dynamic scaling. Defining the temperature of the maximum in $\chi''(\omega)$ as the freezing temperature, T_f , at a specific observation time ($t \approx 1/\omega$), a fit to conventional critical slowing down of the relaxation times,

$$\frac{\tau}{\tau_0} \propto \left(\frac{T_f - T_{RSG}}{T_{RSG}} \right)^{-z\nu} \quad (1)$$

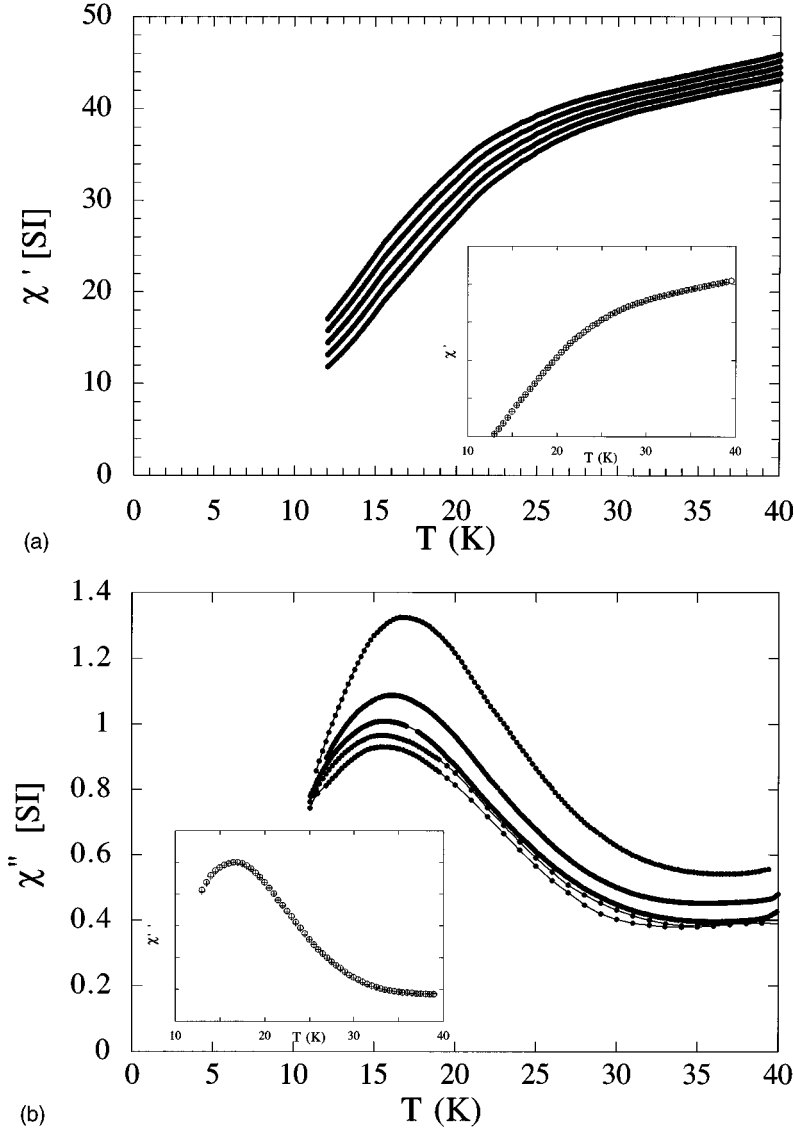


FIG. 4. $\chi'(\omega)$ (a) and $\chi''(\omega)$ (b) vs T for $\omega/2\pi = 0.051, 0.51, 5.1, 51,$ and 510 Hz [top to bottom (a) and bottom to top (b)], $11 \leq T \leq 40$ K, and $h = 2 \times 10^{-4}$ G. The insets show the behavior at $\omega/2\pi = 170$ Hz at two different magnitudes of the ac field, $h = 2 \times 10^{-4}$ and 5×10^{-5} G.

gives a nice fit. A best fit of data in the interval $51 \text{ mHz} \leq \omega/2\pi \leq 1.7 \text{ kHz}$ is shown in Fig. 5, yielding $z\nu = 7.9$ and $T_{\text{RSG}} = 14.7$ K. This value of $z\nu$ is similar to what has been found in ordinary paramagnetic to spin-glass transitions.¹⁷ We have also tried to fit the data to Eq. (1) with $T_{\text{RSG}} = 0$ and to activated dynamic scaling [Eq. (2)] both with T_{RSG} as a free parameter and with $T_{\text{RSG}} = 0$:

$$\ln(\tau/\tau_0) \propto \frac{1}{T_f} \left(\frac{T_f - T_{\text{RSG}}}{T_{\text{RSG}}} \right)^{-\psi\nu}. \quad (2)$$

Neither Eq. (1) nor Eq. (2) fit the data well using $T_{\text{RSG}} = 0$. A best fit to Eq. (2) using $\tau_0 = 10^{-12}$ s gives the values $\Psi\nu = 0.28$ and $T_g = 14.6$ K, and is of comparable quality to the fit in Fig. 5. Thus, dynamic scaling suggests that there is a divergence of the spin-glass relaxation times at a finite transition temperature, but the data cannot distinguish between activated and conventional critical slowing down. A divergence of the spin-glass relaxation times at a finite temperature is in agreement with a true reentrant phase transition.¹⁸

C. Aging

To reveal a possible aging phenomenon we performed time-dependent zero-field-cooled (ZFC) magnetization measurements. The sample was then cooled from a reference temperature (in our case 120 K) to a measurement temperature (T_m) and kept there a wait time (t_w). Thereafter a dc field (h) was applied and the magnetization (m) of the sample was recorded as a function of time. As in the case of ac-susceptibility measurements a linear response to the applied field is found for small enough fields ($h < h_0$).¹⁹ In Fig. 6(a), m is plotted vs $\log t$ at $T_m = 13$ K, $h = 0.2$ G, and $t_w = 100, 1000,$ and $10\,000$ s, (at 13 K, $h_0 > 0.2$ G). The corresponding relaxation rate ($S = 1/h \, dm/d \log t$) is plotted in Fig. 6(b). A strong wait time dependence of the response is observed. The m vs $\log t$ curves have an inflection point at an observation time nearly equal to t_w and the relaxation rate attains a corresponding maximum. A similar aging phenomenon is well known from ordinary spin glasses²⁰ and has earlier also been observed in other RSG systems.²¹ The observed aging behavior does not end at T_{RSG} , but sustains into the ferromagnetic phase.²²

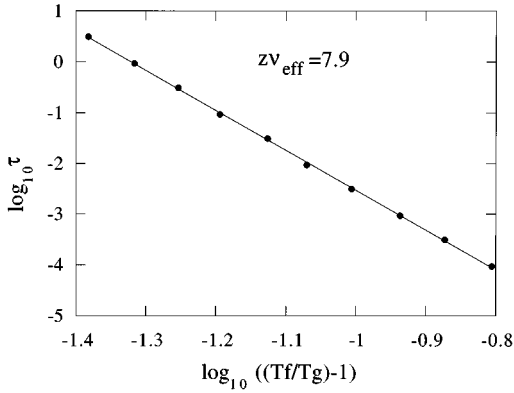


FIG. 5. The best fit to $\tau/\tau_0 = [(T_f - T_g)/T_g]^{-z\nu}$ of the measured freezing temperatures T_f yields $T_g = 14.7$ K and $z\nu_{\text{eff}} = 7.9$.

V. CONCLUSION

The dynamics of the reentrant ferromagnet $(\text{Fe}_{0.20}\text{Ni}_{0.80})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ has been studied. In particular it is shown that susceptibility and magnetization measurements can be used to probe the intrinsic dynamics of reentrant ferromagnets if a probe field $h < h_0$ is used. Thus, there exists an experimental probe—susceptibility and magnetization measurements in low enough fields—which has high accuracy and may readily be exploited to cover more than ten decades of time (10^{-6} – 10^5 s) by combining ac-susceptibility and ZFC-magnetization measurements. In earlier investigations of the intrinsic dynamics of reentrant ferromagnets one has been limited to neutron-scattering time scales ($\tau < 10^{-9}$ or 10^{-8} s) and accuracy.²³

Using ac-susceptibility results, we have analyzed the dynamics around the reentrant spin-glass transition. Maybe most importantly, we find a weak, earlier unresolved or overlooked, frequency-dependent maximum in the out-of-phase component of the ac susceptibility at low temperatures that is the real signature of an emerging reentrant spin-glass phase. Dynamic scaling suggests that there is a spin-glass relaxation time which diverges at a finite transition temperature with a

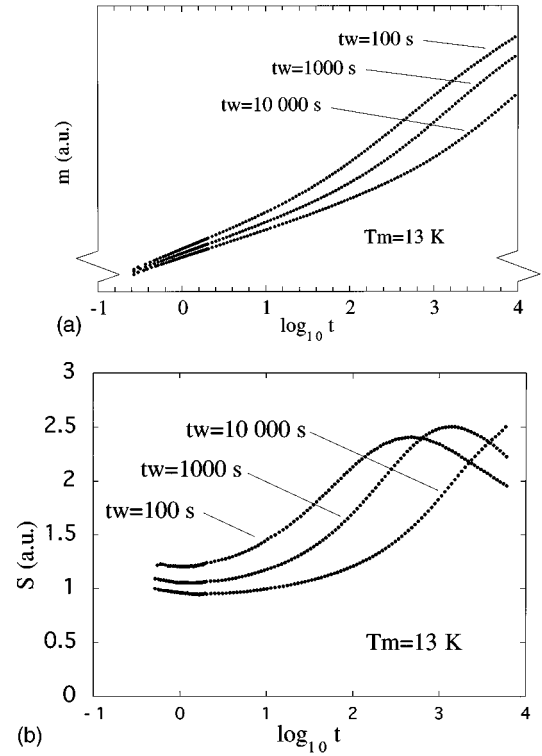


FIG. 6. ZFC magnetization m (a) and corresponding relaxation rate $S = 1/h \, dm/d \log_{10} t$ (b) vs $\log t$ for $t_w = 100, 1000,$ and $10\,000$ s, using $h = 0.2$ G. $T_m = 13$ K.

dynamic exponent similar to what is found near an ordinary PM-SG transition, i.e., there is a true reentrant spin-glass transition. This result is in accord with the neutron scattering⁹ implication that long-range ferromagnetic order disappears at T_{RSG} . We have also verified that aging, an established spin-glass feature, is present in the RSG phase.

ACKNOWLEDGMENT

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