Chaos in the Ferromagnetic Phase of a Reentrant Ferromagnet

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The inherent dynamic properties of the ferromagnetic phase of a reentrant ferromagnet, $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$, have been experimentally investigated by low field magnetic relaxation and ac susceptibility measurements. A prominent aging behavior and an apparent fatal fragility to temperature fluctuations imply that the ferromagnetic phase of the reentrant system has a chaotic nature similar to the spin glass phase, but in striking contrast to the robust nature of the regular ferromagnetic phase. [S0031-9007(96)01179-9]

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Random magnets have been intensively studied in the last couple of decades. An important class of random systems is the bond disordered system in which the magnetic interactions are taken randomly from a distribution of both positive (ferromagnetic) and negative (antiferromagnetic) interactions. If all the interactions are ferromagnetic (FM) the low temperature ordered phase is the ferromagnetic phase. If a few interactions, taken at random, are changed to be antiferromagnetic (AF), the low temperature phase will still have long range ferromagnetic order but some magnetic moments will be frustrated. On increasing the concentration of random AF interactions, frustration increases and, above some concentration, long range ferromagnetic order is no longer favorable. In such a bond disordered system the low temperature phase is the spin glass phase [1].

Experimentally, there are numerous examples of compounds where the amount of AF and FM interactions can be continuously varied by changing the concentration of a magnetic ion. One such example is $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$ [2]. This system is an amorphous metallic glass with RKKY type of 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. For concentrations $x < 0.17$ the system is a spin glass and for $x > 0.17$ the system shows ferromagnetism. In a range of concentrations $0.17 < x < x_{RSG}$ the system shows reentrant behavior, i.e., on lowering the temperature the sequence of phases paramagnetic—ferromagnetic—spin glass occurs.

In this Letter we study the intrinsic dynamics of the ferromagnetic phase of the reentrant ferromagnet $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$, using low field magnetic relaxation experiments. The main conclusion from the investigation is that the ferromagnetic phase of a reentrant ferromagnet is chaotic, in a similar way as the spin glass phase [3], but in contrast to the robust nature of a nonfrustrated ferromagnetic phase.

To recall the characteristic temperature dependence and dynamic nature of the susceptibility of a reentrant ferromagnet the ac susceptibility, $\chi(T,\omega)$, of $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$ is shown in Fig. 1. The main figure displays the out of phase component $\chi''(T,\omega,h)$, and the inset shows the in phase component $\chi'(T,\omega,h)$, both measured at two different magnitudes of the probing ac field $h = 2\times10^{-4}$ and 2×10^{-5} G, but at one and the same frequency $\omega/2\pi = 1.25$ Hz. The sharp maximum at high temperature in both $\chi'(T)$ and $\chi''(T)$ is associated with the onset of ferromagnetism ($T_c \approx 92$ K). These maxima are broadened and pushed toward lower temperature with increasing ac field [4]. The low temperature maximum in $\chi''(T)$ and the corresponding knee in $\chi'(T)$ signals the reentrant spin glass transition. A recent dynamic scaling analysis of the freezing temperatures $T_f(\omega)$, defined from the maximum in $\chi''(\omega,T)$ at the ferromagnetic to reentrant spin glass transition yielded $T_{RSG} = 14.7$ K [4].

Figure 1 also illustrates that the magnetic response to two weak but significantly different ac fields is linear well into the ferromagnetic phase. It is only very close to T_c that any nonlinearity can be resolved. That the magnetic response of $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$ to a field perturbation $h(\omega)$ is linear at low enough temperatures provided $h(\omega) < h_0(\omega,T)$ has earlier been

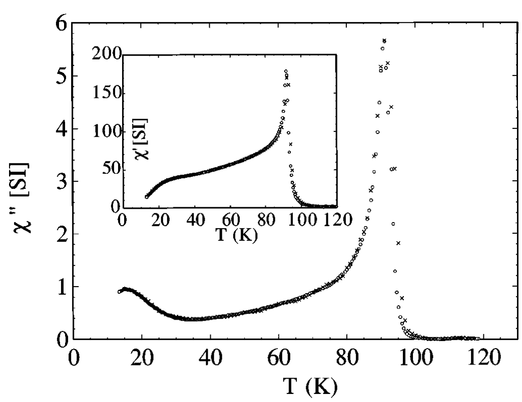

FIG. 1. $\chi''(\omega)$ and $\chi'(\omega)$ (inset) vs T measured in an ac field of frequency $\omega/2\pi = 1.25$ Hz and at two different amplitudes $h_{ac} = 2\times10^{-4}$ (open circles) and 2×10^{-5} G (crosses).

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experimentally established [4]. The limiting field $h_0(\omega, T)$ decreases with increasing temperature. The property of linear response is of crucial importance since it allows direct measurements of zero field dynamics both in the reentrant spin glass region and in the ferromagnetic phase by low field ac susceptibility and dc magnetization experiments.

In a slowly relaxing system exhibiting linear response, the zero field response function is directly reflected in time dependent zero-field-cooled (ZFC) magnetization measurements. To reveal any nonstationarity of the response function of our system, ZFC measurements were performed according to the following procedure: The sample is cooled in zero field from a reference temperature $T_{\text{ref}} = 100$ K, above T_c to the measurement temperature T_m, and kept there a wait time t_w. A small dc field h is then applied and the magnetization m is recorded as a function of time t elapsed after the field application. In Fig. 2(a), m vs $\log t$ is plotted for $t_w = 100$, 1000, and 10 000 s at $T_m = 40$ and 70 K. The corresponding relaxation rate $S = (1/h)dm/d\log_{10} t$ is plotted in Fig. 2(b). The dc field used was $h = 0.02$ G, which at our observation times is well below $h_0(T)$ for the lower and in the vicinity of $h_0(T)$ for the higher measurement temperature. The figure illustrates a strong wait time dependence of the magnetic response at two quite different temperatures in the ferromagnetic phase. The m vs $\log t$ curves have an inflection point at $\log t \approx \log t_w$ and $S(t)$ attains a corresponding maximum. A similar wait time dependence of the magnetic response, reflecting an intrinsic aging process, is inherent to spin glass dynamics [5].

Looking closer into the here observed slow dynamics ($10^{-1} < t < 10^4$ s) of the reentrant ferromagnetic and spin glass phases, some quantitative and qualitative differences are conspicuous. The equilibrium ($t < < t_w$) relaxation rate or equivalently the amplitude of $\chi''(\omega, T)$ is, assuming thermally activated dynamics, one order of magnitude larger in the spin glass region than in the ferromagnetic region [compare, e.g., the magnitude of $\chi''(\omega)$ at $T = 13$ and 40 K in Fig. 1]. The influence of the aging on the relaxation rate is more pronounced in the ferromagnetic than in the spin glass region, e.g., for curves measured at $t_w = 1000$ s, the ratio between the maximum relaxation rate at $t \approx t_w$ and the rate at $t = 1$ s amounts to about 7 in the ferromagnetic phase at 40 K (see Fig. 2) compared to a corresponding ratio of about 2 in the spin glass region at 13 K [4]. When entering into the ferromagnetic regime the measured $\chi''(\omega, T)$ goes through a maximum followed by a shallow minimum at $T \approx 30$ K (Fig. 1). However, in the ferromagnetic region there remains an increasing "fast" contribution to the susceptibility that rather rapidly reaches a "saturated" value at temperatures above T_{RSG}. This contribution relaxes on shorter time scales than our experimental observation time window (the measured out-of-phase component decreases whereas the in-phase component continuously increases with increasing temperature).

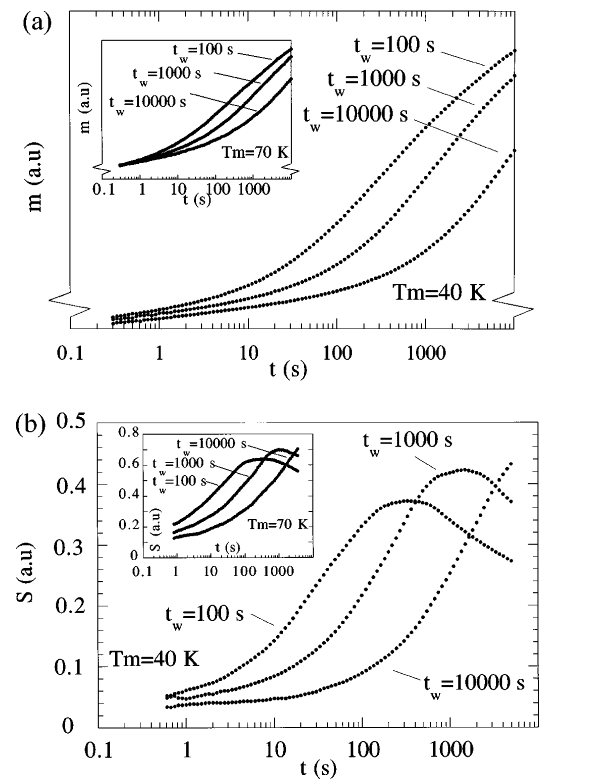

FIG. 2. ZFC magnetization, $m(t)$ vs $\log t$ (a), and relaxation rate $S(t) = (1/h)\partial m/\partial \log_{10} t$ vs $\log t$ (b), for $t_w = 100$, 1000, 10 000 s, at $T_m = 40$ K and $h = 0.02$ G. The insets show the corresponding curves at $T_m = 70$ K.

In spin glasses, the magnetic relaxation curves are informatively affected by temperature cyclings or shifts after the initial wait time. Changes in the behavior which convincingly confirm that measurable aging effects emanate from a chaotic nature of the spin glass ordering [6]. To accordingly examine the here observed aging behavior in the ferromagnetic phase of $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$, temperature cycling experiments were performed. The system is then cooled in zero field from a reference temperature $T_{\text{ref}} = 100$ K above T_c to the measurement temperature T_m. After a wait time t_w, the system is subjected to a temperature cycling of magnitude ΔT and duration $t_{\Delta t}$ at the cycling temperature (the heating and cooling time is not included in this measure). When the temperature T_m is recovered, the probing magnetic field is applied and the relaxation of the magnetization is recorded.

Figure 3 shows the relaxation rate $S(t)$ measured at $T_m = 40$ K for different magnitudes of negative ΔT's and corresponding results using positive ΔT's in the inset. The curves were recorded using $t_w = 3000$ s, $t_{\Delta T} = 10$ s, and $h = 0.02$ G. The curve marked $\Delta T = \infty$ in the graph corresponds to a ΔT that takes the system to

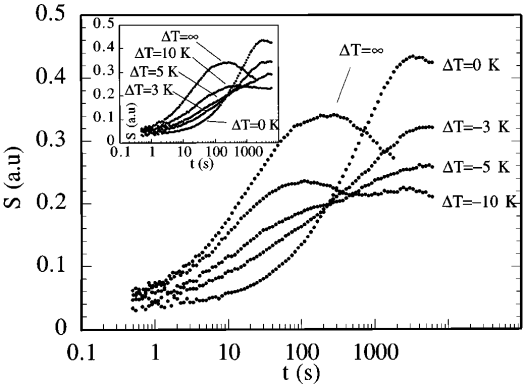

FIG. 3. The relaxation rates $S(t) = (1/h)\partial m/\partial \log_{10} t$ of the zero field cooled magnetization vs $\log t$ at $T_m = 40$ K, $t_w = 3000$ s, and $h = 0.02$ G. Immediately prior to the field application the sample was subjected to a negative temperature cycling $-\Delta T$. The inset shows the result after positive temperature cyclings.

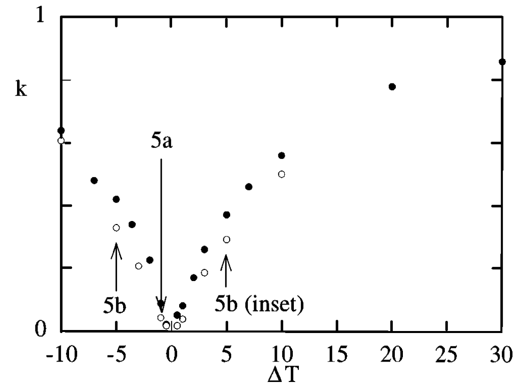

FIG. 4. Fraction k plotted vs the magnitude of the temperature cyclings ΔT. The open circles correspond to $t_{\Delta T} = 10$ s and $t_w = 3000$ s. The filled circles correspond to $t_{\Delta T} = 3000$ s and $t_w = 7000$ s for the positive cyclings and $t_{\Delta T} = 7000$ s and $t_w = 3000$ s for the negative cyclings. The arrows indicate the values of ΔT used in the susceptibility measurements shown in Fig. 5.

a temperature above T_c, and the curve marked $\Delta T = 0$ is the relaxation rate of an ordinary ZFC magnetization curve measured at $t_w = 3000$ s. With increasing $|\Delta T|$, the amplitude of the maximum in $S(t)$ at $t \approx 3000$ s decreases and a second maximum gradually develops at a shorter observation time. The new maximum at short observation times indicates a partial reinitialization of the system during the temperature cycling. The position of this maximum reflects an "effective" wait time governed by the heating (or cooling) rate and the time ($\approx$20 s) allowed for temperature stabilization when T_m is recovered. The appearance of two maxima suggests a coexistence of two characteristic "ages" in the system, one reflecting the original wait time $t_w = 3000$ s prior to the temperature cycling, and the other reflecting the reinitialization.

The relaxation rate curves in Fig. 3 can be decomposed into two parts: a fraction k ascribed to the reinitialized part of the system (giving the maximum at short observation times) and an unperturbed fraction $1 - k$ (yielding a maximum corresponding to $t_w \approx 3000$ s). In Fig. 4 the fraction k is plotted vs the magnitude and sign of the temperature cycling ΔT. The curve is nearly symmetrical around T_m. For small $|\Delta T|$ ($<$0.3 K), $k \approx 0$ and virtually no reinitialization occurs. For larger $|\Delta T|$, k increases with increasing $|\Delta T|$. In Fig. 4, a second series of temperature cycling measurements is also included. In this series the different wait times were $t_w = 7000$ s and $t_{\Delta T} = 3000$ s for the positive temperature cycles and $t_w = 3000$ s and $t_{\Delta T} = 7000$ s for the negative temperature cycles. Essentially this series differs from the first series in that the duration of the temperature cycle $t_{\Delta t}$ is longer. As seen in Fig. 4, $k(\Delta T)$ does not depend very strongly on $t_{\Delta T}$ although the series with the longer duration of the cycling falls slightly above the other.

The observed behavior in the cycling experiments is qualitatively very similar to what has been observed in ordinary spin glasses. However, one difference is that for larger and larger magnitude of a negative ΔT, a logarithmically longer and longer duration of the temperature perturbation is necessary to achieve significant reinitialization of aging in spin glasses [6].

In slowly relaxing systems, the ZFC magnetization and the ac susceptibility are related by $\chi'(\omega) \approx m(t)/h$ and $\chi''(\omega) \approx (\pi/2)S(t)$ at $t \approx 1/\omega$ [7]. In ac susceptibility, magnetic aging is observed as a decay of both $\chi''(\omega)$ and $\chi'(\omega)$ with time spent at constant temperature, or age of the system t_a. Reinitialization due to a temperature cycling should appear as an "instant" increase of the in-phase and out-of-phase components of the susceptibility followed by a continuous slow decay when the measurement temperature is recovered.

In the measurements reported here, an ac field of frequency $\omega/2\pi = 170$ mHz and magnitude $h_{\mathrm{ac}} = 4 \times 10^{-3}$ G was used. The sample is cooled in zero field from the reference temperature $T_{\mathrm{ref}} = 100$ K, above T_c, to the measurement temperature $T_m = 40$ K, where the two components of the ac susceptibility are recorded as a function of time elapsed after reaching T_m. After 3000 s (corresponding to the wait time used in the ZFC experiments), the system is subjected to a temperature cycling ΔT and when T_m is recovered the recording of the susceptibility is continued. In Fig. 5(a), $\chi''(\omega, t_a)$ and $\chi'(\omega, t_a)$ are plotted vs t_a, both components decay with increasing t_a which is the anticipated effect of the aging process. After 3000 s, a temperature cycling $\Delta T = -1$ K is effectuated, and when T_m is recovered the recording is continued. The immediate increase of the magnitude of both $\chi''(\omega, t_a)$ and $\chi'(\omega, t_a)$ after the cycling reflects a partial reinitialization of the system. In Fig. 5(b), the corresponding $\chi''(\omega, t_a)$ vs t_a plots for both positive and negative ΔT using a larger magnitude, 5 and -5 K, are shown. A significantly larger reinitialization

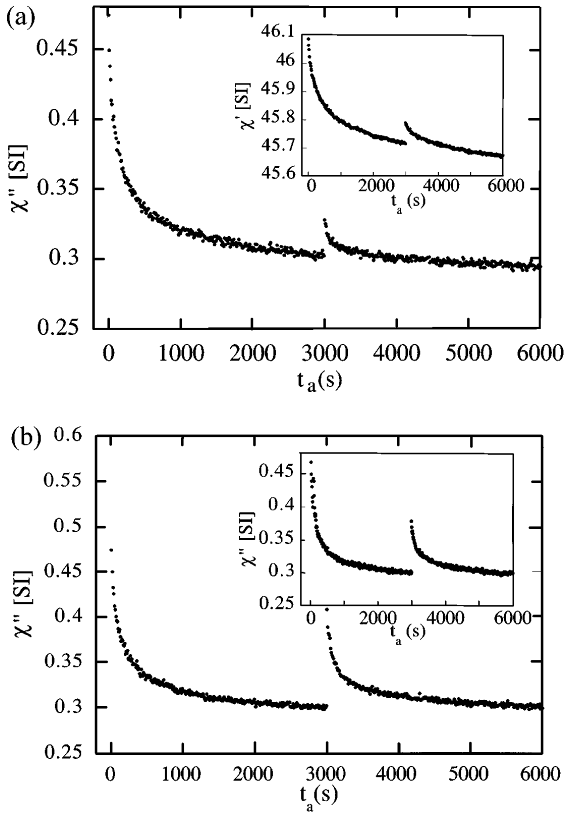

FIG. 5. Susceptibility vs time at $T_m = 40$ K measured by an ac field of frequency $\omega/2\pi = 170$ mHz and amplitude $h_{\mathrm{ac}} = 4$ mG. (a) $\chi''(\omega)$ and $\chi'(\omega, T)$ (inset) vs t_a (time spent at T_m). At time $t_a = 3000$ s, the sample was subjected to a negative temperature cycling $\Delta T = -1$ K of duration $t_{\Delta T} = 10$ s. (b) $\chi''(\omega)$ vs t_a. At the time $t_a = 3000$ s the sample was subjected to a negative temperature cycling $\Delta T = -5$ K and $t_{\Delta T} = 10$ s. In the inset $\chi''(\omega)$ vs t_a is shown for a positive temperature cycling $\Delta T = 5$ K and $t_{\Delta T} = 10$ s.

than in the $\Delta T = -1$ K case is observed, in agreement with the ZFC results.

The experiments show that both aging and the possibility to reinitialize aging by a temperature perturbation are inherent properties of the ferromagnetic phase of a standard reentrant ferromagnet $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$. These properties have earlier been observed only in the spin glass phase and in phases where the dynamics is governed by spin glass dynamics. Aging after a fast temperature quench has earlier been observed in the two dimensional random ferromagnet $Rb_2Cu_{0.89}Co_{0.11}F_4$ [8]. The results were explained in terms of the severely slowed down domain growth predicted in current models of random ferromagnets, and also compared to aging in spin glasses. However, in contrast to spin glasses, the results indicated a relaxation towards a unique ground state at all temperatures below T_c, i.e., a chaotic nature of the ferromagnetic phase was not observed.

We are not aware of any theoretical work on ferromagnetism that could be used to interpret our experimental results on the nonstationary dynamics of the ferromagnetic phase. However, the observed behavior resembles that of spin glasses. One description of low dimensional spin glasses that accounts for chaos is the droplet model [3,9]. In the droplet model, the spin glass phase is chaotic in the sense that at every temperature the system evolves towards a different free energy minimum, and at two different temperatures the free energy minima have identical spin configurations only up to a certain overlap length scale $L^*(\Delta T)$ [3]. The larger the temperature difference ΔT, the smaller the overlap length $L^*(\Delta T)$. The apparent chaotic nature of the reentrant ferromagnetic phase might require a cognate description.

Many features associated with the hysteresis loop (i.e., when the dynamics is driven by a magnetic field) of the ferromagnetic phase of reentrant systems have large similarities with soft ferromagnets. In contrast, the intrinsic dynamics of the reentrant ferromagnetic phase is here found to be very different from that of an ordinary ferromagnet and instead to have a striking resemblance to spin glass dynamics. The results suggest that the ferromagnetic phase is chaotic and that chaos in magnetic systems is directly associated with disorder and frustration and not only with spin glass behavior.

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