Nonresonant Spectral Hole Burning in a Spin Glass

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Large-amplitude low-frequency magnetic fields are found to burn spectral holes in the response of a 5% Au:Fe spin glass. There is negligible overlap between the degrees of freedom influenced by the highest and lowest frequencies. For sufficiently large fields, selected parts of the sample can be heated above the transition while the rest of the sample remains a spin glass. The return to equilibrium after heating indicates that the slow degrees of freedom are coupled to the thermal bath, but not strongly to each other. All prominent features are modeled by a distribution of independent domains, with the same relaxation time for local response and recovery.

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The two most distinctive features which define spin-glass response are the nonexponential relaxation after changing the magnetic field and its aging dependence after changing the temperature [1]. There is general agreement that when cooled below the spin-glass temperature (T_g) in an applied field (h), the thermoremanent magnetization as a function of time $[(M(t)]$ exhibits nonexponential relaxation that depends on the time spent in the field-cooled state before h was removed. However, the connection between $M(t)$ and its wait-time behavior, and whether their time dependences should be approximated by a logarithm [2], power law [3], stretched exponential [4], or some combination [5,6], remains a topic of debate. Still a more fundamental issue is whether $M(t)$ is dominated by intrinsically nonexponential response, or a heterogeneous distribution of relaxation times. Indeed, some measurements indicate a serial hierarchy of time-dependent relaxation rates [7,8], while others favor an effectively static distribution of parallel processes [9–11]. Similarly, some models for spin-glass dynamics are based on homogeneous interactions [12] which yield local relaxation rates that change during the response [13,14], while others start with the picture of independent droplets [15] or domains [10,16]. At least part of the ambiguity arises from the Boltzmann superposition principle which states that it is impossible to distinguish between homogeneous and heterogeneous scenarios with any linear macroscopic measurement. Thus, conclusive evidence must come from more direct techniques.

Recently the technique of nonresonant spectral hole burning (NSHB) was developed to investigate the nonexponential relaxation of glass-forming liquids [17]. NSHB utilizes a large-amplitude low-frequency pump oscillation, followed by a probe step, with a phase-cycling scheme. The technique requires that the sample absorb sufficient energy during the pump oscillation to modify its spectrum of response and that this modification persist long enough to be measured. Often the absorbed energy can be thought of in terms of an excess heat which accelerates the response. For homogeneous response [Fig. 1(a)], all parts of the sample are heated equally, so that the entire spectrum is simply shifted to shorter times; whereas for heterogeneous response [Fig. 1(b)], only the degrees of freedom that respond near to the pump frequency absorb a significant amount of energy and a spectral hole develops. Because it is easy to adjust the pump frequency, pump amplitude, and probe delay, NSHB is a powerful technique for investigating linear response and thermal recovery. Furthermore, because the responding degrees of freedom act as their own local probe, NSHB is the most direct technique available for investigating the net nonexponential relaxation of bulk materials.

We measure magnetic NSHB using a SQUID magnetometer that is optimized for fast response. The 5%

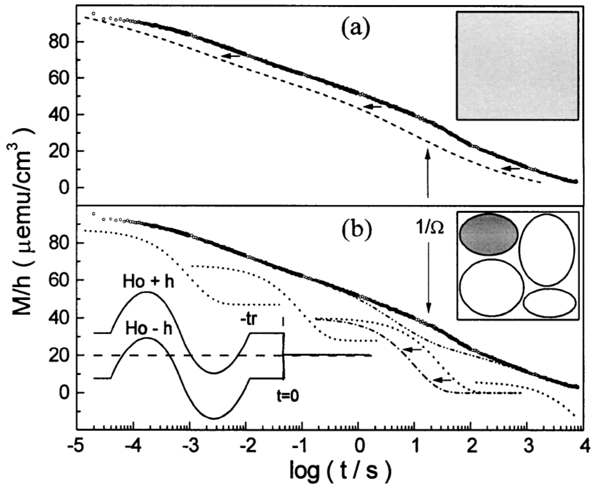

FIG. 1. Sketch of (a) homogeneous and (b) heterogeneous scenarios for NSHB. The lower left inset shows the pump oscillation at frequency Ω and amplitude H_0, with the probe step of amplitude $+h$ after a recovery time t_r, and the phase-cycled step of $-h$. The dashed curve in (a) shows how the response of a homogeneous sample would be uniformly shifted to shorter times. The dotted curves in (b) depict the distribution of relaxation times of a heterogeneous system, and the dot-dashed curves show how the pump oscillation would selectively modify the response.

Au:Fe sample was prepared using standard procedures [18] by melting pure (>99.997%) constituents in an induction furnace, annealing at 900 °C in an Ar/H atmosphere for one day, then holding at 550 °C for $\frac{1}{2}$ h, before drop-quenching into liquid nitrogen. The inset in Fig. 2 shows the sharp cusp in the magnetization which defines $T_g = 21.6$ K. The relaxation data were obtained by [inset in Fig. 1(b)] applying a magnetic field of $+h = 8$ Oe at a temperature of 25 K, field-cooling to a measurement temperature $T < T_g$, waiting 120 s in the field-cooled state, applying a pump oscillation of frequency Ω and amplitude H_0, waiting a recovery time t_r, then removing $+h$ and measuring the response as a function of time. The sequence is repeated with an identical pump oscillation but negative step $-h$, so that the difference between the two spectra removes the larger but less interesting linear response from the pump oscillation, leaving the step response of a sample that was selectively modified, which is compared to a similar sequence of measurements with no pump oscillation. The process is repeated with the sample in the counterwound coil of the flux transformer, so that the difference between the two sets of spectra removes any background signal from the magnetometer. Each group of four modified spectra is shifted to coincide with the group of unmodified spectra at $t = 1$ ms or 100 s, depending on the location of the spectral hole. Thus, each difference spectrum (ΔM) comes from eight separate relaxation spectra.

Figure 2(a) is a semilogarithmic plot of several relaxation spectra taken at $T = 18.8$ K. The open circles show the unmodified response, which starts in the μs time range, is roughly logarithmic over the ms time range, then becomes a stretched exponential after 10 s. More importantly, the other symbols show the response after modification by an oscillation with $H_0 = 96$ Oe at 30 Hz, 1 Hz, and 30 mHz. Even in the raw data it is possible to see that the 30 Hz oscillation modifies most at short times, the 30 mHz oscillation modifies most at long times, while the 1 Hz oscillation modifies most in the middle. The difference spectra [Fig. 2(b)] show that the peak modification closely corresponds to the time scale of each pump frequency and that there is essentially no overlap in the modification between the 30 Hz and 30 mHz oscillations. This clearly establishes that the different oscillations have modified distinct degrees of freedom and provides direct evidence that the bulk spectrum is heterogeneously broadened. The solid curves come from a model of simple exponential relaxation and recovery, with an amplitude factor as the only adjustable parameter, to be described below.

Figure 3 shows the modified response and difference spectra at $T = 19.6$ K after a pump oscillation of 0.1 Hz with several different amplitudes. The inset shows that for $H_0 < 80$ Oe the size of the spectral hole is proportional to the energy absorbed from the pump oscillation, $Q = \frac{1}{2}\chi'' H_0^2$, confirming that this NSHB is nonlinear in H_0 as needed to distinguish the nature of the response. At $H_0 \approx 80$ Oe there is an abrupt change in the slope. The cause of this change is evident from the relaxation spectra,

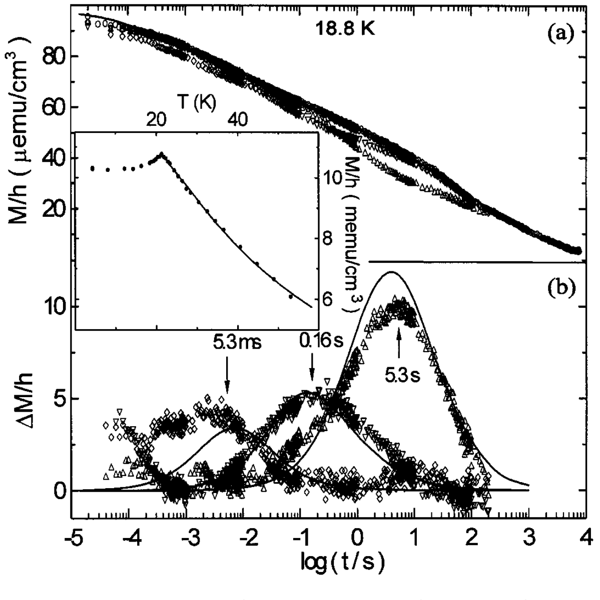

FIG. 2. Magnetization of 5% Au:Fe as a function of temperature (inset) shows a sharp cusp at $T_g = 21.6$ K. (a) Equilibrium magnetic relaxation (○), and relaxation after modification by a single oscillation of $H_0 = 96$ Oe at $\Omega/2\pi = 30$ Hz (◇), 1 Hz (▽), and 30 mHz (Δ). (b) Each difference spectrum shows a peak modification near the time corresponding to the oscillation, $1/\Omega$. The solid curves are from a model for a distribution of simple relaxation times shown in Fig. 4.

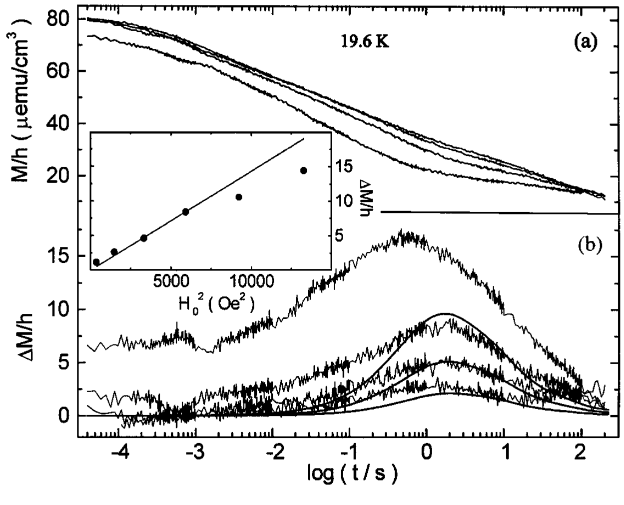

FIG. 3. (a) Magnetic relaxation and (b) difference spectra for pump amplitudes of $H_0 = 38$, 58, 77, and 115 Oe. The inset shows that the hole depth increases quadratically with field for $H_0 < 80$ Oe (solid line), but less rapidly for $H_0 > 80$ Oe. After the largest pump oscillation, a constant offset in the spectra at short times indicates that some of the slow response is missing from the experimental time window, while the rest of the sample remains a spin glass. The solid curves in (b) are from the model shown in Fig. 4.

which show a constant offset at short times, indicating that some slow response has been abruptly eliminated from the experimental time window. Thus, the pump oscillation is no longer a small perturbation on domains with fixed energy barriers. Indeed, selected degrees of freedom which used to relax at $t > 1$ s now relax at $t < 10$ μs, suggesting that they have been heated at least 2 K to above T_g. These domains now exhibit quasiequilibrium paramagnetic behavior, while the rest of the sample remains a spin glass; and this selective local heating lasts for hundreds of seconds. Of course, due to thermal diffusion and contact to the cryostat, the heat does not significantly change the phonon temperature but can be thought of as a persistent change in a local effective temperature $T_{s,i}$, comparable to a local spin temperature.

Measurements as a function of t_r explore the recovery of the specific degrees of freedom that were heated out of equilibrium, similar to a selective waiting-time dependence. Figure 4 shows the recovery at $T = 19.6$ K after two different pump frequencies. First note that the spectral holes do not broaden significantly during recovery, indicating that the modified degrees of freedom do not couple strongly to their neighbors, suggesting that the recovery is governed primarily by coupling to a common thermal bath. Next note that the peak modification recovers on a time scale that is about an order of magnitude slower than the characteristic time for the pump oscillation, empirically similar to a stretched exponential (solid curve in the inset). However, because the peak shifts toward longer times as it recovers, the behavior can be accounted for by a model of intrinsically exponential relaxation and recovery, shown by the smooth curves in (a) and (b).

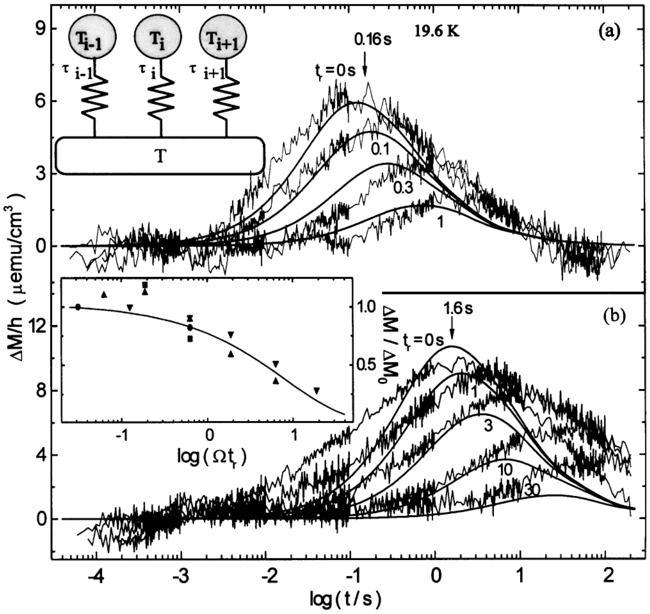

FIG. 4. Difference spectra for various recovery times after pump oscillations of (a) 1 Hz and (b) 0.1 Hz. The inset in (b) shows the normalized peak in the spectral hole as a function of scaled recovery time for pump oscillations of 100 Hz (■), 10 Hz (◆), 1 Hz (▲), and 0.1 Hz (▼). A stretched exponential $\Delta M \sim \exp[-(t_r\Omega/8)^{0.6}]$ (solid curve) mimics the recovery. The inset in (a) is a sketch of a model for independent domains, each with a local effective temperature $T_{s,i}$, and a local relaxation rate $1/\tau_i$ that is equal to the local recovery rate for heat flow to a thermal bath at temperature T. The solid curves in (a) and (b) are from this model.

The model [inset in Fig. 4(a)] is similar to that used for glass-forming liquids [19]. Each domain (i) is described by a local relaxation time τ_i, a local spin temperature $T_{s,i}$, and a local recovery rate $1/\tau_i$ via heat flow to the thermal bath at temperature T. The equilibrium response is characterized by a distribution (g_i) of simple exponentials, $M(t) = \sum_i g_i e^{-t/\tau_i}$, with 30–40 different time constants used to ensure smooth response. The g_i are found by fitting to the unmodified relaxation using a specific theory [20] for finite-sized clusters with a Gaussian distribution of energy levels, yielding, e.g., the solid curve in Fig. 2(a), but any distribution of exponentials that provides similar agreement with $M(t)$ would suffice. Over the temperature range of interest (21–19 K), the logarithm of the net relaxation time is found to increase gradually with decreasing temperature, while the spectrum spreads out from a fixed point at $\tau_0 \approx 30$ μs. Assuming that the local relaxation times exhibit a temperature dependence that is similar to the bulk, their behavior is approximated by the empirical expression $\Delta \ln(\tau_i) = -\Delta T_{s,i} B \ln(\tau_i/\tau_0)$, with $B \approx 0.2$ K^{-1}. Changes in the local effective temperature ($\Delta T_{s,i} = T_{s,i} - T$) are determined by a separate differential equation for each domain,

$$d\Delta T_{s,i}/dt = (\Omega/\Delta c_p)\chi_i''\{H_0 \sin[\Omega(t + t_r)]\}^2 - \Delta T_{s,i}/\tau_i \,, \tag{1}$$

where the sinusoidal term [which exists only during the field cycle, $-2\pi < \Omega(t + t_r) < 0$] gives the energy absorbed from the field, and the subtracted term gives the net energy flow to the thermal bath. The local magnetic absorption comes from the Fourier transform of the equilibrium response $\chi_i'' = g_i \Omega \tau_i/[1 + (\Omega\tau_i)^2]$, leaving the excess specific heat per Fe atom (Δc_p) as the only parameter that is not yet determined by independent measurements. Good overall agreement is obtained with $\Delta c_p/k_B = 5 \times 10^{-7}$. Such a small value for Δc_p explains why NSHB is possible with $H_0 <$ 100 Oe, and why no clear signature for the transition has ever been seen in the specific heat of Au:Fe. Even after assuming that only 1% of the Fe atoms participate in the slow response near T_g [as suggested by the relative magnitudes of the in-field susceptibility and zero-field remanence shown in Fig. 2(a)], this is still 3 orders of magnitude smaller than the excess specific heat per Mn atom measured for a 0.28% Cu:Mn sample $\Delta c_p/k_B = 3 \times 10^{-2}$ [21]. However, at least part of this difference is expected for the more highly ordered Ising-like spins in our relatively concentrated Au:Fe sample.

The solid curves in Figs. 2–4 show that the model gives generally good agreement with all prominent features in the response and recovery. The main discrepancy is that the measured spectral holes are consistently broader than the model. Much of this broadening can be attributed to known effects, such as the spectral width of the pump oscillation and changes in χ'' as the hole develops, which are neglected in the basic model presented here.

A remaining question is how to reconcile previous measurements favoring hierarchical dynamics with our results. For example, aging measurements after a temperature jump (where the dynamics is monitored as excess heat flows into or out of the slow degrees of freedom) have been interpreted in both the hierarchical [8] and droplet [9] pictures, whereas NSHB (which differs mainly in that the dynamics is monitored as excess heat flows out of *selected* slow degrees of freedom) shows that the response is accurately described by a static distribution of single-exponential relaxation times. One answer is that different spin-glass systems may exhibit different dynamics, and indeed Au:Fe has been found to have relatively long-lived local relaxation rates [22]. Another explanation may be the inadequacy of the picture of droplets in their ground state, suggesting the importance of considering clusters with internal degrees of freedom such as hierarchical droplets [9] or dynamically correlated domains [10]. Specifically, the distribution of relaxation times used to obtain the solid curves in Figs. 2–4 comes from a theory for the mean-field behavior of finite clusters [20], which combines some aspects of both the hierarchical and domain pictures. The final answer will require additional measurements on several systems.

In summary, nonresonant spectral hole burning has been achieved in a spin glass. As a function of pump frequency, NSHB establishes that the response is heterogeneously broadened. As a function of amplitude, the pump process is shown to be consistent with selective local heating. As a function of recovery time, the monotonic return to equilibrium of the selected slow degrees of freedom indicates that they couple to a common thermal bath, with negligible direct coupling to the other slow degrees of freedom. All salient features can be accounted for by a model of discrete domains, with a distribution of local relaxation rates that are equal to the local recovery rates. Thus, the model has the appealing features that the local response rate is equal to the rate at which energy flows into and out of a domain and that each local domain is governed by simple exponential relaxation for all of its slow response and recovery.

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