

Tracking spin-glass barriers versus field and temperature in  $a\text{-Gd}_{0.19}\text{Si}_{0.81}$ 

Vladimir Orlyanchik and M. B. Weissman

*Department of Physics, University of Illinois at Urbana-Champaign, 1110 West Green Street, Urbana, Illinois 61801-3080, USA*

E. Helgren,\* L. Zeng, and F. Hellman

*Department of Physics, University of California, Berkeley, California 94720-7300, USA*

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Detailed signatures of the states of the low-temperature spin-glass phase in  $a\text{-GdSi}$  films were followed using resistance fluctuations. Two-state switches between different spin-glass configurations were unambiguously identified by the field dependences of the occupation ratios of the states. The temperature dependences of the rates and occupation ratios were tracked in order to determine thermodynamic parameters of the switchers and the temperature dependences of barriers. A reasonable distribution of energy and entropy differences between states was found. Most importantly, the kinetic barriers were found to show no sign of divergence with lowering temperature, contrary to expectations based on prior theoretical and experimental works.

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## I. INTRODUCTION

Within the spin-glass phase, there are slow dynamics due to switching of spin configurations between different metastable states. The most basic properties of these metastable states remain poorly characterized even after many decades of research.<sup>1-4</sup> One key question concerns whether the kinetic barriers between different states strongly grow (perhaps diverging) upon lowering temperature, as suggested by the interpretation of some aging experiments on bulk materials (e.g., Ref. 5) and by some theoretical pictures.<sup>2</sup> In some descriptions, supported by somewhat indirect experimental evidence on field dependence, the growth of barriers on aging is associated with the gradual spatial growth of spin-glass domains.<sup>5-7</sup> On the other hand the hierarchical picture of temperature-dependent barriers (e.g., Ref. 2) is primarily inspired by the properties of the Parisi solution of the infinite-range model<sup>8,9</sup> for which spatial patterns are irrelevant.

Mesoscopic resistance fluctuation experiments on the spin-glasses AuFe (Ref. 10) and on CuMn doped with Au (Ref. 11) have found individual switchers with the Arrhenius kinetics, i.e., fixed barriers, within the temperature range in which most of the resistance fluctuations are due to spin-glass configuration fluctuations.<sup>12</sup> Since none of these individual switchers was also tracked versus field, their identification as individual spin-glass fluctuators was not quite certain enough to justify re-examining the explanations for the aging experiments. In this paper, we report detailed field and temperature dependences of the kinetic and thermodynamic properties of switchers in films of a nearly insulating spin glass, finding that clearly identified spin-glass switchers show no sign of diverging barriers.

## II. EXPERIMENTAL TECHNIQUES

We studied films of  $a\text{-Gd}_x\text{Si}_{1-x}$  with  $x=0.19$  and thickness  $d=100$  nm, made by electron-beam coevaporation under UHV conditions onto amorphous Si-N coated Si substrates held at room temperature, as described elsewhere.<sup>13,14</sup> Previ-

ous studies<sup>13-15</sup> showed that for  $0.04 < x < 0.19$  such films are classical spin glasses with sharp cusps in susceptibility [at about  $T=6.5$  K for  $x=0.19$  (Ref. 13)] driven by mixed ferromagnetic and antiferromagnetic Gd-Gd interactions mediated by conduction electrons.<sup>16</sup> Although neutron-scattering studies indicated some short-range ferromagnetic correlations,<sup>16</sup> neither extended x-ray-absorption fine structure<sup>17</sup> nor high-resolution TEM (Ref. 18) find any Gd clustering for  $x < 0.25$ . Transport measurements in  $a\text{-Gd}_x\text{Si}_{1-x}$  films revealed both insulating and metallic properties for  $x < 0.13$  and  $x > 0.14$ , respectively.<sup>19</sup> We chose this material because its resistivity ( $\sim 2.5$  m $\Omega$  cm at low  $T$ ) is high compared to more standard metallic spin glasses, which makes it more suitable for transport noise experiments.

The samples were patterned into a six-probe configuration containing two arms, each of length 1.5 mm and width 0.5 mm usable in a Wheatstone bridge configuration. A central part of one of the arms was narrowed down using a focused ion beam to form a small constriction with width about 0.5  $\mu\text{m}$  and length about 1.7  $\mu\text{m}$  containing about  $10^9$  Gd atoms. We generally used an ac noise measurement technique<sup>20</sup> (employing five contacts of the pattern) with a driving frequency of 2140 Hz. The other two bridge arms consisted of two external resistors, one of which was connected in parallel to a capacitor for phase adjustment. Careful adjustment of the external resistors and capacitor allowed us to null the bridge imbalance voltage to about  $10^{-5}$  of the total voltage drop. The imbalance-voltage noise was dominated by the fluctuations of the *resistance* ( $R$ ) of the narrow constriction since the other arm of the bridge has much larger width. To prevent effects due to Joule heating we used a current density of  $\sim 10^4$  A/cm<sup>2</sup> in the narrow constriction and checked to make sure that heating produced an imbalance of less than about 0.02% of  $R$ , corresponding to a maximum increase of about 20 mK in  $T$  of the constriction. We also checked the noise data using dc and a four-probe configuration, finding initially similar noise results except that the samples degraded over the course of days under the higher dc bias.

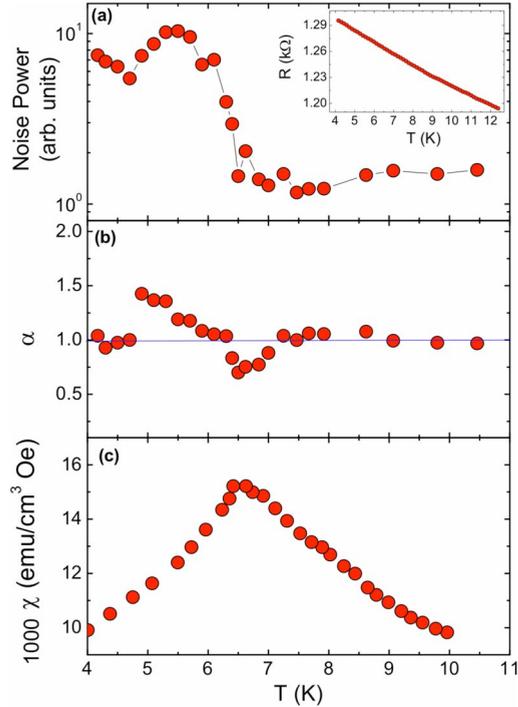


FIG. 1. (Color online) Temperature dependence of (a) noise power  $S_R(f)$  integrated over the frequency range of  $0.59 < f < 10.38$  Hz, inset:  $R$  vs  $T$  of the bridge arm including a small constriction. (b) Noise spectral slope  $\alpha$  (see text for definition) and (c) magnetic susceptibility measured at 4 Oe and 135 Hz.

The imbalance voltage was measured using low-noise voltage preamplifiers followed by a homemade lock-in amplifier<sup>21</sup> with very low phase noise. To measure the spectral density  $S_R(f)$  of the resistance noise, i.e., the squared Fourier transform of the time-dependent  $R$ , the lock-in output was passed through an ac-coupled low-pass antialias filter (SRS 640) before digitizing at rates from 200 to 800 Hz, giving usable voltage-noise spectral data in the frequency range from 0.2 Hz up to 400 Hz depending on the sample and temperature. Background voltage noise spectra were taken with no current and subtracted from the measured voltage spectra to obtain data which could be converted to  $S_R(f)$ . The dc-coupled output of the lock-in was also recorded to obtain lower-frequency data.

All measurements were performed with the sample in an evacuated can immersed in liquid He either in a standard cryostat with a superconducting magnet or in a storage dewar for longer-time measurements. A metallic arm connected to the can provided a strong coupling between the sample holder and the liquid-He bath. Good temperature stability ( $|\Delta T| < 0.3$  mK) was consistently measured at the Cernox thermometer.

### III. RESULTS AND ANALYSIS

We begin describing the results with data showing that the low- $T$  noise in  $R$  is dominated by fluctuations of the spin-glass magnetic configurations, as in other materials.<sup>10–12,21–27</sup>

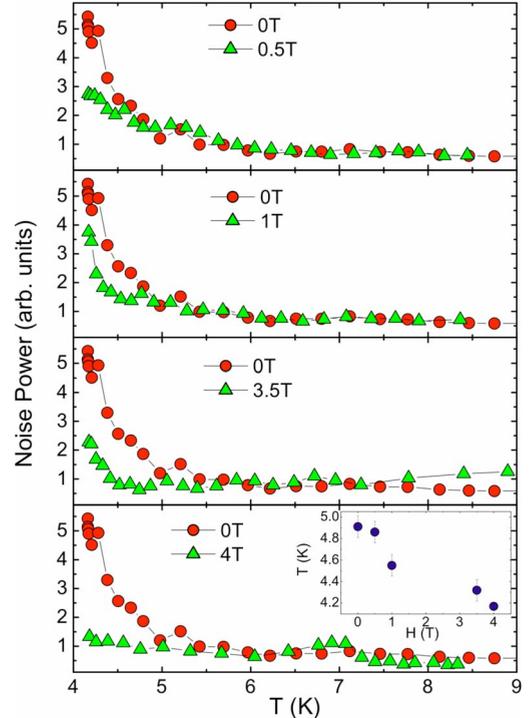


FIG. 2. (Color online) Noise power, from 1.3 to 27.1 Hz, versus temperature at different magnetic fields. Each panel compares  $H = 0$  and  $H > 0$  results. Data were taken on cooling with in-plane field orientation at  $dT/dt \sim 0.2$  K/min. The inset shows the temperature at which the power crosses an arbitrary threshold, 1.5 in these units. Error bars on the  $H = 4$  T point extend to  $T = 0$ .

Figures 1(a) and 1(b) show the dependence of  $S_R(f)$  and noise-spectral slope  $\alpha \equiv -\frac{\partial \ln S_R(f)}{\partial \ln(f)}$  on  $T$ . Above  $T \approx 6.5$  K we found ordinary  $1/f$ -like ( $\alpha \approx 1$ ) noise with small temporal variations in the noise power. Around  $T \approx 6.5$  K,  $S_R(f)$  increases considerably and  $\alpha$  reaches its minimum ( $\alpha \approx 0.5$ ). For  $T < 6.5$  K,  $S_R(f)$  remains more than an order of magnitude bigger than at higher  $T$ , while  $\alpha$  shows large variations. These abrupt changes in  $S_R(f, T)$  below  $T \approx 6.5$  K contrast with very smooth behavior of  $R(T)$  itself (see inset). The sharp increase in  $S_R$  occurs in the vicinity of the temperature of the cusp in ac magnetic susceptibility [Fig. 1(c)] near the transition to a spin-glass phase at  $T_g$ .

The strong dependence of the noise amplitude on magnetic field,  $H$ , as shown in Fig. 2, confirms that the increased  $S_R$  in the spin-glass regime is in fact due to magnetic fluctuations. In this experiment the sample was heated from 4.2 to  $T > 9.5$  K (well above  $T_g$ ) and  $H$  was applied. Then  $S_R(f)$  was measured while slowly cooling the system in constant  $H$ . The whole procedure was repeated for different values of  $H$ . Larger  $|H|$  generally resulted in lower temperatures for the onset of increased noise, similar to previous results on a standard metallic spin glass.<sup>25</sup> In the range of  $|H| \leq 4$  T we confirm<sup>14</sup> that the changes in magnetoresistance are well under 1% and hence are irrelevant to the dramatic  $H$  dependence of the noise. The decrease in the noise onset  $T$  with increase in  $H$  roughly resembles theoretical predictions for the field-dependent  $T_g$  (Ref. 1) and the experimentally observed kinetic crossover in conventional metallic spin glasses

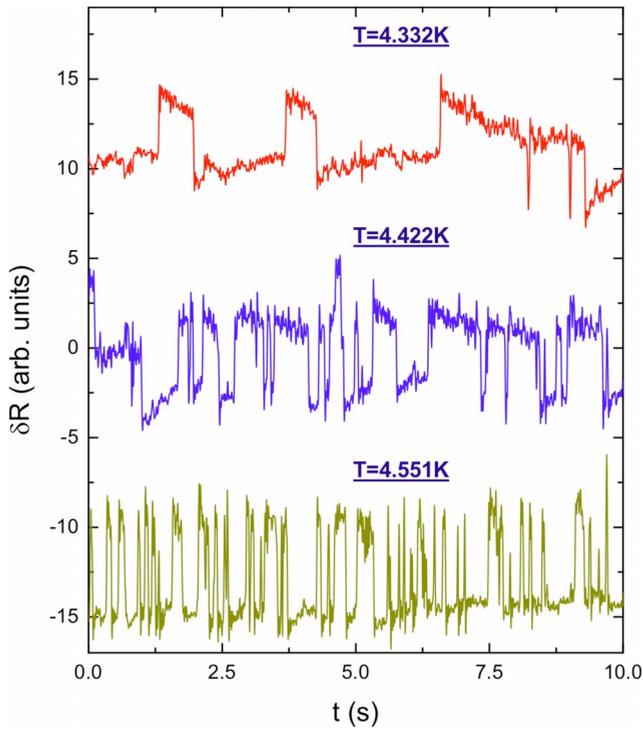


FIG. 3. (Color online) Imbalance voltage of an individual switcher at several temperatures.

(e.g., Ref. 28). This confirms again the spin-glass origin of the noise.

The noise in these samples was distinctly non-Gaussian including many discrete switchers, some of which stood out enough from other noises to be tracked individually versus  $H$  and  $T$ . Figure 3 illustrates the raw time traces of one such switcher. When, as in this case, two clear levels can be discerned, the ratio ( $r$ ) of the time spent in the two states gives the free-energy difference  $\Delta F = k_B T \ln(r)$ , where  $k_B$  is Boltzmann's constant. Standard thermodynamic relations<sup>29</sup> then allow the extraction of the differences between the states' magnetic moments ( $\Delta\mu$ ), energies ( $\Delta U$ ), and dimensionless entropies ( $\Delta\sigma$ ) from the  $H$  and  $T$  derivatives of  $\ln(r)$ . These relations allow one to check that a given fluctuator has reasonable properties to be a magnetic spin-glass configuration change.

Figure 4 shows examples of the  $T$  dependences of several fluctuation properties for two fluctuators. Figure 5 shows  $H$  dependences. The evidence that essentially the same fluctuator is involved throughout the  $H$  and  $T$  ranges in these cases is that there are no gaps or two-switcher overlap regions in these dependences and that the magnitude of the switches in  $R$  remains approximately constant over the whole range. When checked, the values of  $\ln(r)$  return to the starting value when  $H$  or  $T$  is returned to its starting value. In long runs at fixed conditions, no drifts in  $\ln(r)$  outside the error bars were found so that, within this accuracy, the dependences on  $T$  and  $H$  can be assumed to be thermodynamic. In each case the dependence of  $\ln(r)$  on  $1/T$  is linear to within experimental accuracy so the fluctuators seem to be well characterized by single values of  $\Delta U$  and  $\Delta\sigma$ . In some cases the field dependences are linear and thus can be fit with fixed  $\Delta\mu$  but in

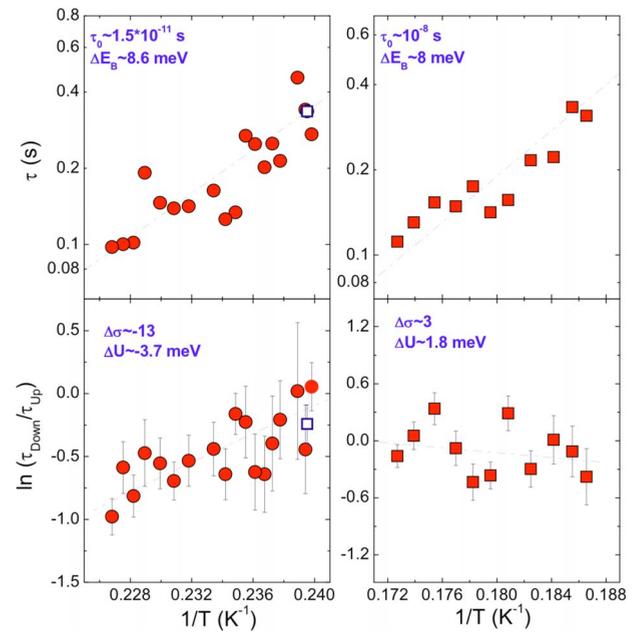


FIG. 4. (Color online) The characteristic net switching time and the natural logarithmic of the ratio of the times spent in the two states are shown as a function of inverse temperature for two different switchers along with inferred kinetic and thermodynamic parameters, as described in the text. Open symbols show data taken on return to the initial  $T$ .

other cases deviations from linearity are evident, as in Fig. 5. These variable  $\Delta\mu$  indicate that at least one of the two metastable states is in fact an ensemble of configurations with a range of different moments.

The thermodynamic barriers ( $\Delta E_B$ ) to rearrangement are, for the Arrhenius fluctuators with typical attempt rates  $\tau_0$ , around  $10^{12}$  Hz and fluctuation rates in our experimental range (e.g., 10 Hz) of order  $25k_B T$ . Since these barrier heights are large compared to the observed  $\Delta U$  values, it is a decent approximation to treat the characteristic fluctuation rate (the sum of the forward and backward rates) as itself having a simple Arrhenius form characterized by a single barrier height and attempt rate. Examples of these rates are also shown in Figs. 4 and 5 as a function of  $H$  and  $T$ .

Table I shows the collection of all measured thermodynamic and kinetic dependences of switchers in these samples. In some cases we do not have clear enough data to determine thermodynamic properties of a switcher, but the characteristic frequency of its Lorentzian contribution to  $S(f)$  can still be tracked, so just the kinetic properties are listed. In one case, which is not listed, only the  $H$  dependence of the rate was measured, from which it was inferred that the transition state had a moment some  $750\mu_B$  different from the metastable states.

In all cases the measured  $H$  dependences indicate moment differences in the range of  $(130-800)\mu_B$  corresponding to some 20–120 Gd moments, roughly as one would expect for configuration changes involving the order of  $3 \times 10^3$  randomly oriented moments. The entropy differences are small but in some cases not within the error bars of zero, again as anticipated for fluctuations between states with no simple

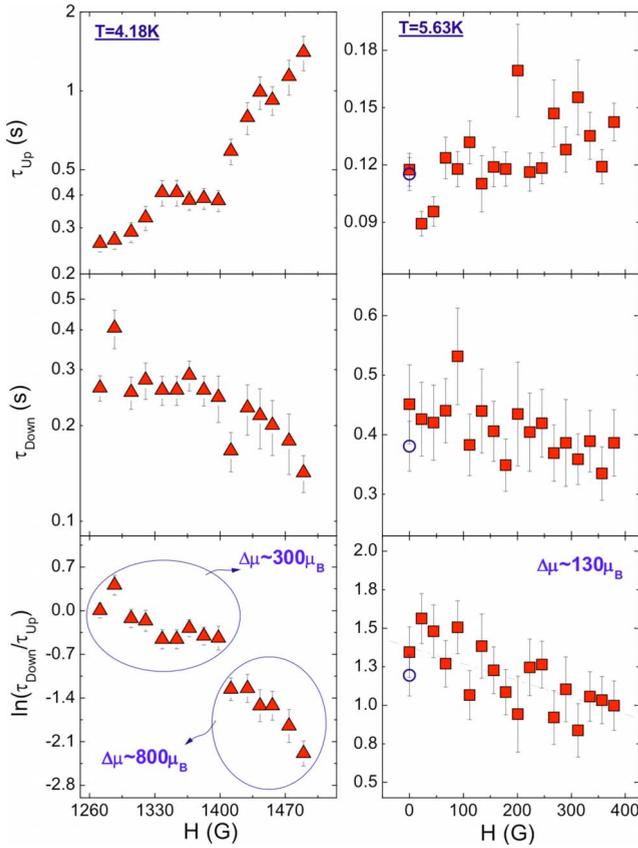


FIG. 5. (Color online) The characteristic times spent in each state and the ratios of those times are shown as functions of the applied magnetic field along with the inferred moment differences between the two states. Open symbols show points taken on return to  $H=0$ .

symmetry relation. In principle such data could be used to determine ratios of exponents for scaling of entropy and energy differences with fluctuator size but only with much more extensive sets.

Although, as noted above, the switchers are in many cases not simple two-state systems, they all share approximately

Arrhenius temperature-dependent kinetics. Although noise data allow tracking over only a small log range of frequencies and hence can obscure departures from the simple Arrhenius form, growth of barriers as  $T$  is lowered would show up also as anomalously strong  $T$  derivatives of rates. In the Arrhenius fits that effect gives anomalously large attempt rates. We found no such anomalies.

The slow spin-glass kinetics in  $a$ -GdSi films show no sign of diverging barriers as  $T$  is lowered. The kinetics in fact closely resemble the more fragmentary data on AuFe (Ref. 10) and CuMnAu (Ref. 11) spin glasses in which the magnetic nature of the individual Arrhenius switchers was not directly shown. Therefore we conclude that diverging barriers are absent in a fairly wide variety of mesoscopic spin-glass films.

IV. DISCUSSION

Our data are not sufficient to resolve between two possible interpretations. One would be that diverging barriers (never directly observed) were not really required to explain the strong asymmetry in temperature dependences found in aging experiments on bulk materials.<sup>5</sup> Another would be that the finite thicknesses of the mesoscopic film samples suppress the barrier divergence by preventing the growth of long-range correlations.<sup>6,7,30</sup>

The onset of  $1/f$  noise in our experimental frequency range near  $T=6$  K indicates that, even for fixed barriers, the range of characteristic times at 4.2 K would extend far beyond the longest times involved in any aging experiments. Thus if finite-size effects account for the lack of barrier growth in the mesoscopic samples, they do so by some more subtle path than simply limiting the maximum barrier height. This conclusion may be related to the observation that the growth of correlation volumes inferred from aging kinetics does not follow the Arrhenius temperature dependence of simple thermally activated processes,<sup>6</sup> unlike the kinetics directly appearing in the magnetic susceptibility or resistance noise. The key experiments to test the role of finite-size effects in limiting barrier growth would still be aging experi-

TABLE I. The measured thermodynamic and kinetic parameters of the fluctuators along with approximate error bars. For fluctuator (1) the  $H$  dependence was measured several hours apart from the  $T$  dependence but the rates and magnitude indicated it was the same fluctuator. The high- $R$  state is arbitrarily defined as the base for each of the two-state differences.

	$\tau_0$ (s)	$\Delta E_B (\pm 0.5)$ (meV)	$\Delta\sigma (\pm 7)$	$\Delta U (\pm 0.5)$ (meV)	$\Delta\mu (\pm 50)$ ( $\mu_B$ )
1	$1.5 \times 10^{-11}$	8.6	-13	-3.7	400 <sup>a</sup>
2	$10^{-8}$	8	3	1.8	130
3	$10^{-12}$	9.5	-15	-6.5	
4	$10^{-14}$	10.8			
5	$10^{-13}$	10.3			
6	$10^{-14}$	12.3	0	0	
7	$3 \times 10^{-10}$	10	-2	-1	200
8					300-800

<sup>a</sup>For fluctuator (1).

ments on material with finite-size effects, as proposed earlier,<sup>7</sup> but such experiments are quite difficult to perform on films. Meanwhile, our data show that within mesoscopic regions, the sort of regions for which an infinite-range model might be a decent approximation, the kinetic barriers show no signs of divergence as the temperature is lowered.

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\*Present address: Department of Physics, California State University, East Bay.

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