Effect of anisotropy on $1/f$ noise measurements of CuMn spin glasses

David C. Harrison,[1] E. Dan Dahlberg,[1] and Raymond L. Orbach[2]
[1]School of Physics and Astronomy, The University of Minnesota, Minneapolis, Minnesota, 55455, USA
[2]Texas Materials Institute, The University of Texas at Austin, Austin, Texas 78712, USA

(Received 25 July 2019; revised manuscript received 5 August 2019; published 19 August 2019)

The effect of systematic Au doping on the onset of enhanced $1/f$ noise in the electrical resistance fluctuations of CuMn spin-glass alloys is reported. The purpose of the Au doping is to add a unidirectional anisotropy to that already present from the Mn in Cu. We find that the ratio of the noise onset temperature to the spin-glass temperature is not affected by the increase in anisotropy.

DOI: 10.1103/PhysRevB.100.064411

I. INTRODUCTION

The fundamental nature of the spin-glass transition for Heisenberg spin systems remains one of the most vexing questions in condensed matter physics. For example, there is no question that the canonical spin-glass CuMn is a Heisenberg system, as any single-ion anisotropy would be small. However, for a Heisenberg system, the lower critical dimension is close to 3 [1] and the glass temperature T_g is expected to be very small. This is consistent with very large lattice simulations [2] that show "No matter how small the anisotropy, the asymptotic critical exponents are those of the Ising-Edwards-Anderson model." Thus, the spin-glass transition requires some anisotropy, and it could come from the magnetic ions themselves generating a unidirectional anisotropy through the Dzyaloshinsky-Moriya (DM) interaction [3,4], arising from the spin orbit coupling contrast of the Mn to the Cu host matrix. Thus, any metallic spin glass would be expected to exhibit Ising-like behavior, leading to the observed T_g values. This has been supported by measurements of the critical exponents in CuMn that show good agreement with simulations for Ising spin glasses [5,6].

An intriguing question remains: If the DM anisotropy results in Ising-like behavior near T_g for a Heisenberg spin system, then what happens to the transverse moments? Elderfield and Sherrington [7] derived a phase diagram for spin glasses with uniaxial anisotropy, schematically reproduced in Fig. 1. They found a phase diagram that exhibits a regime of longitudinal freezing immediately below T_g. For small uniaxial anisotropy, they predicted that the transverse moments would freeze at a temperature near, though slightly lower than, T_g. As the uniaxial anisotropy increases, the temperature difference between the two phase transition lines increases.

Experimentally, we cannot systematically add a uniaxial anisotropy to CuMn, but we can increase the unidirectional anisotropy by doping with Au, soluble in Cu. Adding Au to CuMn substantially increases the DM anisotropy because of the much larger spin orbit coupling contrast of Au as compared to Mn. An early example is found in de Courtenay *et al.* [6] where they measured the nonlinear magnetization of CuMn and AgMn alloys doped with varying amounts of Au impurities.

In addition, Prejean *et al.* showed that Au increases the anisotropy of CuMn by measurements of the magnetic hysteresis taken in the spin-glass state [8]. In this case, the addition of 0.15 at.% Au to a sample with 1 at.% Mn broadens the width ΔH of the hysteresis cycle from 200 Oe to 1000 Oe [8]. The width of the hysteresis loop is proportional to the anisotropy; the effect of the Au is, therefore, to increase the anisotropy. Additional measurements on CuMn alloys doped with other impurities (Al, Ag, and Pt) with different atomic masses show that the spin-orbit interaction is the unambiguous origin of this anisotropy, consistent with DM anisotropy.

Although expected, the freezing of the transverse moments in spin glasses has never been experimentally observed. Of the traditional measurement techniques that show a reasonably sharp signature associated with the spin-glass phase, all are sensitive to only the longitudinal freezing. We wished to see if this was also true for measurements of the $1/f$ noise, i.e., whether the noise was associated with the transverse or longitudinal freezing. An additional advantage of our transport measurements is that they can be conducted in zero applied magnetic field.

Previous work has demonstrated that there is a somewhat abrupt rise in the magnitude of the $1/f$ noise in CuMn and CuMnAu systems near the transition temperature [9–11]. However, the Au doping in those studies was not systematic and was over a relatively narrow range of Au concentrations. Additionally, direct measurements of the thickness-dependent freezing temperature, T_f, were not always made. In our work, we fabricate and perform measurements on samples systematically varying the Au concentration. By depositing large-area thin films, suitable for more conventional magnetometry, simultaneously with samples suitable for $1/f$ noise measurements, we are able to directly compare T_f (measured with conventional magnetometry) to the temperature at which the noise begins to rise, T_{noise}, in samples with varying anisotropy.

Previous publications ascribe the enhanced $1/f$ noise in the spin-glass state to universal conductance fluctuations (UCF), which couple the slow magnetic fluctuations (known to be $1/f$) of a sample in the spin-glass state to the electrical resistance of the sample [9–11]. In CuMn, the electrical

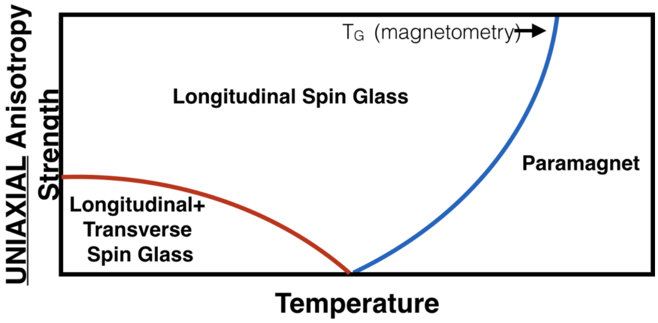

FIG. 1. Elderfield and Sherrington computed a phase diagram for spin glasses with uniaxial anisotropy, schematically reproduced below. The longitudinal transition occurs at T_g. When the uniaxial anisotropy is low, they predict the transverse moments will freeze at a similar, though slightly lower temperature. As the uniaxial anisotropy increases, the temperature difference between the two phase transitions increases.

resistance noise grows continuously as the temperature is reduced in the spin-glass state, expected (for CuMn) in UCF theory. The magnetic fluctuations are related to the imaginary part of the ac susceptibility by the fluctuation dissipation theorem. However, Israeloff noted that the resistance fluctuations are much less strongly affected by the application of magnetic fields than the imaginary part of the longitudinal susceptibility,[1] χ'' [9]. He noted that the relevant spin correlations are of fourth order and higher, and experiments sensitive to fourth-order correlations—such as EPR linewidths—are typically much less sensitive to the application of magnetic fields. In simple terms, this is because some fluctuations will appear in the noise but not in the magnetization. Weissman [12] makes a detailed argument for why this is the case. However, since the $1/f$ noise is related to the imaginary part of the susceptibility but shows behavior that differs from the imaginary part of the longitudinal susceptibility, we were motivated to test the possibility that the $1/f$ noise in the resistance is sensitive to transverse freezing.

II. SAMPLE PREPARATION

Samples were dc sputtered in Ar at a pressure of 2 mtorr. Multiple sputtering targets were used: $Cu_{86.5}Mn_{13.5}$, $Cu_{73}Mn_{13.5}Au_{13.5}$, and $Cu_{79.75}Mn_{6.75}Au_{13.5}$. All were stated to be 99.95% pure.[2] A fourth set of samples was produced by cosputtering from the $Cu_{86.5}Mn_{13.5}$ and $Cu_{73}Mn_{13.5}Au_{13.5}$ targets to produce an approximately $Cu_{73}Mn_{13.5}Au_{6.75}$ sample. Sample compositions, as determined by energy dispersive x-ray spectroscopy (EDS), differed slightly from the concentrations of the targets. The sputtering system reached a base pressure of under 10^{-7} torr. Deposition rates were approximately 0.5 nm/s (1 nm/s for the cosputtered sample). The system used 3″ diameter guns, angled toward a rotating stage, on which the sample substrates were placed off axis.

[1]We know of no measurements of the imaginary part of the transverse susceptibility, χ'' in CuMn spin glasses.

[2]Purchased from ACI Alloys.

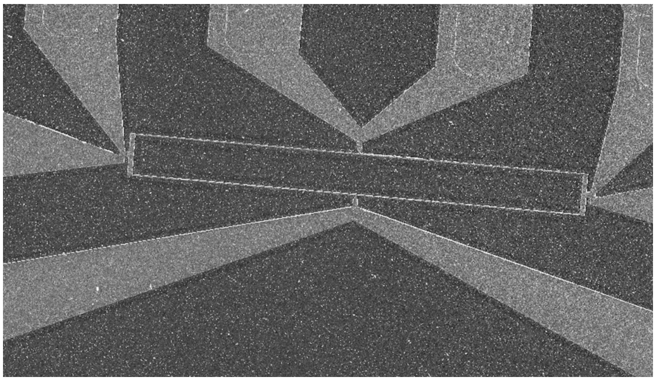

FIG. 2. SEM image showing sample layout. The width of each arm of the sample is approximately 300 nm and the total length of the sample is approximately 100 μm. Eight leads are seen attaching to the sample at four points.

This allowed the deposition of films with areas of larger than 18 square inches with an overall variation in thickness less than 10%.

For each Mn and Au concentration under consideration, we simultaneously deposited two sets of samples, each approximately 80 nm thick. One set was deposited onto six 1″ × 3″ glass slides which had been coated in MicroChemicals AZ 1505 Photoresist. The photoresist was then dissolved from glass slides, and the resulting metallic flakes were used for magnetometry measurements.

Simultaneous with the deposition of the first set of films, our $1/f$ noise samples were deposited onto Si_3N_4 substrates. After deposition, these samples were coated in PMMA resist and baked for 2 min at 180°C. We used electron-beam lithography to pattern our samples as shown in Fig. 2. We then deposited an aluminum hard mask and ion milled. Last, we dissolved the aluminum hard mask in KOH. It is worth noting that while the large area magnetometry films had a thickness variation of less than 10%, given the smaller dimensions of the noise measurements samples, they are expected to be very uniform.

III. MEASUREMENT TECHNIQUES AND RESULTS

We used the first set of films to determine T_f for our samples from the onset of irreversibility in our zero-field-cooled (ZFC) and field-cooled (FC) magnetization measurements. Using a Quantum Design MPMS system, we cooled from at

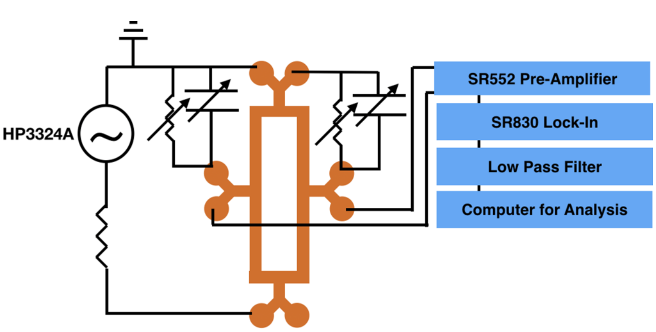

FIG. 3. Diagram of experimental setup.

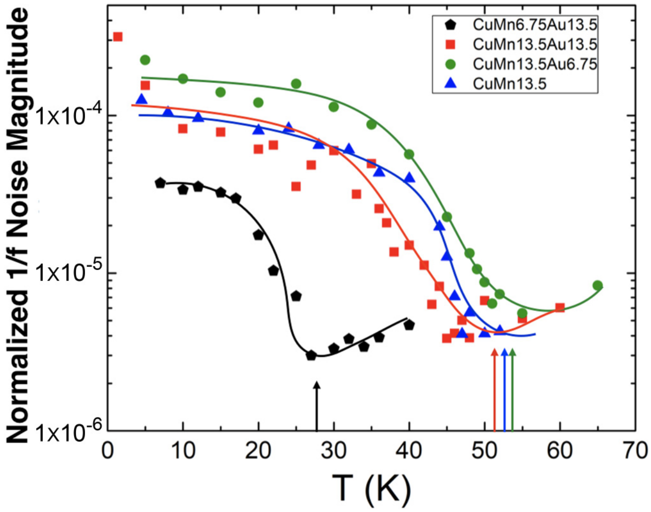

FIG. 4. $1/f$ noise magnitude as a function of temperature. The solid lines are to guide the eye, and the arrows along the abscissa indicate T_f, as determined by the onset of irreversibility in FC/ZFC measurements.

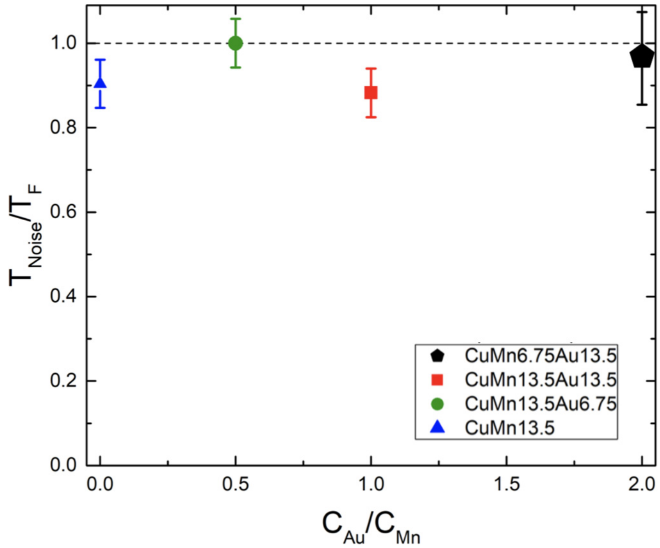

FIG. 5. T_{noise}/T_f vs $C_{\mathrm{Au}}/C_{\mathrm{Mn}}$. We see no systemic dependence on Au doping. (Applying the Elderfield and Sherrington theory for uniaxial anisotropy, we would expect a decrease in T_{noise}/T_f with an increase in $C_{\mathrm{Au}}/C_{\mathrm{Mn}}$.)

least 10 K above T_g to a temperature at least 10 K below T_f. For the ZFC magnetization measurements, we cooled in zero applied field and then applied a 100-G field and measured the magnetization on warming. For the FC measurements, a 100-G field was turned on prior to cooling from at least 10 K above T_g to a temperature at least 10 K below T_f and left unchanged. Again, we recorded the magnetization on warming. The onset of irreversibility, the temperature at which the FC and ZFC magnetizations begin to differ defines T_f [13].

We used the second set of samples to perform our transport measurements with an ac technique described in detail elsewhere, omitting the transformer [9–11]. In brief, by applying an ac current and demodulating using a lock-in amplifier, we moved near the minimum in the noise contour of an SR552 preamplifier, away from the low-frequency $1/f$ noise present in our preamplifier and all other electronics. By using a sample patterned as a Wheatstone bridge with the arms spaced closely together, we mitigated the effects of local temperature fluctuations. External balancing resistors and capacitors were used to null the voltage across the bridge. A diagram of our experimental setup is presented in Fig. 3.

In all of our noise samples, we observed a relatively sharp rise in the magnitude of the $1/f$ noise in the sample resistance. We call the temperature at which the noise increases in magnitude the noise onset temperature, T_{noise}. These results are consistent with previously published data [9–11] and are shown in Fig. 4 for the four samples investigated. Also shown in this figure is the magnetometry measured freezing temperature. While we present only the results of the four samples for which we simultaneously deposited large area films, we have seen the relatively sharp rise in the $1/f$ noise magnitude, consistent with the four samples shown, in more than 20 samples.

Because we were able to perform both noise and magnetometry measurements on films from a single deposition to determine both T_{noise} and T_f, we have confidence in the ratio T_{noise}/T_f, exhibited in Fig. 5 versus the ratio of the Au to Mn concentrations, $C_{\mathrm{Au}}/C_{\mathrm{Mn}}$. As can be seen from this data, we do not observe a dependence of the onset temperature of the $1/f$ noise with the addition of Au, with the concomitant increase in the DM anisotropy strength in this system.

We are able to use Eq. (11) from de Courtenay *et al.* [6] to calculate the anisotropy parameter used in the Elderfield and Sherrington phase diagram. In another work [14], Sherrington and Cragg compute a quantitative phase diagram to which we may more directly compare. For our largest anisotropy value ($C_{\mathrm{Au}}/C_{\mathrm{Mn}} = 2$), we would have expected a reduction in T_{noise} of 27% from the undoped T_f, well outside of our error bars. This does not take into account the additional increase in T_f predicted by Sherrington and colleagues [7,14], which we do not observe. This is consistent with de Courtenay *et al.* who measured only a 4% increase in T_f from an undoped CuMn sample to a sample with with a $C_{\mathrm{Au}}/C_{\mathrm{Mn}} = 1.85$ [6], where the Sherrington phase diagram predicts a 25% increase.

IV. CONCLUSIONS

As seen in Fig. 5, we find no systemic variation of T_{noise}/T_f with Au doping. The application of a magnetic field is expected to affect only the Ising component of the spin glass and has no significant effect on $1/f$ noise, which was the major reason to believe the $1/f$ noise would be sensitive to the transverse freezing. This null result lends further support to the suggestion [9] that the weak magnetic field dependence of the onset of enhanced $1/f$ noise in metallic spin-glass films is related to the dependence of the electrical resistance noise on spin correlation functions of at least fourth order. If this is not the case, then the phase diagram computed by Elderfield and Sherrington for a uniaxial anisotropy is not appropriate for systems with a unidirectional anisotropy.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, under Award No. DE-SC0013599. Parts of this work were carried out in the Characterization Facility, University of Minnesota, which receives partial support from NSF through the MRSEC program. Portions of this work were conducted in the Minnesota Nano Center, which is supported by the National Science Foundation through the National Nano Coordinated Infrastructure Network, Award No. NNCI-1542202. Part of this work was performed at the Institute for Rock Magnetism (IRM) at the University of Minnesota. The IRM is a U.S. National Multi-user Facility supported through the Instrumentation and Facilities program of the National Science Foundation, Earth Sciences Division, under Award No. 1642268, and by funding from the University of Minnesota.

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