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Field-pulse memory in a spin-glass

Devin C. Schmitt,[1] Joseph C. Prestigiacomo,[2] Philip W. Adams,[2] David P. Young,[2] Shane Stadler,[2] and Julia Y. Chan[1,2]
[1]*Department of Chemistry, Louisiana State University, Baton Rouge, Louisiana 70803, USA*
[2]*Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803, USA*

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We report a magnetic field-pulse memory effect in the temperature-dependent magnetization of $Tb_{30}Ru_{6.0}Sn_{29.5}$, a spin-glass material having a newly identified structure type. $Tb_{30}Ru_{6.0}Sn_{29.5}$ exhibits a glassy magnetization component of ~2290 emu/mol-Tb, which represents over 84% of its total saturation magnetization. We show that when a magnetic field pulse of a few hundred gauss is applied to the sample as it is cooling, the system retains a memory of the temperature at which the pulse was applied. Upon warming, the imprinted memory is observed as a precipitous drop in magnetization at the pulse temperature. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4818262]

An intense effort has been devoted to the classification and understanding of spin-glass materials over the past several decades.[1–15] Spin-glasses behave as non-equilibrium systems below the glassy transition temperature, leading to exotic magnetic properties including isothermal stretched-exponential relaxation[16] and frequency-dependent spin dynamics.[17] These slow dynamics have precluded their use as functional materials since spin-glass memory effects typically require hours of waiting time to achieve a measureable partial equilibrium state.[14,18,19] Here, we present nonequilibrium magnetization data on the newly discovered spin glass[20] $Tb_{30}Ru_{6.0}Sn_{29.5}$. We show that this material, which has a complex crystal structure containing many inequivalent magnetic sites, exhibits a non-equilibrium magnetic field pulse memory effect that is analogous to the memory-dip effect observed in other spin glass systems. We demonstrate that the low temperature metastability of the pulse memory is sufficient to store information that can be read upon warming.

X-ray diffraction data were collected on a single crystal of $Tb_{30}Ru_{6.0}Sn_{29.5}$ at 90 K using monochromatic Mo Kα radiation. See supplementary material for crystallographic and atomic parameters.[28] Crystal growth and structural details can be found in Ref. 20. Magnetization measurements were performed with H applied parallel to the crystallographic a-direction. Temperature- and field-dependent DC magnetization data were collected using a Quantum Design Magnetic Property Measurement System (MPMS) in fields up to 500 Oe. Temperature-dependent alternating current (AC) magnetization data were collected using a Quantum Design Physical Property Measurement System (PPMS) with an AC field of 10 Oe.

We have grown high quality single crystals of $Tb_{30}Ru_{6.0}Sn_{29.5}$ which exhibit a spin-glass transition at $T_g = 15$ K. Figure 1(a) shows a typical thermoremanent magnetization (TRM) measurement conducted at a temperature below T_g. A single crystal of $Tb_{30}Ru_{6.0}Sn_{29.5}$ was field-cooled at 500 Oe to 12 K at a rate of 0.3 K/min and held at that temperature for 1 h. The field was then reduced to zero, and the magnetization was measured as a function of time. As can be seen in Fig. 1(a), a sizable fraction of the magnetization evolved slowly with time. This time dependent fraction was well fit by the stretched exponential form given in the caption of Fig. 1. We estimate the maximum glassy component of the magnetization to be ~2290 emu/mol-Tb. The baseline magnetization immediately prior to field removal in the TRM measurement was 2715 emu/mol-Tb; thus, the time-dependent component of the magnetization accounts for over 84% of the total field-induced magnetization at 12 K. Figure 1(b) displays AC susceptibility measurements of $Tb_{30}Ru_{6.0}Sn_{29.5}$ which show a frequency dependence of the spin dynamics, indicated by a shift in the maximum of the data to higher temperatures with increasing frequency. These data represent definitive evidence that $Tb_{30}Ru_{6.0}Sn_{29.5}$ is a spin-glass system with a large glassy component.

Glassy magnetic behavior is typically attributed to magnetic frustration, either due to the disorder related to random couplings in a dilute magnetic system, such as described by the Heisenberg model,[21] or from the degeneracy caused by a geometrically frustrated antiferromagnetic ground state. These latter systems lack the ability to align antiferromagnetically with all nearest neighbors (Ising model).[21–23] Interestingly, $Tb_{30}Ru_{6.0}Sn_{29.5}$ does not possess any obvious structural source of geometric frustration nor is the level of disorder of our samples, as determined by x-ray analysis, sufficient to account for the glassy behavior. Alternatively, we speculate that the glassy behavior may be a result of the highly complex unit cell, shown in Fig. 2, which contains 11 inequivalent magnetic Tb sites.

In a canonical spin-glass, an interesting memory effect can be observed by taking advantage of non-equilibrium spin dynamics.[24] In a typical memory-dip experiment, a reference measurement is obtained by zero-field cooling (ZFC) a sample to a temperature below T_g. The sample is then warmed in a 10 Oe field, and the temperature-dependent magnetization is measured. The procedure is then repeated except that the sample is held for a period of time at a specific waiting temperature (T_w) before continuing down to the base temperature. Upon warming, a memory dip forms at T_w, graphically depicted in Fig. 3(a), in the temperature-dependent magnetization. This memory dip occurs due to the formation of local ordering (during the wait time) with energy-specific correlation lengths.[5,19] Multiple dips can be "written" upon

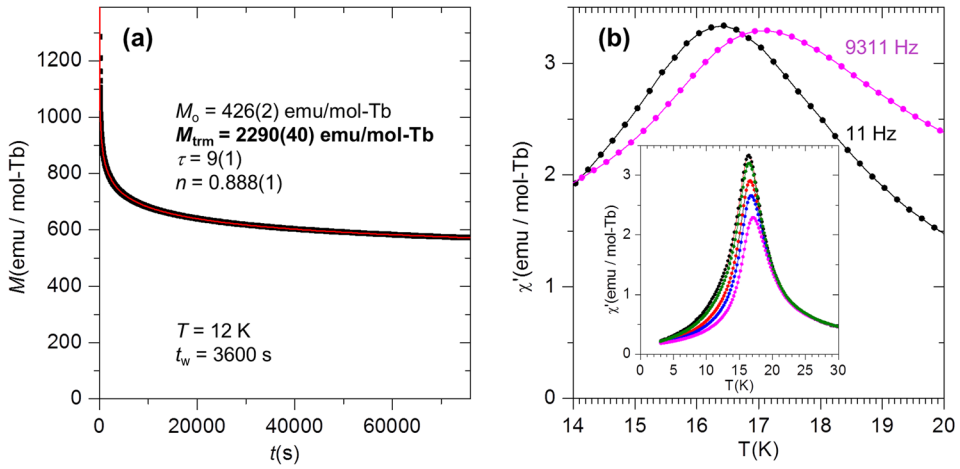

FIG. 1. (a) Thermoremanent magnetization (time-dependent magnetization) of $Tb_{30}Ru_{6.0}Sn_{29.5}$ collected at $T = 12$ K with $t_w = 3600$ s under field-cooled conditions of $H = 500$ Oe. Data are fit to the equation $M(t) = M_0 + M_{trm}{}^* e\,[-(t/\tau)\,(1-n)]$, where M_0 is the time-independent remanent magnetic contribution, M_{trm} is the glassy component of the magnetization, t is time, τ is the characteristic time constant, and n is the characteristic stretching-exponent. (b) Temperature-dependent AC susceptibility of $Tb_{30}Ru_{6.0}Sn_{29.5}$ at 11 Hz (black) and 9311 Hz (purple) normalized to 11 Hz. The inset shows temperature-dependent AC susceptibility collected at frequencies, from left to right, of 11, 57, 579, 2311, and 9311 Hz.

cooling, as wait times at successively colder temperatures do not affect the previous wait times at higher temperature. It is possible to store and read several memory dips by exploiting this phenomenon.[18] In fact, a thermal memory cell was designed which is capable of imprinting and recovering 8-bits of data;[14] however, the necessary long wait times prohibit the use of this effect as a viable memory storage technology.

Miyashita and Vincent describe the memory of previous (higher temperature) length correlations as "frozen impurities," which melt at temperatures above T_w.[19] Mathieu *et al*. found that applying a small magnetic field ($H = 0.5$ Oe) during the wait time of a memory dip experiment causes a significant drop in the measured DC magnetization at T_w during the warming process.[15] Combining these findings, we surmised that temperature-specific magnetization drops can be rapidly frozen-in through a field pulse without waiting (i.e., a constant cooling rate) during an uninterrupted initial cooling cycle. Upon warming from low temperature, the magnetization should begin at a maximized value and, as the frozen components approach the temperature of the corresponding field-pulse(s), a melting should occur, causing an effective magnetization reduction. This scenario is graphically depicted in Fig. 3(b).

Figure 4 shows the results of the field-pulse experiment with magnetic field pulses applied parallel to the crystallographic *a*-direction at 11, 9, 7, 5, and 3 K and field pulse magnitudes of 50, 100, 150, 250, and 400 Oe, respectively. All data are collected while warming at 0.3 K/min with a 10 Oe positive field bias. Figure 4(a) shows the background data set (M_b), measured without field pulses, the field-pulse data set (M_p), and $M_p - M_b$. Remarkably, the spin distribution perturbations shown in Fig. 4(b) (blue curve) as $d(M_p - M_b)/dT$ versus temperature, are preserved for all field pulses. The widths of the peaks in Fig. 4(b) broaden with increasing pulse duration and the temperature at which the pulse was

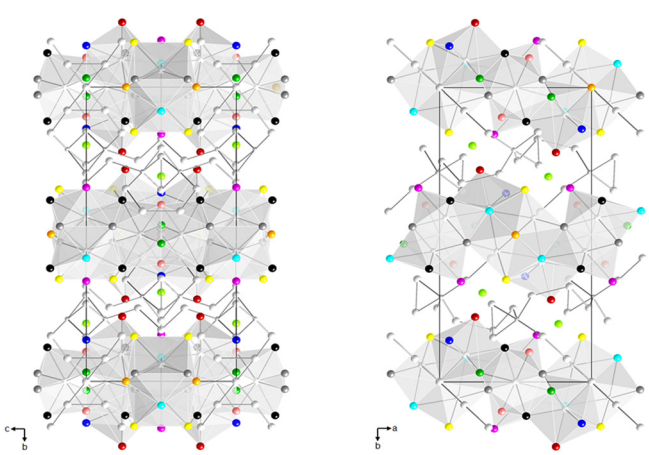

FIG. 2. Crystal structure of $Tb_{30}Ru_{6.0}Sn_{29.5}$ with individual Tb (Tb1–Tb11) sites highlighted as different colored spheres. Sn and Ru sites are shown as large and small white spheres, respectively, with polyhedra representing polar Sn-centered sites surrounded by Tb atoms.

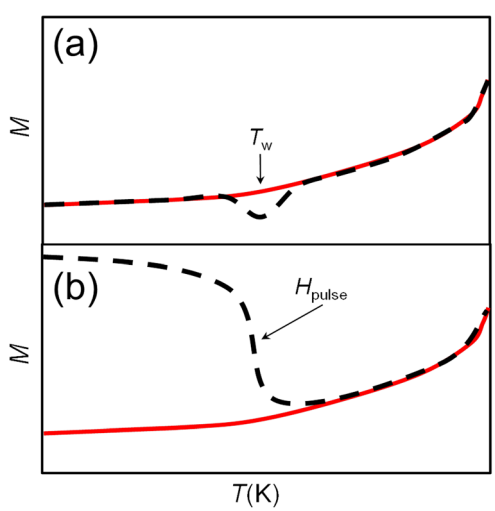

FIG. 3. A graphical depiction of the temperature-dependent magnetization of (a) a typical memory-dip experiment and (b) the proposed field-pulse experiment. The red lines represent the background magnetization profile of the spin glass sample. The background is obtained by cooling in a small applied field and then measuring magnetization as the sample is warmed. The dashed lines represent the corresponding magnetization profiles when a wait time or field pulse is applied at temperature T_W during the cool-down.

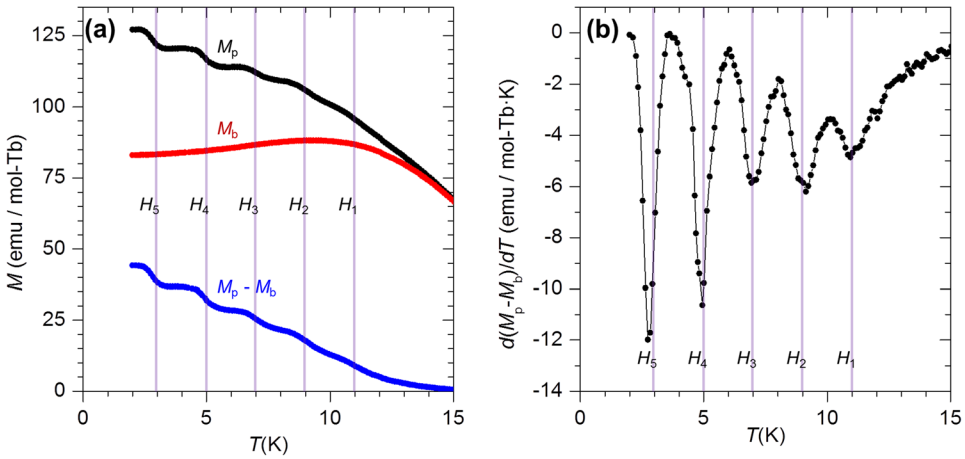

FIG. 4. (a) Temperature-dependent magnetization of $Tb_{30}Ru_{6.0}Sn_{29.5}$ collected upon warming with temperature-specific field-pulses (H_p) at 11, 9, 7, 5, and 3 K (black), the blank run (M_b) without initial field-pulses (red), and $M_p - M_b$ (blue). (b) The derivative of $M_p - M_b$ as a function of temperature showing slope minima at temperatures corresponding to the field-pulses. The warming and cooling ramp rates were fixed at 0.3 K/min, and the initial field was set to $H_i = 10$ Oe

applied, while at lower temperatures, higher pulse magnitudes are necessary, as expected, to achieve comparable ΔM values.

Figure 5 gives an indication of the degree of volatility of the pulse memory. The data in this figure were obtained by imprinting the sample with two field pulses, one applied at 9 K and the other at 5 K. The sample was then cooled to 1.8 K, where it was held for a specified time before executing a read cycle. The figure shows the resulting magnetization traces after being held at 1.8 K for wait times of 120 s (red), 28 800 s (black), and 86 400 s (blue). The traces are nearly identical, indicating that very little relaxation occurred at this low temperature. These data suggest that the non-equilibrium dynamics of this system make long-term data storage a feasible possibility. Once the data are written to a sample, it can be recovered via a once-only read cycle.

One can envision a data storage device capable of surpassing the superparamagnetic limitations[25–27] of widely used magnetic film media. The strategy would be to write data via localized heat pulses rather than field pulses. For example, one could design a spin-glass platter that is ZFC to well below the glass transition temperature. Then with a modest bias field applied to the rotating platter, a laser could write bits of data by simply locally heating parcels of the platter surface. Because the spin-glass response is not limited by a minimum magnetic domain size, one, in principle, could realize an extremely high bit-density. The resulting bits could then be read using standard magnetic read head technologies

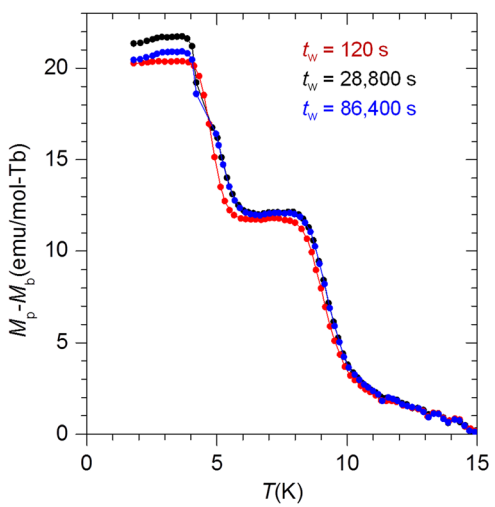

FIG. 5. Temperature-dependent magnetization ($M_p - M_b$) of $Tb_{30}Ru_{6.0}Sn_{29.5}$ with field pulses at 9 and 5 K with the sample held at 1.8 K for 120 s (red), 28 800 s (black), and 86 400 s (blue) prior to data collection upon warming. The warming and cooling ramp rates were fixed at 1 K/min.

Although it is apparent that spin-glasses with an appropriate set of glass dynamics can be technologically useful, to date there have not been any particular applications of these unusual systems. Two important factors in determining their viability as an information storage medium are, first, the degree to which a quasi-static and controllable spin state can be realized in a particular system and, second, the practicality of actually reading such a state. Clearly, increasing the magnitude of the glassy component of a system and maximizing the system's glass transition temperature are obvious long-term goals. In addition to $Tb_{30}Ru_{6.0}Sn_{29.5}$,[20] we have found spin-glass behavior in other analogs of the same structure type, such as $Dy_{30}Ru_{4.57}Sn_{30.72}$. Indeed, this structure type may have many more undiscovered spin-glass analogs. We believe that spin-glass behavior arises in these compounds as a result of competing interactions between the many non-equivalent magnetic sites in their structure. Unlike more traditional spin-glasses, the dynamics do not appear to be related to geometric frustration and/or structural disorder.

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[1]A. P. Ramirez, Nature **421**, 483 (2003).
[2]D. S. Fisher and D. A. Huse, Phys. Rev. Lett. **56**(15), 1601–1604 (1986).
[3]A. J. Bray and M. A. Moore, Phys. Rev. Lett. **58**(1), 57–60 (1987).
[4]D. S. Fisher and D. A. Huse, Phys. Rev. B **38**(1), 373–385 (1988).
[5]D. S. Fisher and D. A. Huse, Phys. Rev. B **38**(1), 386–411 (1988).
[6]A. P. Ramirez, Annu. Rev. Mater. Sci. **24**, 453–480 (1994).
[7]J. E. Greedan, J. Mater. Chem. **11**(1), 37–53 (2001).
[8]J. Snyder, J. S. Slusky, R. J. Cava, and P. Schiffer, Nature **413**(6851), 48–51 (2001).

[9]S. Ghosh, R. Parthasarathy, T. F. Rosenbaum, and G. Aeppli, Science **296**(5576), 2195–2198 (2002).
[10]S. Nakatsuji, Y. Nambu, H. Tonomura, O. Sakai, S. Jonas, C. Broholm, H. Tsunetsugu, Y. M. Qiu, and Y. Maeno, Science **309**(5741), 1697–1700 (2005).
[11]R. Moessner and A. R. Ramirez, Phys. Today **59**(2), 24–29 (2006).
[12]E. A. Goremychkin, R. Osborn, B. D. Rainford, R. T. Macaluso, D. T. Adroja, and M. Koza, Nature Phys. **4**(10), 766–770 (2008).
[13]E. M. Benbow, N. S. Dalal, and S. E. Latturner, J. Am. Chem. Soc. **131**(9), 3349–3354 (2009).
[14]J. Dolinsek, M. Feuerbacher, M. JagodiC, Z. JagliCic, M. Heggen, and K. Urban, J. Appl. Phys. **106**(4), 043917 (2009).
[15]R. Mathieu, M. Hudl, and P. Nordblad, Europhys. Lett. **90**(6), 1–6 (2010).
[16]R. V. Chamberlin, G. Mozurkewich, and R. Orbach, Phys. Rev. Lett. **52**(10), 867–870 (1984).
[17]J. L. Tholence, Solid State Commun. **35**(2), 113–117 (1980).
[18]J. P. Bouchaud, V. Dupuis, J. Hammann, and E. Vincent, Phys. Rev. B **65**(2), 024439 (2001).
[19]S. Miyashita and E. Vincent, Eur. Phys. J. B **22**(2), 203–211 (2001).
[20]D. C. Schmitt, N. Haldolaarachchige, J. Prestigiacomo, A. Karki, D. P. Young, S. Stadler, R. Jin, and J. Y. Chan, J. Am. Chem. Soc. **135**(7), 2748–2758 (2013).
[21]K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801–976 (1986).
[22]S. F. Edwards and P. W. Anderson, J. Phys. F: Met. Phys. **5**, 965–974 (1975).
[23]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. **35**, 1792–1796 (1975).
[24]V. Dupuis, E. Vincent, J. P. Bouchaud, J. Hammann, A. Ito, and H. A. Katori, Phys. Rev. B **64**(17), 174204 (2001).
[25]S. Chikazumi, *Physics of Ferromagnetism* (Oxford University Press, New York, 1997).
[26]D. A. B. Thompson and J. S. Best, IBM J. Res. Dev. **44**, 311–322 (2000).
[27]D. M. Weller and A. Moser, IEEE Trans. Magn. **35**, 4423–4439 (1999).
[28]See supplementary material at http://dx.doi.org/10.1063/1.4818262 for tables containing crystallographic and atomic position parameters.