

Spin-glass-like state in GdCu: Role of phase separation and magnetic frustration

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We report investigations on the ground-state magnetic properties of the intermetallic compound GdCu through dc magnetization measurements. GdCu undergoes first-order martensitic-type structural transition over a wide temperature window of coexisting phases. The high-temperature cubic and the low-temperature orthorhombic phases have different magnetic character and they show antiferromagnetic and helimagnetic orderings below 145 and 45 K, respectively. We observe a clear signature of a glassy magnetic phase below the helimagnetic ordering temperature, which is marked by thermomagnetic irreversibility, aging, and memory effects. The glassy magnetic phase in GdCu is found to be rather intriguing, with its origin lying in the interfacial frustration due to the distinct magnetic character of the coexisting phases.

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I. INTRODUCTION

Spin glass is an intriguing example of a nonergodic state marked by several physical properties, such as thermomagnetic irreversibility, slow dynamics, nonexponential decay, magnetic memory effect, etc.¹⁻⁵ The nonequilibrium state arises from *frustration* due to competing magnetic interactions among atomic spins as well as *disorder* capable of pinning the spins. Recently, spin-glass-like nonequilibrium dynamics and time-dependent phenomena have been observed in several magnetic systems, where the basic building blocks responsible for the “glassy” behavior are not really the atomic spins but rather a spin cluster or a bigger spin entity.⁶ In particular, such behavior has been widely observed in phase-separated manganites⁷⁻¹¹ and cobaltites.^{12,13} The observation of slow dynamics resembling classical spin glass is also extended to intermetallic alloys¹⁴⁻¹⁶ showing first-order magnetostructural transition, and also to magnetic nanoparticles.¹⁷ Since the basic building blocks are a bigger spin entity, they are often referred to as cluster glass, superspin glass, or magnetic glass.

The coexisting magnetic phases related to glassiness in manganites and other bulk solids are often related to first-order phase transition (FOPT).¹⁸ In the presence of static disorder, FOPT can lead to the coexistence of high-temperature (T) parent and low- T product phases within the region of transition. Under certain circumstances, there can also be structural freezing of the parent phase below the transition point.^{14,15,19} The glassiness in such a phase-separated system can have two likely origins: first, the slow dynamics of the coexisting phases due to their metastability, and secondly, the frustration arising from the magnetic interaction between two clusters having a distinct magnetic nature. The spin-glass-like state (often called cluster glass) in phase-separated manganites has often been attributed to the coexisting ferromagnetic (FM) and charge-order antiferromagnetic (AFM) phases of micrometer size due to FOPT.^{9,18} It is worth noting that in the case of a polydispersive noninteracting nanoparticle system (where the magnetic interaction between two particles is negligible), a simple model based on the distribution of superparamagnetic relaxation time can explain the observed glassy magnetic behavior including the memory effect.²⁰⁻²³

Considering the fact that a large number of bulk phase-separated materials show glassy magnetic behavior, it is

pertinent to investigate the role of intercluster magnetic interaction in the observed state. However, unlike magnetic nanoparticles,²² one cannot adjust the strength of the magnetic interaction in a bulk material. More importantly, the FOPT in such systems is of magnetostructural type with a strong interplay between magnetic and structural degrees of freedom. As a result, the structural and magnetic transitions occur below the same T , and their individual role in the glassiness becomes difficult to differentiate.

Here we report the magnetic investigation on the GdCu intermetallic compound, which was reported to show phase coexistence due to first-order martensitic transition (MT).²⁴⁻²⁷ What motivated us to investigate GdCu is the fact that the magnetic and structural transitions occur at distinctly different T . Therefore, there is an opportunity to investigate the role of magnetic interaction and structural phase separation in determining the ground-state magnetic character.

The RCu (where R denotes rare earth) series of compounds with heavy rare earths ($R \geq Gd$) crystallize in the cubic CsCl-type structure (hereafter called the C phase) at high T .²⁸ Some of the members ($R = Gd, Tb, \text{ and } Y$) show lattice instability and undergo MT at low T to an orthorhombic FeB-type structure (hereafter called the O phase). GdCu shows long-range AFM ordering below T_N^C in the C phase, while it undergoes a second transition below T_N^O to a helimagnetic (HM) spin structure in the low- T O phase. Our investigation shows the existence of an unconventional glassy magnetic phase in GdCu below T_N^O , which presumably arises out of the magnetic frustration from coexisting phases.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of GdCu and DyCu were prepared by argon arc melting with the constituent elements of purity better than 99.9 wt. %. The ingots were homogenized at 800 °C for 120 h. Room-temperature powder x-ray diffraction (XRD) patterns were recorded using a Bruker AXS diffractometer (Cu $K\alpha$ radiation, 2θ range from 20° to 80° with a step size of 0.02° and 5 s/step counting time). The collected powder patterns were used for phase identification of the given compound using the GSAS software package.²⁹ Rietveld refinement data along with XRD patterns of GdCu are shown in the inset of Fig. 1. Lattice parameter ($a = 3.518 \text{ \AA}$) and unit-cell

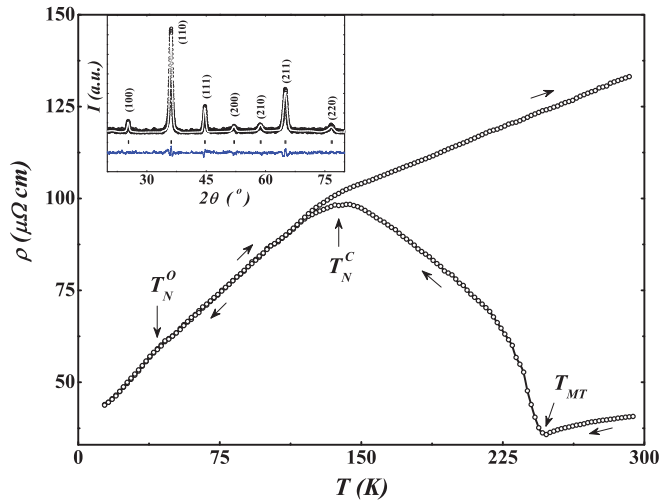


FIG. 1. (Color online) The main panel shows the zero-field resistivity as a function of temperature for cooling and subsequent heating legs for GdCu. The sample was cooled from 300 K down to 5 K and heated back to 300 K again. The inset shows the Rietveld refinement of the x-ray powder-diffraction pattern of the GdCu compound. The open circles represent observed data and the lines drawn through the data points correspond to the calculated patterns.

volume ($V = 43.54 \text{ \AA}^3$) are in good agreement with the previously reported data. The analysis indicates that the annealed samples are single phase with cubic CsCl structure at room temperature.

The T variation of resistivity (ρ) in zero field as well as in the presence of external magnetic field (H) were measured down to 5 K in a commercial cryogen free high magnetic field system from Cryogenic Ltd. UK. Magnetization (M) was measured using a commercial Quantum Design SQUID magnetometer (MPMS XL Ever Cool model).

III. RESULTS

Figure 1 represents the T dependence of ρ of GdCu measured during cooling and heating. A sharp anomaly is observed around $T_{MT} = 250$ K in the cooling data, which signifies the MT from the high- T C phase to low- T O phase as reported in previous studies.^{24,25,27} Only a part of the thermal hysteresis loop associated with the MT is visible in the measured T range, because the transition extends up to 650 K in the high- T side.²⁴ In the low- T side, the loop closes at 140 K.^{24,25} Similar hysteresis is also present between the field-cooling (FCC) and subsequent field-cooled heating (FCH) legs of the M^{-1} versus T data [Fig. 2(a), main panel]. A small anomaly is observed both in $\rho(T)$ and $M^{-1}(T)$ near 145 K, which agrees well with the AFM transition temperature T_N^C of the austenitic C phase reported earlier.²⁵ A second magnetic anomaly is observed in $\rho(T)$ and $M^{-1}(T)$ data around 45 K (denoted by T_N^O), which indicates the onset of helical magnetic ordering of the martensitic O phase present in the sample.²⁵

In Fig. 2(b), we have shown M versus T data measured in the zero-field-cooled heating (ZFCH) and FCH protocols with $H = 1$ kOe. A clear bifurcation is observed between ZFCH and FCH magnetizations [Fig. 2(b)] below T_N^O , signifying the existence of thermomagnetic irreversibility associated with

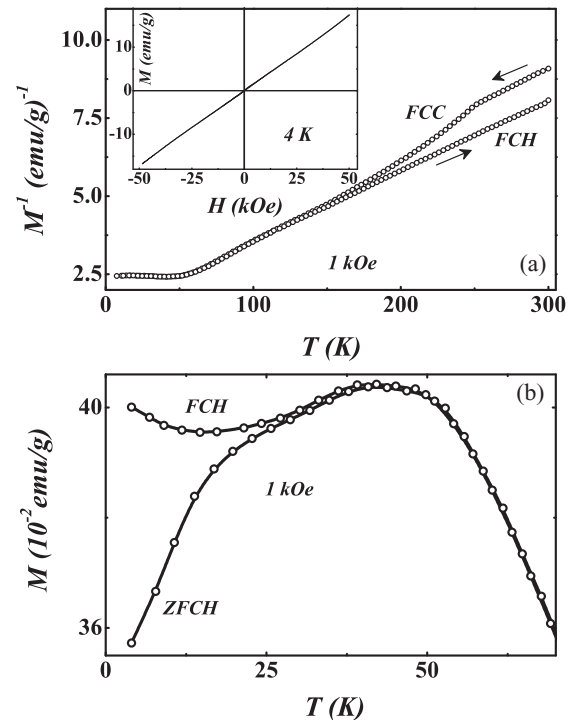


FIG. 2. (a) Inverse magnetization as a function of temperature measured in the FCC and subsequent FCH protocols for GdCu. The FCC and FCH data show thermal hysteresis down to about 140 K. The inset shows the magnetization as a function of field recorded at 4 K. (b) ZFCH and FCH magnetizations as a function of temperature in 1 kOe of applied field.

the HM transition. The ZFCH magnetization drops with decreasing T , while the FCH counterpart shows a sluggish upturn. The feature in the $M(T)$ curve near T_N^O is found to be rather broad. Such a broad feature associated with thermomagnetic irreversibility can be an indication of a disordered and/or glassy magnetic phase below T_N^O .

The inset of Fig. 2(a) shows the M - H curve of GdCu measured at 4 K, which is found to be linear without any signature of field-induced transition. The ρ versus H curves (not shown here) also do not show any signature of metamagnetism. The magnetoresistance (MR) (which is equal to $[\rho(H) - \rho(0)]/\rho(0)$) at 4 K is found to be very small and positive (about 4% for $H = 50$ kOe), which is common among bulk AFM materials.³⁰ Therefore, one can rule out the possibility of any major role of H in stabilizing different magnetic and structural phases in GdCu. This is possibly indicative of a weak magnetostructural coupling in GdCu, unlike manganites³¹ or magnetic shape memory alloys.¹⁶

A. Magnetic relaxation

Considering magnetic irreversibility in GdCu, the time (t) evolution of M was investigated at low T . The measurement was performed in two protocols, namely ZFC and FC as described in Figs. 3(a) and 3(b). The t dependence of M for both protocols was measured under 100 Oe of applied field [Figs. 3(a) and 3(b)]. The ZFC measurement was performed at different temperatures above and below T_N^O . However, only a strong relaxation effect was observed below T_N^O . The $M(t)$

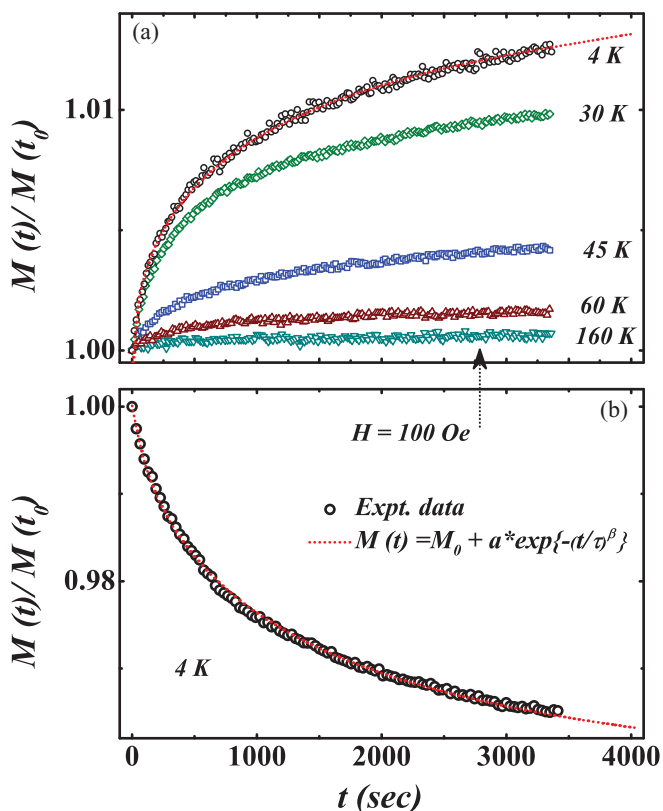


FIG. 3. (Color online) (a) Time dependence of magnetization at several constant temperatures measured in the ZFC state with an applied field of 100 Oe for GdCu. The ZFC state is achieved by cooling the sample from room temperature to desired measurement temperatures in the absence of field. (b) Similar measurement in the FC mode at 4 K, where the FC mode is achieved by cooling the sample from room temperature to 4 K in $H = 10$ kOe and the time dependence was measured after reducing the field to 100 Oe. The solid lines through 4 K data (both ZFC and FC modes) are fittings with a stretched exponential function (see text). Here magnetization has been normalized with its initial value at t_0 (the starting time for the relaxation measurement).

data collected in the ZFC mode at 30 K (below T_N^O) show a 1% change in 3600 s, while at 60 K (just above T_N^O) the change is only 0.1%.

The time evolution of M has been analyzed on the basis of various available models of slow dynamics applicable to a magnetic system. It can often show a power law ($\sim t^{\pm\alpha}$) or a Kohlrausch-Williams-Watt (KWW) stretched exponential behavior ($\sim \exp[-(t/\tau)^\beta]$) (Refs. 32 and 33) where τ is the characteristic relaxation time and β is the shape parameter. We have used both relations to fit our data, and found that the best fit is obtained with the KWW model. Such a model was widely used to analyze the data for spin glass and other disordered magnets.³⁴ In this model, β lies between 0 and 1 for a different class of disordered materials. The value of β was found to be 0.51 and 0.59 for the ZFC and FC data, respectively, at 4 K. The exponent β in the KWW model signifies the number of intermediate states through which the system should evolve, and it approaches 1 when the number of such intermediate states diminishes.³⁵ Glassy magnetic systems are found to show β values over a wide range between 0.2 and 0.6 below the

freezing temperature (T_f). For example, the spin-glass alloy La-Fe-Mn-Si (Ref. 36) has a β value close to 0.5, well below T_f , while CuMn (4.0 at. %) (Ref. 37) shows β to be between 0.2 and 0.4 depending on the temperature of measurement.

B. Aging

We studied the aging effect in GdCu below T_N^O (see Fig. 4). The sample was first zero-field-cooled down to 15 K and was allowed to age there for a certain waiting time of t_w . Subsequently M was measured as a function of time in the presence of $H = 100$ Oe. The measurement was performed for three different values of t_w , namely 600, 2000, and 5000 s. From the t variation of M in the resulting states, we have calculated the magnetic viscosity [$S(t) = \frac{1}{H} \frac{\partial M(t)}{\partial \ln t}$] as shown in Fig. 4. It is clear that the $S(t)$ behavior is strongly influenced by the waiting time. The $S(t)$ plots show a clear peak at the respective t_w values, which is typical experimental evidence of the nonequilibrium states in a classical spin-glass system.² It is worth noting that such a perfect match between t_w and the peak position of $S(t)$ in the case of GdCu is absent in several glassy magnetic materials, indicating a deviation from the classical spin-glass behavior (see, for example, Refs. 38 and 39).

C. Memory in the temperature variation of M

We probed the memory in dc magnetization in GdCu through different modes of measurements.^{4,17,22,38,39} First we depict the memory in the T dependence of M measured in the FC state following the protocol by Sun *et al.*¹⁷ [see Fig. 5(a)]. Chronologically, the measurement was performed in the following steps: (i) the sample was cooled in $H = 100$ Oe from 100 to 2 K with intermediate stops of duration $t_w = 1$ h each at $T_{\text{stop}} = 30, 20,$ and 10 K. During each stop, T was kept constant while H was reduced to zero. After each stop, we

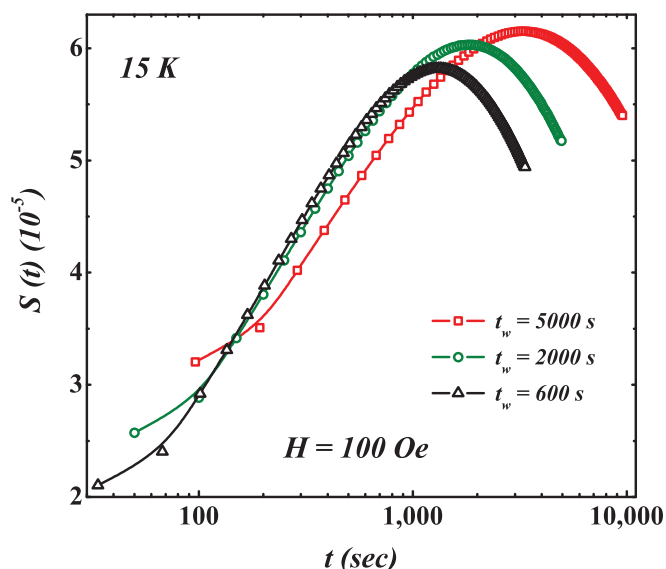


FIG. 4. (Color online) The waiting time (t_w) dependence of magnetic viscosity for GdCu recorded at 15 K. The relaxation was measured in the zero-field-cooled state at $H = 100$ Oe for different values of t_w .

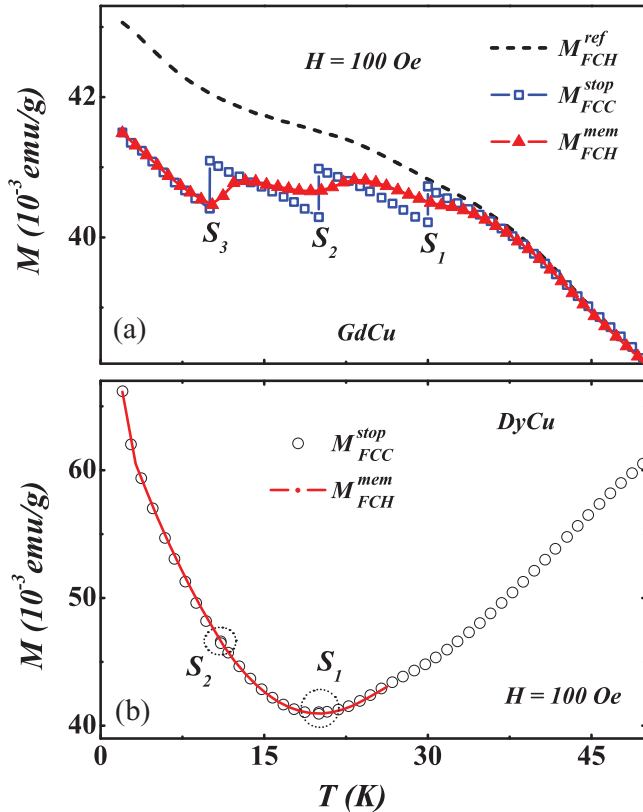


FIG. 5. (Color online) (a) FC memory effect in the temperature (T) variation of dc magnetization of GdCu where the curves were obtained by cooling the sample in 100 Oe with intermediate zero-field stops at $T = 30, 20$, and 10 K (M_{FCC}^{stop}) and then subsequent field heating (M_{FCH}^{mem}). The reference curve (M_{FCH}^{ref}) was measured on heating after the sample was field-cooled in 100 Oe without intermediate stops. (b) A similar memory experiment in DyCu.

reapplied H and resumed cooling, which resulted in a steplike M - T curve (M_{FCC}^{stop}). (ii) After reaching 2 K, the sample was heated back in $H = 100$ Oe, which produces the so called ‘‘memory’’ curves (M_{FCH}^{mem}). (iii) A reference curve was also recorded by simply allowing the sample to heat in $H = 100$ Oe after being field-cooled in the same field without any stop (M_{FCC}^{ref}). The T ramp rate in all the measurement (as well as during field cooling) was kept fixed at 1 K/min. While heating [curve M_{FCH}^{mem} in Fig. 5(a)], we observe a striking memory effect signifying nonergodic behavior of the low- T phase. At each T_{stop} , the sample produces a clear upturn revealing the previous history of stops in zero field at that T . This is a manifestation of memory in the low- T phase. However, it is interesting to note that *no memory was observed above* T_N^O (i.e., $T_{stop} > T_N^O$), which once again proves the role of HM ordering for the observed arrested dynamics in GdCu.

We also looked for memory in DyCu, which belongs to the same RCu series of compounds. However, unlike GdCu, DyCu does not undergo MT and it remains in the C phase down to low T . DyCu undergoes AFM ordering below 63 K,⁴⁰ and shows a second magnetic transition below 20 K. We performed the same memory experiment on DyCu, and no signature of magnetic memory was observed [see Fig. 5(b)]. This is in

contrast with the behavior of GdCu, and presumably related to the absence of MT-related phase coexistence.

Memory in the FC magnetization can also occur due to a distribution of relaxation time for a polydisperse nanoparticle system. GdCu being a phase-separated system, a similar effect may also arise from the independent relaxation of metastable phase clusters. A probable way to rule out such a possibility is to investigate the memory in the ZFC magnetization, which is an unequivocal signature of a glassy magnetic state originating from cooperative spin-spin interaction.²² Here the protocol is the same as that of FC memory measurement, only the sample was cooled in zero field and allowed to relax at selected stops (here 20 and 12 K) in zero field only. While heating in 100 Oe, the sample shows characteristic features at the point of stops. This is clearer in Fig. 6(b), where we have subtracted the reference ZFC curve (M_{ZFC}^{ref} , measured without stops) from the memory curve (M_{ZFC}^{mem} , measured with stops). The present observation clearly strengthens the view of a spin-glass-like state in GdCu. Notably, the reference and the ‘‘memory’’ curves coincide exactly at the low- and high- T end points, which is

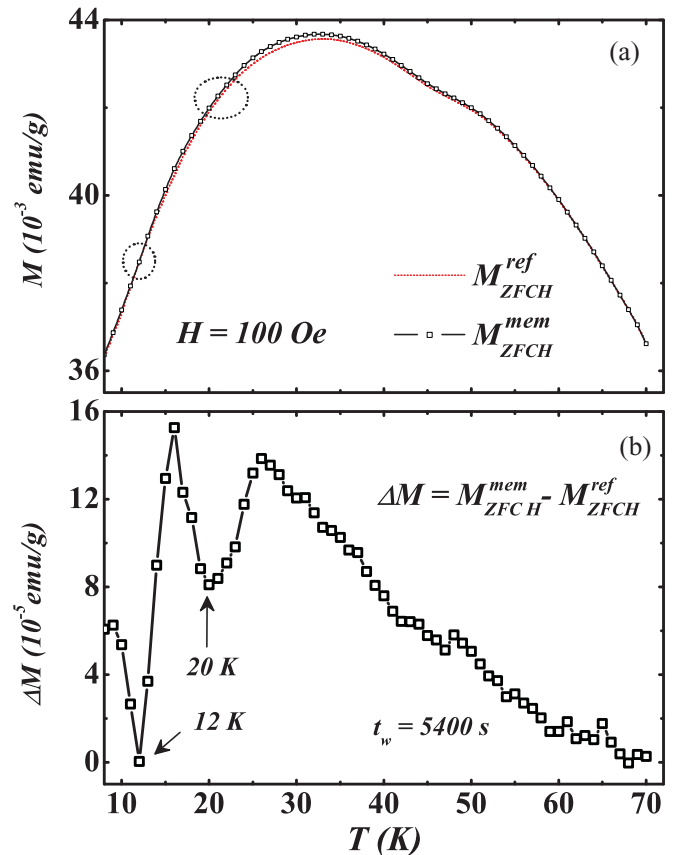


FIG. 6. (Color online) (a) The memory measurement in the zero-field-cooled condition recorded on dc magnetization vs temperature data for GdCu. The sample was first cooled in $H = 0$ down to 4 K, with intermediate stops at 20 and 12 K for 1.5 h each. The sample was then reheated in $H = 100$ Oe up to 70 K (M_{ZFC}^{mem}). A zero-field-cooled reference curve (M_{ZFC}^{ref}) without intermediate stops during heating is also shown as a dotted line. (b) The difference between the magnetization between the memory curve (with intermediate stops) and the reference curve.

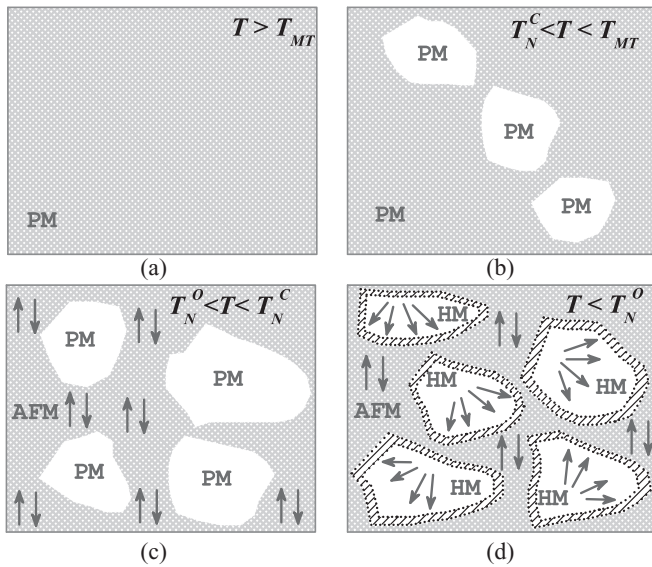


FIG. 9. A cartoon of the magnetic and structural phase separation in GdCu at different temperatures. The meshlike region and the white region denote the C phase and the O phase, respectively. PM, AFM, and HM, respectively, denote paramagnetic, antiferromagnetic, and helimagnetic character of the coexisting phases. The hatched phase boundary in (d) represents the interfacial glassy layer due to spin frustration.

has two magnetic transitions. The failure to observe memory in DyCu indicates that the low- T glassy magnetic phase is somehow related to the FOPT. In the case of GdCu, the thermal hysteresis is present over a very wide T range, starting from 650 to 140 K. This is the region of MT where both the O phase and the C phase can coexist. However, phase coexistence down to very low T (~ 4 K) was confirmed by neutron-diffraction experiment. The existence of the high- T C phase well below the region of MT indicates the *arrested kinetics of the FOPT*,^{14,15,19,44} which allows the metastable C phase to coexist with the low- T O phase.

The C phase becomes antiferromagnetically ordered below about 145 K, and the slow dynamics can originate from the time dependent magnetization of the frozen phase. Arrested dynamics in the structurally frozen phase has been observed in Ti-Ni shape memory alloys.⁴⁵ However, our relaxation and memory experiments identify the glassy phase only below $T_N^O = 45$ K, which is the onset point of HM order of the O phase. Therefore, it is not the metastability of the structurally frozen phases but rather the cooperative magnetic interaction that determines the glassiness. Otherwise, one would expect the glassy behavior to originate right below T_{MT} out of the frozen dynamics of the coexisting structural phases.

Our data also rule out the possibility of a glassy state originating from a distribution of relaxation time of noninteracting spin clusters, as observed in weakly interacting nanoparticles. Had this been the scenario, we would not expect memory in the ZFC measurement (see Fig. 6). Notably, unlike nanoparticles, the magnetic anisotropy is rather weak in a Gd-based system with total orbital quantum number $L = 0$.

The likely mechanism behind the observed glassiness in GdCu lies in the magnetic correlation. Since it occurs only

below T_N^O , the low- T HM state plays a deciding role. The glassy phase below T_N^O resembles the classical spin-glass state, particularly from the evidence of ZFC memory and aging (Figs. 6 and 4, respectively). In GdCu (both in the O and C phases), there is only one crystallographic site²⁵ for Gd, which may not be sufficient for competing magnetic interaction among atomic spins leading to frustration. In addition, geometrical frustration can be ruled out in a cubic or orthorhombic crystal structure (both orthogonal). The only possibility that seems viable in the present case is the coexisting structural phases with contrasting magnetic character, which introduces frustration and disorder leading to a spin-glass-like state.

We propose the following scenario for the observed glassiness, which has been depicted in Fig. 9. As is evident, GdCu has several magnetic and structural phases. Above T_{MT} (see Fig. 1), the system is purely cubic and paramagnetic (PM) [see Fig. 9(a)]. According to the terminology of MT in metallic alloys, T_{MT} is called the martensitic start temperature, below which the low- T martensite starts to nucleate. As the system goes below T_{MT} , the O phase develops within the matrix of the C phase, however both are PM in nature. When T is lowered further, the fraction of O phase increases. Due to arrested kinetics, the complete transformation of C phase \rightarrow O phase does not take place and a fraction of the C phase exists even at the lowest T . Now below T_N^C , the C phase orders antiferromagnetically, but the O-phase fraction remains PM, and eventually it orders below T_N^O . Within the temperature region $T_{MT} < T_N^O$, the mixed phase prevails in the sample. However, unless one goes below T_N^O , the O phase remains PM and cannot introduce magnetic frustration. Eventually at $T < T_N^O$, both coexisting phases become magnetically ordered, and interfaces between the two phases give rise to frustration due to their conflicting magnetic character. The effect is analogous to the surface spin-glass state in interacting γ - Fe_2O_3 nanoparticles⁴⁶ with phase boundaries in GdCu playing the part of surface layers of the nanoparticles. The interesting point seen in the case of GdCu is that the spin-glass-like state does not originate right below the first-order transition, rather it develops below the low- T HM transition temperature point. The presence of C-phase and O-phase clusters may give rise to a frozen local strain order as observed in Ti-Ni alloys,⁴⁵ but that does not give rise to a magnetic glassy phase as long as $T > T_N^O$. It is to be noted that a previous heat capacity study²⁶ showed a broad peak around the HM transition point, which is also a signature of the onset of a glassy phase in a material.

In conclusion, we observed a glassy magnetic phase in GdCu marked by slow dynamics and a clear memory effect. Our results suggest that cooperative magnetic correlation between the coexisting phases is the primary cause of the observed spin-glass-like state. Such an understanding can be quite useful in interpreting the spin-glass-like state in other phase-separated systems including manganites.

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- ¹K. Binder and A. P. Young, *Rev. Mod. Phys.* **58**, 801 (1986).
- ²J. A. Mydosh, *Spin Glasses* (Taylor and Francis, London, 1993).
- ³M. B. Weissman, *Rev. Mod. Phys.* **65**, 829 (1993).
- ⁴K. Jonason, E. Vincent, J. Hammann, J. P. Bouchaud, and P. Nordblad, *Phys. Rev. Lett.* **81**, 3243 (1998).
- ⁵F. Lefloch, J. Hannan, M. Ocio, and E. Vincent, *Eur. Phys. Lett.* **18**, 647 (1992).
- ⁶R. Mathieu and Y. Tokura, *J. Phys. Soc. Jpn.* **76**, 124706 (2007).
- ⁷A. Maignan, C. Martin, F. Damay, B. Raveau, and J. Hejtmanek, *Phys. Rev. B* **58**, 2758 (1998).
- ⁸I. G. Deac, J. F. Mitchell, and P. Schiffer, *Phys. Rev. B* **63**, 172408 (2001).
- ⁹F. Rivadulla, M. A. López-Quintela, and J. Rivas, *Phys. Rev. Lett.* **93**, 167206 (2004).
- ¹⁰L. Ghivelder and F. Parisi, *Phys. Rev. B* **71**, 184425 (2005).
- ¹¹K. De, M. Patra, S. Majumdar, and S. Giri, *J. Phys. D* **40**, 7614 (2007).
- ¹²Y.-k. Tang, Y. Sun, and Z.-h. Cheng, *Phys. Rev. B* **73**, 012409 (2006).
- ¹³M. Patra, S. Majumdar, and S. Giri, *J. Phys. Condens. Matter* **22**, 116001 (2010); *J. Appl. Phys.* **107**, 033912 (2010).
- ¹⁴M. K. Chattopadhyay, S. B. Roy, and P. Chaddah, *Phys. Rev. B* **72**, 180401(R) (2005).
- ¹⁵S. B. Roy, M. K. Chattopadhyay, P. Chaddah, J. D. Moore, G. K. Perkins, L. F. Cohen, K. A. Gschneidner Jr., and V. K. Pecharsky, *Phys. Rev. B* **74**, 012403 (2006).
- ¹⁶S. Chatterjee, S. Giri, S. Majumdar, and S. K. De, *Phys. Rev. B* **77**, 224440 (2008).
- ¹⁷Y. Sun, M. B. Salamon, K. Garnier, and R. S. Averback, *Phys. Rev. Lett.* **91**, 167206 (2003).
- ¹⁸E. Dagotto, *New J. Phys.* **7**, 67 (2005).
- ¹⁹P. Chaddah, K. Kumar, and A. Banerjee, *Phys. Rev. B* **77**, 100402(R) (2008).
- ²⁰M. Sasaki, P. E. Jönsson, H. Takayama, and P. Nordblad, *Phys. Rev. Lett.* **93**, 139701 (2004).
- ²¹R. K. Zheng, H. Gu, and X. X. Zhang, *Phys. Rev. Lett.* **93**, 139702 (2004).
- ²²M. Sasaki, P. E. Jönsson, H. Takayama, and H. Mamiya, *Phys. Rev. B* **71**, 104405 (2005).
- ²³S. Chakraverty, M. Bandyopadhyay, S. Chatterjee, S. Dattagupta, A. Frydman, S. Sengupta, and P. A. Sreeram, *Phys. Rev. B* **71**, 054401 (2005).
- ²⁴J. C. M. van Dongen, T. T. M. Palstra, A. F. J. Morgownik, J. A. Mydosh, B. M. Geerken, and K. H. J. Buschow, *Phys. Rev. B* **27**, 1887 (1983).
- ²⁵J. A. Blanco, J. I. Espeso, J. García Soldevilla, J. C. Gómez Sal, M. R. Ibarra, C. Marquina, and H. E. Fischer, *Phys. Rev. B* **59**, 512 (1999).
- ²⁶J. C. Ho, S. He, C. Wu, T.-S. Chein, and A. S. Pavlovic, *Ch. J. Phys.* **29**, 147 (1991).
- ²⁷V. G. Sathe, R. Rawat, and A. Dubey, *Phys. Rev. B* **74**, 052101 (2006).
- ²⁸A. Iandelli and A. Palenzona, in *Handbook on the Physics and Chemistry of Rare Earths* (Ref. 1), p. 1.
- ²⁹A. C. Larson and R. B. Von Dreele, 2004 General Structure Analysis System (GSAS), Los Alamos National Laboratory Report LAUR 86-748.
- ³⁰H. Yamada and S. Takada, *J. Phys. Soc. Jpn.* **34**, 51 (1973).
- ³¹See, for example, D. A. Filippov, R. Z. Levitin, A. N. Vasil'ev, T. N. Voloshok, H. Kageyama, and R. Suryanarayanan, *Phys. Rev. B* **65**, 100404(R) (2002).
- ³²A. Ito, H. Aruga, E. Torikai, M. Kikuchi, Y. Syono, and H. Takei, *Phys. Rev. Lett.* **57**, 483 (1986).
- ³³R. V. Chamberlin, G. Mozurkewich, and R. Orbach, *Phys. Rev. Lett.* **52**, 867 (1984).
- ³⁴J. C. Phillips, *Rep. Prog. Phys.* **59**, 1133 (1996).
- ³⁵X. Du, G. Li, E. Y. Andrei, M. Greenblatt, and P. Shuk, *Nat. Phys.* **3**, 111 (2007).
- ³⁶F. Wang, J. Zhang, Y.-f. Chen, G. -j. Wang, J. -r. Sun, S.-y. Zhang, and B.-g. Shen, *Phys. Rev. B* **69**, 094424 (2004).
- ³⁷D. Chu, G. G. Kenning, and R. Orbach, *Phys. Rev. Lett.* **72**, 3270 (1994).
- ³⁸S. Hari Krishnan, S. Rö sler, C. M. N. Kumar, Y. Xiao, H. L. Bhat, U. K. Rö sler, F. Steglich, S. Wirth, and S. Elizabeth, *J. Phys. Condens. Matter* **22**, 346002 (2010).
- ³⁹V. Markovich, I. Fita, A. Wisniewski, G. Jung, D. Mogilyansky, R. Puzniak, L. Titelman, and G. Gorodetsky, *Phys. Rev. B* **81**, 134440 (2010).
- ⁴⁰M. Amara, P. Morin, and F. Bourdarot, *J. Phys. Condens. Matter* **9**, 7441 (1997).
- ⁴¹C. Dasgupta and O. T. Valls, *J. Phys. Condens. Matter* **12**, 6553 (2000).
- ⁴²J. Dolinsek, J. Slanovec, Z. Jagličić, M. Heggen, S. Balanetsky, M. Feuerbacher, and K. Urban, *Phys. Rev. B* **77**, 064430 (2008).
- ⁴³P. E. Jönsson, R. Mathieu, P. Nordblad, H. Yoshino, H. A. Katori, and A. Ito, *Phys. Rev. B* **70**, 174402 (2004).
- ⁴⁴P. A. Sharma, S. El-Khatib, I. Mihut, J. B. Betts, A. Migliori, S. B. Kim, S. Guha, and S.-W. Cheong, *Phys. Rev. B* **78**, 134205 (2008).
- ⁴⁵S. Sarkar, X. Ren, and K. Otsuka, *Phys. Rev. Lett.* **95**, 205702 (2005).
- ⁴⁶B. Martínez, X. Obradors, Ll. Balcells, A. Rouanet, and C. Monty, *Phys. Rev. Lett.* **80**, 181 (1998).