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## Jamming Behavior of Domains in a Spiral Antiferromagnetic System

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Using resonant magnetic x-ray photon correlation spectroscopy, we show that the domains of a spiral antiferromagnet enter a jammed state at the onset of long-range order. We find that the slow thermal fluctuations of the domain walls exhibit a compressed exponential relaxation with an exponent of 1.5 found in a wide variety of solidlike jammed systems and can be qualitatively explained in terms of stress release in a stressed network. As the temperature decreases, the energy barrier for fluctuations becomes large enough to arrest further domain wall fluctuations, and the domains freeze into a spatial configuration within 10 K of the Néel temperature. The relaxation times can be fitted with the Vogel-Fulcher law as observed in polymers, glasses, and colloids, thereby indicating that the dynamics of domain walls in an ordered antiferromagnet exhibit some of the universal features associated with jamming behavior.

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Recently, there has been much interest in jamming transitions [1,2], which were originally studied as a way to discuss the behavior of granular materials [3] or dense assemblies of particles but have been eventually proposed as a more general and unified way of looking at dynamics of systems as diverse as structural glasses [4], entangled polymers [5,6], colloidal gels [7], supercooled liquids [8], and the like. Jamming occurs when the particle densities or entanglements are such that the motions of the individual particles become very restricted so that they slow down. The fluctuations which occur in a jammed system, e.g., a colloidal gel, with long-range interactions are cooperative fluctuations where a local displacement causes an inhomogeneity which is correlated with a displacement in another region. A theory based on elastic strains as the source of long-range interactions has been shown [9,10] to yield an intermediate scattering function  $F(Q, t)$  which decays as a compressed exponential  $\exp(-t/\tau)^\beta$ , where the exponent  $\beta$  is  $\sim 3/2$ . This has been seen in colloidal gels via dynamical light scattering and x-ray photon correlation spectroscopy (XPCS), as well as in other soft matter systems [10,11]. Interestingly, similar behavior was observed in antiferromagnetic Cr metal by using XPCS [12]. Up to now, domain wall dynamics under a magnetic field have been extensively studied in ferromagnets [13,14]. However, purely thermally driven domain wall fluctuations in antiferromagnetic systems have been scarcely investigated, while these might be of importance for the discussions of noise issues in magnetic nanodevices. Unlike domains in a ferromagnetic material, the energetics of domain rearrangements in an antiferromagnet are not affected by macroscopic magnetic field energies but are complicated by the breaking of translational symmetry.

We show in this Letter that the domain walls of the spiral antiferromagnet dysprosium undergo slow fluctuations

near the Néel temperature which show essentially the same quantitative relaxational behavior as in Cr. As we discuss below, the magnetic domains that form when an antiferromagnet orders are essentially in a jammed state, similar to particles with a repulsive interaction in a jammed granular system. We also observe that the domain dynamics freeze about 10 K below the Néel temperature and the behavior shows remarkable similarity to glassy behavior observed in other systems. A very similar system, namely, the spiral antiferromagnet holmium, was recently investigated by using XPCS [15], and similar slow fluctuations were observed. However, the Ho film was only 11 monolayers thick, and no quantitative analysis was carried out on the measured  $F(Q, t)$ .

In this study, we used an epitaxially grown yttrium(Y)/dysprosium(Dy)/yttrium(Y) sample with the 500-nm-thick Dy being the active layer [16,17]. Dy has a hexagonal close packed structure, is paramagnetic above 180 K ( $T_N$ ), and exhibits an incommensurate helical spin structure in the temperature range 89–180 K along the  $c$  axis [see Fig. 1(a)] with ferromagnetic alignment of the moments in the basal plane [18]. The helical magnetic phase is due to indirect exchange interactions between the Dy moments, and in real space these can be represented by exchange interactions which oscillate in sign versus distance: positive for nearest neighbors, but negative for next-nearest neighbors. At 89 K, bulk Dy undergoes a first-order antiferromagnetic (AFM)-ferromagnetic (FM) phase transition when the magnetostrictive energy overtakes the exchange energy. In a thin film, however, this can be reduced or enhanced depending on the film thickness, strains, etc. [19,20].

Coherent x-ray scattering experiments were carried out at beam line 12.0.2.2 of the Advanced Light Source, Lawrence Berkeley National Laboratory. A linearly polarized incident x-ray beam was tuned to the Dy  $M_5$  edge at

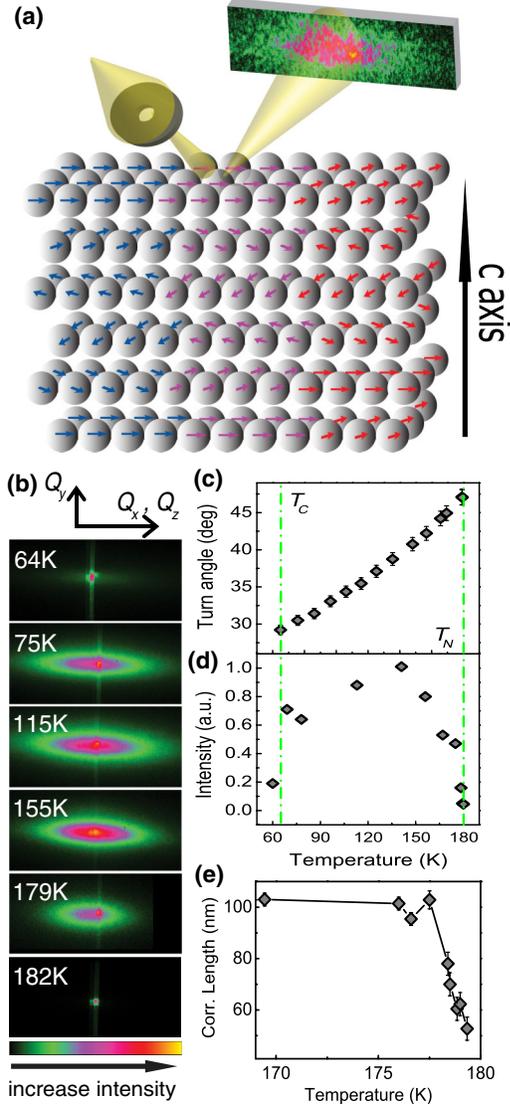


FIG. 1 (color online). (a) Schematic diagram of the spiral spin structure along the  $c$  axis. Three types of spin structure are shown that give rise to domains in the Bragg plane. (b) The  $(0, 0, Q_m)$  antiferromagnetic Bragg peak measured with a noncoherent beam at representative temperatures of 64, 75, 115, 179, and 182 K. The signal disappears at 182 K, as the sample is paramagnetic. The Bragg peak also disappears below 64 K, as the sample becomes ferromagnetic. (c) Evolution of the spiral turn angle as a function of temperature. (d) Integrated intensity as a function of temperature. The green vertical lines in (c) and (d) indicate the transition temperatures. (e) Lateral correlation length as a function of temperature.

1297 eV to access the charge-forbidden  $(0, 0, Q_m)$  AFM Bragg peak via resonant magnetic scattering in reflection geometry. A  $5 \mu\text{m}$  diameter pinhole was put in the beam path to establish transverse coherence of the incident x-ray beam. Speckle patterns were recorded on a charge coupled device detector 0.45 m away. For each measurement we have raised the temperature above  $T_N$ , then reduced it to the measurement temperature, and waited for one hour

until the temperature was stabilized within 10 mK. A two-hour-long movie of the evolving speckle pattern was subsequently taken with a frame time resolution of 5 sec [17].

The helical spin structure gives rise to magnetic satellite peaks along the specular direction at  $Q = (0, 0, l \pm Q_m)$ , in units of reduced reciprocal lattice vectors, where  $l$  is an even integer. Figure 1(b) shows the evolution of the  $(0, 0, Q_m)$  AFM Bragg peak as a function of temperature which shows  $T_N = 180$  K and  $T_C = 63$  K. The position of the magnetic satellite at  $Q_m$  varies with temperature, because the turn angle between spins in adjacent basal planes changes [Fig. 1(c)]. To determine the lateral correlation length  $\xi$ , we fit the envelope of the Bragg peak along  $Q_y$  with a Lorentzian to the  $3/2$  power as theoretically expected for random 2D domains [17,21]:

$$I(Q_y) \propto \frac{\xi^2}{(1 + Q_y^2 \xi^2)^{3/2}}. \quad (1)$$

The magnetic domain size is  $\sim 6\xi$ . The temperature-independent charge scattering due to the surface roughness was fitted with a separate Lorentzian that yielded a correlation length of 400 nm. No significant change in the correlation length or the dynamics was observed as a function of time over a period of 2 hours at each temperature, indicating that aging effects were too slow to observe and that the domains were in at least metastable equilibrium during the measurements. We found that the magnetic lateral correlation length increases rapidly below the ordering temperature but saturates at a value of 100 nm for  $T < 177$  K [Fig. 1(e)]. We note that neutron [22] and x-ray [23] imaging of similar spiral magnetic domains in Ho yield domain sizes of the order of microns. However, these imaging measurements were sensitive only to the change of chirality but not other defects, such as spin slip [24], which probably limit the lateral spiral spin correlations to the smaller values observed in our study and in other studies of Dy magnetic domains based on scattering techniques [25]. The correlation length along the  $c$  axis (perpendicular to the film surface) can also be determined, and it is limited by the penetration depth of soft x rays into Dy ( $\sim 20$  nm). Thus we are sensitive only to the domains close to the top Y/Dy interface.

XPCS is a technique for using a coherent x-ray beam to obtain the real time intermediate scattering function  $F(Q, t)$ , i.e., the Fourier transform of  $S(Q, \omega)$  for long times, analogous to what is done in dynamical light scattering. The coherence of the x-ray beam results in the interference pattern of the scattered x rays manifesting itself as speckle, which represents a fingerprint of the scattering system. The scattered intensity around the  $(0, 0, Q_m)$  satellite position essentially disappears below  $T_C$  and above  $T_N$ , which means that the speckle we observe in the AFM spiral phase is primarily magnetic in origin and arises due to fluctuations of the spin arrangements out of

the perfect spiral order [15,26] with some additional weak scattering from the surface roughness. By taking  $N$  successive images with a shutter time of  $\Delta t$  for a time span of  $S$ , where  $S \gg \Delta t$ , a movie of the speckle pattern can be created. Analyzing the time series allows us to calculate the autocorrelation function and determine the temporal evolution of the fluctuating domain walls for varying time delays ( $t_n - t_0$ ),  $n = 0, 1, 2, \dots, N$  over a time span  $S$ .

In Fig. 2(a), we show snapshots of the speckle pattern at a fixed temperature of 178.50 K. Inspection shows that the speckle pattern is changing with time, which is indicative of fluctuating domain walls. In contrast to driven dynamics, as would be the case if the domains were driven with an external stimulus [13,14], such as a magnetic field, the dynamics that we observe are self-sustained and thermally activated. To quantify the dynamic behavior we calculate the intensity-intensity autocorrelation function  $g_2$  from the time series speckle data which is defined as [27]

$$g_2(Q, t) = \frac{\langle I(Q, t')I(Q, t' + t) \rangle_{t'}}{\langle I(Q, t') \rangle_{t'}^2} = 1 + A|F(Q, t)|^2, \quad (2)$$

where  $I(Q, t')$  is the intensity at wave vector  $Q$  at a time  $t'$ ,  $t$  is the time delay, and angular brackets represent averages over the subscripted variable.  $F(Q, t)$  is the intermediate scattering function, and  $A$  is the speckle contrast.

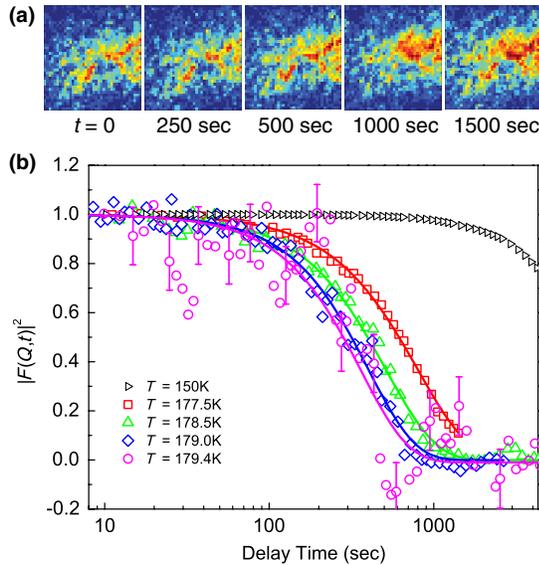


FIG. 2 (color online). (a) Time snapshot of the speckles at 178.50 K. The changing speckle pattern with time indicates dynamics. (b) Normalized autocorrelation function as a function of temperature. The correlation is calculated from the time series of the  $(0, 0, Q_m)$  Bragg peak intensity. All speckles in the Bragg peak are taken into account. The increasingly faster dynamics towards  $T_N$  is evident. A compressed exponential function was used to fit the data. Solid lines are fit to the data. Error bars are not shown when they are comparable to or smaller than the size of the points.

The  $|F(Q, t)|^2$  [Fig. 2(b)] shows strong temperature dependence. We observe dynamics only near  $T_N$ . The dynamics are fastest near  $T_N$  and gradually get arrested within 10 K below  $T_N$ . For  $T < 175 \text{ K}$  [e.g., at 150 K as shown in Fig. 2(b)], the autocorrelation curve is almost constant within the same time span of measurement, and the time constant is beyond the instrumental stability. The dynamics that we observe are not due to critical spin fluctuations which occur at a few tens of nanometer length scales at gigahertz frequency or greater [28]. We observe a smooth decaying curve which is not a simple exponential but a compressed exponential, indicative of the presence of a distribution of decay times due to fluctuating walls in a disordered domain wall network.

In the present case the intermediate scattering function  $F(t)$  has been fitted with the expression

$$|F(t)| = \exp[-(t/\tau)^\beta], \quad (3)$$

where  $\tau$  is the decay constant and  $\beta$  is the stretching exponent. We found  $\beta \approx 1.5$  for all the curves at all measured temperatures [Fig. 3(a), inset]. A value of  $\beta > 1$  signifies collective motion, which means that the domain walls in Y/Dy/Y exhibit cooperative behavior most likely due to the long-range nature of the Ruderman-Kittel-Kasuya-Yosida interaction ( $J_{\text{RKKY}} \propto 1/r^3$ ), which plays a significant role in establishing the spiral structure in this material. More importantly, an exponent of  $\sim 1.5$  has been found in the jammed state of a wide variety of systems including colloidal gels [10], concentrated emulsions, dense ferrofluids [29], and Cr [12]. In these disordered systems, freezing of the fluctuations takes place, and the dynamic behavior corresponds to nondiffusive motion that can be semiquantitatively explained in terms of the relaxation of long-range elastic strains in a stressed network [10]. We note that this explanation may also apply to Cr, since the Cr spins interact via a long-range exchange interaction as well, dominated by Fermi surface effects and the conduction band structure. This will yield the  $\beta \sim 1.5$

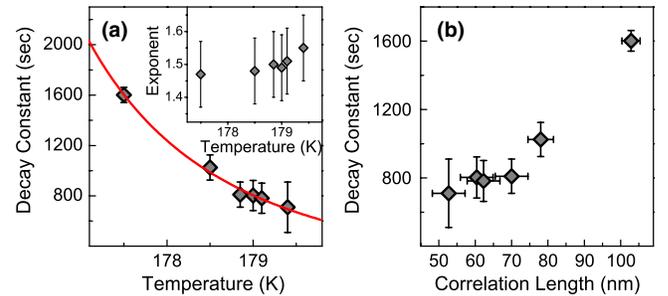


FIG. 3 (color online). (a) Decay constants obtained from fitting the autocorrelation curves in Fig. 2. The red line is a Vogel-Fulcher function fit to the data. Within experimental accuracy we found  $\beta = 1.5$  for all the temperatures (inset). (b) The decay constant versus correlation length. The dynamics slow down as the correlation length increases.

observed in the temperature range just below  $T_N$ . Domain dynamics in Cr are, however, drastically different at low temperature, where they are driven by quantum tunneling down to 4 K. Such a phenomenon is not observed in the case of Dy, most likely because of the localized nature of the magnetic moments in this material.

The physical processes underlying this behavior are as follows: As one approaches the Néel temperature from above ( $T_N = 180$  K), the spin-spin correlation length diverges, but the antiferromagnetic fluctuating clusters which form may have different spin orientations in the basal plane, or opposite chiralities. Below  $T_N$ , these clusters become domains and are head to head with each other leading to frustration; thus, the domains are in a jammed state as soon as the long-range order sets in. In this regime there are normal spin wave excitations, and there are also slow thermal fluctuations of the spins in the domain walls which form a disordered network. These thermally excited spin fluctuations set up a stress field in the domain wall network which are released by other spin fluctuations at a later time  $t$ . The fluctuations thus involve correlated fluctuations of the spins in the domain walls due to the long-range nature of the Ruderman-Kittel-Kasuya-Yosida interaction and result in the compressed exponent 1.5. In most of the jammed systems, there is also a  $Q^{-1}$  dependence of the time constant [10], but we were not able to study the  $Q$  dependence because of the limited range due to the diffraction geometry. We observe the dynamics only when the temperature is very close to  $T_N$ . As the temperature is lowered, the energy required to flip spins and the anisotropy energy increase, which results in reducing domain wall fluctuations. At  $\sim 177$  K the time constant for the fluctuations becomes extremely large, reminiscent of a glassy state.

In analogy to glasses or jammed systems, for gradually freezing domain dynamics we expect that the decay constant should show super Arrhenius behavior [30]. In Fig. 3(a), we plot the decay constant as a function of temperature and fit the data with the Vogel-Fulcher function of the form

$$\tau^{-1} = \tau_0^{-1} \exp[-DT_0/(T - T_0)]. \quad (4)$$

We obtain a good fit with  $T_0 = 171$  K and the fragility parameter  $D = 0.14$ . We note that for pure Arrhenius behavior the fragility parameter should approach infinity. However, another way of looking at Eq. (4) is simply that we have a strongly temperature-dependent activation energy. Figure 3(b) shows the decay constant as a function of correlation length. The dynamics do get slower as the correlated regions grow in size, which suggests that the freezing behavior depends on the size of the domains [31]. A close relation between the energy barriers for domain wall fluctuations and for domain growth is indeed expected.

In summary, we provide evidence of the jamming behavior of domains in a single crystal spiral antiferromagnet. We observe that the domains enter a jammed state as soon

as the long-range order sets in. The dynamics show a collective behavior as evidenced by compressed exponent 1.5 and exhibit a super Arrhenius behavior that can be well described by a Vogel-Fulcher function with a divergence of relaxation time. We also show that the decay constant  $\tau$  is tied to the domain size. For temperature just below  $T_N$ , the domains fluctuate but increasingly get frozen as the temperature shifts towards  $T_0$ . We speculate that this behavior is the origin of the blocking temperature that is often observed in antiferromagnets [32]. The theoretical treatment of jamming transitions, together with computer simulations and experiments on model systems, is quite robust [1,7,31,33,34] and explains many aspects of kinetic freezing type phase transitions. The concept of a jamming transition has not hitherto been applied to magnetic systems but appears to manifest itself in the slow collective dynamics of domain walls below the ordering temperature. Our study shows that domain dynamics near the Néel temperature show many aspects similar to those in supercooled liquids, polymers, colloidal gels, glasses, etc., and bolsters the evidence for the universal nature of the jamming transition in discussing the freezing behavior of disordered and frustrated systems. Because of the restriction in time scales for the experiments, the data are somewhat limited in the range of time scales which could be probed for this system. Future experiments are planned on other systems which will enable us to look for universal behavior associated with domain growth in antiferromagnetic systems when they order via a continuous transition from a paramagnetic state.

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