Domain wall dynamics in a spin-reorientation transition system Au/Co/Au

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We report measurements of domain wall dynamics in an ultrathin Au/Co/Au system that exhibits a spin reorientation phase transition as a function of temperature. The domain walls exhibit cooperative motion throughout the temperature range of 150 - 300 K. The decay times were found to exhibit a maximum at the transition temperature. The slowdown has been explained as due to formation of a double well in the energy landscape by the different competing interactions. Our results show that the complex, slow dynamics can provide a more fundamental understanding of magnetic phase transitions.

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Ultrathin magnetic systems provide a way to physically realize and study the fundamentals of magnetic ordering in low dimensions and can differ drastically from macroscopic systems [1]. The magnetization direction and domain formation is governed by a complex interplay among anisotropy, exchange and dipolar interaction energies. By precise control and tailoring of different parameters such as, thickness and temperature [2, 3], crystal symmetry [4], strain [5], surface adsorption [6], interface roughness [7] and underlayer/overlayer material [3, 8], it is now possible to fine tune the anisotropy that leads ferromagnetic systems to exhibit a spin-reorientation transition (SRT) - where the equilibrium magnetization direction changes from in-plane to out-of-plane and vice versa [1, 9].

Fluctuations play an important role in systems that have reduced dimensionality and complex phases [10, 11]. In magnetic system, fluctuations - technically related to magnetic noise - can affect domain growth and annihilation processes and thus can have implications in technological development of low-dimensional magnetic devices. Although complex, understanding the effect of fluctuations can provide fundamental insight into the competition between anisotropies. In the SRT system, the behavior of the domain wall fluctuation as a function of temperature is not obvious because of the possible presence of non-trivial domain states as the system goes from one stable ferromagnetically ordered state at 300 K to another ferromagnetic equilibrium domain state at low temperature [2, 12–15]. The question of the time correlation of domain fluctuations and their evolution through the transition temperature in SRT system is an important one that has not been addressed yet, and is the topic of this Letter.

By combining the techniques of resonant x-ray magnetic scattering and x-ray photon correlation spectroscopy we directly address the question of domain wall fluctuations in an ultrathin single crystalline Au/Co/Au thin film system. The system exhibits a thermally driven SRT as the temperature is decreased. The study was performed using a coherent x-ray beam with energy resonantly tuned to the L_3 edge of Co to get an element specific speckle pattern. By monitoring the intensity variation within the speckle as a function of time and temperature, the dynamic behavior of the domain walls was determined. We observed collective domain wall dynamics at all temperatures with the time scale of fluctuations becoming longer as the SRT temperature (T_{SRT}) is approached from either side. The decay time near the transition temperature was found to be 2-3 times larger than the decay time at the extrema of the measured temperature range.

The sample used in the experiment was epitaxial Co whose structure is Si(111)/Cu 4 nm/Au 2 monolayer/Co 8 monolayer/Au 3.5 nm grown via molecular beam epitaxy. Further details of the sample can be found elsewhere [8]. The room temperature remanent magnetization direction is determined by the thickness of the Au underlayer. The magnetic characterization has been done using a superconducting quantum interference device. Fig. 1 (b) shows the magnetization as a function of temperature at an applied field of 10 Oe. For a 2 monolayer underlayer thickness, the magnetization principally lies in-plane at or above T = 300 K. The sample shows a broad reorientation transition (T_{SRT}=230 K) with the magnetization rotating out-of-plane as the temperature decreases.

X-ray photon correlation spectroscopy (XPCS) measurements were carried out at beamline 12.0.2.2 of the Advanced Light Source at Lawrence Berkeley National Laboratory. A diagram of the scattering geometry is shown in Fig. 1. The experiment was conducted in a

small angle geometry with the $2\theta = 18$ degrees. The magnetic sensitivity is brought about by tuning the energy of the incoming linear σ-polarized x-rays to the L_3 edge of Co (780 eV) [16, 17]. At such a resonant condition the net scattered intensity from the incoming σ light has contributions from both the charge and magnetic scattering. An energy scan across the Co $L_{2,3}$ edges for the sample at 300 K is shown in Fig. 1 (a). The transverse coherence in the beam was established by placing a ~10 μm pinhole approximately 3 mm in front of the sample. A CCD placed 0.45 m away was used as a detector.

Diffuse scattering of coherent x-rays give rise to speckle due to interference of scattered wavefronts that are randomly phase-shifted by the morphology of the sample, such as surface roughness. For magnetic systems, the speckle pattern will have an additional contribution from magnetic heterogeneity, i.e., domains [18–20]. This magnetic contribution to the intensity distribution in the speckle pattern is enhanced due to the resonance condition. Any change in the morphology results in a change in the speckle pattern. The principle of the experiment is to monitor the variations in the scattered intensity as a function of time and then correlate the intensities. The experiment was conducted by taking a long series of images at a fixed temperature to monitor changes of the speckle pattern and scattered light intensity. Although the intensity within the speckle is a combination of charge and relatively weaker magnetic scattering, we assume that the charge scattering remains static in time [21]. No attempt was made to remove the static charge background features, and any time variations were related to the dynamic feature of the magnetic domains. The analysis was performed at different wavevector $\mathbf{q} = 2\pi/\mathbf{d}$ to determine the dynamic behavior at different length scales $\mathbf{d}$. The above process was repeated for different temperatures. No magnetic field was applied during the experiment.

The intensity-intensity temporal autocorrelation function g_2 is defined as:

$$g_2(\mathbf{q}, \Delta t) = \frac{\langle I(\mathbf{q}, t) I(\mathbf{q}, t + \Delta t) \rangle_t}{\langle I(\mathbf{q}, t) \rangle^2_{\mathbf{q},t}} = 1 + A|F(\mathbf{q}, t)|^2, \quad (1)$$

where $I(\mathbf{q})$ is the intensity at wavevector $\mathbf{q}$, Δt is the time delay, and brackets represents averages over the subscripted variable. F($\mathbf{q}$,t) is the intermediate scattering function and A is the speckle contrast that is determined by the beam coherence, among other things.

The observed speckle pattern in Fig. 1 (a) is rich in structure which signifies the presence of charge and magnetic features at the relevant length scales. A linecut of the data (not shown here) obtained after time averaging along the $\mathbf{q_x}$ direction at $T = 300$K (in-plane magnetization) and 150 K (out-of-plane magnetization) shows the curves retain speckle features due to the constant charge scattering signal. Overall however, they decay smoothly

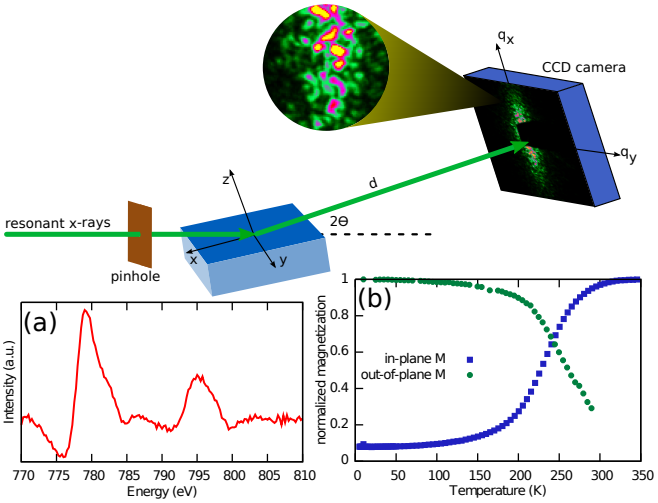

FIG. 1: (color online) The geometry for the resonant x-ray scattering experiment. Incident x-rays are filtered with a ~10 μm pinhole, creating a spatially coherent light source. The scattering pattern is collected with a CCD camera at an angle of $2\theta = 18$ degrees. Detail of the speckle pattern is shown in the inset. The energy scan of the sample in (a) shows the Co L_3 and L_2 edges. The spin-reorientation of the sample is illustrated by the SQUID curves in (b), where the magnetization changes from out-of-plane to in-plane as the temperature is increased.

as a function of $\mathbf{q}$ at both temperatures, as would be obtained for disordered domains. A Lorentzian fit to the curves show that the width of the curve at $T = 150$K is larger than the width at $T = 300$K (FWHM at 300 K is $2 \times 10^{-4} \mathring{A}^{-1}$; FWHM at 150 K is $6 \times 10^{-4} \mathring{A}^{-1}$). In the context of a static charge signal, the increase of the FWHM means that the size of domains become smaller when the magnetization is out-of-plane. The interpretation of this is that the stability of the out-of-plane magnetization state increases by forming a larger number of small domains so as to decrease the dipolar energy [22].

The top panel of Fig. 2 shows the speckle detail and time evolution of the speckle pattern taken at T = 279.48 K. Each image is a 10 sec. exposure of the same region and separated in time by 410.5 sec. The changes in the speckle pattern with time are indicative of slow fluctuations in the magnetic domain structure on the order of hundreds of seconds. The plot in Fig. 2 shows the calculated intensity-intensity temporal autocorrelation function from Equation (1) for $\mathbf{q} = 2.58 \times 10^{-4} \mathring{A}^{-1}$ as a function of temperature. Repeated attempts to fit the data with a power law or logarithmic time decay that are signature of glassy dynamics and domain growth [23, 24] were unsuccessful, rather the data were found to agree very well with a stretched exponential of the form:

$$|F(\mathbf{q}, t)|^2 \propto \exp\left(-(t/\tau)^{\beta}\right)^2, \quad (2)$$

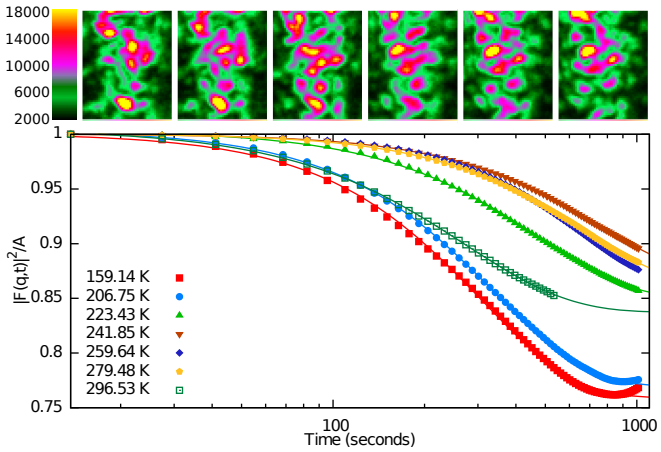

FIG. 2: (color online). Top Panel: False color plot of the speckle pattern. The images are separated in time by 410.5 seconds. Plot of the intermediate scattering function F(**q**,t) as a function of temperature for $\mathbf{q} = 2.58 \times 10^{-4} \mathring{A}^{-1}$. The curves are normalized by the speckle contrast A. The lines are fits to the data. The form of the fit is a stretched exponential and is discussed in the text.

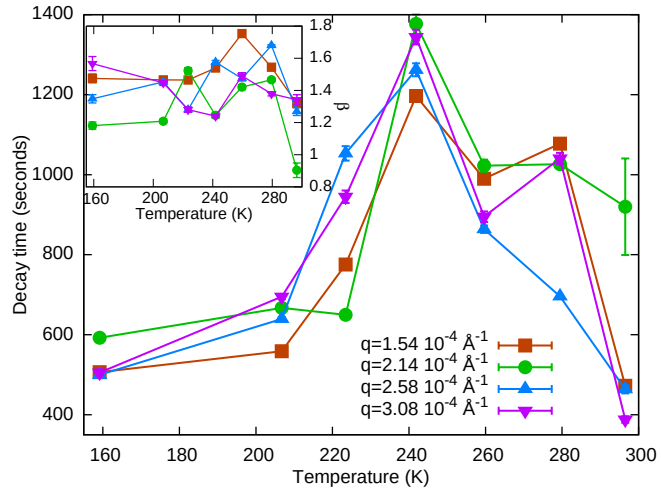

FIG. 3: (color online). Extracted decay times as a function of temperature. The lines are a guide for the eye. There is an increase in the decay time at the spin-reorientation transition. Inset shows the stretching exponent β as a function of temperature.

where τ is the characteristic decay constant and β is the stretching exponent. Within our measurement time, we have not seen the autocorrelation curve go to a fully decorrelated state. This is because a significant amount of speckle intensity is due to the frozen-in charge disorder which is always self-correlated.

Fig. 3 shows the decay constants extracted from the fitted g_2 functions for four different **q** values. The decay constant, on the order of several hundred seconds, shows that the domain walls exhibit slow fluctuations and, interestingly enough, is even longer near T_{SRT}. At the transition the decay time is 2-3 times as large as the decay time above and below the transition. The slowdown is reminiscent of the critical slowdown of fluctuations observed for systems exhibiting second order phase transition [11]. However, we note that we have not found any systematic **q** dependence of the decay times. At this stage we are not sure of the exact reason for such, but it is possible that the frozen-in disorder has some effect on how the domains evolve and fluctuate at a particular length scale at each temperature. The obtained value of the stretching exponent β (inset of Fig. 3) is typically greater than unity but does not show any systematic variation as a function of temperature. Since a value of $\beta > 1$ signifies solid-like collective motion, our results show that the slow fluctuations do not involve independent, uncorrelated events; rather the domain dynamics are collective for the entire measured temperature and wave vector ranges [25].

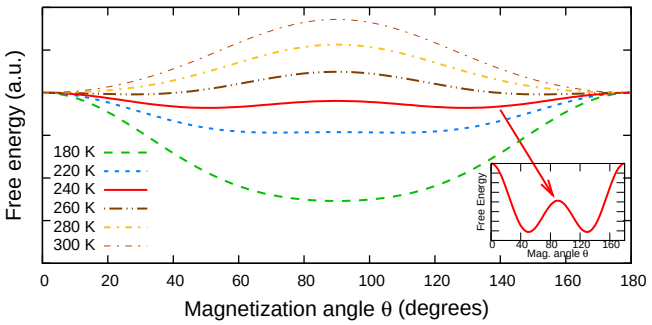

FIG. 4: (color online). Normalized free energy as a function of angle θ (measured from the film plane) for different temperatures. The calculation is described in the text. Above (below) T_{SRT} the energy minimum is such that the magnetization lies in-plane (out-of-plane). Near the transition temperature a double-well potential forms (shown in the inset).

To provide further insight into our results, we calculated the free energy of the SRT system using the first two terms of the anisotropy. We take the free energy here to consist of the first and second order anisotropy constants and the demagnetization energy since there is no external magnetic field in this experiment. It can then be written as [26]:

$$E(T) = \left[2\pi M_s(T)^2 - K_1(T) + 2K_2(T)\right] \sin^2\theta + K_2(T) \sin^4\theta, \quad (3)$$

where $K(T)$ are the temperature-dependent anisotropy constants, $M_s(T)$ is the temperature-dependent saturation magnetization and θ is the angle of the average magnetization measured from the film plane. The temperature dependence of $M_s(T)$ for a two-dimensional system was used [27], along with the measured value of $K_1(T)$ (11.9×10^6 erg/cm^3 at 300K) [8] and the literature value for a similar system for $K_2(T)$ [28]. It is well known [29]

that the Curie temperature of a thin film is smaller than that of the bulk, and our value of $T_C = 900$K was set to match the observed SRT temperature. Fig. 4 illustrates the free energy as a function of angle for different temperatures near T_{SRT}. Although simplified, the energy diagram captures the main aspects of our result. It clearly shows the formation of a well defined energy minimum for $T \gg T_{SRT}$ and $T \ll T_{SRT}$ that shifts gradually from 90° at low temperature to 0° (180°), thereby explaining the spin reorientation transition. At or near T_{SRT} the energy minimum is relatively shallow, and, the competing energies of in-plane and out-of-plane anisotropies balance each other and a double well is formed (inset of Fig. 4). The system is then at a metastable state so that the fluctuations are long lived. An increase in the decay time is consequently observed as T_{SRT} is approached. It is also worth noting that the magnetization is at angle of 45° and 135° for the double well to form. If now the magnetization can take any possible angle in the plane of the sample, then our result is in agreement with the recently proposed and experimentally observed canted and cone state domains at the transition [14, 30, 31].

In conclusion, we have measured domain wall dynamics in a spin reorientation transition system. We found that within the measured temperature range the domain growth dynamic is collective in nature. As the temperature approaches the spin reorientation transition temperature, the fluctuations in the system slows down, resulting in an increase of the decay times.

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