Final Technical Progress Report
NANOSTRUCTURED MAGNETIC MATERIALS

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I. INTRODUCTION

This report describes progress made during the final phase of our DOE-funded program on Nanostructured Magnetic Materials. This period was quite productive, resulting in the submission of three papers and presentation of three talks at international conferences and three seminars at research institutions. Additional new results that will lead to further papers as soon as the data are analyzed were obtained thanks in part to a year-long visiting professor from Chengdu University (China), a month-long visiting professor from Pusan University (Korea), and the current work of a visiting scientist from Singapore. Also, further papers will result from data obtained from ongoing collaborations with researchers at Argonne, Ames Laboratory, the National Institute of Standards and Technology, Hitachi Research and Sandia National Laboratory,

Following this Introduction, the remaining sections of this report contain a description of the progress with DOE funding during the final period of this program; a description of the status of the personnel who worked on this project (Section II); lists of the publications, invited talks, seminars and colloquia crediting DOE sponsorship (Section III); and a summary of current and pending federal support (Section IV). The References are at the end.

Our DOE-funded research efforts were directed toward studies of magnetism at surfaces and interfaces in high-quality, well-characterized materials prepared by Molecular Beam Epitaxy (MBE) and sputtering. We have an exceptionally well-equipped laboratory for these studies, with:

Thin film preparation equipment: three heavily-instrumented MBE machines, two computer-controlled, multi-target sputtering machines, a large rf sputtering machine, and an electron beam gun evaporator.

Characterization equipment: various x-ray diffraction instruments including an 18 kW rotating anode x-ray source with Bragg-Brentano θ -2 θ , low-angle, wide film Debye-Scherrer (Read), Laue, and Seemann-Bohlin; Scanning Tunneling and Atomic Force Microscopy (STM and AFM); Auger, X-ray Photoelectron Spectroscopy (XPS) Ion Scattering Spectrometry (ISS), and Secondary Ion Mass Spectrometry (SIMS); Reflected High and Low Energy Electron Spectroscopy (RHEED and LEED); and Scanning and Transmission Electron Microscopy (SEM and TEM);

Equipment to study magnetic properties of surfaces and ultra-thin magnetic films and interfaces in multi-layers and superlattices: *in situ* (to 2.2 kOe at 10^{-10} torr) and *ex situ* (to 15 kOe)

Surface Magneto-Optic Kerr Effect (SMOKE); variable-temperature Vibrating Sample Magnetometer (VSM); and multi-pass Fabry-Perot interferometer (*in situ* as well as *ex situ*) for spin wave studies of samples as thin as one atomic layer using Brillouin Light Scattering (BLS).

II. RESEARCH ACCOMPLISHMENTS

A. Domain wall dynamics in the spin-reorientation transition system Au/Co/Au

This research was conducted in collaboration with K.A. Seu, S. Roy, J. J. Turner, S. Park, and S. D. Kevan, and took advantage of the facilities of the Advanced Light Sources at LBL. It resulted in the publication of a manuscript in the Physical Review.

We used ultrathin magnetic systems to physically realize and study the fundamentals of magnetic ordering in low dimensions¹. The magnetization direction and domain formation is governed by the interplay among anisotropy, exchange and dipolar interaction energies. By using Molecular Beam Epitaxy to precisely tailor certain physical parameters,^{2,3} crystal symmetry,⁴ strain,⁵ surface adsorption,⁶ interface roughness⁷ and underlayer/overlayer material,^{3,8} we were able to 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.^{10,11} In magnetic system such fluctuations can affect domain growth and annihilation processes, and thus can have implications in technological development of low-dimensional magnetic devices. Although complex, an understanding of fluctuations can provide fundamental insights into the competition between anisotropies. In a spin-reorientation transition (SRT) system, the behavior of domain wall fluctuations 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 a different ferromagnetic equilibrium domain state at low temperature.^{2,12–15} During this past year our work addressed for the first time the question of the time correlation of domain fluctuations and their evolution through the transition temperature in an SRT system.

By combining the techniques of resonant x-ray magnetic scattering and x-ray photon correlation spectroscopy we were able to directly study domain wall fluctuations in an ultrathin single crystalline Au/Co/Au thin film system. This 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 L3 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, we determined the dynamic behavior of the domain walls. We observed collective domain wall dynamics at all temperatures with the time scale of fluctuations becoming longer as the SRT temperature (T_{SRT}) was approached from either side. We found the decay time near the transition temperature was 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 was 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 ($T_{\text{SRT}} = 230$ K) with the magnetization rotating out-of-plane as the temperature decreases.

FIG. 1. Geometry for the resonant x-ray scattering experiment. Incident x-rays are filtered with a $\sim 10 \mu\text{m}$ pinhole to create 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 L3 and L2 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.

X-ray photon correlation spectroscopy (XPCS) measurements were carried out, on samples we prepared specifically for these experiments, at 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 L3 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 L2,3 edges for the sample at 300 K is shown in Fig. 1 (a). The transverse coherence in the beam was established by placing a $\sim 10 \mu\text{m}$ 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 $q = 2\pi/d$ to determine the dynamic behavior at different length scales d . The above process was repeated for different temperatures. No magnetic field was applied during the experiment.

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 q_x direction at $T = 300\text{K}$ (in-plane magnetization) and 150K (out-of-plane

magnetization) shows the curves retain speckle features due to the constant charge scattering signal. Overall, however, they decay smoothly as a function of q at both temperatures, as would be obtained for disordered domains. Our 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

FIG. 2: (Top): False color plot of the speckle pattern. The images are separated in time by 410.5 seconds. (Bottom) Plot of the intermediate scattering function $F(q,t)$ as a function of temperature for $q = 2.58 \times 10^{-4} \text{ \AA}^{-1}$. The curves are normalized by the speckle contrast A . Lines are fits using a stretched exponential.

The top panel of Fig. 2 shows the speckle detail and time evolution of the speckle pattern taken at $T = 279.48 \text{ K}$. 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. Our 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.

FIG. 3: 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.

Within our measurement time, we have not seen the autocorrelation curve go to a fully decorrelated state, indicating 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: 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). The inset shows that near the transition temperature a double-well potential forms.

To provide further insight into our results, we calculated the free energy of the SRT system using the first two terms of the anisotropy²⁶ using the temperature dependence of $M_s(T)$ for a two-dimensional system was²⁷ along with the measured value of $K_1(T)$ and $K_2(T)$.^{28,29} Fig. 4 illustrates the free energy as a function of angle for several temperatures near T_{SRT} . This simplified energy diagram clearly shows the formation of a well defined energy minimum for $T \gg T_{\text{SRT}}$ and $T \ll T_{\text{SRT}}$ 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}

B. Uniaxial Magnetic Anisotropy and Spin Injection Efficiency of the Fe-GaAs(001)

This research was conducted with Philip Ryan, Jong-Woo Kim, Justin M. Shaw, Lahsen Assoufid, Richard A. Rosenberg and David Keavney, and took advantage of the facilities of the Advanced Photon

Source at Argonne. It resulted in the publication of a manuscript in The Physical Review. I only briefly outline the experiment and conclusions here.

We conducted post-growth annealing studies of low coverage, 0.4–7 nm, Fe films on GaAs(001). Our studies revealed the self assembly of structurally ordered, fully strained, Fe₃Ga coherent islands. These islands have a consistent height of 8 monolayers, and their lateral size, shape, and distributional order are elongated along the [110] axis. We found that shear modulus softening along the [110] direction of the Fe₃Ga alloy under biaxial compressive strain is responsible for the shape of these islands. Also, the related magnetic shape anisotropy determines the frequently observed [110] uniaxial magnetic anisotropy of the Fe-GaAs(001) system. We found that the annealing process ($T \geq 200^\circ\text{C}$) concurrently dissociates an interstitially ordered non-ferromagnetic interfacial Fe-As bilayer resulting in a solely ferromagnetic (Fe₃Ga)–semiconducting (GaAs) structure, which reduces interfacial spin scatter.

C. References

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III. STATUS OF RESEARCHERS WORKING ON THIS PROJECT

A graduate student in Optical Sciences, Christopher Zimmer, was funded by DOE for this research starting in November 2008, receiving his M.S. degree in Optical Sciences last year. In addition to his academic skills, he was exceptionally adept at diagnosing and repairing both electronic as well as vacuum problems, which made him a very valuable member of the group.

Between March 2008 and April 2009 Professor Shuang Liu of the University of Electronic Science and Technology of China, Chengdu was in our group. We have one joint publication and another submitted based on her work while she was here. She took several samples and data with her to continue our collaboration so additional publications should result in the future. Also during the summer of 2009 Sungkyun Park from Pusan University in Korea spent a month in our group.

IV. PUBLICATIONS, INVITED TALKS, ABSTRACTS, AND SEMINARS CREDITING DOE SUPPORT 2009–Present

A. Publications Crediting DOE Support, 2009–Present

1. **Cone phase and magnetization fluctuations in Au/Co/Au thin films near the spin-reorientation transition.** K. A. Seu, S. Roy, J. J. Turner, S. Park, Charles M. Falco and S. D. Kevan. Phys. Rev. B 82, 12404 (2010).
2. **Metallic Nanostructures.** Charles M. Falco. in, Proceedings of the Conference on Interacciones en Nanosistemas (Interactions in Nanosystems). Rosario, Argentina. 24 July – 6 August, 2011.
3. **Study of the Multi-Layer Structure of 4nm Pt_xSi film by ARXPS.** Liu Shuang and Charles M. Falco. International J. of Modern Physics B (submitted).

B. Invited Talks at International Conferences Crediting DOE Support 2009–Present

1. **Metallic Nanostructures.** Charles M. Falco. Interacciones en Nanosistemas (Interactions in Nanosystems). Rosario, Argentina. 24 July – 6 August, 2011.
2. **Brillouin Light Scattering Studies of Buried Nanostructures.** Charles M. Falco. Conference on Physics at the Nanoscale. Madrid, Spain. October 20–22, 2011.
3. **Brillouin Light Scattering Studies of Buried Nanostructures.** Charles M. Falco. III International Conference for Young Scientists, Low Temperature Physics (ICYS–LTP–2012). Kharkiv, Ukraine. 14–18 May 2012.

C. Seminars and Colloquia Resulting from our DOE-Funded Research, 2009–Present

1. **Magnetism in Ultra-Thin Films.** Physics Department, University of Arizona. October 2, 2009.
2. **Magnetism in Ultra-Thin Films.** Physics Department. University of Washington. January 24, 2010.
3. **Brillouin Light Scattering from MBE-Grown Ultrathin Films.** Physics Department. Minnesota State University, Moorhead. February 16, 2010.
4. **Brillouin Light Scattering from MBE-Grown Ultra-Thin Films.** Physics Department. Iowa State University, Ames. April 6, 2010.
5. **Brillouin Light Scattering from MBE-Grown Ultrathin Films.** Physics Department. Ohio University, Athens. May 14, 2010.
6. **Brillouin Light Scattering from MBE-Grown Ultrathin Films.** Optical Society of America, Reno, Nevada Chapter, 18 November 2011

V. Current and Pending Support

Name of Investigator: Charles M. Falco

Source of Support	Project Title	Award	Period Covered	Person-Mo [Total]	Location
Current Support					
QNRF	High Performance Electrodes for Solar Cells	1,045,629	10/12 – 9/15	3	UofA
Pending Support					
QNRF	Multispectral Imaging (in preparation)	approx. \$1M	10/13 – 9/16	3	UofA