Nanoscale Dynamical Heterogeneity in Complex Magnetic Materials

Final Report, 2011-15

Stephen D. Kevan, Principal Investigator

Current Grant Number: DE-FG02-11ER46831

Applicant/Institution: University of Oregon
Street Address: Sponsored Projects Services
677 E. 12^{th} Ave, Suite 500
Eugene, OR 97401
Postal Address: 5219 University of Oregon
Eugene, OR 97403-5219

Principal Investigator: Stephen D. Kevan
Professor of Physics
University of Oregon
Eugene, OR 97403-1274
Office: 541-346-4742
FAX: 541-346-3422
e-mail: kevan@uoregon.edu

Admin. Point of Contact: Christopher Pirkle
541-346-3107
postawardteama@uoregon.edu

DOE Program Office: Basic Energy Sciences
Materials Sciences and Engineering Division

DOE Program Managers: Lane Wilson
Lane.Wilson@science.doe.gov

Research Area: Experimental Condensed Matter Physics

I. Research Progress

The current 3-year proposal (plus 1-year no-cost extension) was the first DOE funding for this project, though we had been operating on other funds previously. The stated objectives of the project were:

- Develop protocols to measure field-driven intermittency in magnetic films – Barkhausen cascades – with coherent soft x-rays, and relate these to loss of magnetic domain memory and field-driven changes in local domain symmetry;
- Search for unusual phases and domain intermittency near the thermally-driven spin-reorientation transition in thin magnetic heterostructures;
- Probe local symmetries and domain structures in complex manganites and nickelates in both space and time;
- Further develop methodologies to accomplish diffractive imaging in a Bragg or low angle reflection geometry.

We have made progress on the first and second, and fourth of these, though success in the third and fourth has been limited. We have gotten involved in two new and rapidly developing areas of research in magnetism, skyrmions and spin ice structures, and these will be described briefly below. Skyrmions form a major focus of our renewal proposal, though spin ice structures do not since it lies a little outside our primary area of interest.

I.a Intermittency and hidden symmetries

An important achievement has been observation of intermittent Barkhausen events using coherent scattering, as indicated in Fig. 1 (reproduced from the renewal proposal main text so this three-year report is complete). These cascades occur when a small region of the domain pattern encounters a barrier to reversal that is abruptly overcome as the field is varied, leading to a magnetization avalanche. We observed and characterized these by calculating a 'two-field' correlation function, in analogy with several studies of intermittent cascades in soft and hard systems using hard x-ray coherent scattering using two-time correlation functions.

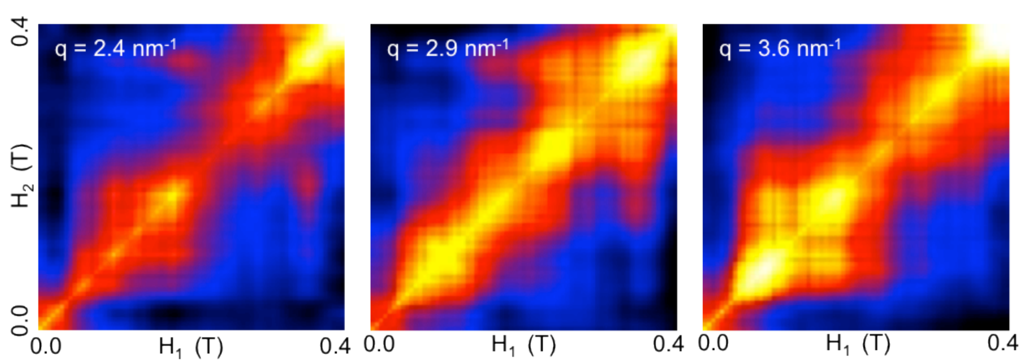

Fig. 1: Normalized two-field correlation functions, $\langle I(H_1)I(H_2)\rangle / \langle I(H_1)\rangle\langle I(H_2)\rangle$, for a CoPd multilayer magnetic film at three different scattering wave vectors. The squarish structures along the diagonal correspond to Barkhausen cascades, often not clearly resolved from one another. We are seeking other correlation functions that will better distinguish individual events.

Are these cascades are related to the hidden rotational symmetries present in Fig. 2? This would be a major step toward developing a microscopic understanding on intermittency. We are also investigating the relationship between these Barkhausen events and microscopic domain memory that we have studied for many years.

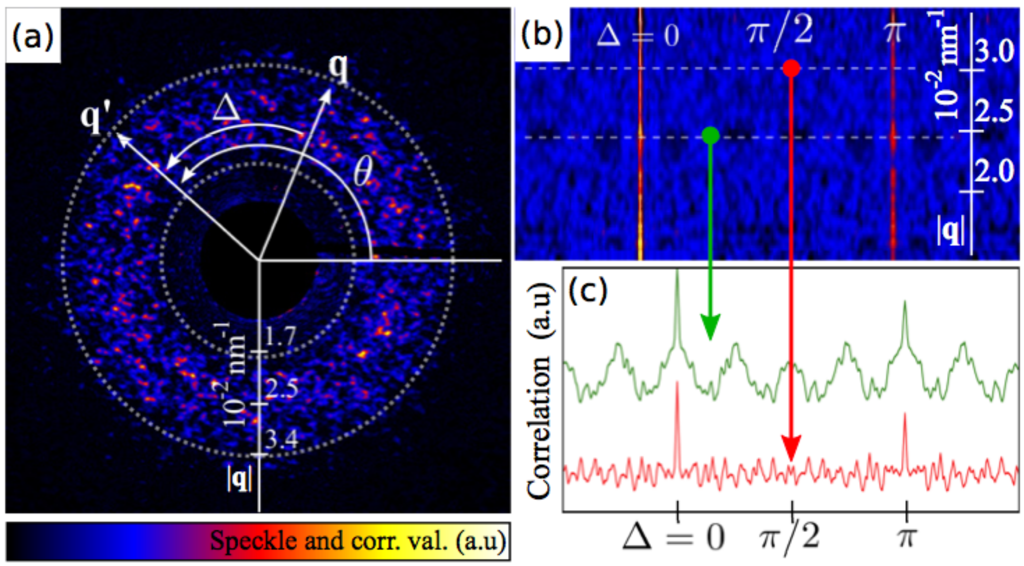

Fig. 2: 'Hidden' rotational symmetries in magnetic domains appear in the rotational autocorrelation functions (b) of speckle patterns (a). We have shown that these are due to finite sampling of the domain structure, but this does not mean they are unrelated to Barkhausen cascades like those in Fig. A.II.1, which also pertain to a local region. Establishing the relationship between these two kinds of correlation functions is a major focus of our renewal proposal.

We have done two other analyses to understand these symmetries. First we have analyzed a large field-of-view magnetic force microscope (MFM) image of a similar domain pattern and searched statistically for hidden symmetries. These were found in the rotational correlation function of the Patterson function (the Cartesian auto correlation of the image). We have shown mathematically the relationship between these real and Fourier space images, and a paper is in preparation to describe these results. The second analysis approach is to calculate three-angle rotational correlation functions of speckle patterns. These 2-D maps shown dramatic arrays of symmetries that have so far eluded understanding. They also show intermittency, and we think this might be a fruitful approach to related local (accidental) symmetries to Barkhausen events.

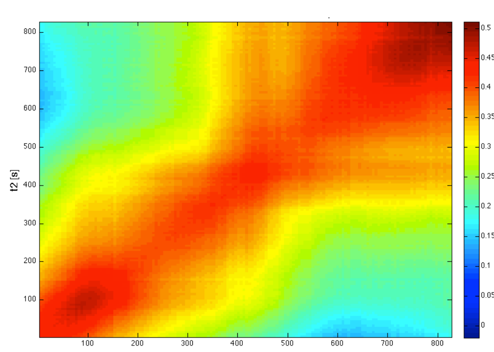

Fig. 3: Wave-vector-resolved two-field correlation functions of soft x-ray speckle patterns collected in transmission from an Au/Co/Au heterostructure thin film. The overlapping square structures along the $H_1 = H_2$ diagonal indicate abrupt decorrelation at certain field values caused by Barkhausen cascades

We have searched for magnetization intermittency in a spin-reorientation system consisting of Au/Co/Au heterostructures. In this case the magnetization is switched from perpendicular to parallel to the film plane bay varying the temperature, not the applied field. We have calculated two-time correlation functions and have found good evidence for such intermittency (Fig. 3), which would then be associated with domain ripening. Again, the square-ish structures along the diagonal in this figure strongly suggest thermally-driven intermittency. We are keen to do more such experiments, but the time scales are quite long and we will wait until the COSMIC end station, which will have much better stability, to pursue these measurements.

We have done a similar analysis to search for thermally-driven intermittency in $Pr_{0.6}Ca_{0.5}MnO_3$ near the orbital-ordering transition. So far we have been unsuccessful, but this transition involves largely static orbital domains in the presence of small fluctuations, which means the intermittency (if it exists) will be very subtle. Again, higher stability and signal on COSMIC and on the NSLS-II CSX beamline will greatly help these measurements.

As part of the experiments on $Pr_{0.6}Ca_{0.5}MnO_3$, we have applied our approach to do coherent diffraction imaging in a Bragg reflection geometry. This has turned out to be a very difficult and ultimately unsuccessful experiment. We are presently trying other modalities to achieve such experiments.

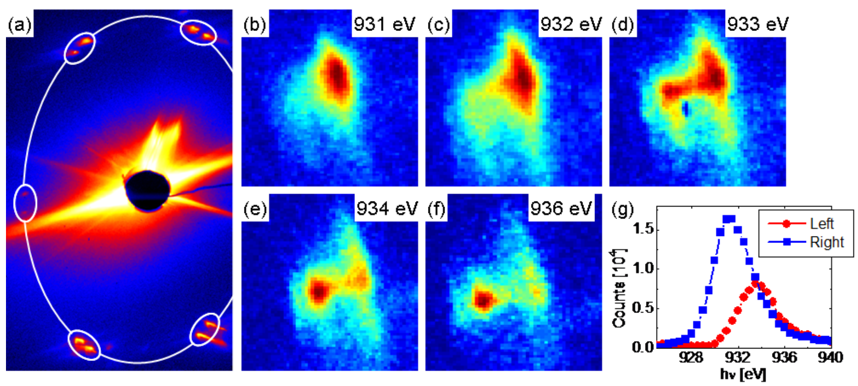

Fig. 4: Scattering off the Skyrmion lattice in Cu_2OSeO_3. a) Diffraction patter near the (100) crystal Bragg peak. 5 of 6 satellite peaks from the hexagonal skyrmion phase are readily apparent. (b)-(e) Evolution of a single split satellite peak as a function of photon energy near the Cu L_3 edge. The two split peaks resonate at different energies.

I.b Coherent soft x-ray scattering results from skyrmion phases

Skyrmions, swirling topological spin defects, have generated enormous interest recently. Aside from their fundamental physics interest, these magnetic objects are stable and loosely coupled to the lattice, and so have been proposed for an array of ultralow power spintronic applications. They were first studied by neutron scattering and Lorentz TEM, but the results in Fig. 4 are the first observation using soft x-rays. As a function of field and temperature, we found an unusual rotational splitting of the skyrmion satellite peaks that indicates the existence of two skyrmion phases rotated with respect to each other. Perhaps more importantly, we can now use the momentum and spatial resolution and temporal sensitivity of coherent soft x-ray beams to probe skyrmions in an entirely new way. This provide the second major focus of our renewal proposal.   The results in Fig. 4 are submitted for publication.

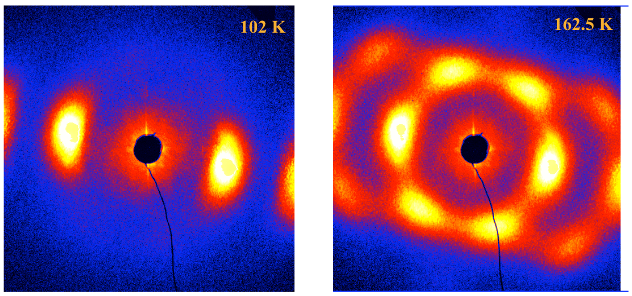

Fig. 5: Speckle patterns collected in resonance with the Fe L-edge in transmission through an Fe:Gd multilayer film. The 6-fold skyrmion phase (right panel) is observed over a narrow range of temperature and applied field close to magnetic saturation. At lower temperature (left panel) a phase with approximately aligned magnetic stripes is observed. This roughly mirrors the helical phases observed near the skyrmion phase in other systems

We are already searching for other skyrmion and possibly skyrmion-like phases, and we might have found one on Fe:Gd multilayers, as indicated in Fig. 5. The phase diagram of the observed hexagonal phase is skyrmion-like, with helical or conical phases at lower and higher temperature, and the skyrmion phases exists at non-zero field. We are actively trying to understand this system by applying other probes and further mapping the phase diagram. We are particularly excited about this thin film system since it will be easier to drive the phase with electrical current.

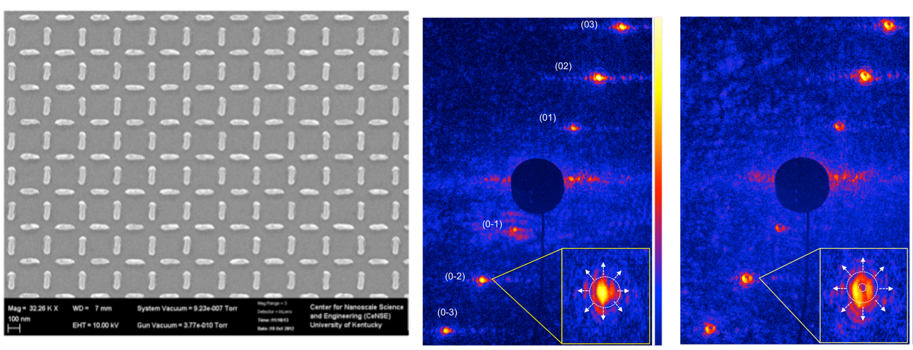

Fig. 6: left: SEM micrograph of a square array of permalloy islands – an artificial spin ice with a highly degenerate ground state magnetization. middle and right: diffraction patterns of the spin ice lattice, collected at the Fe L_3 resonance to achieve magnetic contrast, at remanence (middle) and partly magnetized (right). The Bragg spots on the right develop ring-like structures that are highly suggestive that we have produced beams having orbital angular momentum with a phase singularity in the middle.

I.c Recent coherent soft x-ray scattering results from artificial spin ice structures

Another class of magnetic systems of significant current interest around the world is artificial spin ices. These are lithographically-prepared lattices of magnetic nano-islands in which the ground state is frustrated – like the intrinsic proton disorder in water ice (Fig. 6). Spin ices also have excitations that exhibit some characteristics of magnetic monopoles connected by Dirac strings. We have found an unusual and so far not completely understood effect. When the spin ice is partly magnetized, meaning that the pseudo-monopoles are partly separated by Dirac strings, we observe that the Bragg peaks evolve into circular doughnuts. The effect happens only when the incident beam is tuned to resonance with the Fe L3 edge, and the sample is partially magnetized. This shows that the effect is magnetic in origin. The size of the donut and intensity profiles have the functional form and correct size to be composed of photons that have one unit of orbital angular momentum (OAM). We have good evidence that this is indeed what we have observed, and we are developing a robust model. Beams with non-zero OAM are very much a hot topic at present, though not in magnetism and certainly not in spin ice structures. We are also pursuing XPCS studies on these structures as a function of applied magnetic field. We see that the decay rates depend on the applied field and a dynamic Monte Carlo simulation performed by our collaborators (Patrik Henelius, Royal Institute of Technology, Sweden) indicates that the fluctuations appear due to motion of monopole-like excitation in the sample.

II. Journal publications acknowledging DOE support from current grant:

Published in 2011-2015

1. Su, R., K.A. Seu, J.J. Kan, E.E. Fullerton, S. Roy, and S.D. Kevan, "Emergent rotational symmetries in disordered magnetic domain patterns," *Phys. Rev. Lett.* **107**(25), 257204 (2011). http://dx.doi.org/10.1103/PhysRevLett.107.257204
2. Turner, J.T., X. Huang, O. Krupin, K.A. Seu, D. Parks, S.D. Kevan, E. Lima, K. Kisslinger, I. McNulty, R. Gambino, S. Mangin, S. Roy, and P. Fischer, "X-ray diffraction microscopy of magnetic structures," *Phys. Rev. Lett.* **107**, 033904 (2011). http://dx.doi.org/10.1103/PhysRevLett.107.033904
3. Chesnel, K., J. Nelson, B. Wilcken, and S.D. Kevan "Mapping spatial and field dependence of magnetic domain memory by soft X-ray speckle metrology," *J. of Synch. Rad.* **19**, 293 (2012).
4. Chesnel, K., B. Wilken, M. Rytting, S.D. Kevan and E.E. Fullerton, "Field and temperature dependence of magnetic domain memory induced by exchange couplings," *New J. Phys.* **15**, 023016 (2013). doi:10.1088/1367-2630/15/2/023016
5. Pierce, M.S., J.E. Davies, J.J. Turner, K. Chesnel, E.E. Fullerton, J. Nam, R. Hailstone, S.D. Kevan, J.B. Kortright, Kai. Liu, L.B. Sorenson, B.R. York, and O. Hellwig, "Influence of structural disorder on magnetic domain formation in perpendicular anisotropy thin films," *Phys. Rev. B* **87**, 184428 (2013). http://dx.doi.org/10.1103/PhysRevB.87.184428
6. "Development of coherent scattering and diffractive imaging and the COSMIC facility at the Advanced Light Source", D. Shapiro, S. Roy, R. Celestre, W. Chao, D. Doering, M. Howells, S. Kevan, D. Kilcoyne, J. Kirz, S. Marchesini, K.A. Seu, A. Schirotzek, J. Spence, T. Tyliszczak, T. Warwick, D. Voronov and H.A. Padmore, J. Phys.: Conf. Ser. **425**, 192011 (2013). doi:10.1088/1742-6596/425/19/192011
7. "Altered magnetism and new electronic length scales in magneto-electric $La_{2/3}Sr_{1/3}MnO_3$–$BiFeO_3$ heterointerface", Mazumdar, K. Tarafdar, Lin-Wang Wang, S. D. Kevan, C. Sanchez-Hanke, A. Gupta and S. Roy, New J. Phys. **15**, 113042 (2013). doi:10.1088/1367-2630/15/11/113042
8. "Coupled Skyrmion Sublattices in Cu_2OseO_3", Langner, M.C., S. Roy, S. K. Mishra, J. C. T. Lee, X. W. Shi, M. A. Hossain, Y.-D. Chuang, S. Seki, Y. Tokura, S. D. Kevan, and R. W. Schoenlein, Phys. Rev. Lett., 112, 167202 (2014). dx.doi.org/10.1103/PhysRevLett.112.167202.
9. "Partially Coherent X-ray Diffractive Imaging of Complex Objects". Parks, D.H., X. Shi, and S. D. Kevan, Phys. Rev. A 89, 063824 (2014). DOI: 10.1103/PhysRevA.89.063824.
10. "Pixelated Detector with Photon Address Event Driven Time Stamping and Correlation", Williams, George M., Jehyuk Rhee, Adam Lee, IEEE Transactions on Nuclear Science **61**, 2323 (2014). (paper with Voxtel engineers describing the SOI-based time-stamping detector). DOI: 10.1109/TNS.2014.2327513

11. “Evaluation of Partial coherence correction in X-ray ptychography”, Nicolas Burdet, Xiaowen Shi, Daniel Parks, Jesse N. Clark, Xiaojing Huang, Ross Harder, Sujoy Roy, Steve Kevan, and Ian K. Robinson, Optics Express **XX**, XXXX (2015).

Currently submitted

1. “Controlled transfer of orbital angular momentum eigenstates using magnetic charge defects of artificial spin ice”, S. K. Mishra, J. C. T Lee, Vinayak S. Bhat, X. Shi, D. H. Parks, B. Farmer, Lance E. De Long, I. McNulty, S. D. Kevan, and S. Roy, submitted to Nature Phot.
2. “High resolution magnetic imaging using resonant ptychography with circularly polarized x-rays”, X. Shi, J.C.T. Lee, D. A. Shapiro, M. Farmand, T. Tyliszczak, S. K. Mishra, D. Parks, J. N. Clark, E. E. Fullerton, S. Montoya, P., S. Roy,* and S. D. Kevan, submitted to Phys. Rev. Lett.
3. “Field quench induced nonequilibrium magnetic relaxation in artificial spin ice”, S. K. Mishra, J. C. Andresen, V. S. Bhat, E. Teipel, B. Farmer, J. C. T. Lee, X. Shi, L. E. De Long, P. Henelius, S. D. Kevan, and S. Roy, submitted to Phys. Rev. Lett.

In preparation

1. “Room temperature skyrmions in FeGd films”, J.C.T. Lee, X. Shi, E.E. Fulelrton, S. D. Kevan, and S. Roy, in preparation.

II.3 Presentations related to sponsored research

1. *Complexity and Coherent Soft X-ray Scattering, ALS Cross-cutting Review of Scattering, December, 2011,* talk by Kevan
2. *Hidden Domain Symmetries in Magnetic Films,* Brookhaven National Laboratory, September, 2012, , talk by Kevan
3. *Hidden Symmetries in Magnetic Domains*, ALS Users Meeting, October, 2012, , talk by Kevan
4. *Optics Needs for Coherent X-ray Beams*, DOE Workshop on X-ray Optics Needs, March, 2013, , talk by Kevan
5. *First Experiments at CSX*, NSLS-II First Science Workshop, August 2013, talk by kevan
6. *Toward a Microscopic Understanding of Intermittency*, UC San Diego Physics Colloquium, May, 2013, talk by Kevan
7. *Coherent Soft X-ray Science at the ALS*, DIAMOND Light Source, July, 2013, talk by Kevan
8. *Looking Back, Looking Forward*, after-dinner talk at the Conference of the Spectroscopy of Novel Superconductors, July, 2013, talk by Kevan
9. *An Outside-In Look at ARPES at SRC*, SRC Users Meeting, October, 2013, talk by Kevan
10. *High resolution magnetic imaging using resonant ptychography with circularly polarized x-rays, 59th* Magnetism and Magnetic Materials Conference, November, 2014, delivered by Xiaowen Shi
11. *High resolution magnetic imaging using resonant ptychography with circularly polarized x-rays*, Advanced Light Source User Meeting, October, 2014, delivered by Xiaowen Shi
12. *Soft X-ray ptychography on magnetic nanostructures*, contributed talk at the March Meeting American Physical Society, 2014, delivered by Xiaowen Shi,
13. *Coherent soft x-ray scattering of magnetic systems*, invited talk at the American Physical Society March Meeting, Austin, 2015.
14. *Coherent soft x-ray scattering of magnetic systems*, invited speaker, AVS 61st International symposium and exhibition, Baltimore, 2014, delivered by Sujoy Roy
15. *Coherent soft x-ray scattering of magnetic systems*, invited speaker, Coherence 2014: International

Workshop on Phase Retrieval and Coherent Scattering, Chicago 2014, delivered by Sujoy Roy

16. *Coherent soft x-ray scattering of magnetic systems*, George Arfken Scholar Seminar, Dept. of Physics, Miami University 2014, delivered by Sujoy Roy
17. *Coherent soft x-ray scattering of magnetic systems*, Photon Sciences Seminar Series, SLAC, 2014, delivered by Sujoy Roy
18. *Coherent soft x-ray scattering of magnetic systems* invited speaker, 2012 APS March meeting Baltimore 2013, delivered by Sujoy Roy
19. *Coherent soft x-ray scattering of magnetic systems*, Invited speaker, LCLS User meeting workshop, SLAC, 2013, delivered by Sujoy Roy
20. *Coherent soft x-ray scattering of magnetic systems*, Invited speaker, International Conference on x-ray microscopy, Shanghai 2012, delivered by Sujoy Roy
21. *Coherent soft x-ray scattering of magnetic systems*, Nebraska Center for Materials and Nanoscience, University of Nebraska – Lincoln, April 2012, delivered by Sujoy Roy

II.4 Project personnel partially supported on this grant

1. Keoki Seu, Postdoctoral Associate, was partially paid by this grant in 2011-12, and partially supported by the ALS Postdoctoral Fellowship Program
2. Daniel Parks, UO Physics doctoral student, supported 50% by this grant and 50% by an ALS doctoral fellowship; defended his thesis 5/3/2013.
3. Run Su, UO Physics doctoral student, supported 50% by this grant and 50% by an ALS doctoral fellowship through 2011, then supported by UO funds; defended his theses 11/8/12.
4. James Lee and Xiaowen Shi, Postdoctoral Associates, supported 50% by this grant and 25% by the ALS Postdoctoral Fellowship Program, 25% by University of Oregon funds.

II.5 Estimate of Unexpended Funds

We do not expect to have unexpended funds at end of the budget period.