Nanoscale Intermittency and Emergence in Complex Materials

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Intermittency: an adiabatic change in applied stress that produces a (seemingly) random series of fast responses

(Intermittent?) fluctuations near the spin-reorientation transition

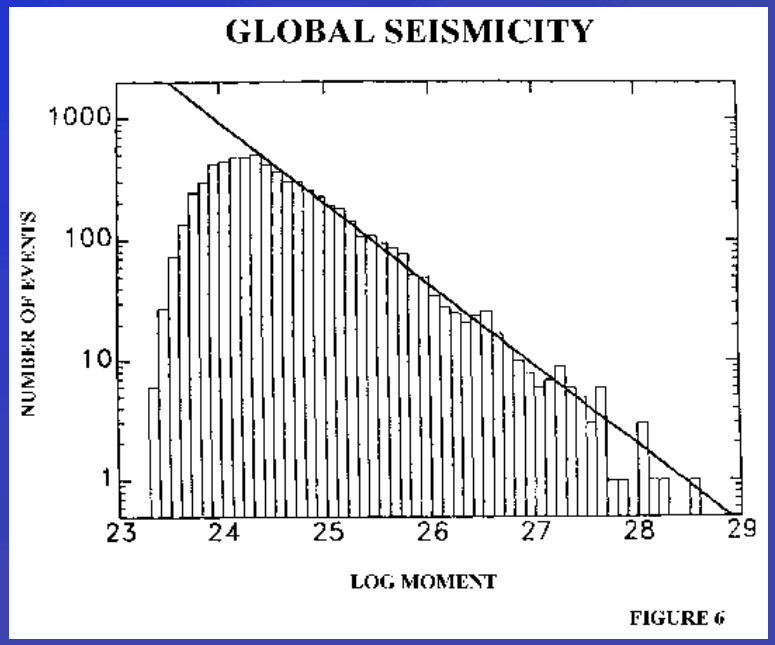
Conclusions

Intermittency and Self Organized Criticality at the Macroscale

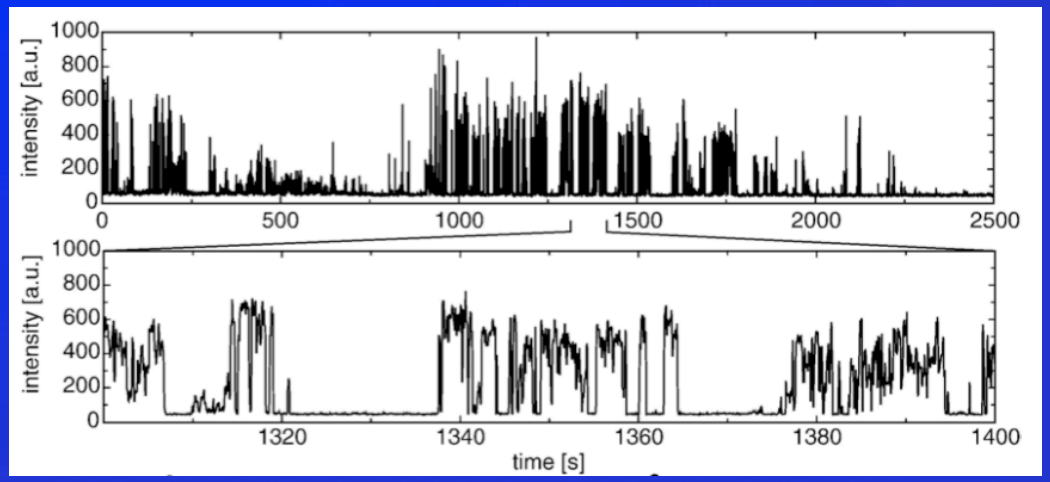
Earthquakes are intermittent, seemingly random events; are they?

Power law spectrum of earthquake magnitudes is presented as a common example of self-organized criticality (though this is apparently not a correct interpretation). This is the Gutenberg-Richter relationship.

[Lay, T., & Wallace, T. Modern Global Seismology, Academic Press, San Deigo, CA, 1995.]



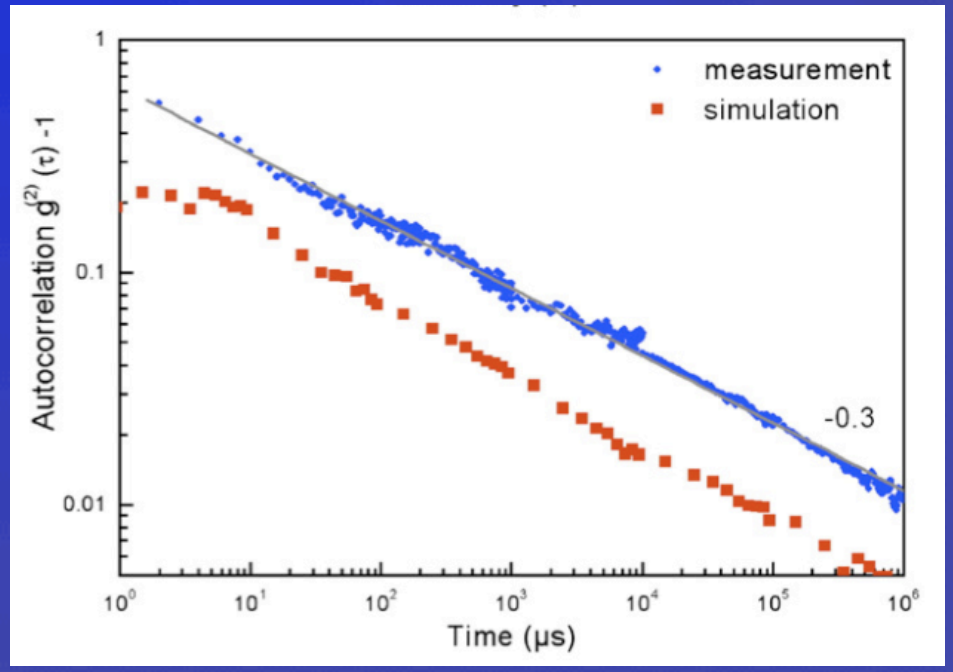
Intermittency in Luminescence from Single Quantum Dots



Time-dependent luminescence from a single CdS nanocrystal shows random telegraph noise.
[R. Verberk, et. al. Phys Rev B 66, 233202 (2002).]

Intensity ACF shows power law behavior over >6 decades of time; exponent depends on environment but not much on temperature.
[C. von Borczyskowski, et. al., Phys Rev B 71, 161302, (2005).]

Long-time averages of on/off periods often shows stretched exponential behavior as well.



Intermittency Near the Colloidal Glass Transition

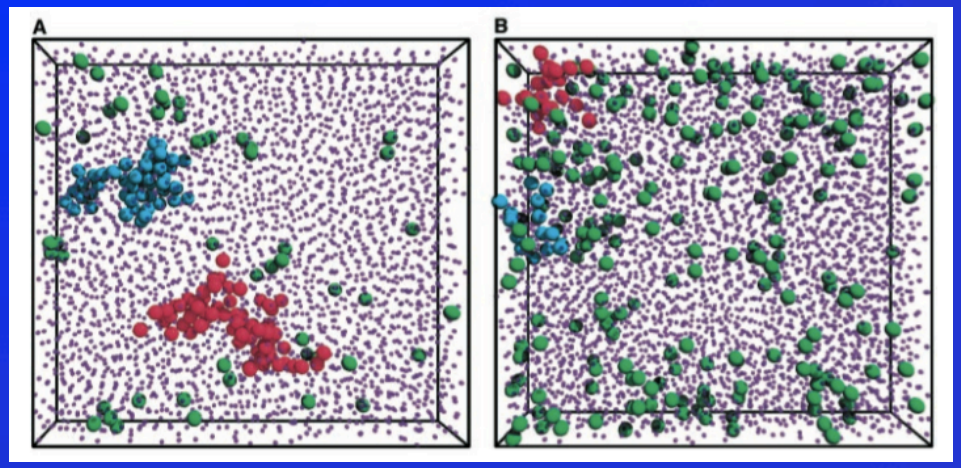
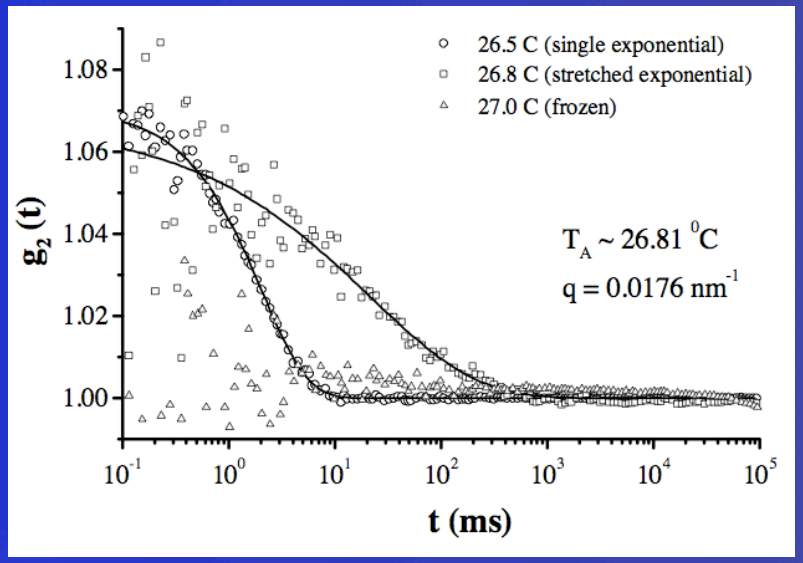
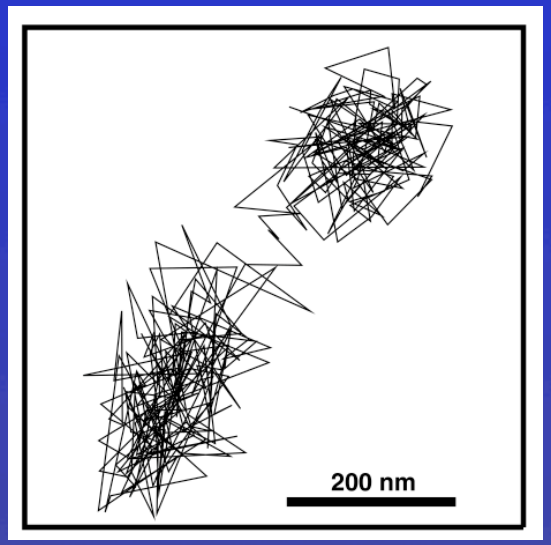


Image of particle trajectories in a colloid near a glass transition showing dynamical heterogeneity; large dots are 'fast' particles. [Weeks, et. al., Science 287, 627 (2000).]

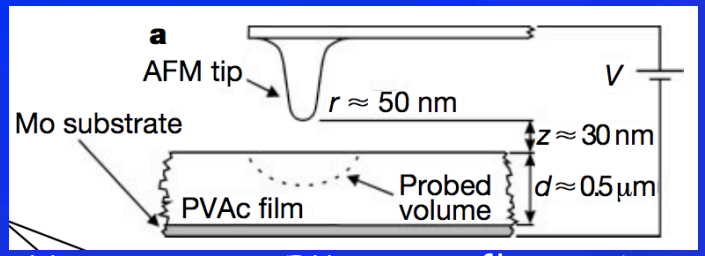


Ensemble diffusive behavior showing stretched exponential decay near the colloidal glass transition, measured with x-ray photon correlation spectroscopy. [Pontani and Narayanan, J. Appl. Cryst. 36, 787 (2003).]

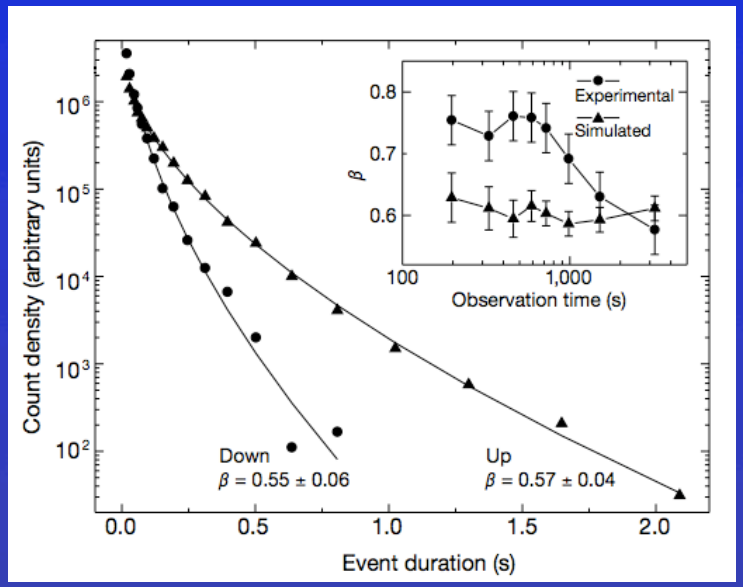


Typical trajectory of a single 'fast' particle near the colloidal glass transition showing intermittent diffusive behavior.

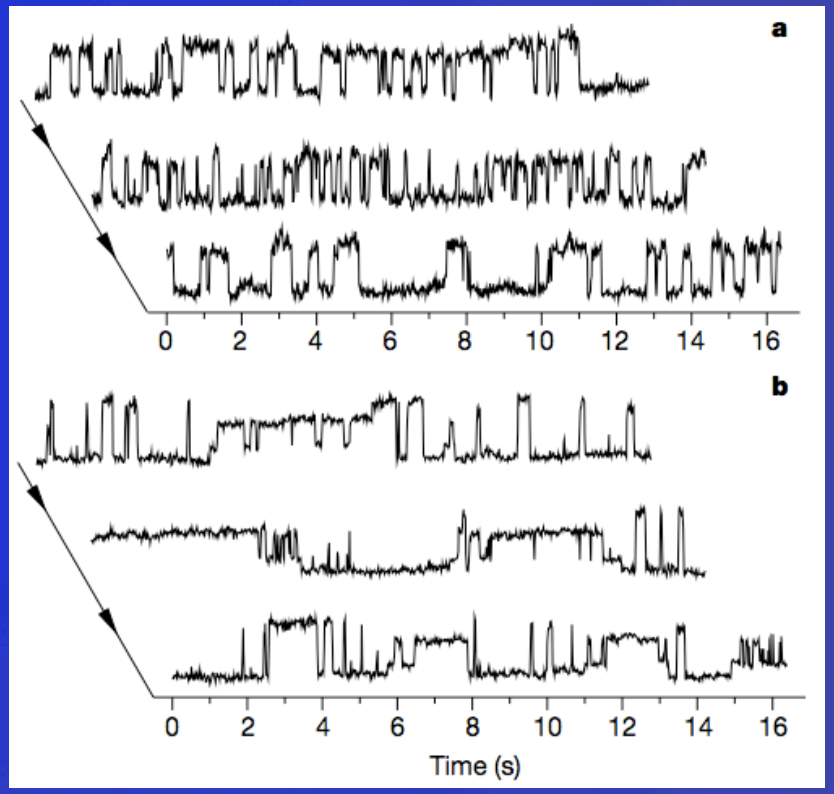
Intermittency in the Dielectric Properties of PVAc Films



Non-contact AFM senses fluctuations in local electrical force and thereby the local dielectric properties.



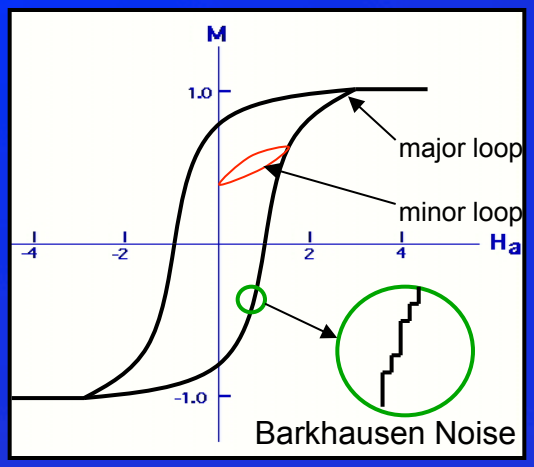
Histogram of 'up' and 'down' states exhibit stretched exponential behavior.

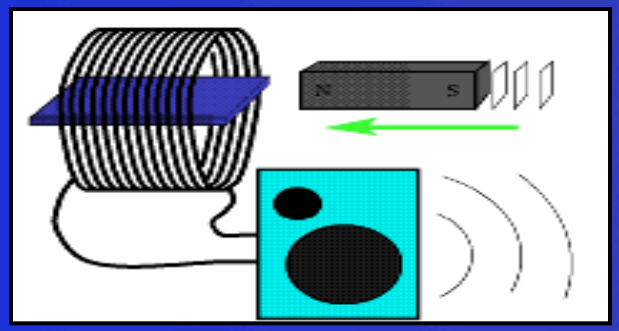


Random telegraph signal suggests 'dynamical heterogeneity' near and below the glass transition. Similar words have been used to describe microphase separation in TMO's.

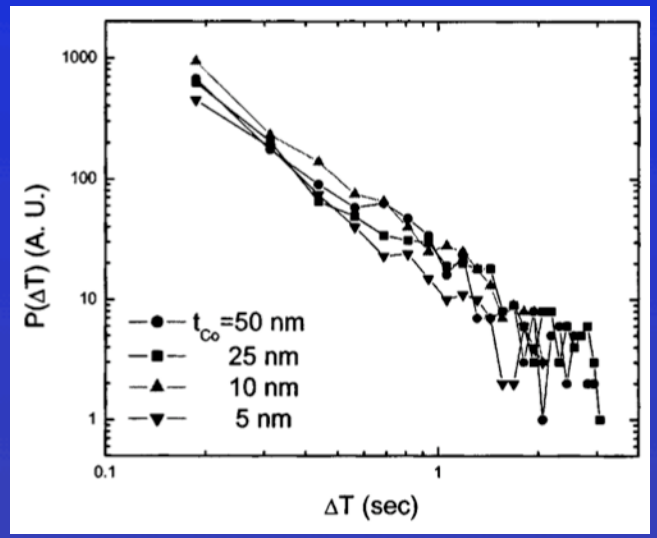
Russell and Israeloff, Nature 408, 695 (2000).

Intermittency Magnetization Loops

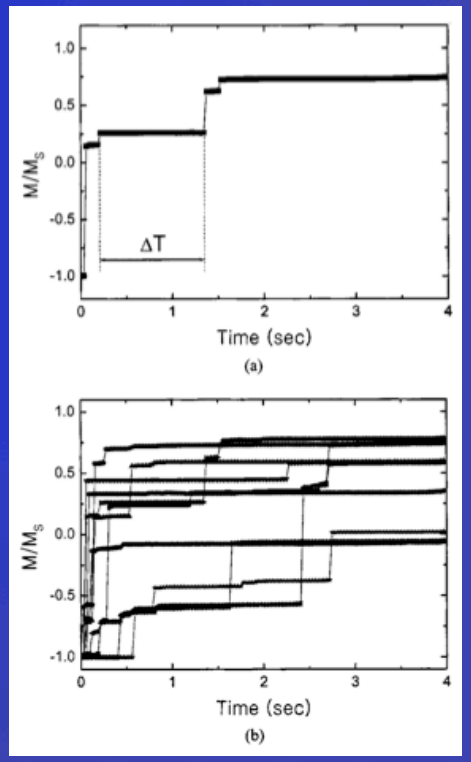




Feynman Lectures on Physics
vol. 2, Fig. 37-11



Histogram of intervals between Barkhausen events showing power law behavior.



Barkhausen events in a Co thin film [Kim and Shin, J. Appl. Phys. 95, (2004).]

Probing Nanoscale Intermittency

Nanoscale intermittency connects (ultra)fast dynamics with slow, complex kinetic behaviors, but it's not clear that pump-probe studies will measure the dynamics that are relevant to intermittency.

Intermittency will impact the performance of many nano-devices. We should find ways to measure and, hopefully, to control it.

This is a classic example where you'd like to measure rare events with microscopy to see what's happening in real space and also to measure a statistical process in reciprocal space.

The difficulty is that there are no characteristic length and time scales, so you need simultaneous and very broad spatial resolution and time resolution, in regimes that are not very well probed by many techniques.

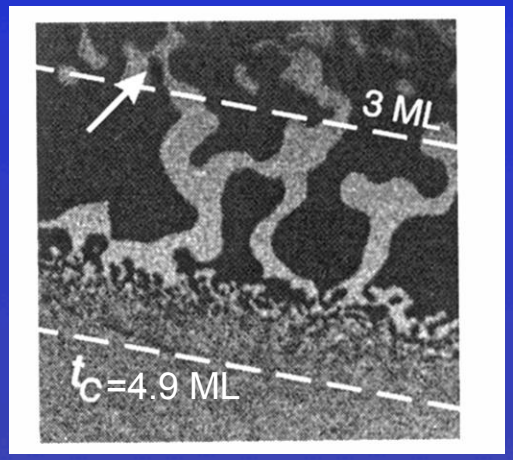
Spin Reorientation Transitions: Can Barkhausen Noise Be Driven Thermally?

Nèel predicted in 1954 that thin films might exhibit anomalous perpendicular anisotropy.

Several studies in the early 1990's demonstrated this behavior.

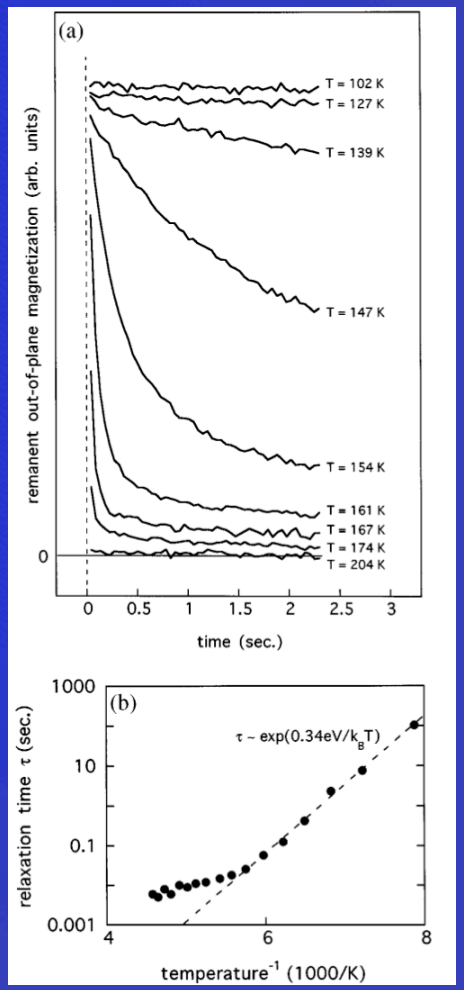
Now this is used in your hard drive.

SEMPA measurements of Co/Au(111) in Speckmann, Oepen, and Ibach, Phys. Rev. Lett. 75, 2035 (1995).



Scale: 100 μm x 100 μm

→→ Thickness increases →→



Simple Arrhenius macroscopic kinetic behavior of domain recovery in Fe/Ag(100).

SMOKE measurements in A. Berger and H. Hopster, PRL 76, 519 (1996).

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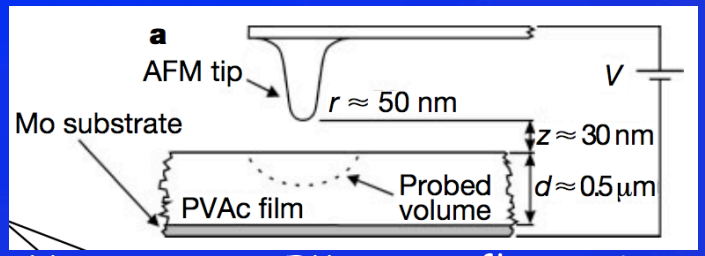

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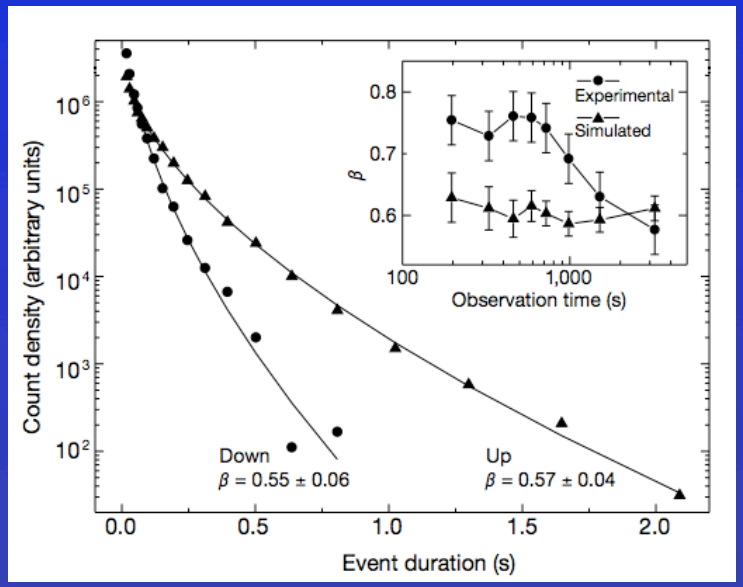

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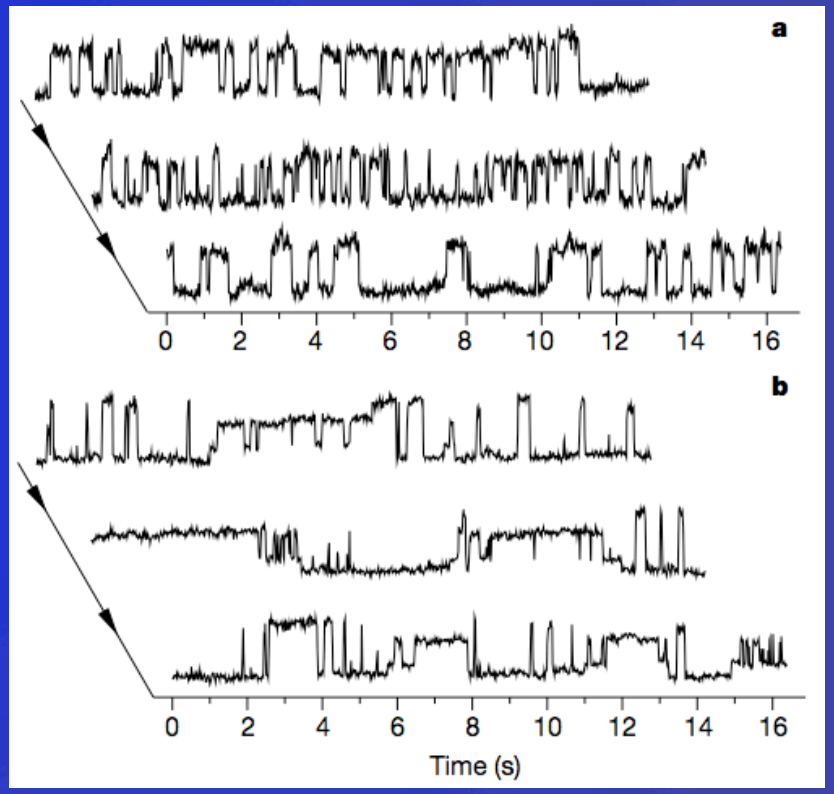

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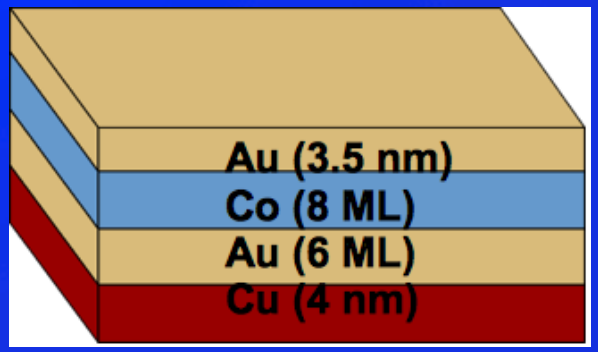
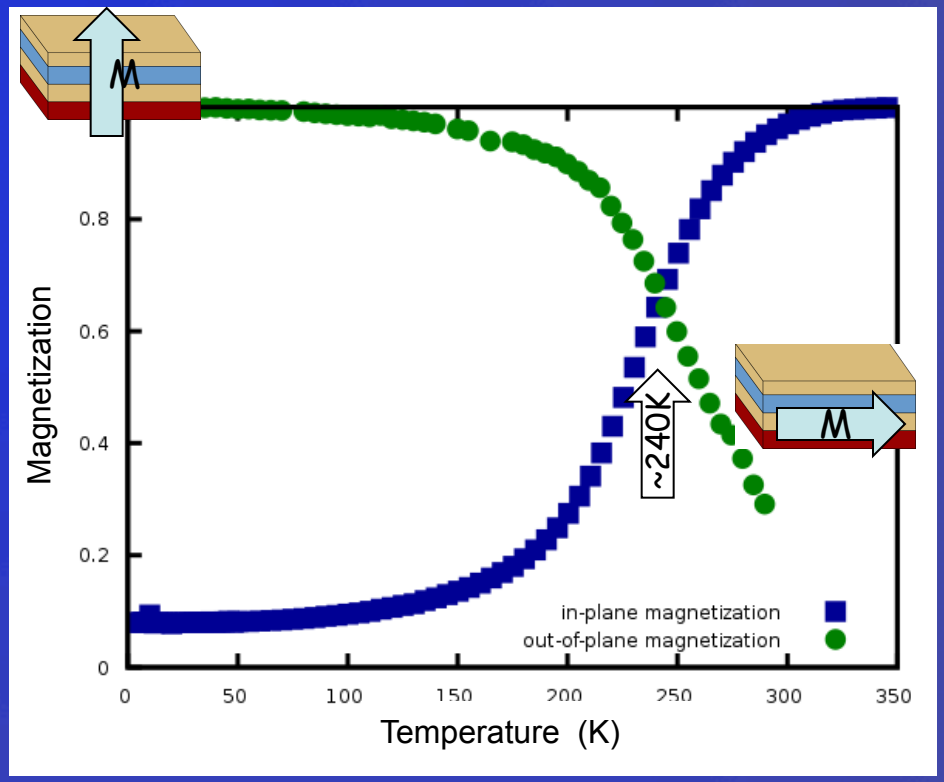
Spin Reorientation Transition in Au:Co:Au



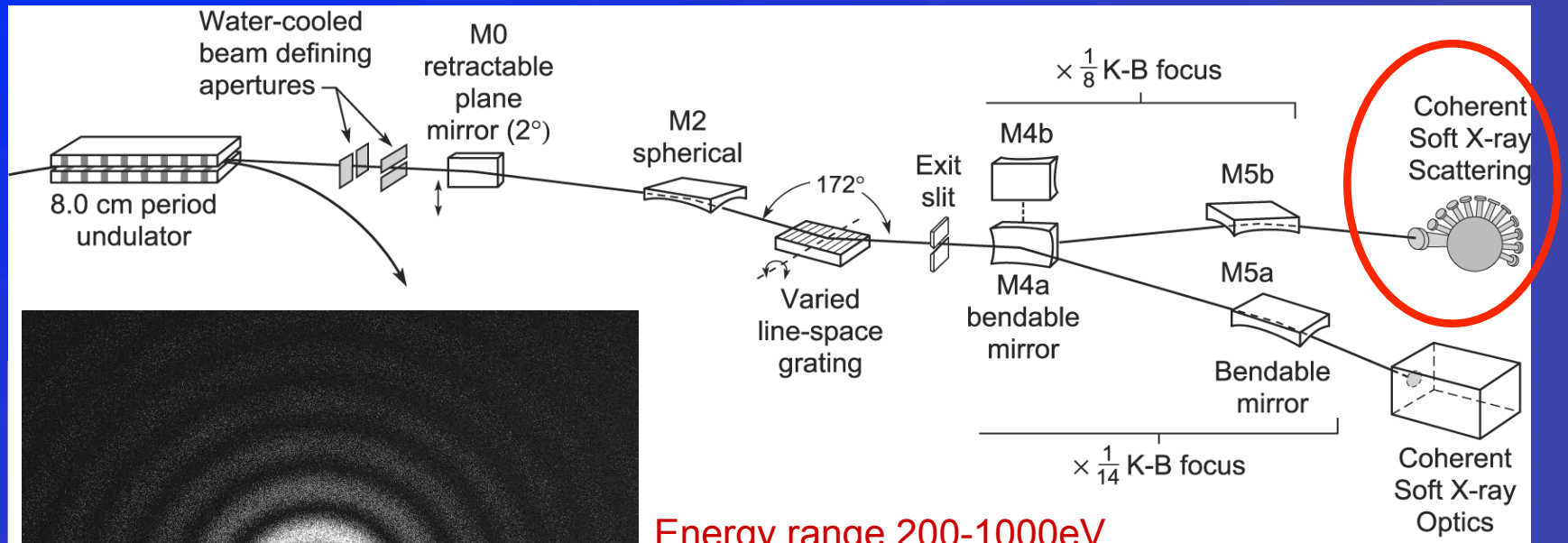
Ultrathin Co layers -> spin-reorientation transition. Preferred magnetization direction is determined by competition between shape and crystalline/surface anisotropy (Pescia *et al. PRL* ***65, 2599).***

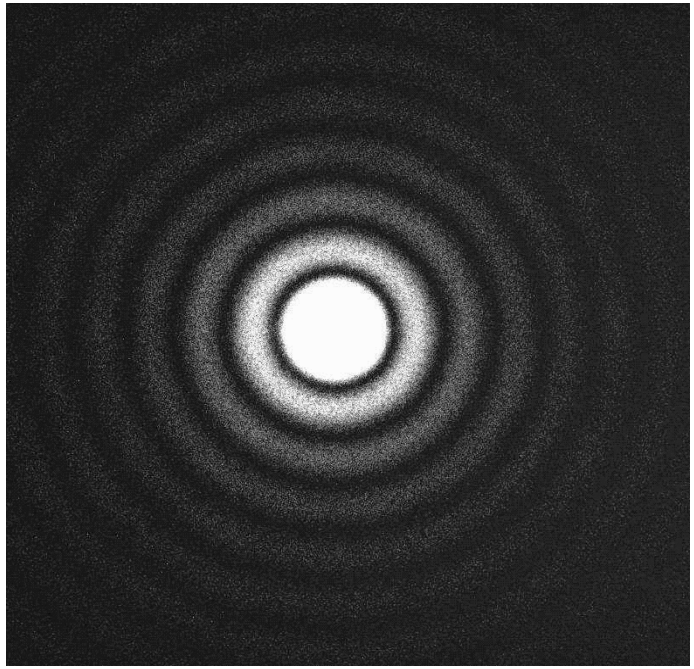
$$E = \left\{K_1(T) - 2\pi M_s(T)^2\right\}\sin^2(\theta) + K_2(T)\sin^4(\theta)$$

Magnetization rotates from out-of-plane to in-plane as a function of increasing temperature (Park *et al. APL 86 042504).*



ALS Coherent Soft X-ray Beamline
(the current generation – stay tuned)





Rosfjord et al. (2004)

Energy range 200-1000eV

Moderate dispersion

8x demagnification of the source

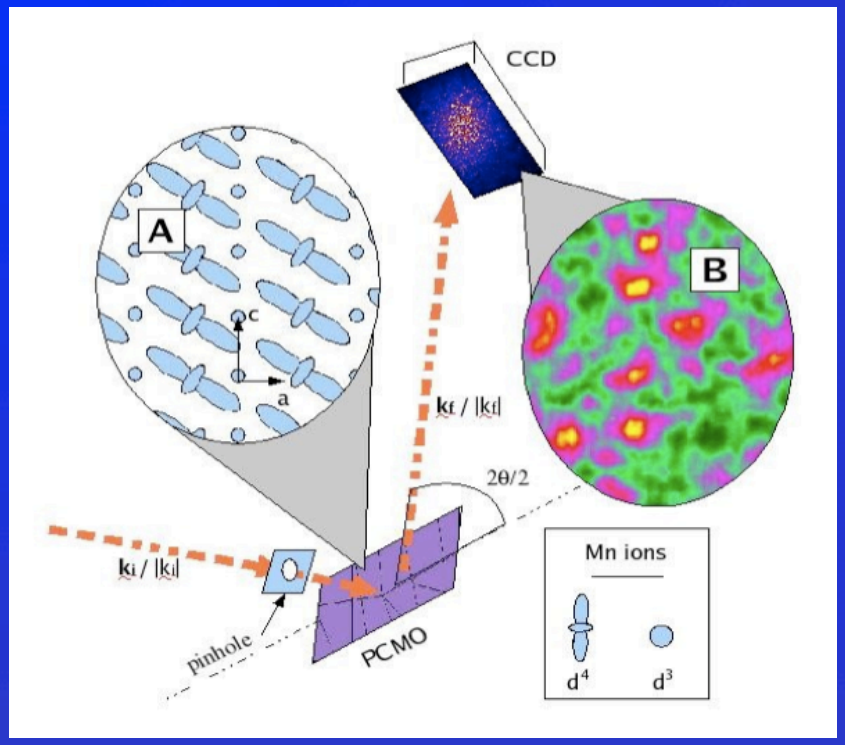
Quality optics to preserve coherence

Coherent flux at 500eV: ~ 5×10^{10} ph/sec/0.1%BW

$\lambda = 2.48$ nm (500 eV)
d = 2.5 µm

Coh_Optics_BLwGraf.4.03.ai

'Imaging' Complexity with Coherent X-rays



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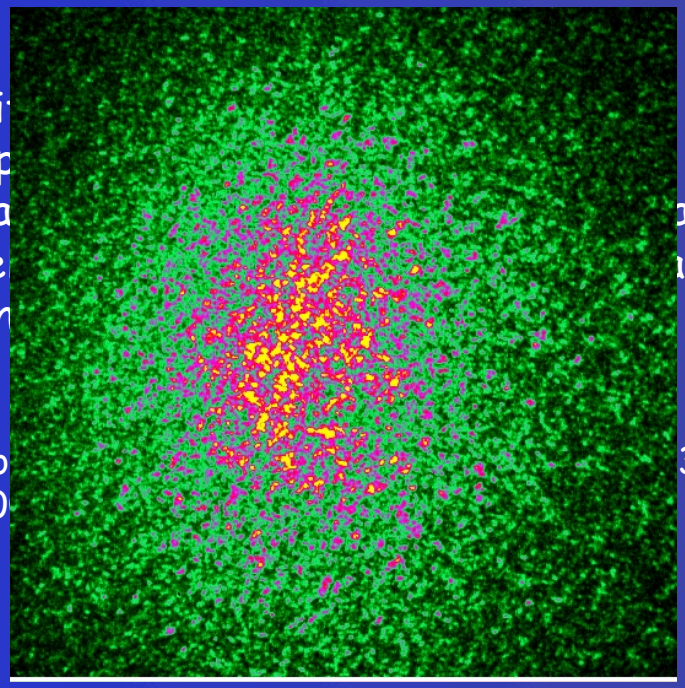
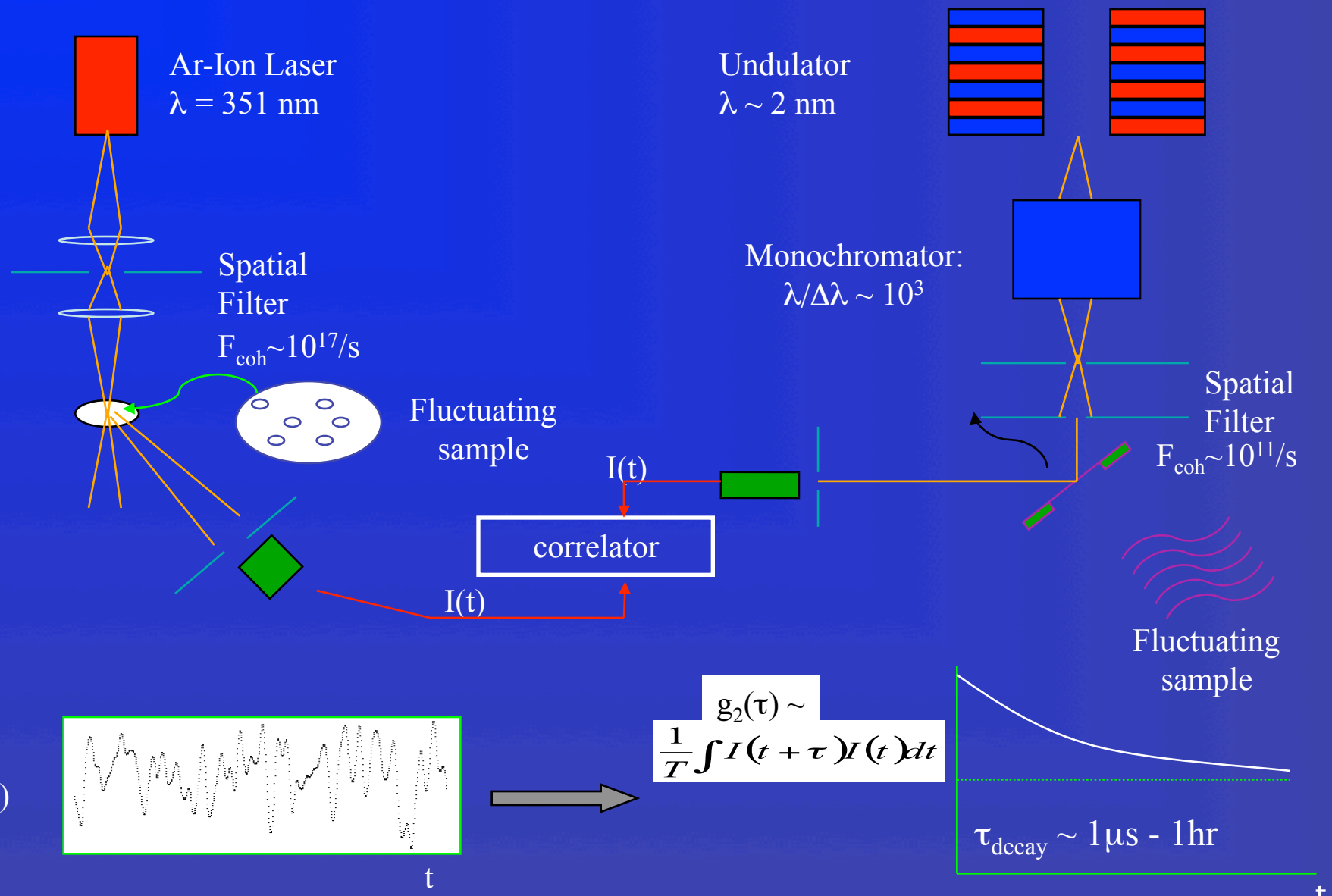


Lowest PCMO Bragg reflection at the Mn L_3 edge: away to image orbital domains?

- Phase retrieval, diffractive imaging, holography
- Speckle metrology, memory effects and external stresses, fields, currents
- Correlation spectroscopy and slow dynamics

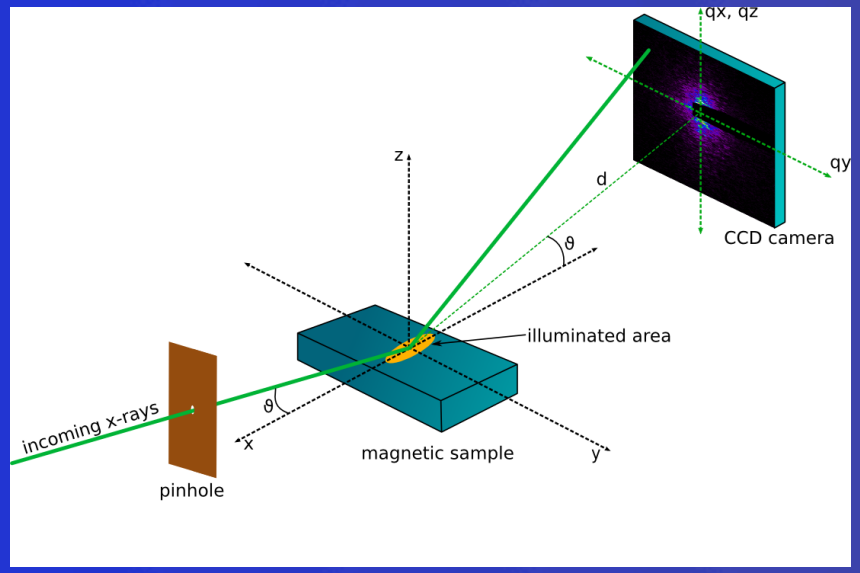
Dynamic Light Scattering

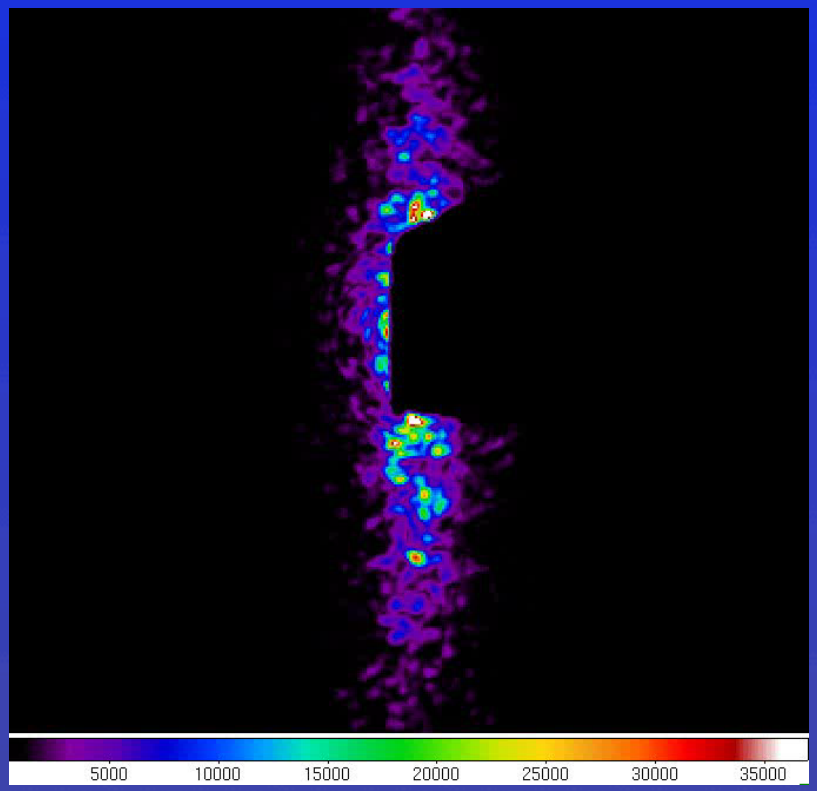


$g_2(t)$ gives the time Fourier transform of the dynamical structure factor, $S(q,\omega)$.

Fluctuations near a Spin Reorientation Transition in Au/Co/Au Heterostructures

- Low angle diffuse reflectivity ($2\theta \sim 18^{o}$)
- Resonant: X-rays tuned to Co L_3 edge to provide elemental and magnetic sensitivity
- Scattered light is measured with a CCD camera: movies with q resolution



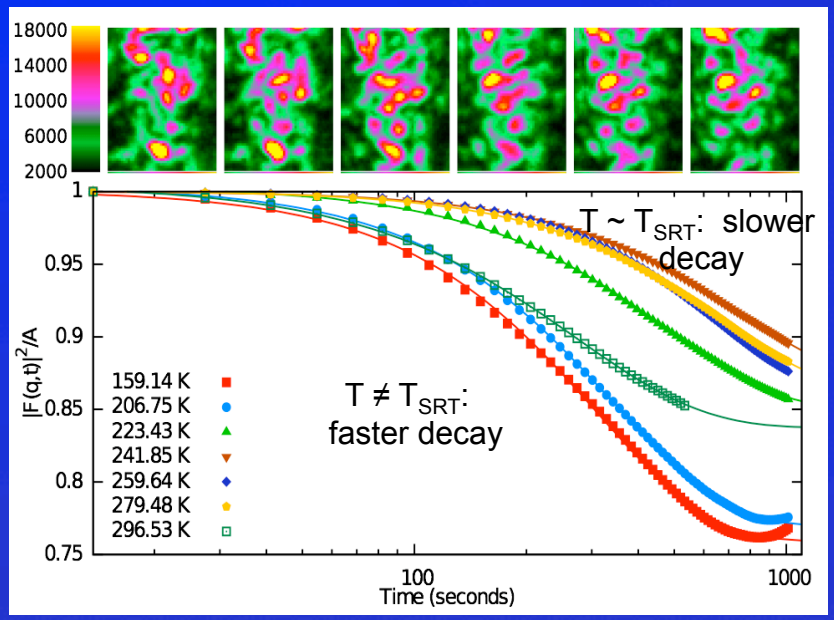


The intensity-intensity time-autocorrelation function directly measures the intermediate scattering function F(**q**,**t**):

$$\frac{\langle I(q,t)I(q,t+\tau)\rangle_t}{\langle I(q,t)\rangle_t^2} = 1 + A\,F(q,\tau)^2$$

Complex Fluctuations near a Spin Reorientation Transition

$q = 2.58 \times 10^{-4}$ Å^{-1}

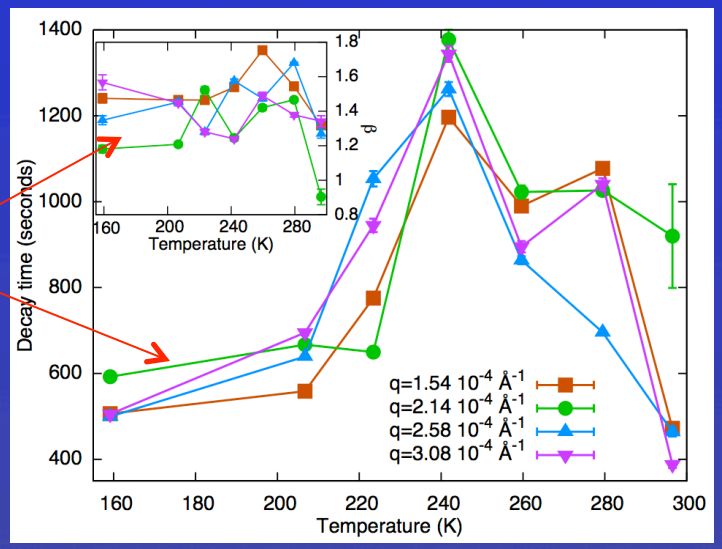


The intermediate structure factor F(q,t) measures the time the SRT system takes to decorrelate due to diffusive motion of the magnetization.

The measurement probes complex magnetization dynamics, possibly with a non-ergodic magnetic component and certainly with a static component due to charge/surface roughness scattering.

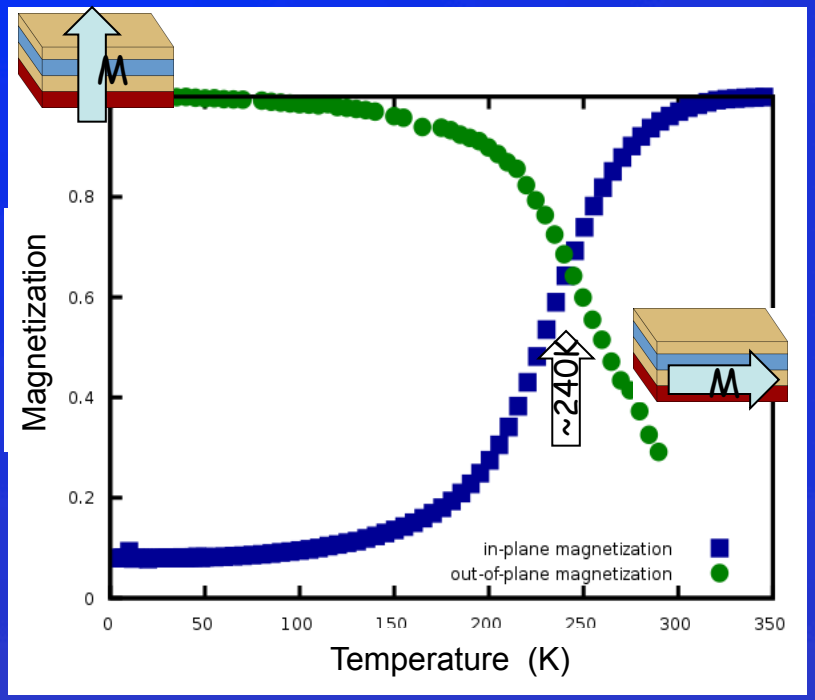
Stretched exponential provides a decent fit. . .

$$F(q,t) \propto e^{-\left(\frac{t}{\tau}\right)^{\beta}}$$



- Increase in decay time at the transition: vestige of critical slowing down?
- Stretching exponent > 1 : collective dynamics; too low for a jamming transition (Nature **447 68**)
- No significant q-dependence? Need larger dynamic range, better pinhole & smaller blocker. . .

Canted Magnetization through the Spin Reorientation Transition

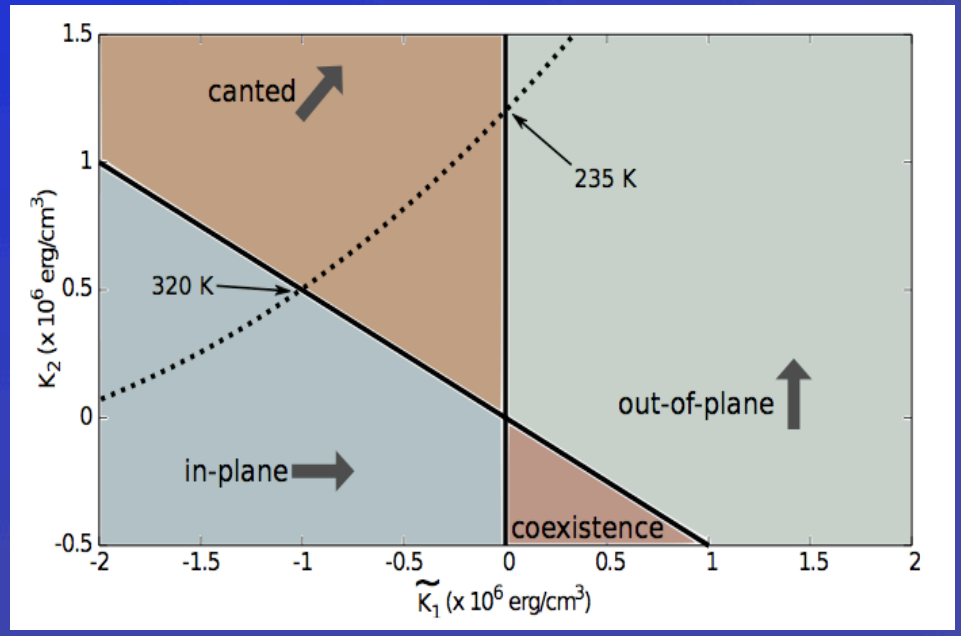


Landau free energy of a uniaxial SRT system:

$$E = \left\{K_1(T) - 2\pi M_s(T)^2\right\}\sin^2(\theta) + K_2(T)\sin^4(\theta)$$

$K_2 < 0$: mixed phase of L and P domains;

$K_2 > 0$: continuous phase with canted magnetization



Why is the transition so broad?

Estimate anisotropies and $M_s(T)$ from

1) measured values at 300K for K_1 and K_2
2) scaling relations to get $K_1(T)$ and $K_2(T)$;
3) T_C tuned to predict T_{SRT}

Predicts $K_2(T) > 0$ through the SRT – and does a decent job explaining the width of the transition.

Nanoscale Intermittancy: Where's the Beef?

- Nanoscale intermittency is often considered to be extrinsic, i.e., driven by environmental factors such as a charged center near an FET gate insulator or changes in the embedding matrix that cause a fluorophore to turn on or off.

- In other cases, the intermittent system *is* the environment, as in dynamical heterogeneity in polymers, TMO's, and magnetic systems.

- IMHO, the pathological sensitivity to the environment of nanoscale systems makes the 'intrinsic' vs. 'extrinsic' argument more or less irrelevant.

- Coherent x-rays allow us to image inhomogeneous structures and to measure statistical properties in space and time with the same technique more or less at the same time; there is a rich diversity of intermittent behaviors that can be usefully probed with these sensitivities.

Collaborators and Funding

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- John Hill

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- Jeff Kortright
- Karine Chesnel

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- Mark Pfeifer
- Josh Turner
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- Dan Parks
- Run Su

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- Charlie Falco

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