



Argonne  
NATIONAL  
LABORATORY

*... for a brighter future*



U.S. Department  
of Energy

UChicago ▶  
Argonne LLC

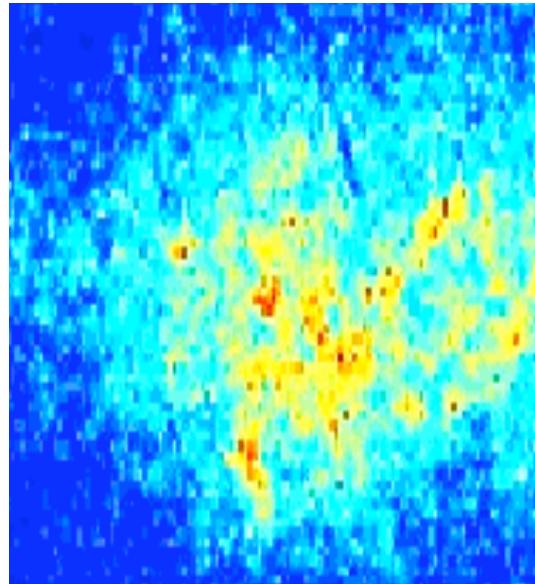


Office of  
Science

U.S. DEPARTMENT OF ENERGY

A U.S. Department of Energy laboratory  
managed by UChicago Argonne, LLC

# Slow dynamics using coherent x-rays



**Eric D. Isaacs**

Center for Nanoscale Materials,  
Argonne National Laboratory  
and The James Franck Institute,  
The University of Chicago

## *Collaborators*



Oleg Shpyrko, Center for Nanoscale Materials, Argonne National Lab  
Jonathan Logan, U. Chicago  
Clarisse Kim, U. Chicago

Prof. Gabriel Aeppli, U. College London

Rafael Jaramillo, U. Chicago  
Dr. Yejun Feng, U. Chicago  
Prof. Thomas Rosenbaum, U. Chicago

Prof. Paul Evans, U. Wisconsin, Madison

Dr. Paul Zschack, Advanced Photon Source, Argonne (33ID & 34ID)  
Dr. Michael Sprung, Advanced Photon Source, Argonne (8ID)  
Dr. Alec R. Sandy, Advanced Photon Source, Argonne (8ID)

**Supported by:**

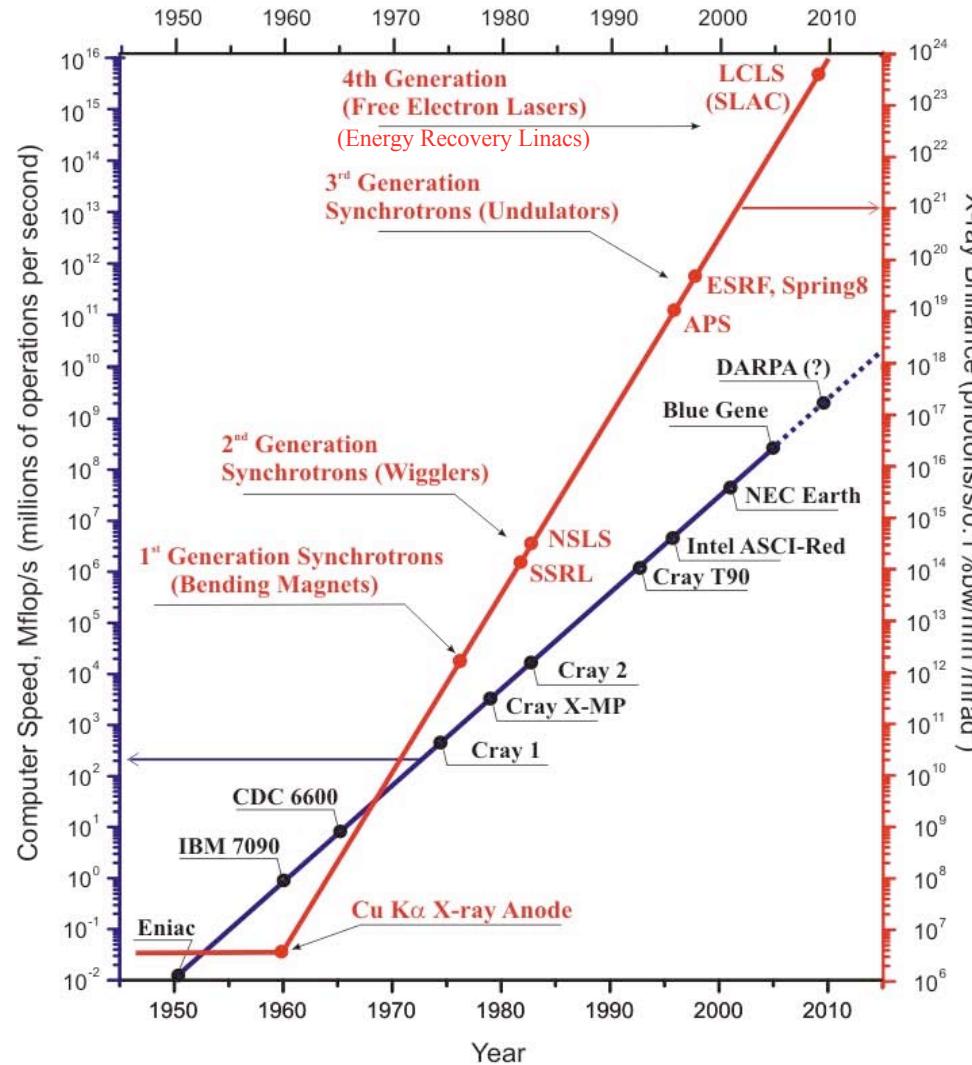


# *Outline*

- Condensed matter physics - texture and multiphase coexistence
- X-ray Photo-correlation spectroscopy - fluctuations of domain walls at the nanoscale
- Wider implications
- What we would really like - spatio-temporal resolution using coherence

# Moore's law for X-ray Sources

12 orders  
of magnitude  
In 6 decades



18 orders  
of magnitude  
In 5 decades!

# *Developments for Nanoscience*

1. Nano-scale focusing - CNM's Hard X-ray Nanoprobe at the APS (Workshop on Thursday)  
- Maser, Stephenson, Streiffer)

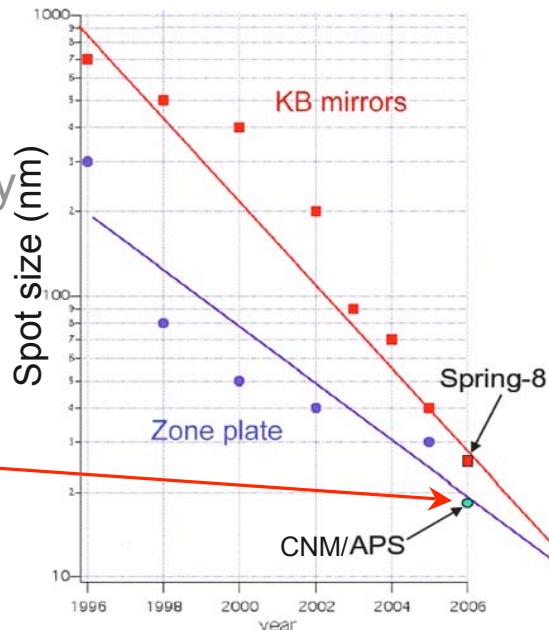


Zone plate



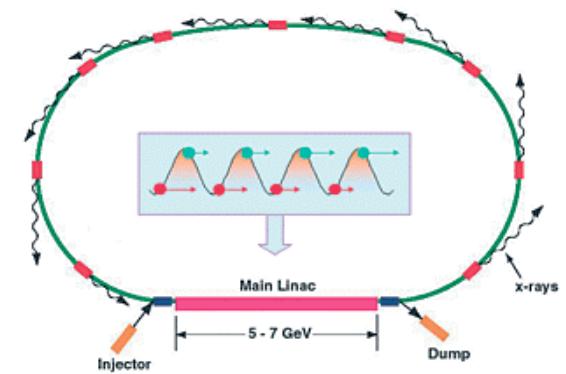
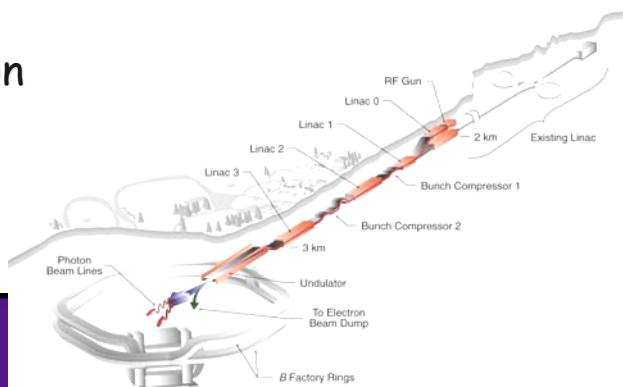
Multi-layer Laue lens

Hard x-ray spot size vs. year



2. Coherence (X-ray “Lasers”)
  - Lens-less imaging
  - X-ray photo correlation spectroscopy

X-ray Free Electron  
Laser at SLAC  
(opening in 2009)

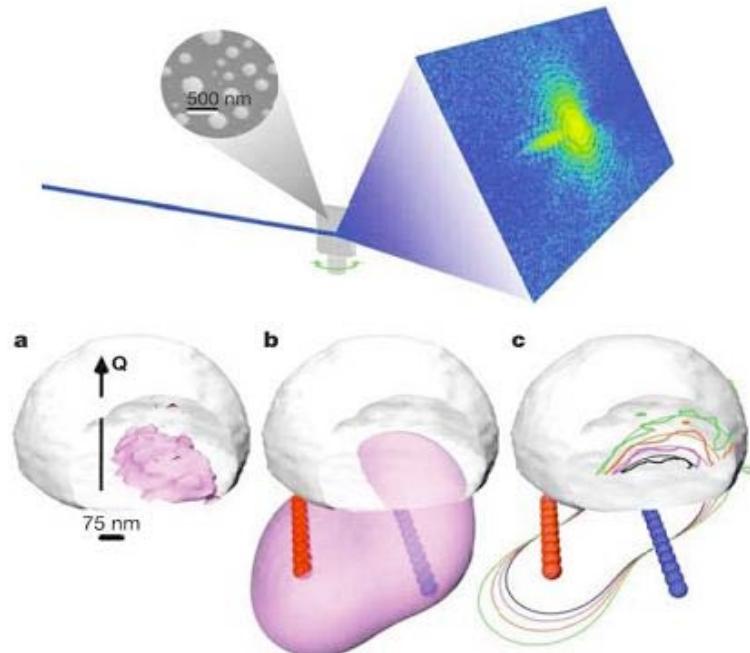


Proposed Energy Recovery Linac at Cornell, Argonne



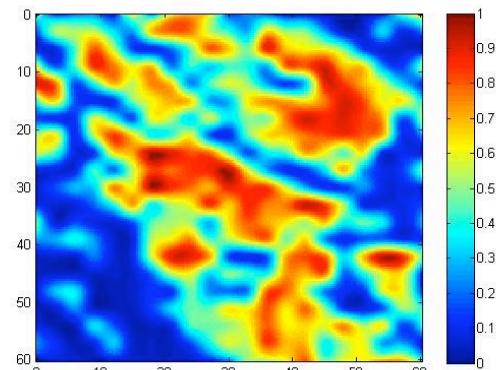
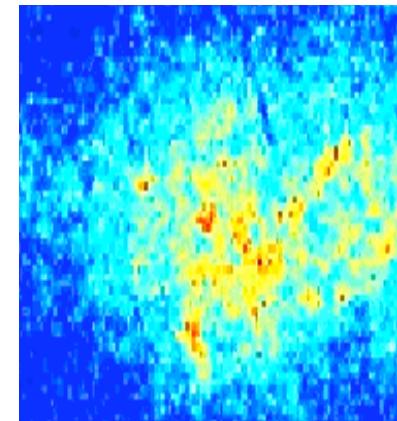
# Coherent X-ray Scattering

Imaging nanoparticles - long-range order no longer required



M. A. Pfeifer et al., *Nature* **442**, 63-66 (2006).  
E.D. Isaacs, *News&Views*, *Nature* **442** (2006).

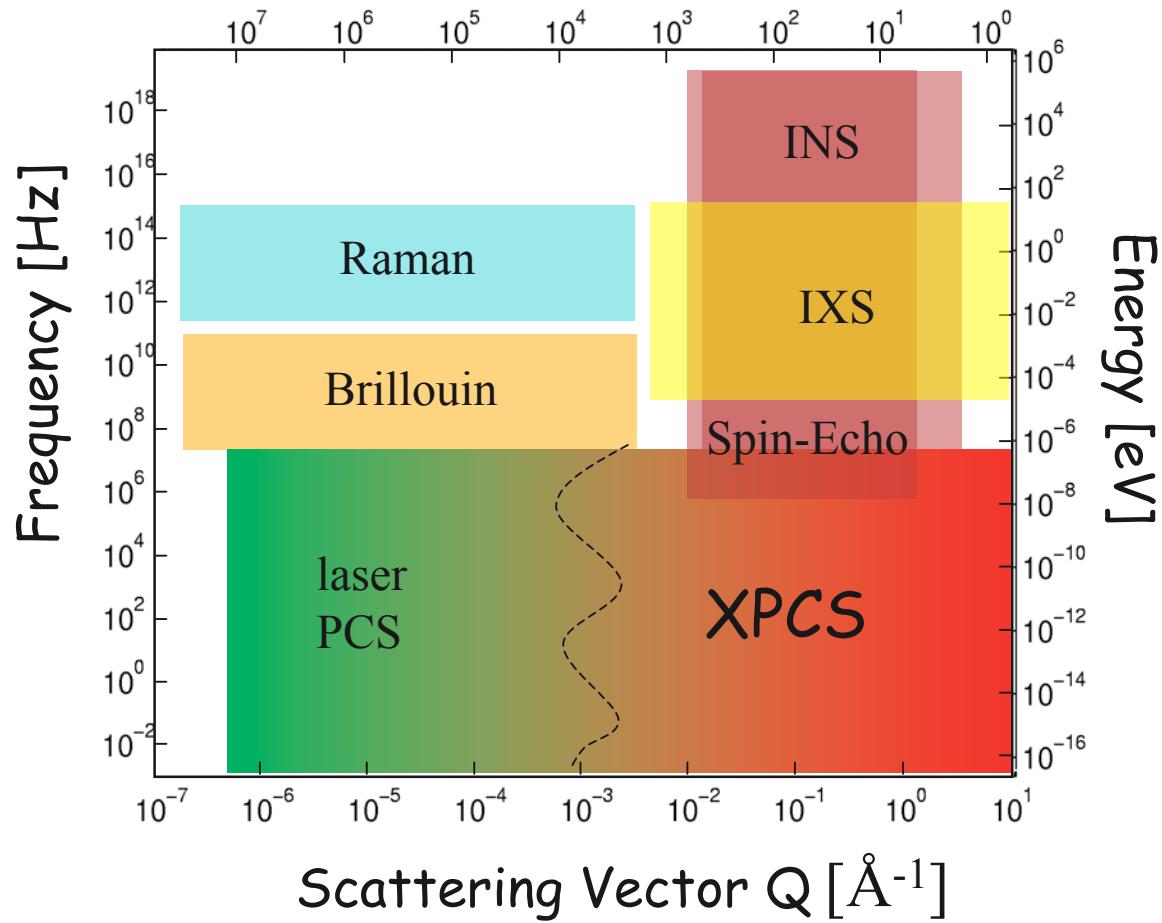
Slow dynamics with high spatial resolution



O. Shpyrko, et al *Nature* **447**, 68-71 (2007)

## XPCS and other spatio-temporal probes

$S(q, \omega)$  map: Length Scale [Å]



### Advantages

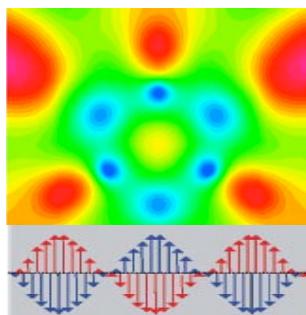
- Shorter lengthscales
- Non-transparent materials
- Charge, Spin, Chemical and atomic structure sensitivity

### Disadvantages

- Need coherent x-ray sources!

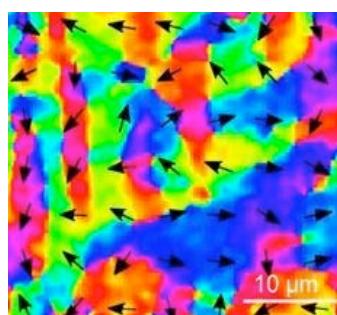
# Collective dynamics in the presence of quenched disorder

Charge-, Spin-density waves  
( $10^{-10}$ - $10^{-7}$  m)



1 nm

Magnetic domains  
( $10^{-8}$ - $10^{-4}$  m)



1 mm

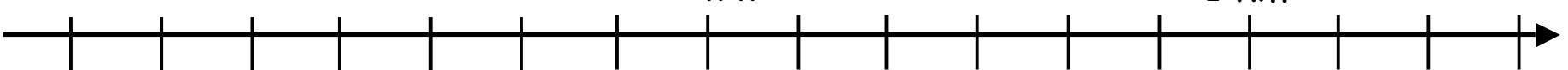
Sandpiles  
( $10^{-3}$ - $10$  m)



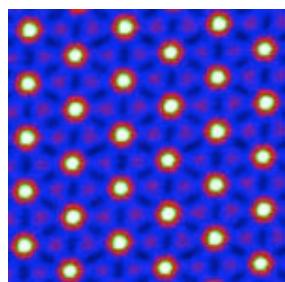
tectonic plates  
( $10^2$ - $10^6$  m)



1 km

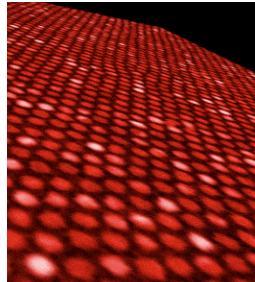


1 Å



Abrikosov vortex lattice  
( $10^{-7}$  m)

1 μm



Jamming, shear flow in granular materials, colloids  
( $10^{-7}$ - $10^{-2}$  m)

1 m



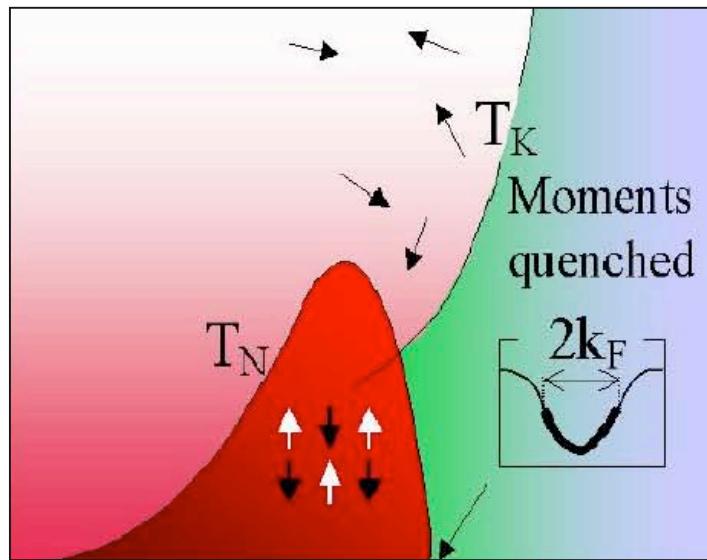
Liquid droplets pinned on rough substrates  
( $10^{-4}$  -  $10^{-2}$  m)

100 km

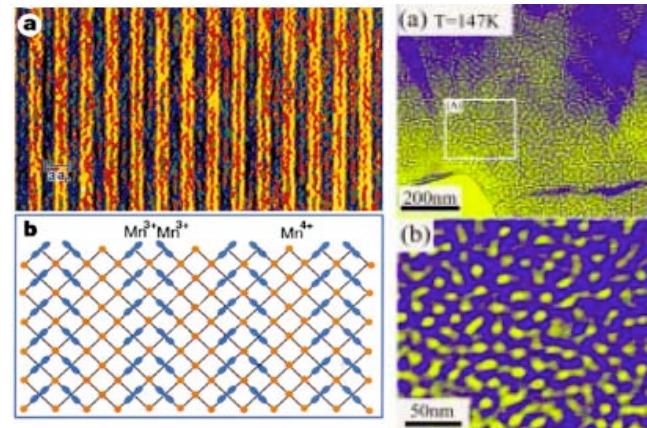


Avalanches  
( $10$ - $10^3$  m)

# Mesoscale Structure in Strongly Interacting Fermi Systems

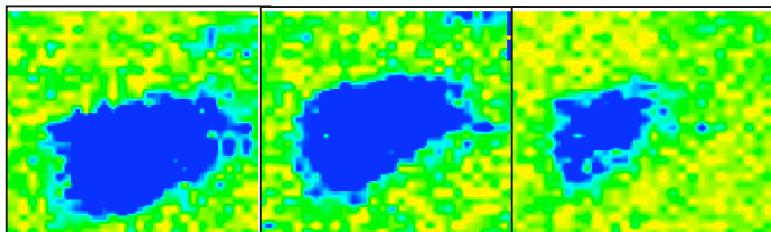


CMR manganites



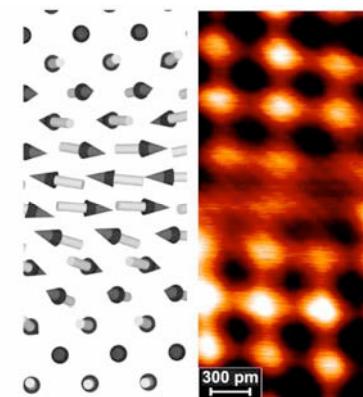
S. Mori et al., *Nature* **392**, 473 (1998)  
M. Uehara et al., *Nature* **399**, 560 (1999)

AF chromium



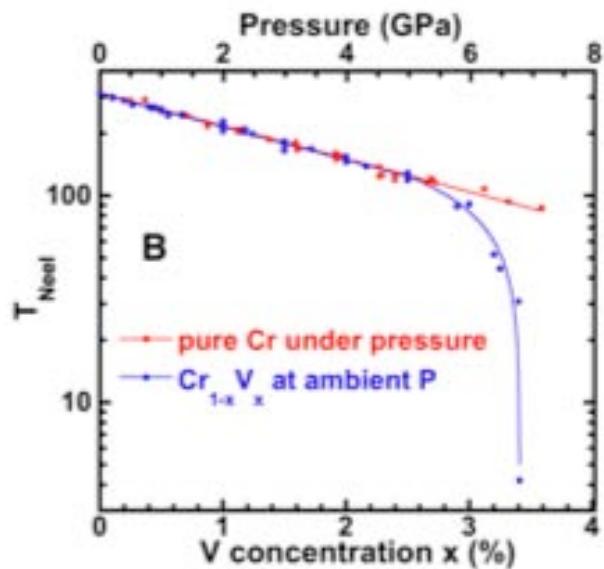
P. G. Evans et al., *Science* (2002)

AF iron

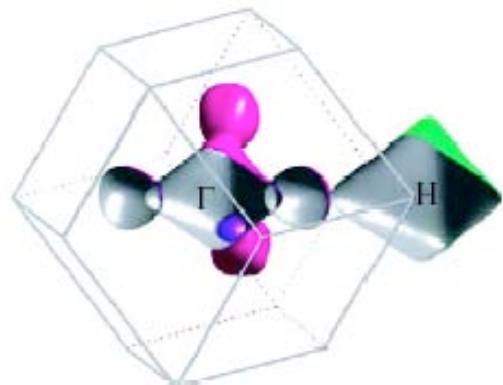


M. Bode, et al., *Nature* (2006)

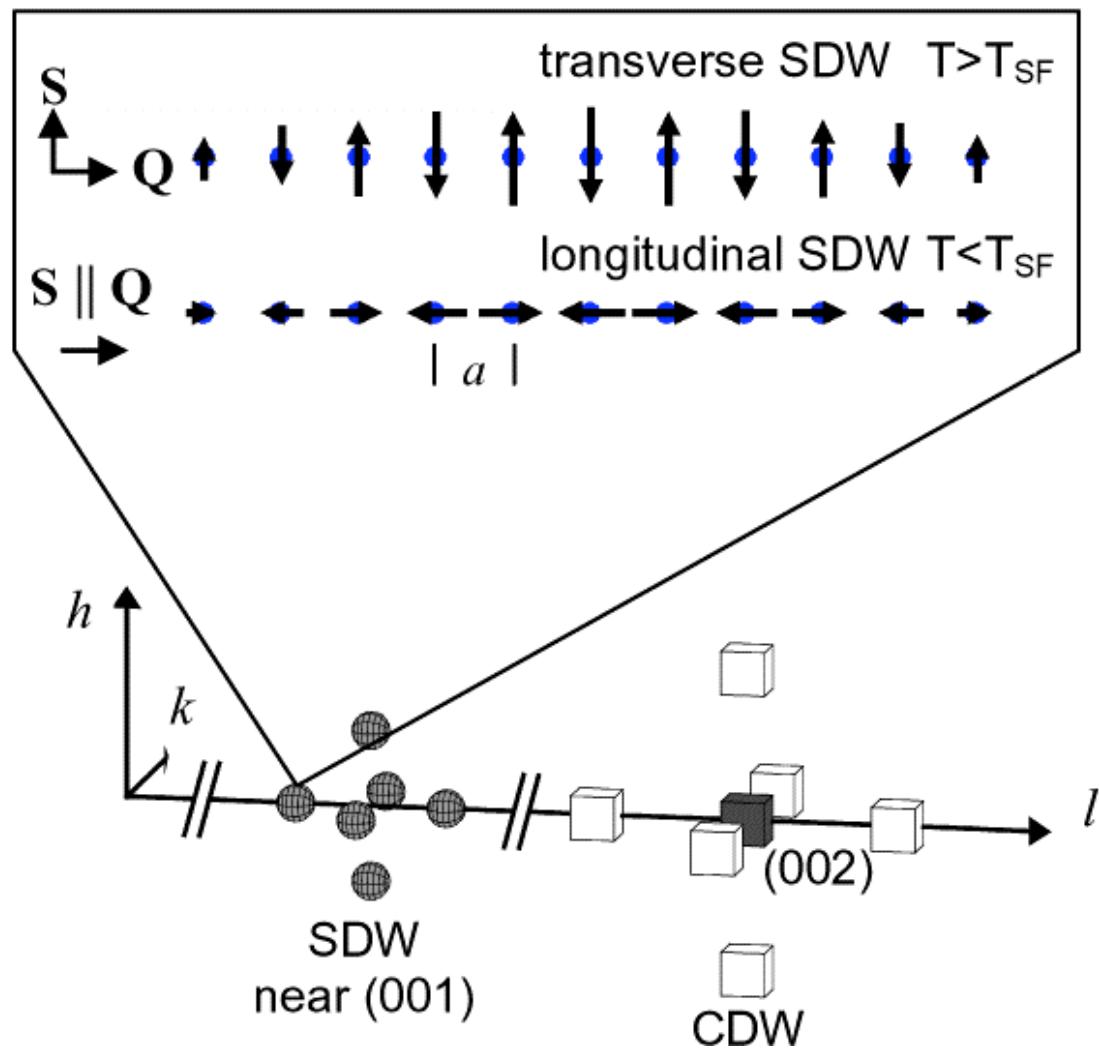
# No need to look at complicated material to find complexity



Chromium and its common alloys are ‘simple’ bcc metals, exhibiting complex behaviors including:



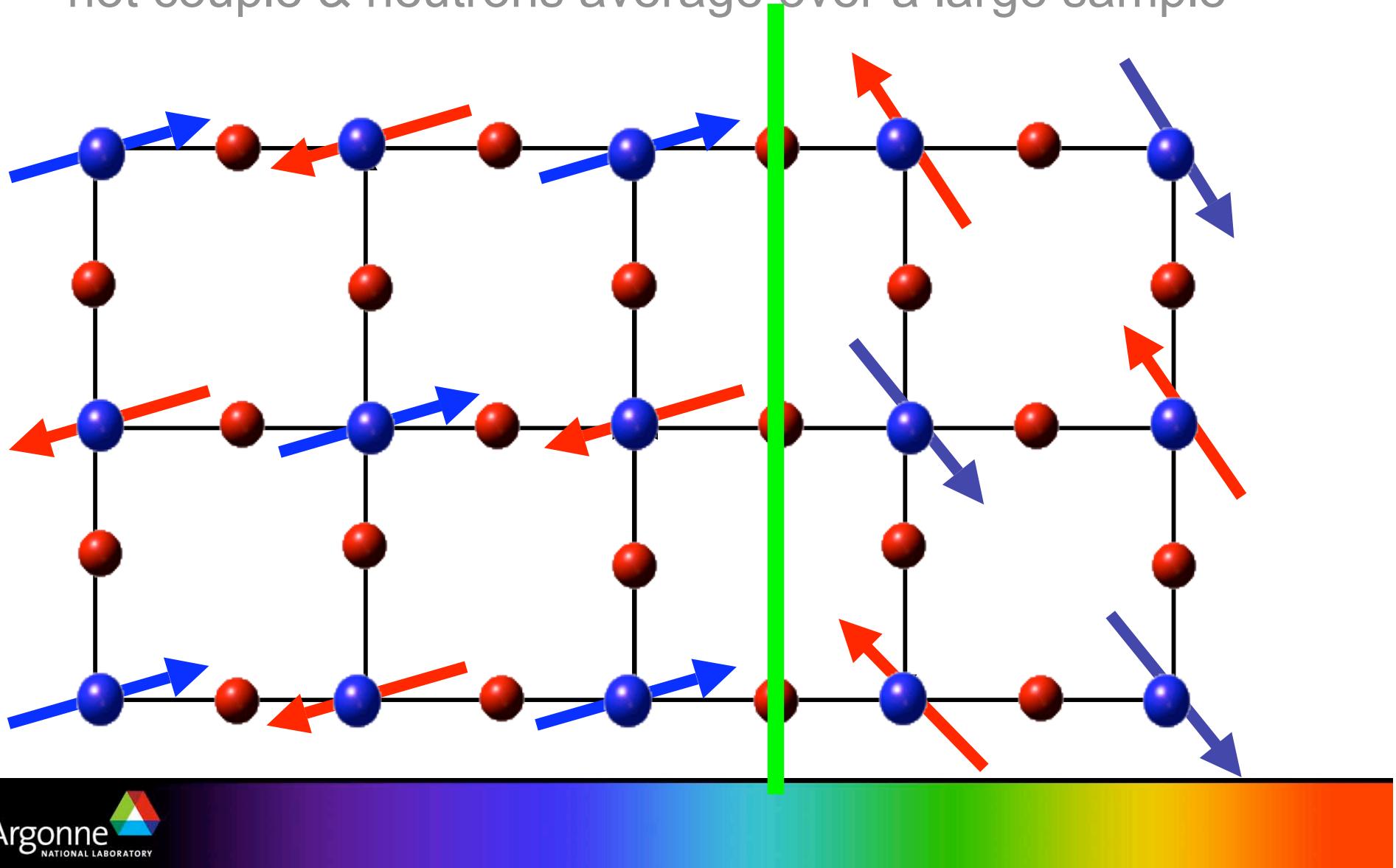
- Only elemental material w/SDW.
- Spin-density wave ground state at 311 K due to Fermi-surface nesting.
- Spin-flip transition at 123 K.
- Quantum critical behavior: drive  $T_N$  to zero by **doping with V** or by **applying pressure**.



# Texture and multiphase coexistence

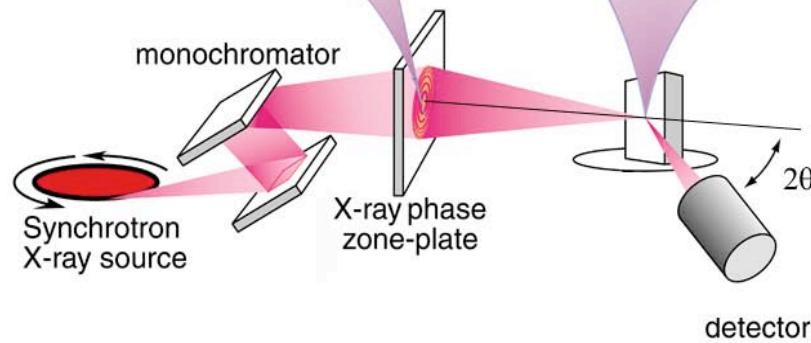
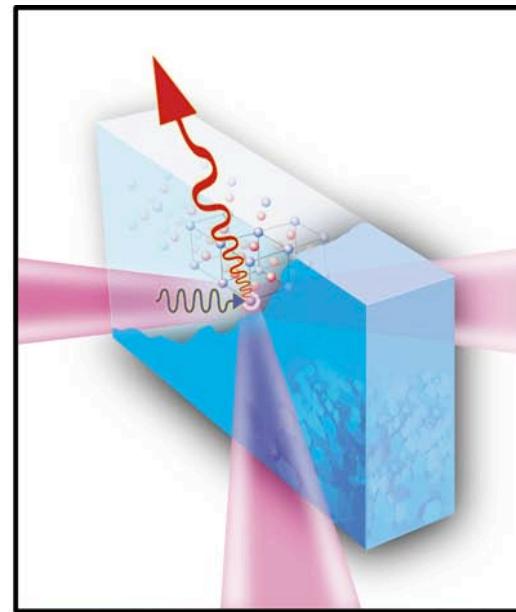
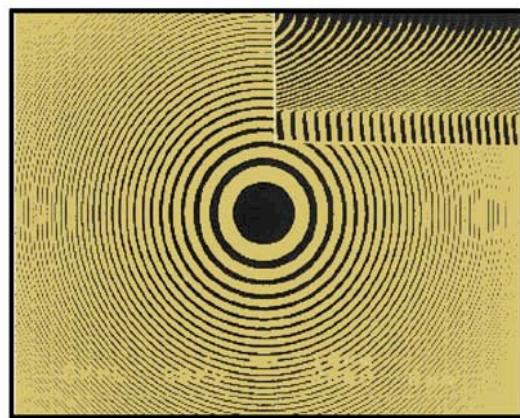


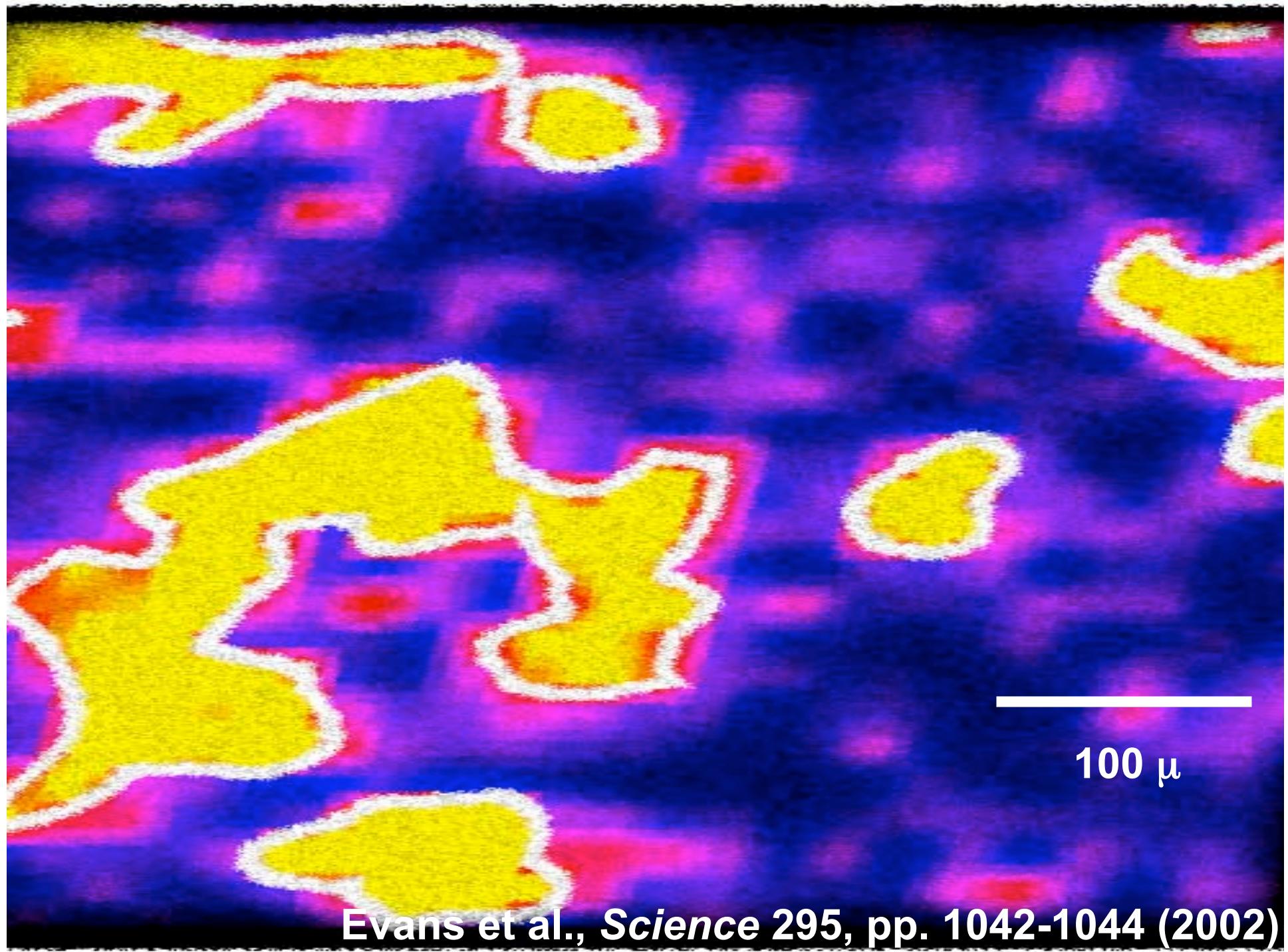
For engineering & science, need to image individual domains & walls between them - but visible light does not couple & neutrons average over a large sample



# *Atomic-scale light - X-rays*

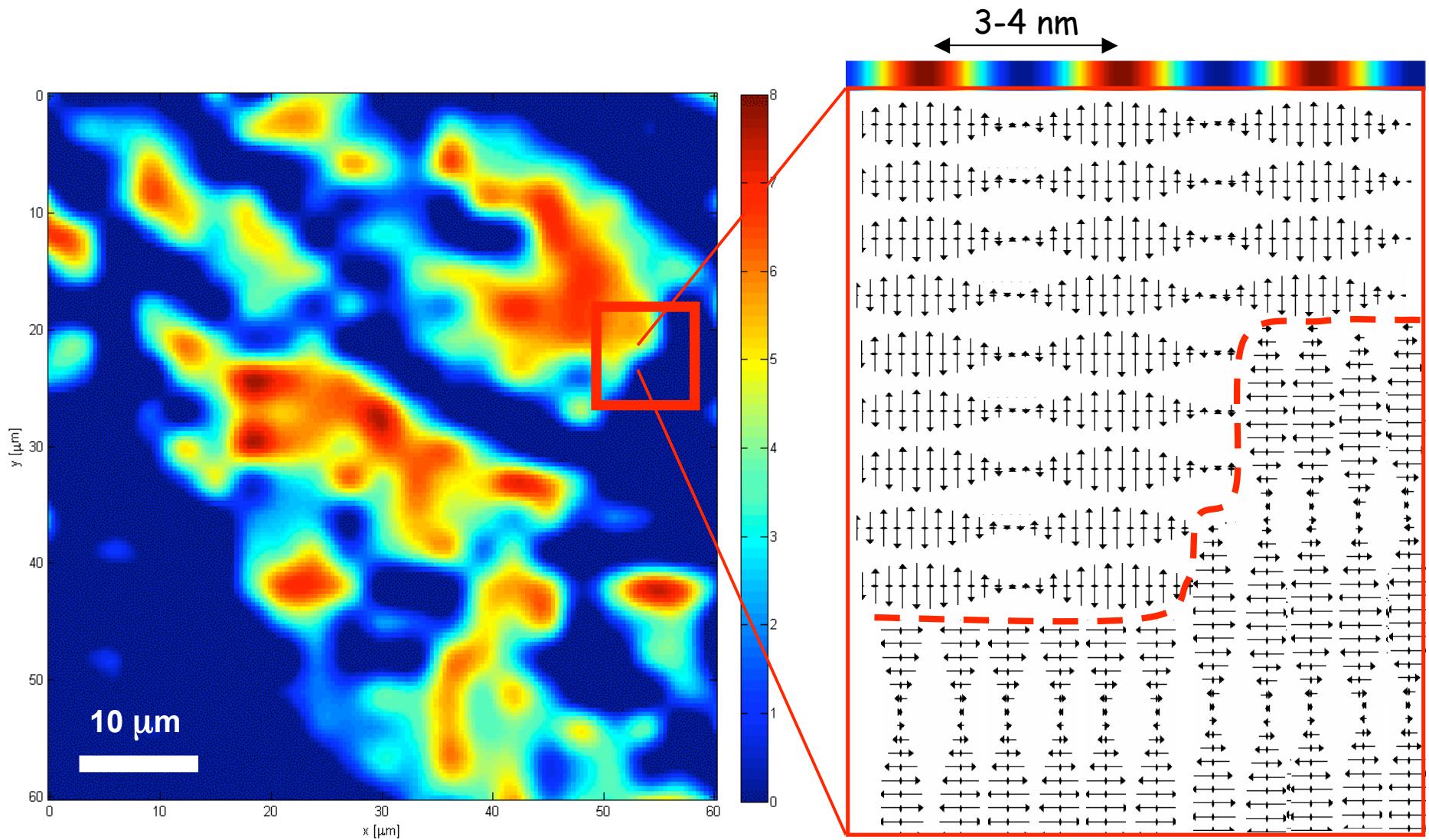
outermost zone  
1992 - 0.5 mm  
now - 20 nm





Evans et al., *Science* 295, pp. 1042-1044 (2002)

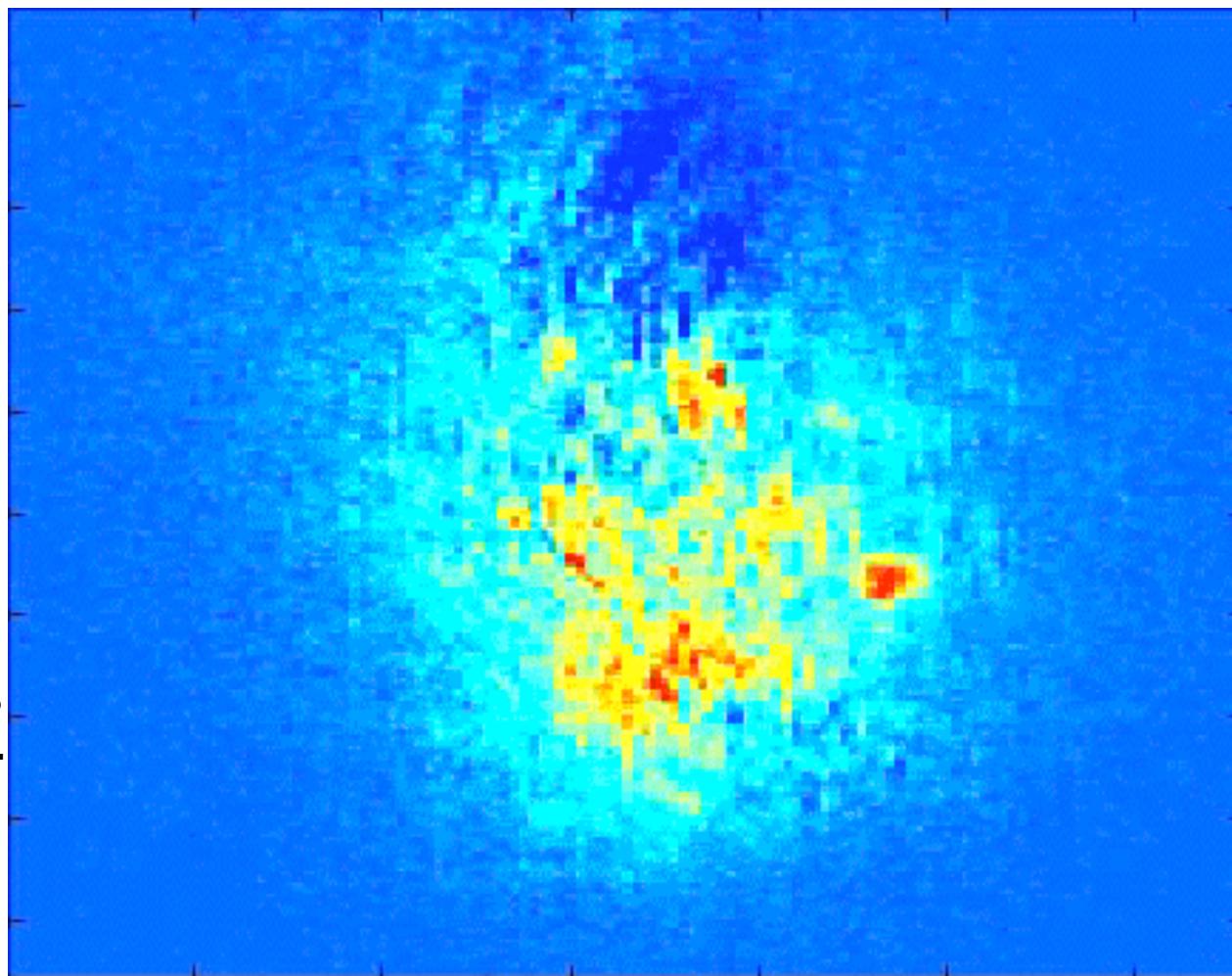
## *Q-domains*



## *Speckle Pattern and Spatial Resolution*

Resolution:  
 $\sim \pi / \Delta Q$   
 $\sim 50\text{nm}$

**Resolution is  
coherent flux-  
limited!**



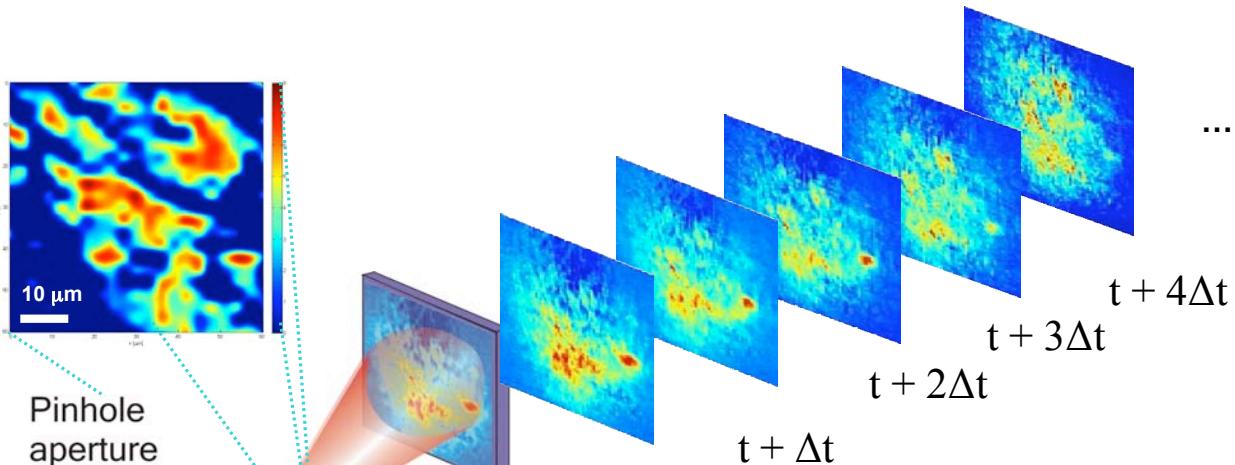
↔

$Q (10^{-2} \text{\AA}^{-1})$

CCD  
image of  
scattered  
x-rays

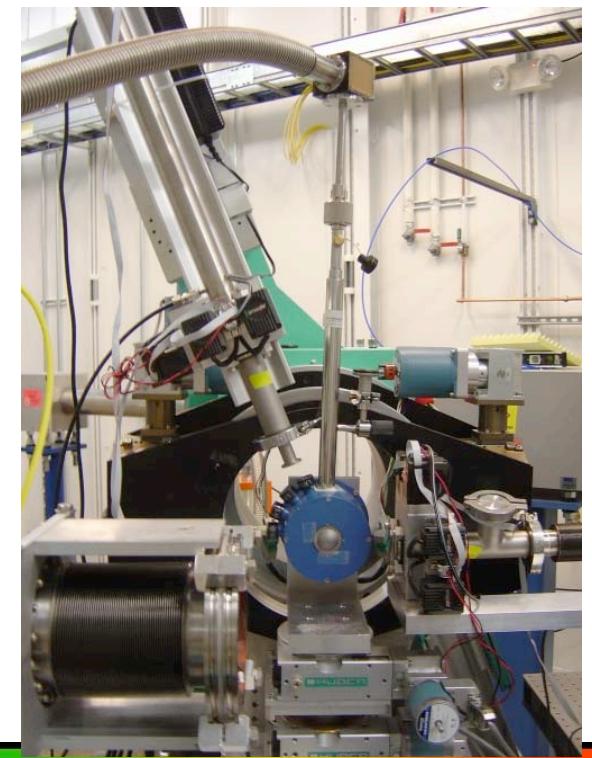
# X-ray Photo Correlation Spectroscopy (XPCS)

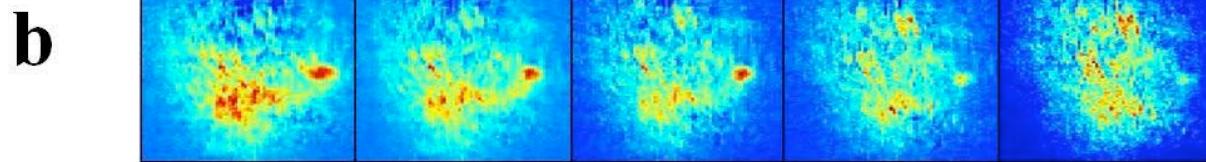
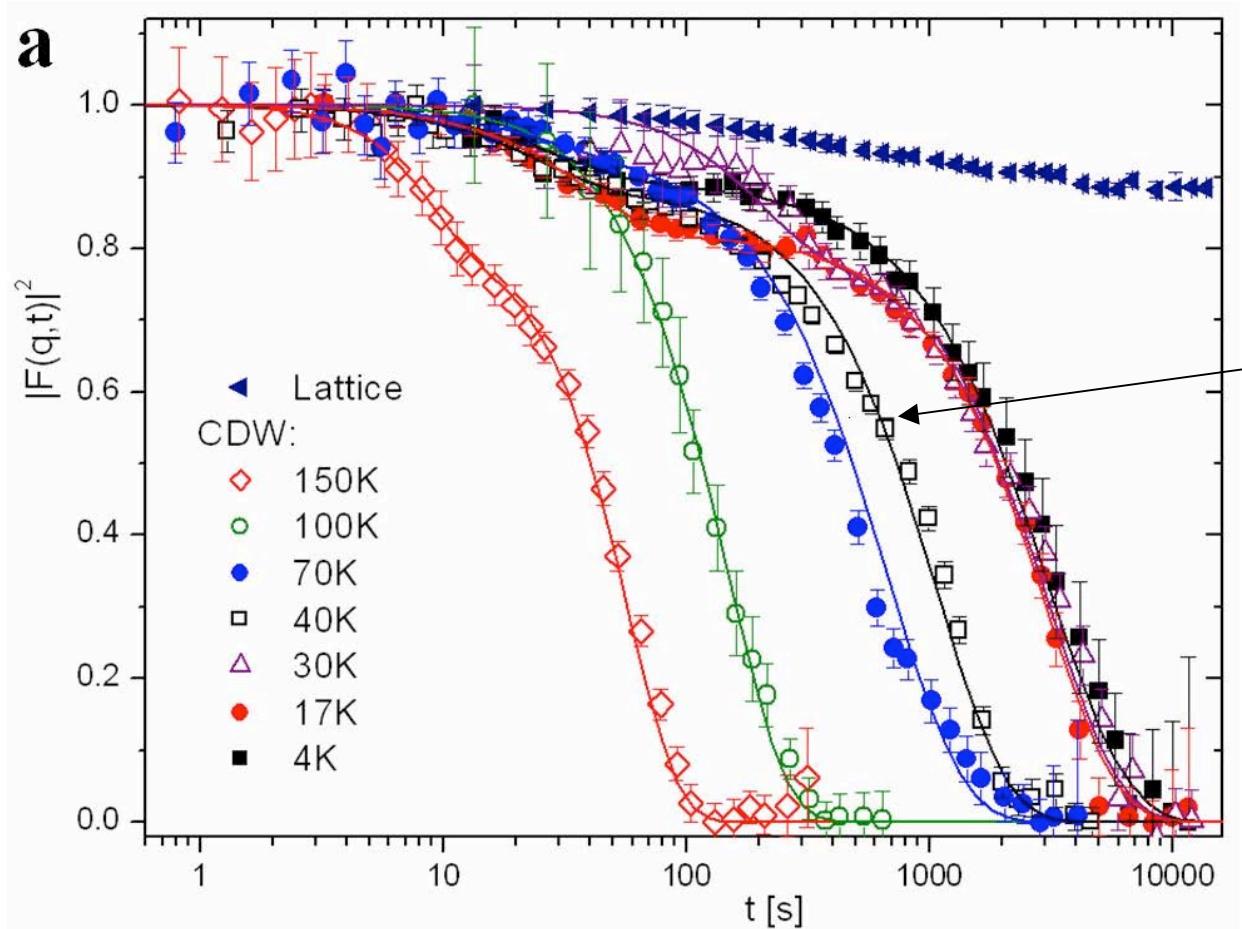
CDW domains  
in real space



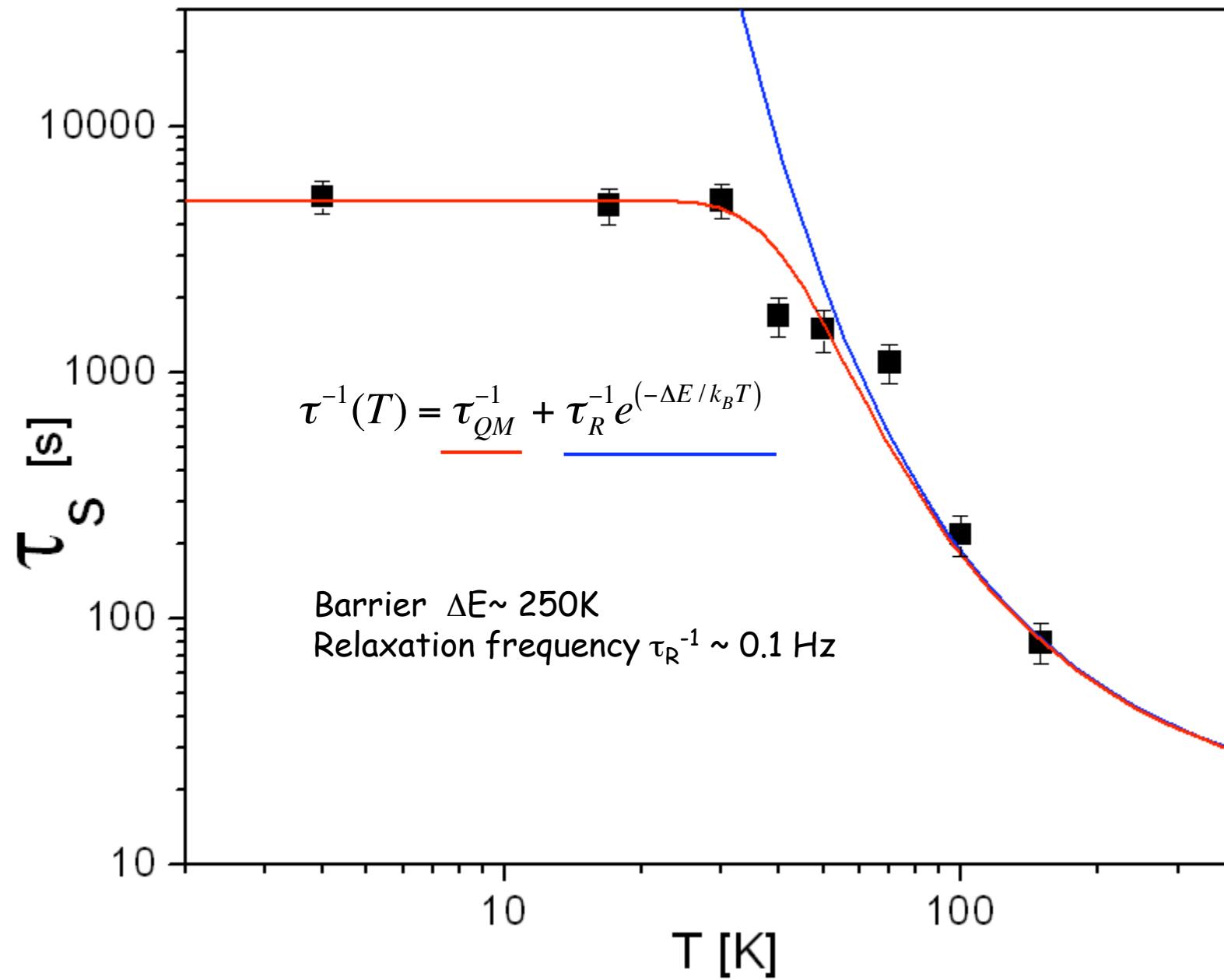
autocorrelation function

$$g_2(\vec{Q}, t) = 1 + A [S(\vec{Q}, t)/S(\vec{Q})]^2 = \frac{\langle I(\vec{Q}, t) I(\vec{Q}, t + \tau) \rangle_\tau}{\langle I(\vec{Q}, \tau) \rangle_\tau^2}$$

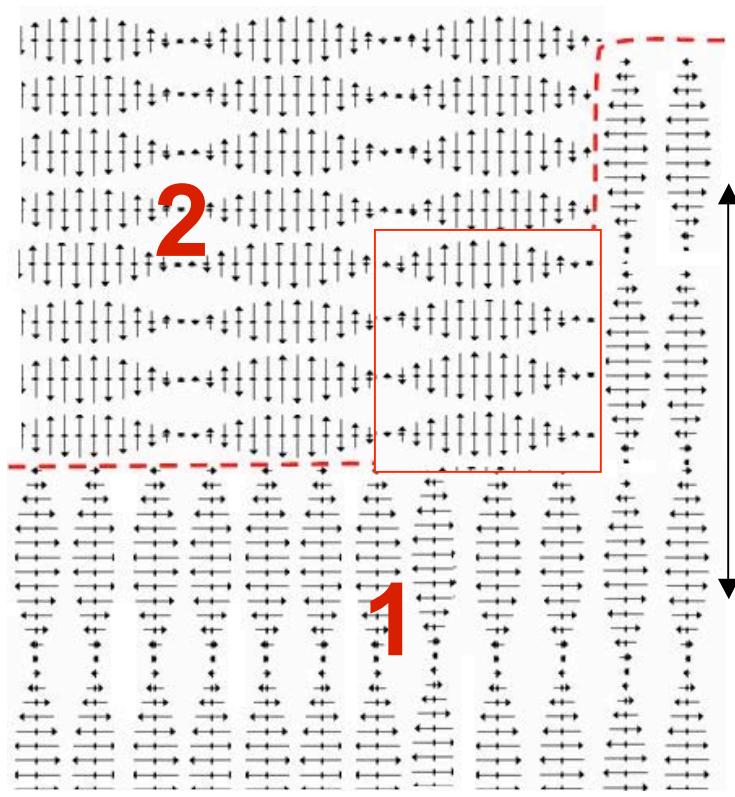




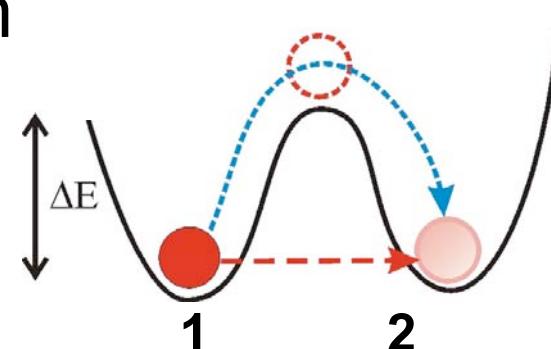
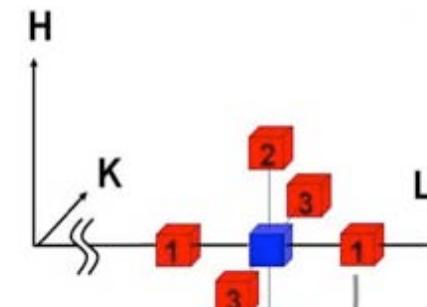
O. Shpyrko et al., Nature, 447, 68 - 71 (2007)



*Where could this come from?*



$$\Lambda \sim 7 \text{ nm}$$



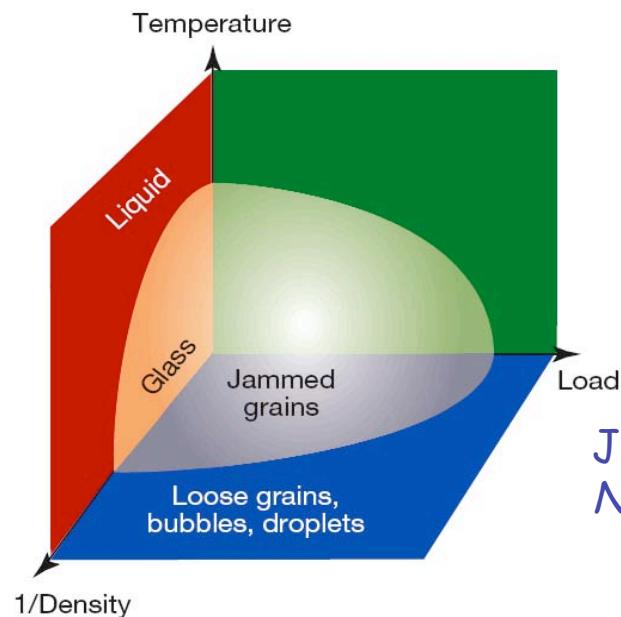
$$\tau^{-1}(T) = \tau_{QM}^{-1} + \tau_R^{-1} e^{(-\Delta E / k_B T)}$$

where  $\Delta E = 25 \text{ meV}$

**Quantum rotor (WKB):**

$\tau_R/\tau_{QT} = \exp-(S/h)$  where  $S=(I \Delta E)^{1/2}$  is the tunneling action

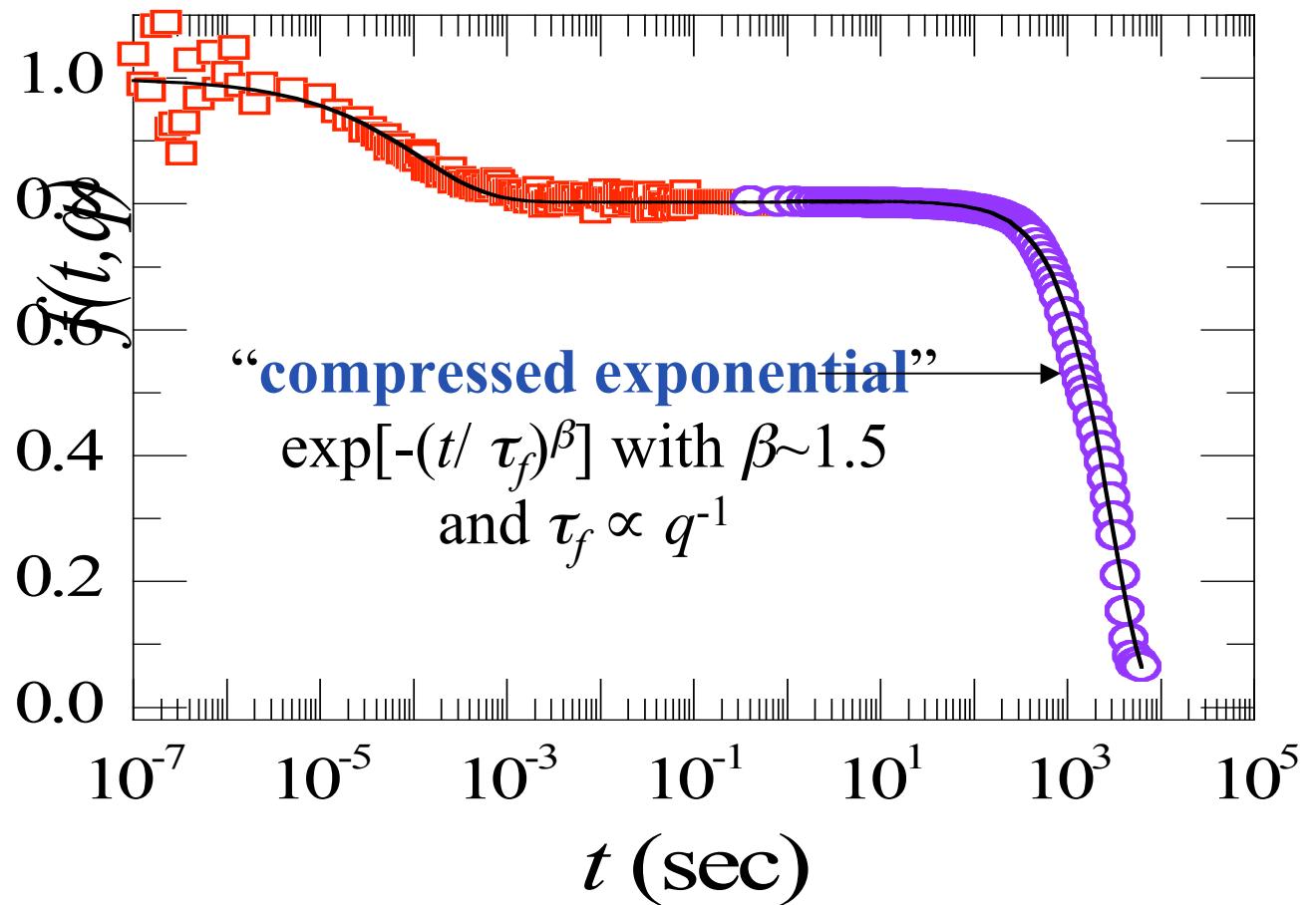
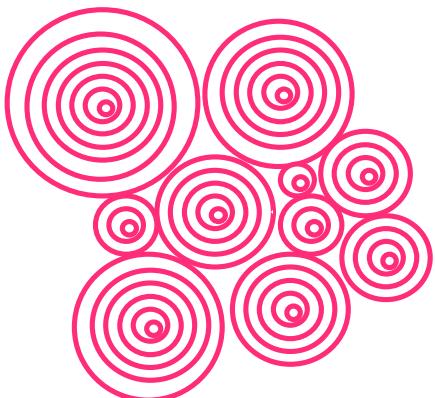
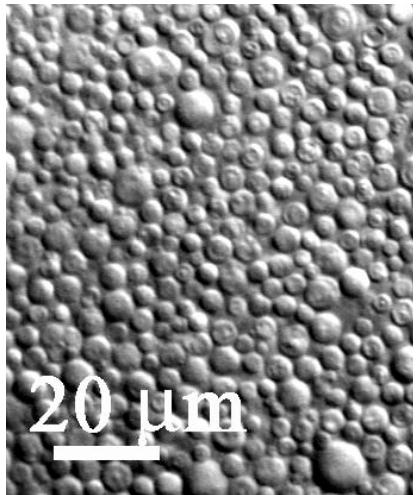
# Wider implications.... dynamics in disordered soft materials



J. Liu and S. Nagel,  
*Nature 396*, 21 (1998)

# *Slow Dynamics in Soft matter*

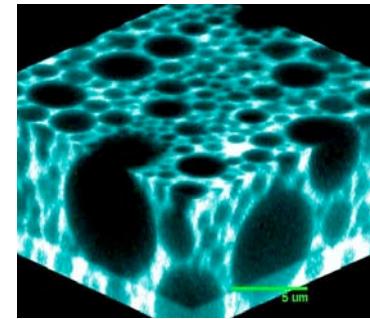
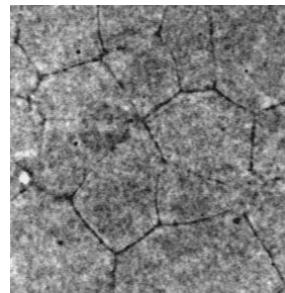
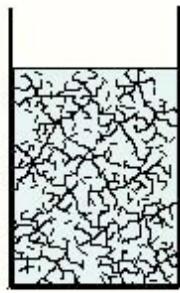
## Onion gel



L. Ramos & L. Cipelletti, PRL 2001

## Other soft systems undergoing ‘jamming’ transitions

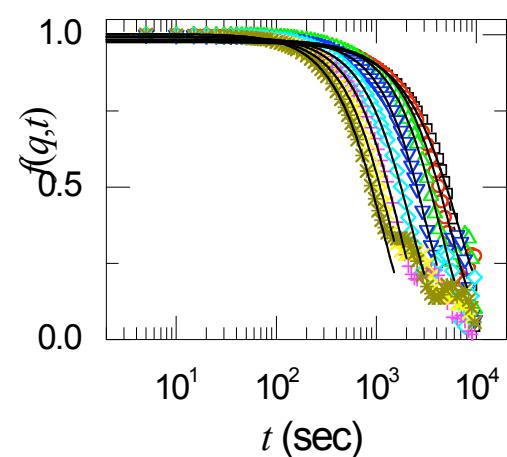
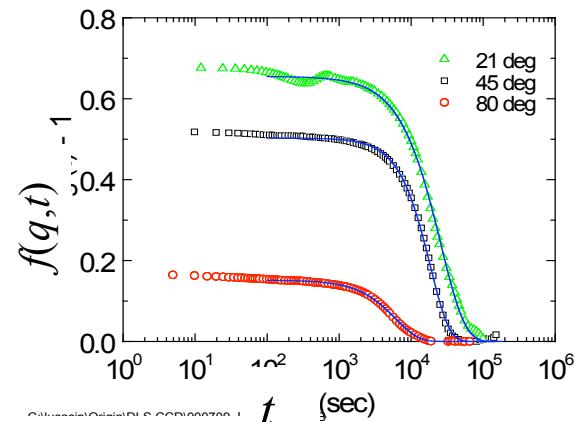
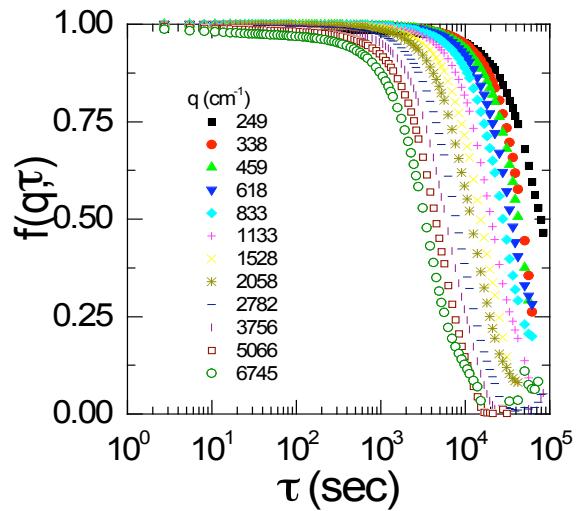
review: Cipelletti *et al.*, Faraday Discuss. **123** (2003)



Colloidal particles gel

Micellar polycrystal

Conc. Emulsion



$$f(q, \tau) \propto \exp[-(t/\tau_f)^{3/2}], \quad \tau_f \propto q^{-1}$$

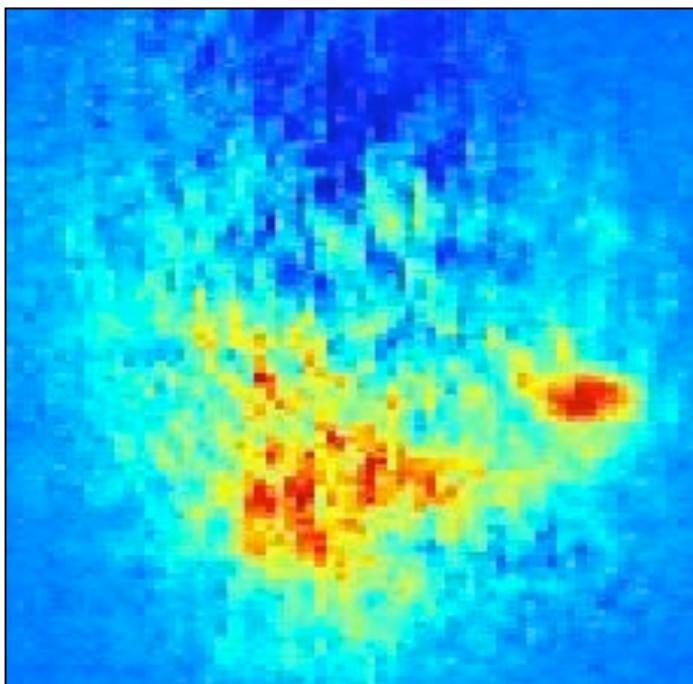
# summary

- X-ray photo correlation spectroscopy (XPCS) allows measurement of dynamics of antiferromagnets at tens of nanometers length scales (finite-q).
- Dynamics strongly reminiscent of ‘glassy’ collective behavior in disordered systems seen universally in soft matter, but, ...
- ... in solid-state system, quantum tunneling provides additional channel for relaxation.
- Beginning of domain wall engineering in antiferromagnets

## *Speckle Pattern and Spatial Resolution*

Spatial resolution:  $\sim \pi / \Delta Q \sim 50 \text{ nm}$

= smallest object that can contribute to speckle



$\Delta Q \sim 10^{-2} \text{ \AA}^{-1}$

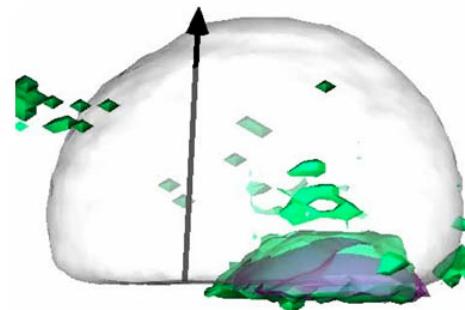
**Resolution is coherent flux-limited:**

$$I_{\text{Bragg}} \sim 1/q^2$$

Current resolution:  $\sim 50 \text{ nm}$

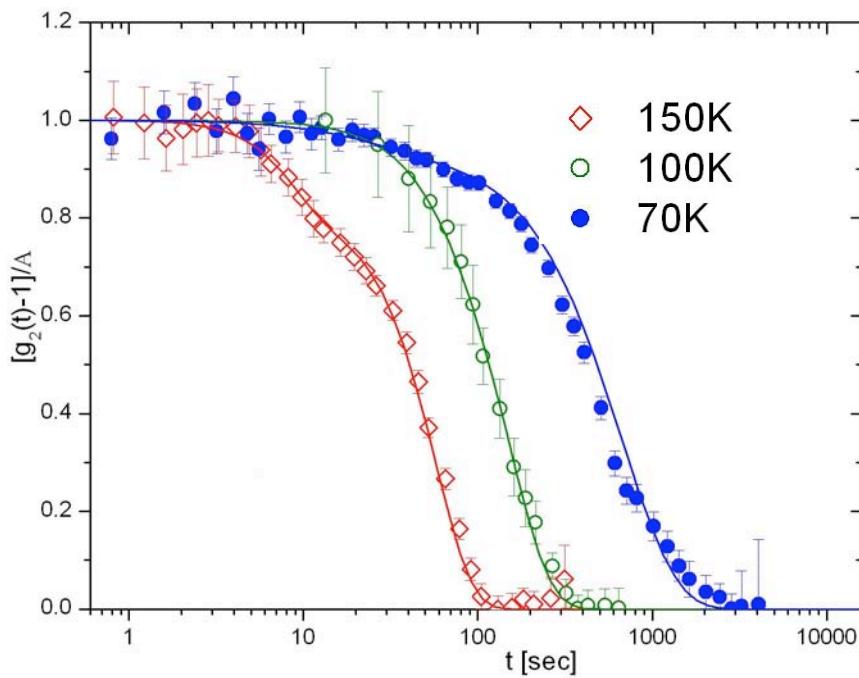
$\Rightarrow$  we need factor of 100 in coherent flux to achieve 5 nm.

Can we image disorder directly?



M. A. Pfeifer et al., *Nature* **442**, 63-66 (2006).

# Autocorrelation Function and Temporal Resolution



**Resolution is coherent flux-limited:**  
 $g_2(t) \sim$  intensity-intensity correlations

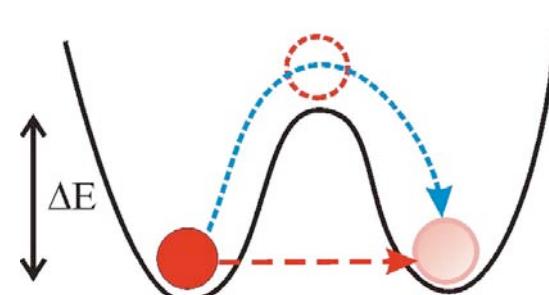
## Statistics:

5% error bars  $\Leftrightarrow$  1 second resolution

$\Rightarrow$  we need factor of 100 in coherent flux to achieve msec resolution.

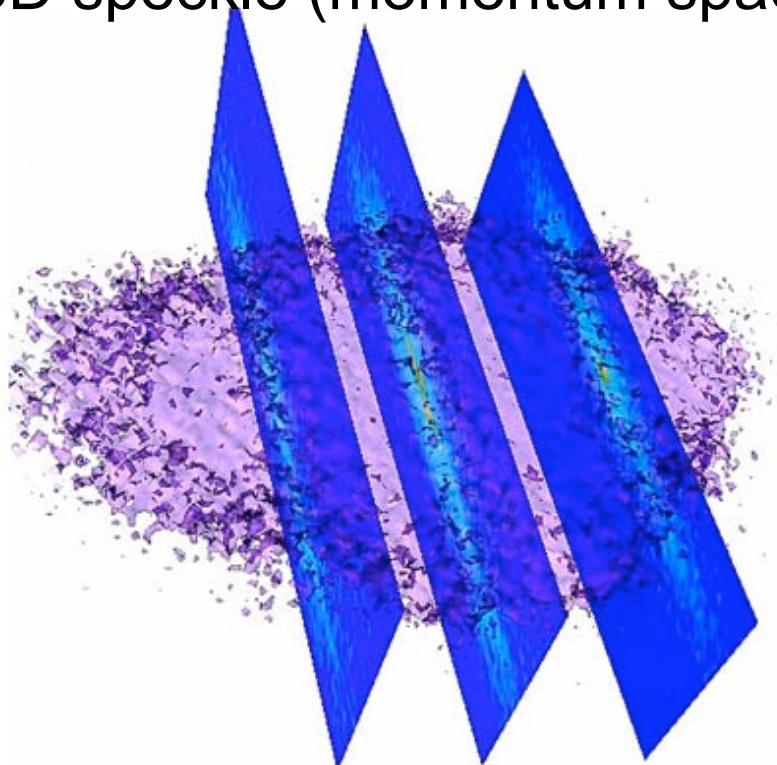
$\Rightarrow$  **spatial + temporal: need 10,000 x**

Can we image quantum tunneling directly?

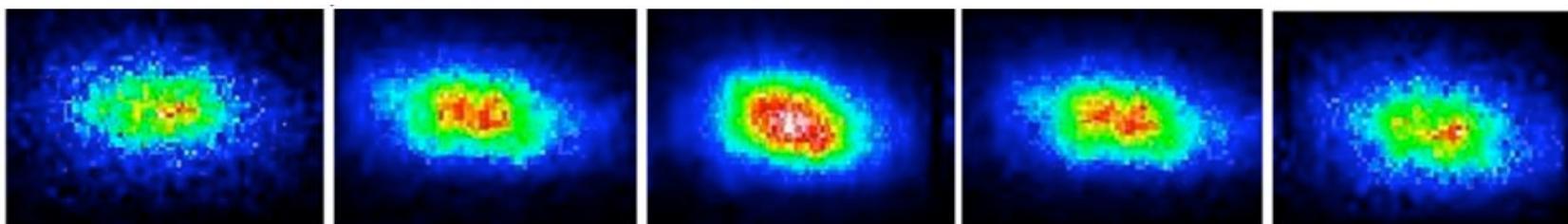


# *Coherent (lens-less) X-ray Imaging*

3D speckle (momentum space): Can we invert speckle of embedded, irregular structures to obtain 3D real-space images? (ptychography)



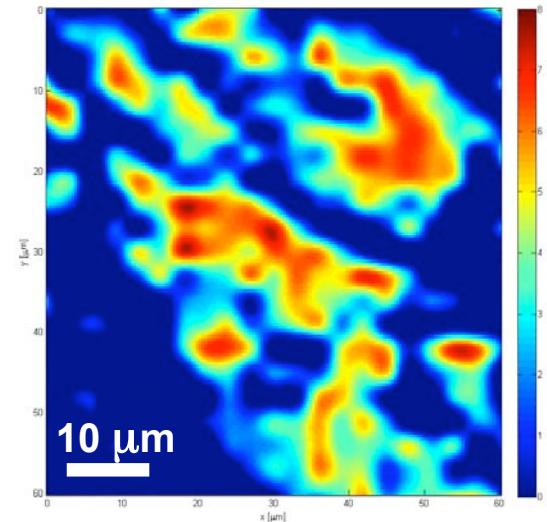
1. Reveal internal CDW phase information (topological defects, phase strain) with  $\sim 50$  nm resolution currently
2. 3D movies of nanoscale fluctuations (FEL, ERL)



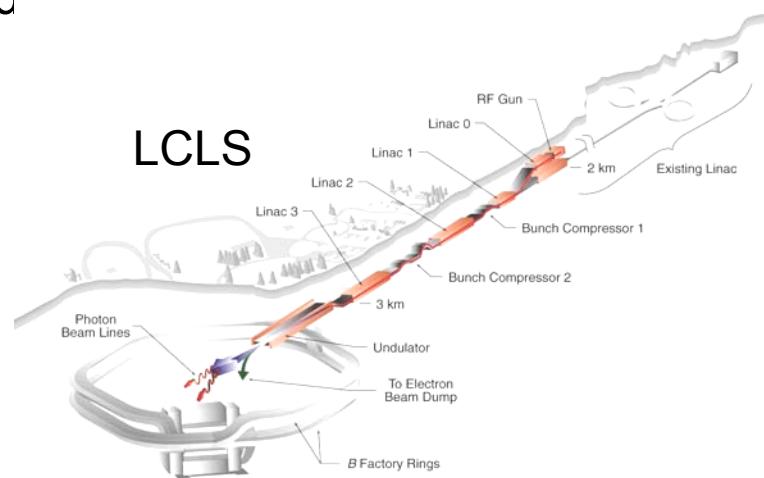
# Conclusions

- Coherence may transform our ability to use x-rays for,
  - Imaging (Nanoprobe, coherent diffraction - lensless imaging of non-periodic structures)
  - Dynamics
- Many interesting problems in strongly correlated Fermi systems, soft materials, phase transitions (eg., jamming)...
  - Many of these are embedded structures
- Dedicated beamline at APS would important start
- Detectors, too!
- Future: FEL, ERL

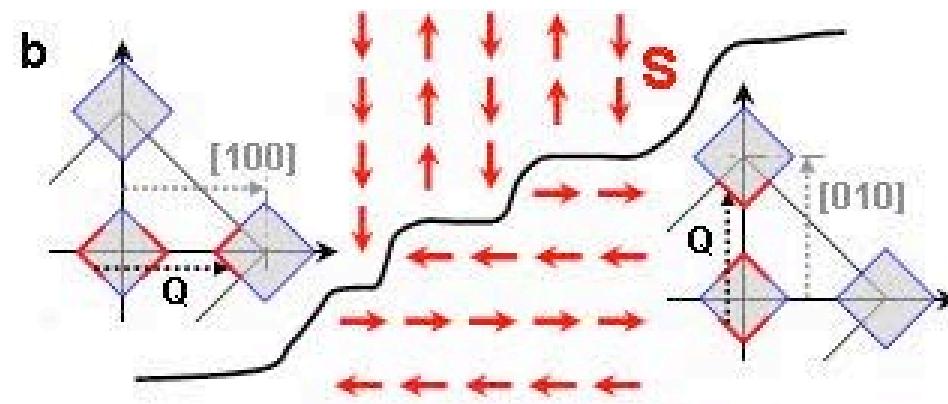
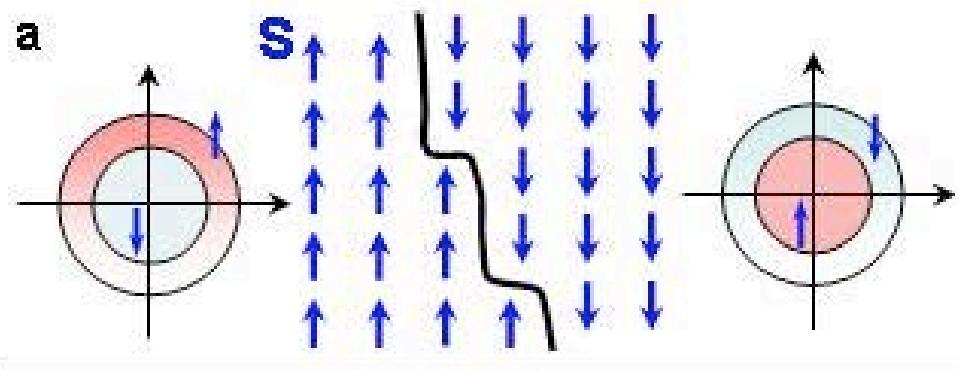
Slow dynamics



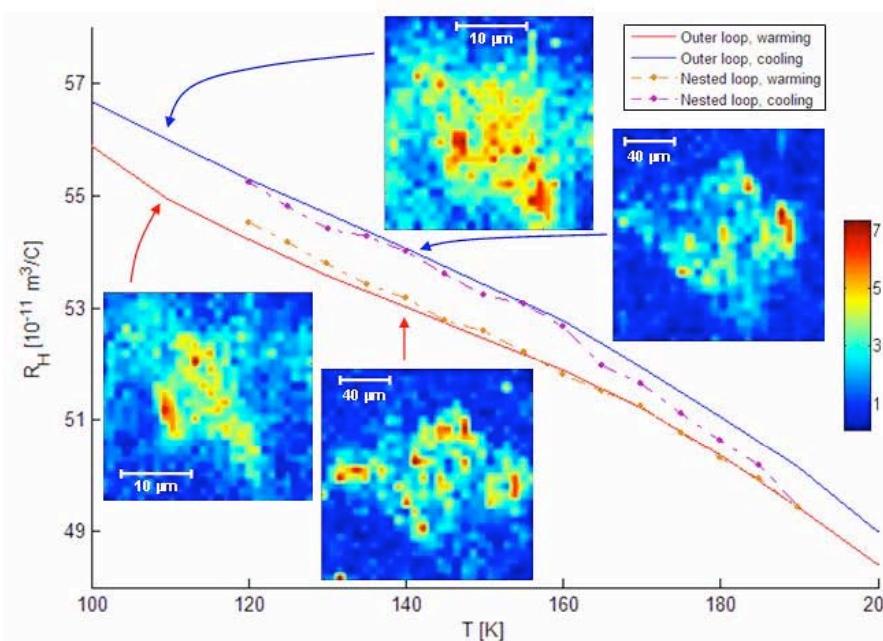
LCLS



Wider implications...  
domain wall resistivity



# Preliminary estimate of domain wall resistivity

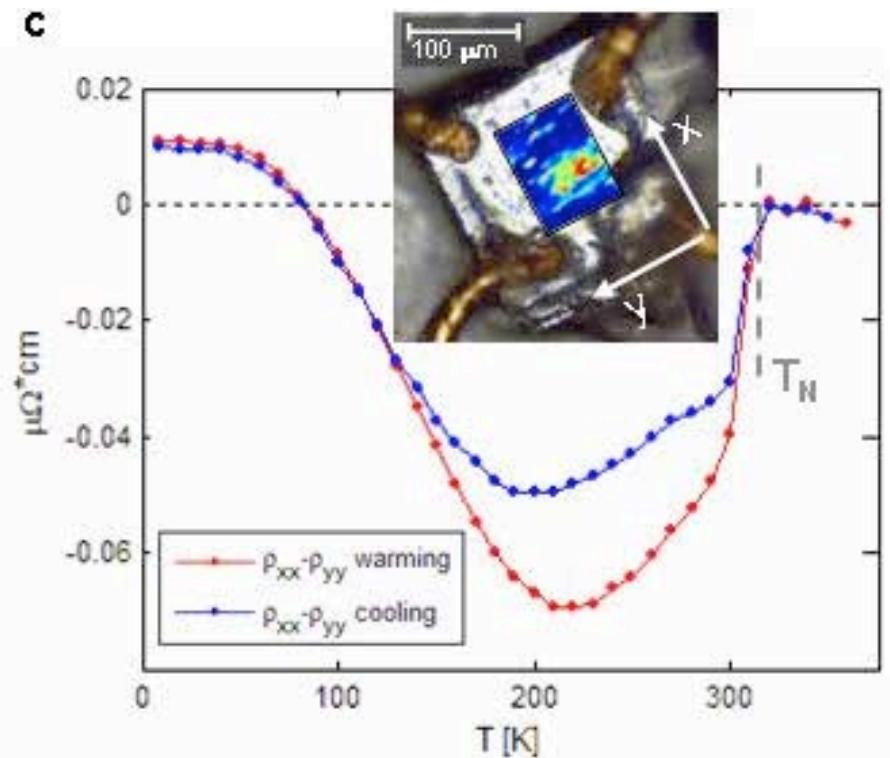


Hysteresis loop size at 100K  $\sim 50$  nanoOhm-cm.

Typical scale of domain wall length change  $\sim 50\%$  of 10 micron.

Therefore,

$$\begin{aligned}\text{resistance of typical wall} &= 2 \times 50 \text{ nanoOhm-cm} \times 10 \text{ micron/Area} \\ &= 10^{-4} \text{ microOhm-cm}^2/\text{Area}\end{aligned}$$



Resistivity 300K - 11.7  $\mu\Omega \cdot \text{cm}$   
10K - 0.24  $\mu\Omega \cdot \text{cm}$

195x180X45  $\mu\text{m}^3$