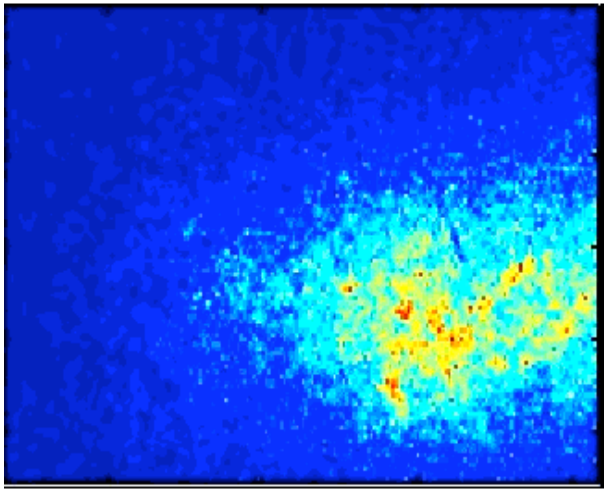
Dynamics at the Nanoscale

Eric D. Isaacs

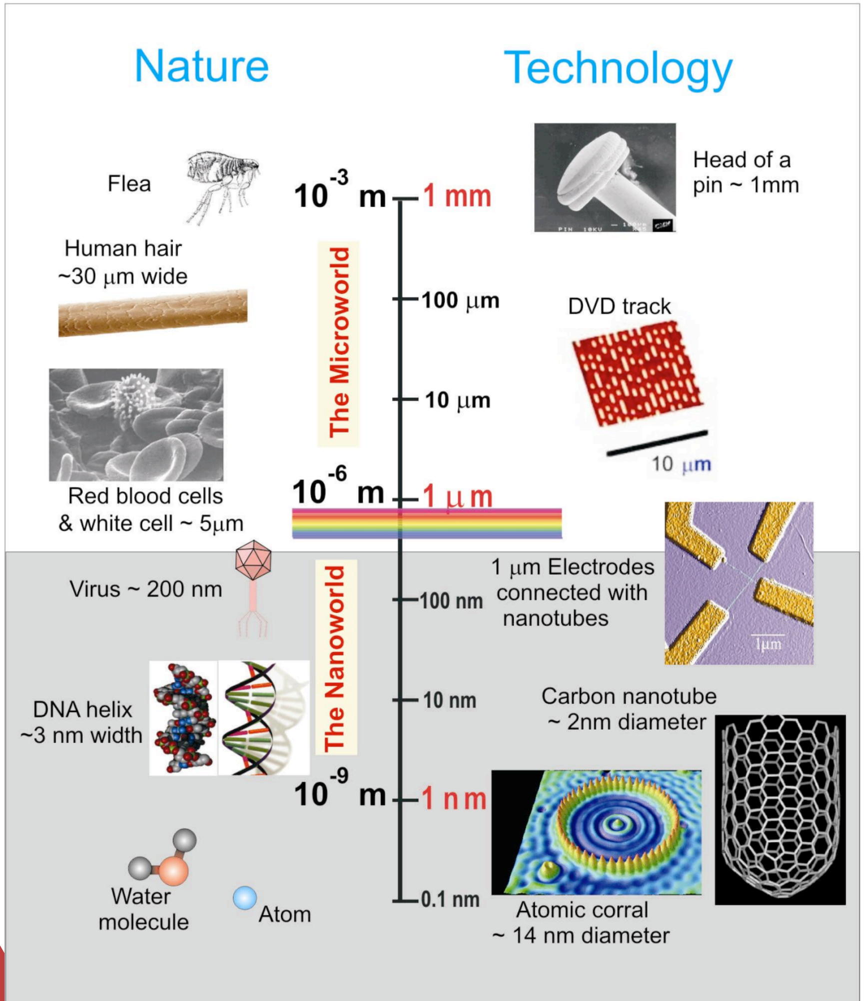


- Introduction: length and time scales.
- Nearly impossible XPCS studies of slow domain wall dynamics.
- Almost impossible experiments for an ERL?
- Ultra-fast structural dynamics in nanoparticles/macromolecules.

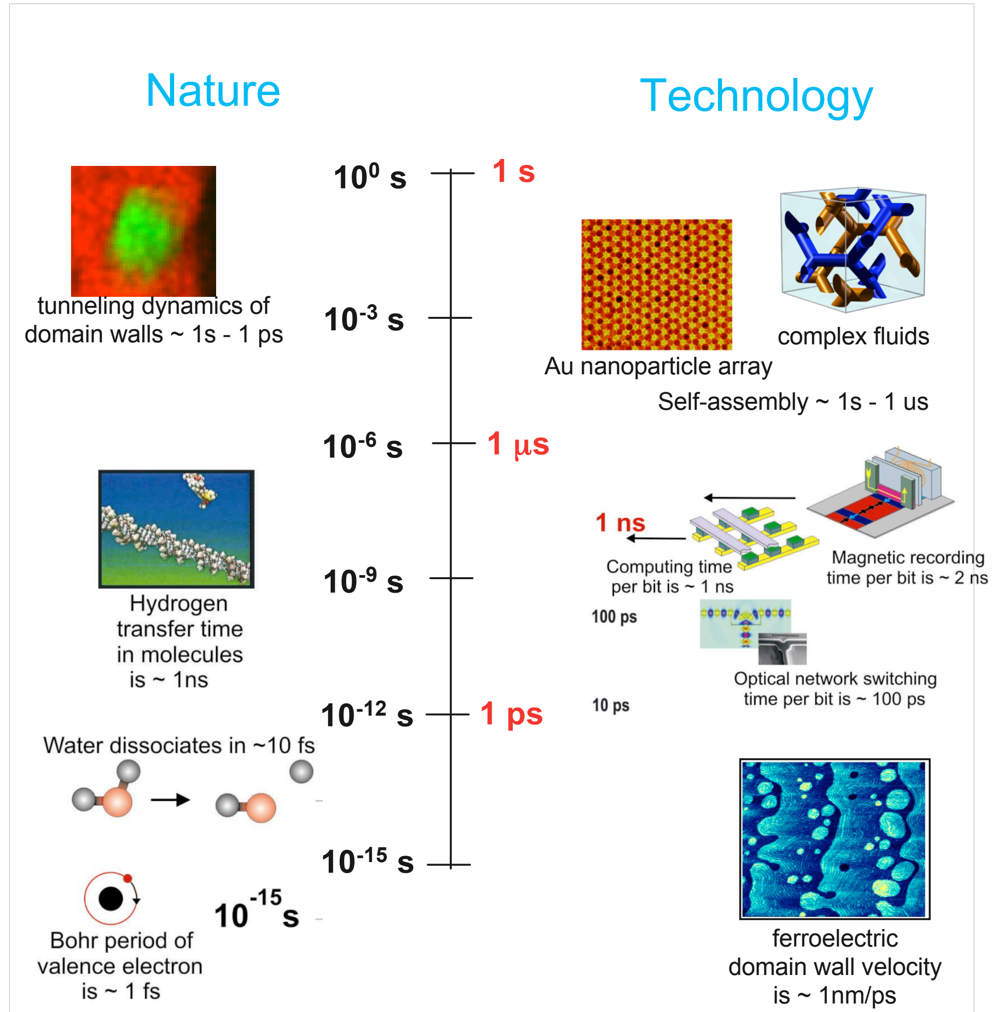
ERL Workshop III, Almost Impossible Materials Science,
Cornell University, June 17, 2006

Structure and dynamics at the nanoscale with X-rays.

Ultra-Small

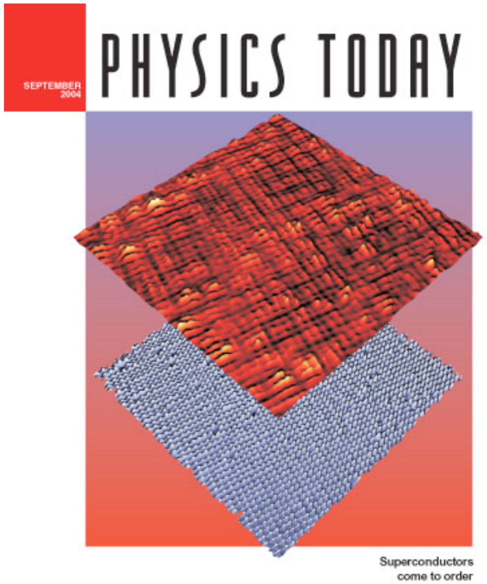


Slow to Ultra-Fast

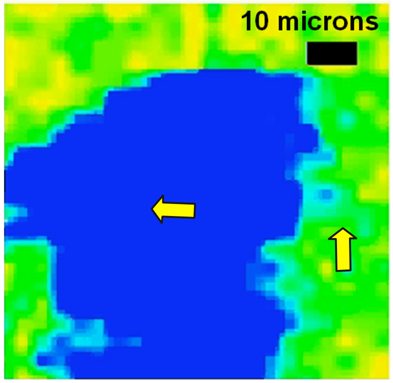


Major Challenges in Condensed Matter Science

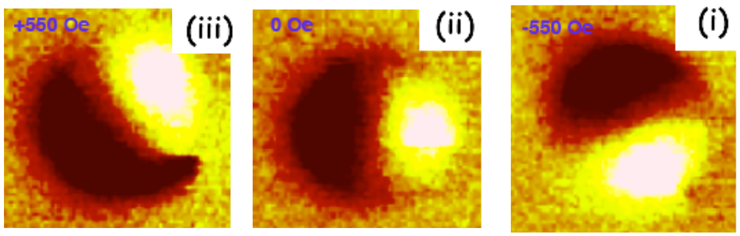
- Competing ground states on the nanoscale.
  - The relationship between mesoscale phases (spin, charge, lattice) and physical properties is a grand challenge in condensed matter physics.
  - The role of ***quantum and thermal spin/charge dynamics***.
- How can we study mesoscale phases in bulk?
  - x-ray microprobe (slow).
  - x-ray photon correlation spectroscopy (faster).



STM: checkerboard pattern in high-T_C superconductor. (Davis, et al).



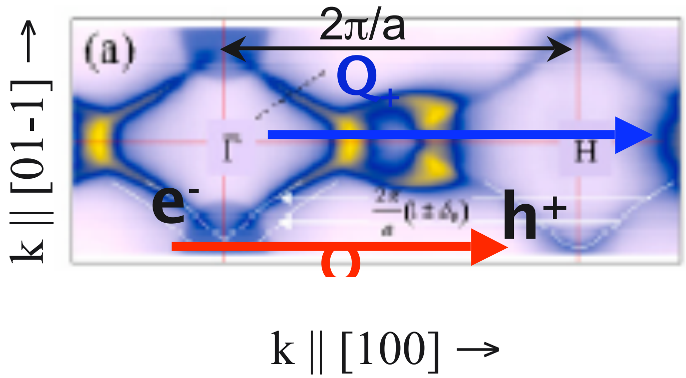
X-ray: spin domains in bulk Cr (Isaacs, et al)

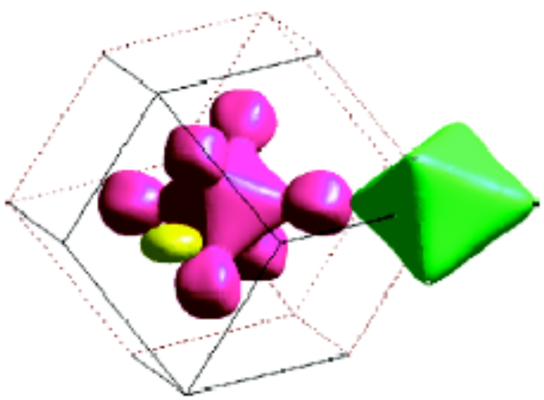


AFM: exchange bias vortices in spin domains in Py (Sort, et, al)

Why Study Chromium?

Photoemission shows Fermi surface





E. Rottenberg et al.,
New Journal of Physics **7** (2005) 114

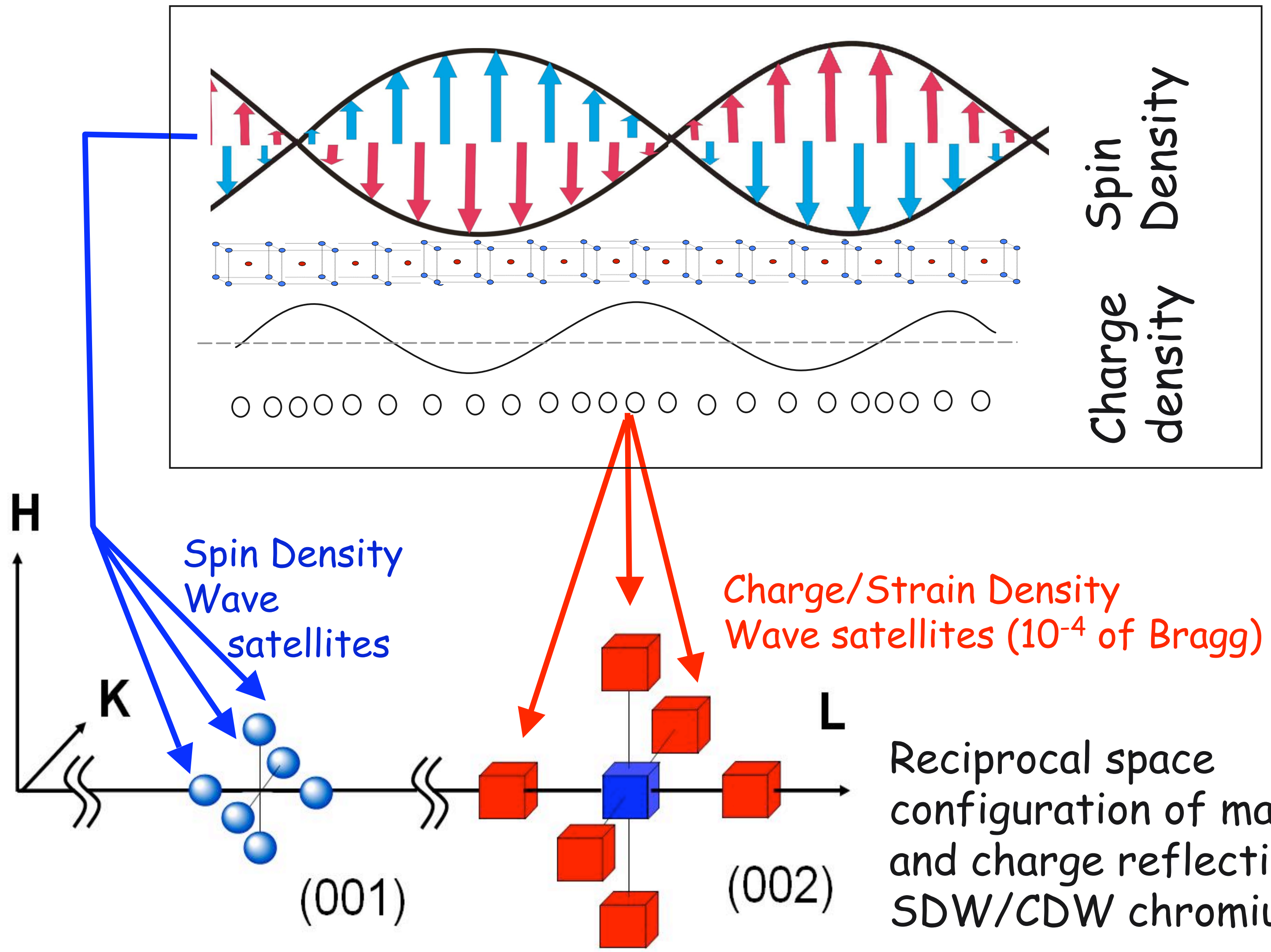
chromium and its common alloys are 'simple' bcc metals, exhibiting complex behaviors including;

- 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** (effectively increasing the size of the hole pocket) or by **applying pressure**.

Spin Density Wave (SDW) accompanied by Charge (Lattice Strain) Waves (CDW)



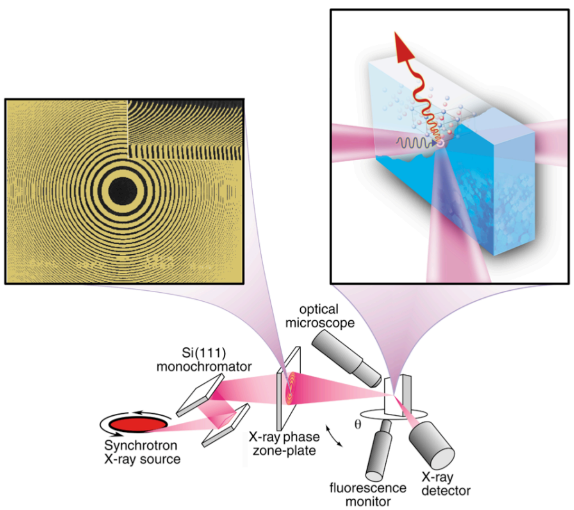
Reciprocal space configuration of magnetic and charge reflection of SDW/CDW chromium

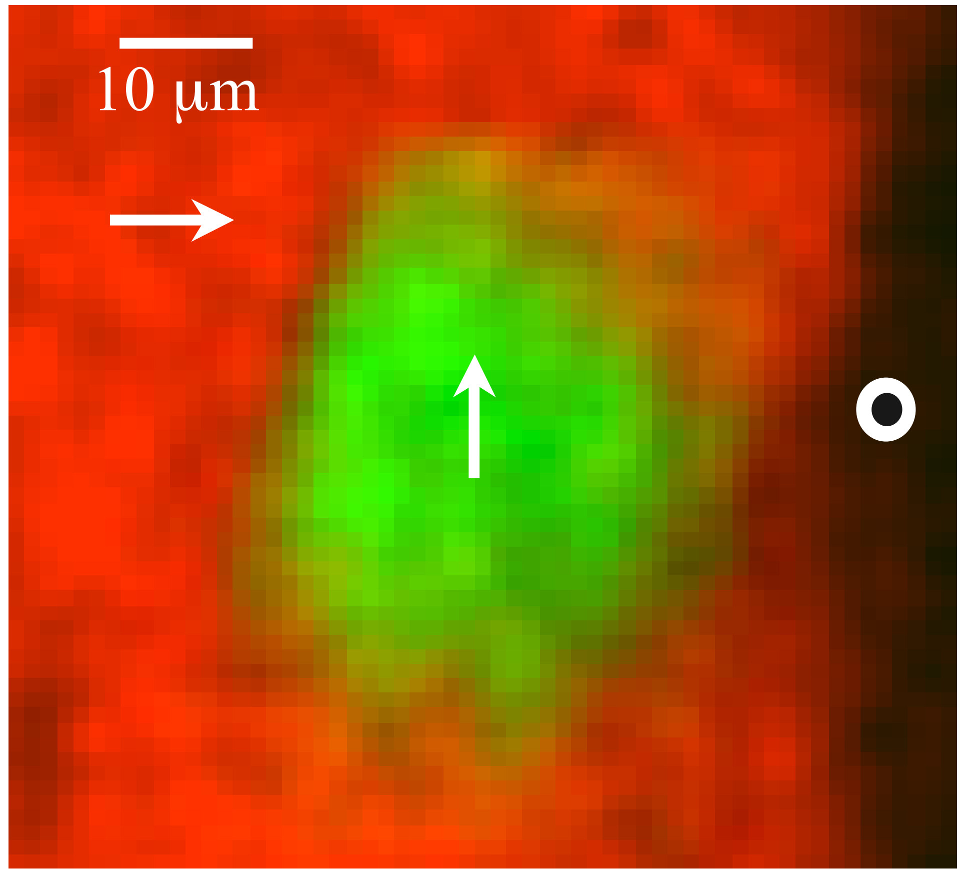
Typical Map of Domains in Chromium

Form images of domains using both SDW and CDW Bragg satellite peaks.

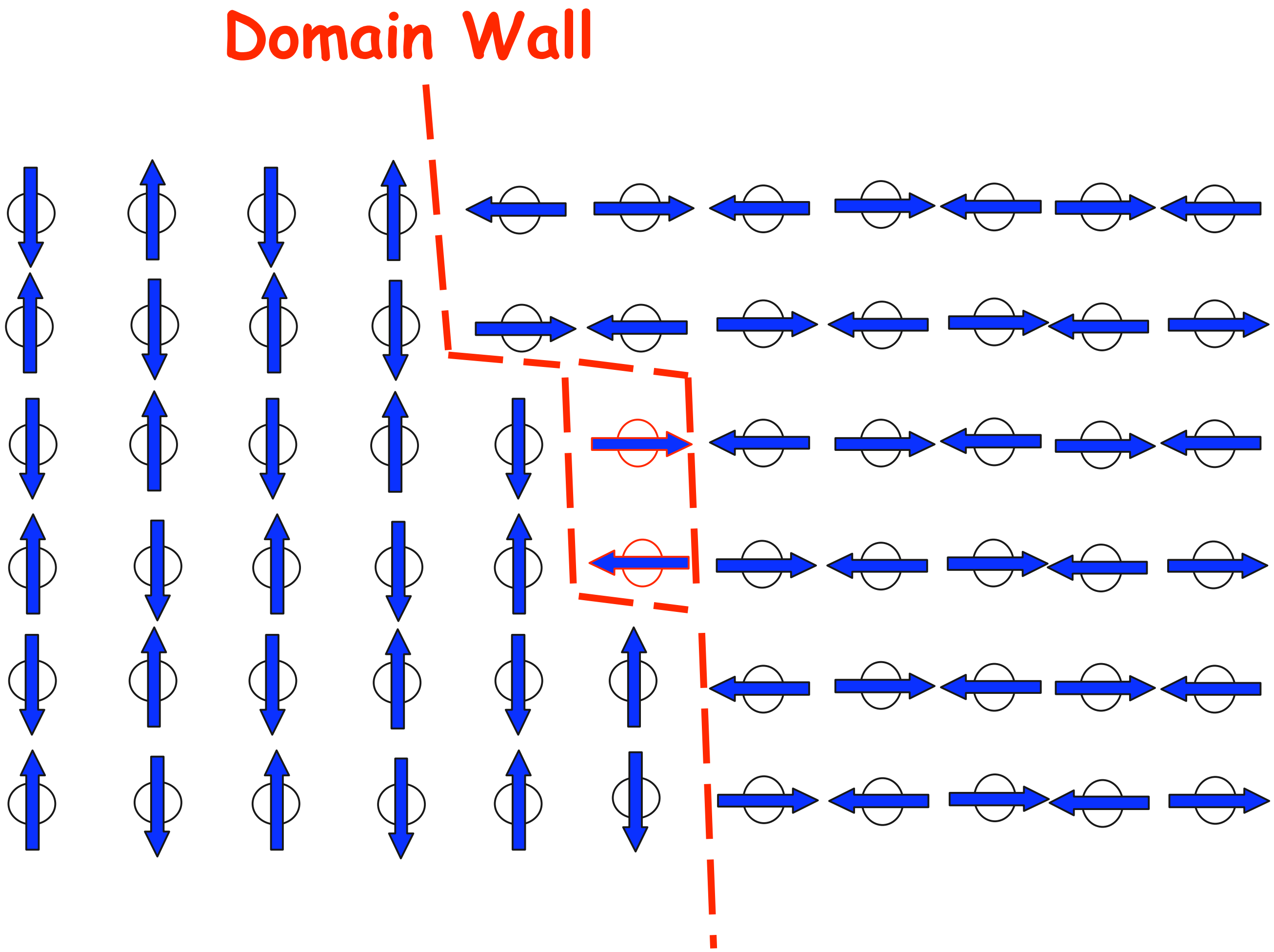
X-ray microprobe image of SDW domains in Cr.

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



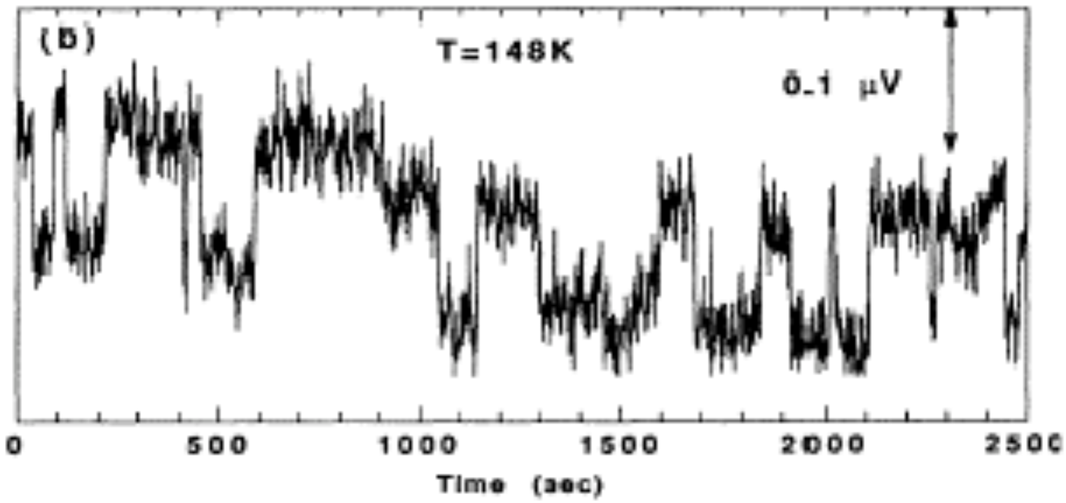


Domain Wall Fluctuations at the Nanoscale



1/f Noise in Thin Cr Films

Is this domain switching?



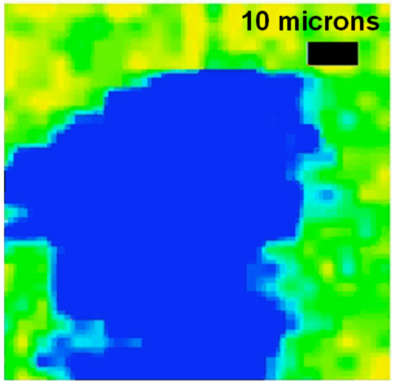
Michel et al., PRB 44, 7413 (1991)

Nanoscale probes of dynamics

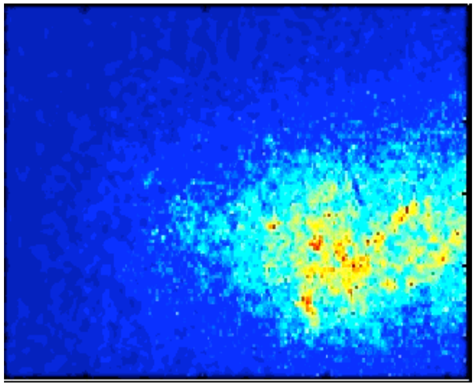
Probes of Domains

- Three x-ray imaging modes: scanning probe, full-field, 'lens-less'
  - Scanning probe: high spatial resolution, but slow (100nm or better)
  - Full-field won't work with diffraction, e.g, for magnetic contrast.

- Need probe with both time and spatial resolution - coherent scattering
  - (< 1 sec, < 1 nm spatial resolution, over macroscopic region of space)



Microprobe image of ppin domains in chromium (~ 3 hrs)



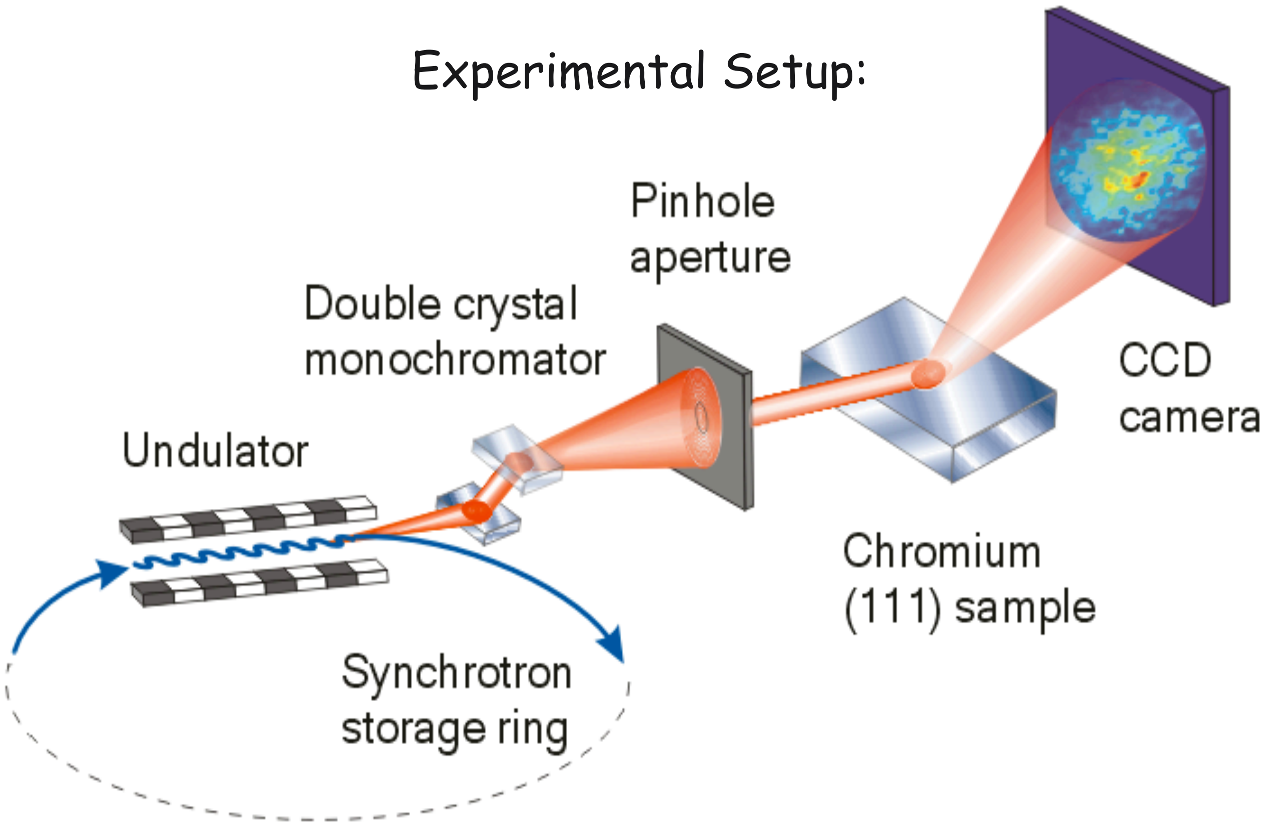
Coherent x-ray speckle dynamics

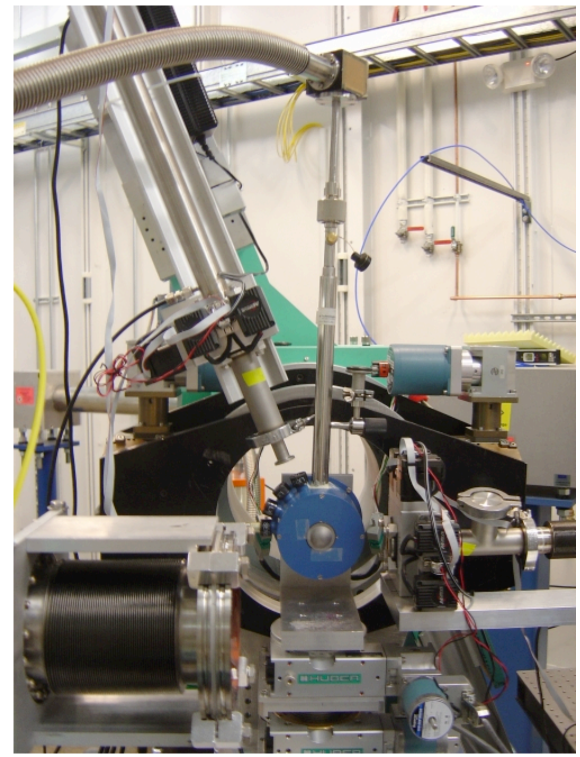
What can we do with x-ray coherence?

- Invert the speckle pattern to get high-resolution image of electron density.
  - 40 nm is best hard x-ray resolution today.
  - 1 nm physically possible!
- **Study time variations of speckle pattern to get information about dynamics of system.**
  - **phase transitions**
  - **complex fluids and glasses**
  - **defects/disorder dynamics**
  - **Interfaces and surfaces**

Coherent X-ray Speckle

X-ray Photo Correlation Spectroscopy (XPCS)



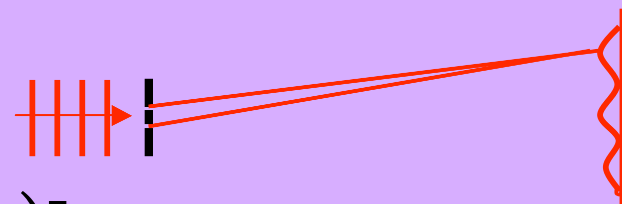


- CCD: 22 μm pixels
- speckle size: λ/d_{coh} * 2 m ~ 40 μm

X-ray Coherence

Young's double slit experiment.

Intensity varies as



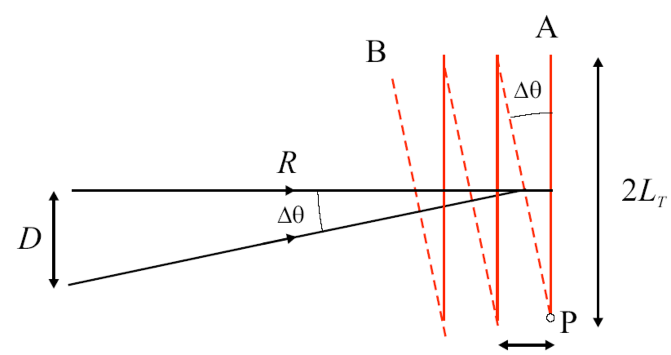
$$I = 2I_0\left[1+\beta\cos\left(2\pi d\sin(\theta)/\lambda\right)\right]$$

Temporal and spatial coherence at the APS.

longitudinal: $\Lambda_L = \frac{\lambda}{2}\frac{\lambda}{\Delta\lambda}$

transverse: $\Lambda_T = \frac{\lambda}{2}\frac{R}{D}$

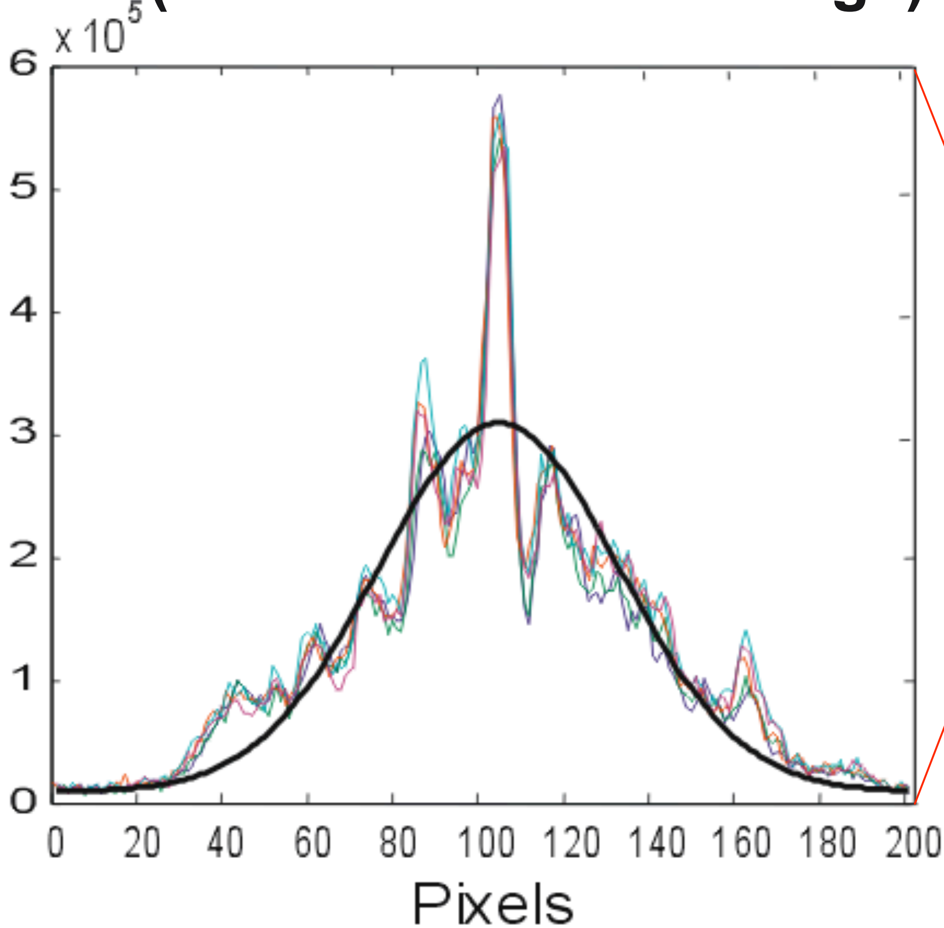
(b) Transverse coherence length, L_T



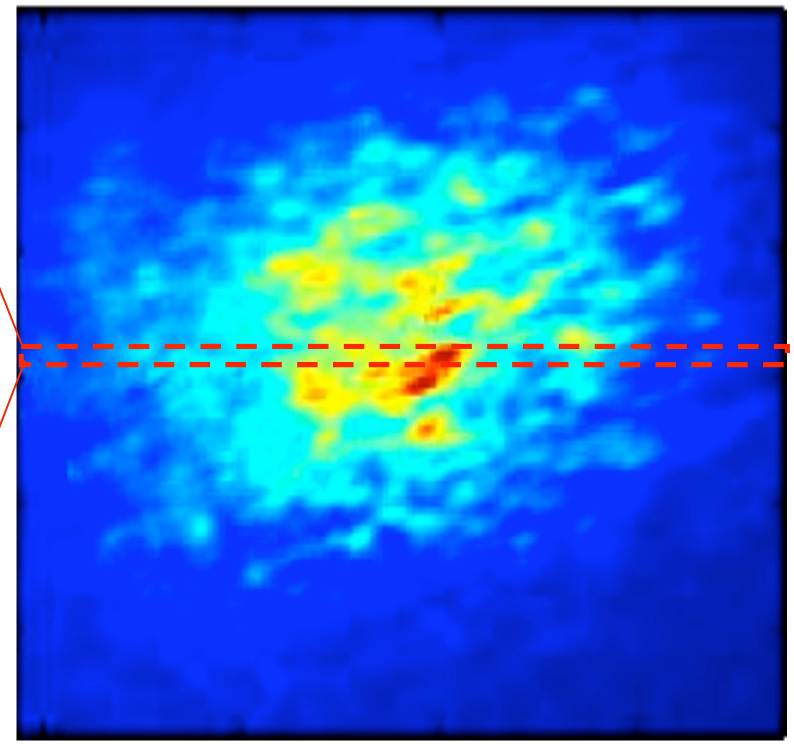
10 x 40 μm^2 pinhole @ APS: ~ $3x10^9$ ph/s, β ~ 15 %

Coherent X-ray Diffraction 'Speckle' Pattern From Bulk Chromium

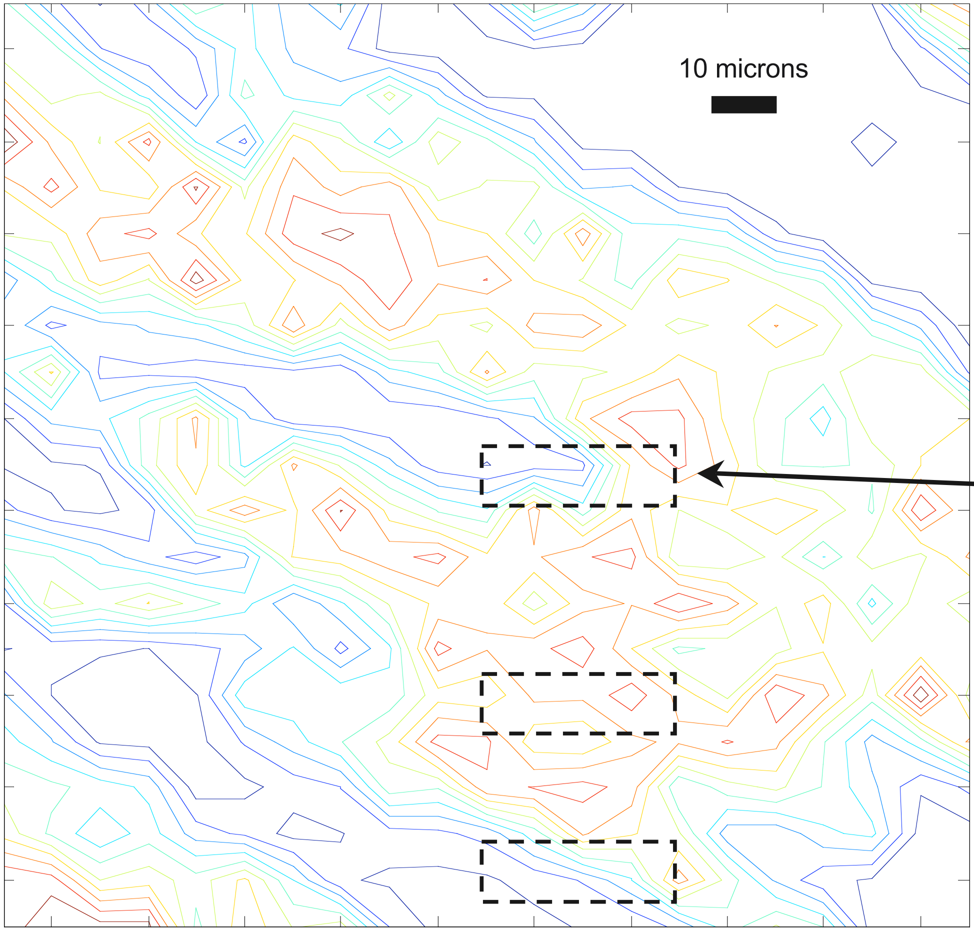
Static Bragg speckle (line scan from 2D image)



Static x-ray speckle pattern



Typical Map of Q-domains in Chromium

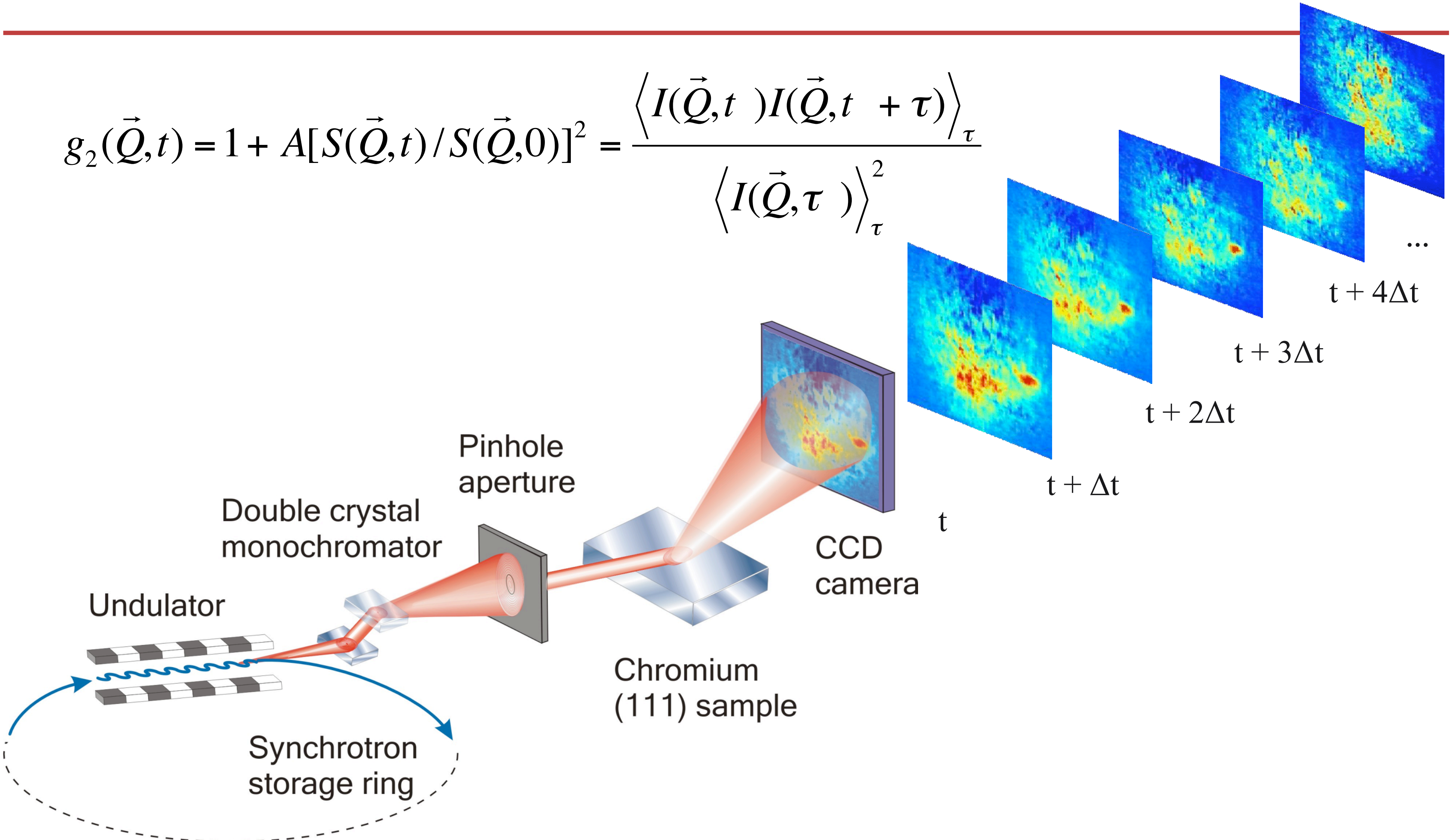


(0,0,2-2δ)

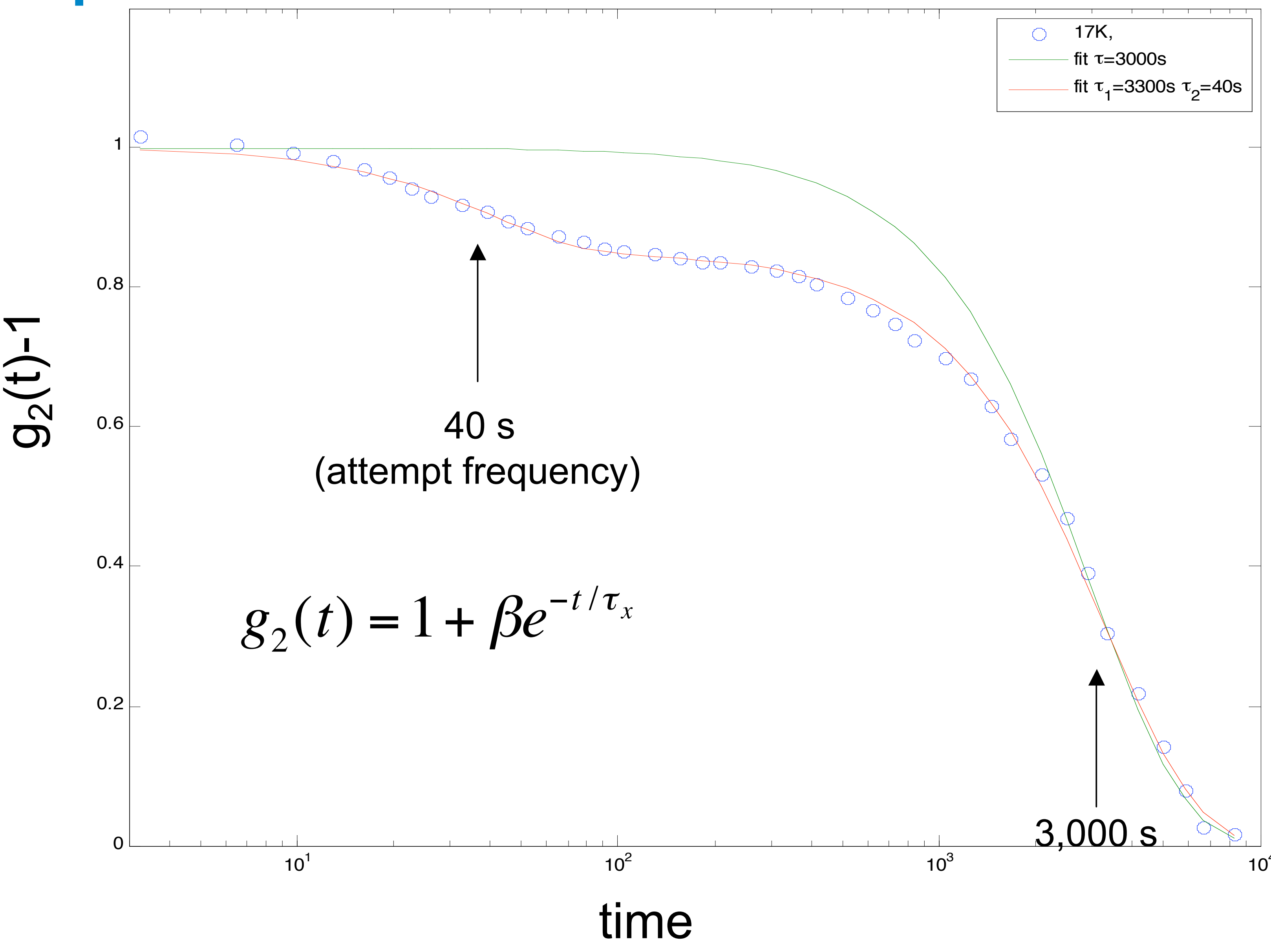
Domain wall

Autocorrelation function, $g_2(t)$:

$$g_2(\vec{Q},t) = 1 + A[S(\vec{Q},t)/S(\vec{Q},0)]^2 = \frac{\langle I(\vec{Q},t)I(\vec{Q},t+\tau)\rangle_\tau}{\langle I(\vec{Q},\tau)\rangle_\tau^2}$$

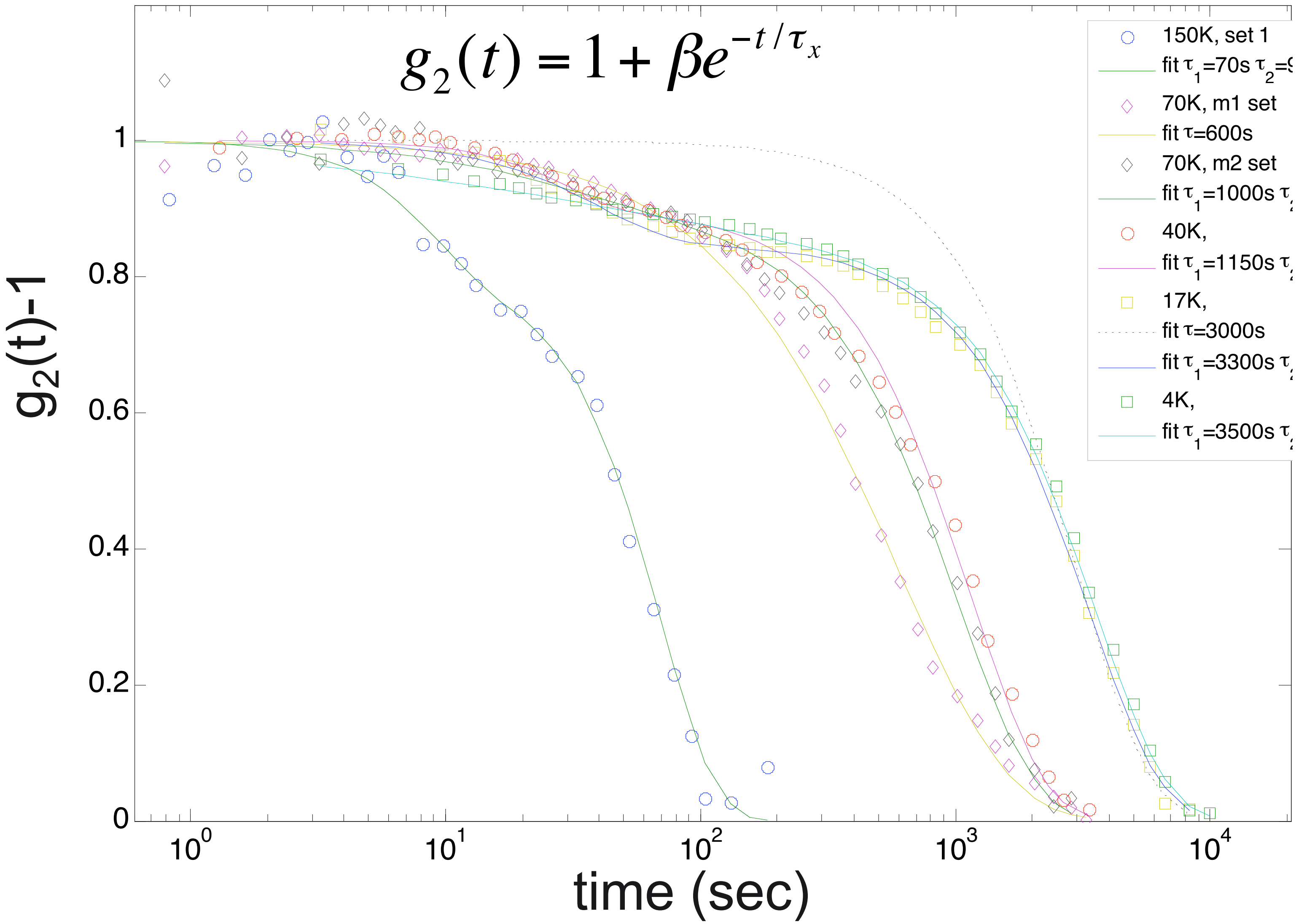


Dynamics with autocorrelation function $g_2(t)$: multiple timescales

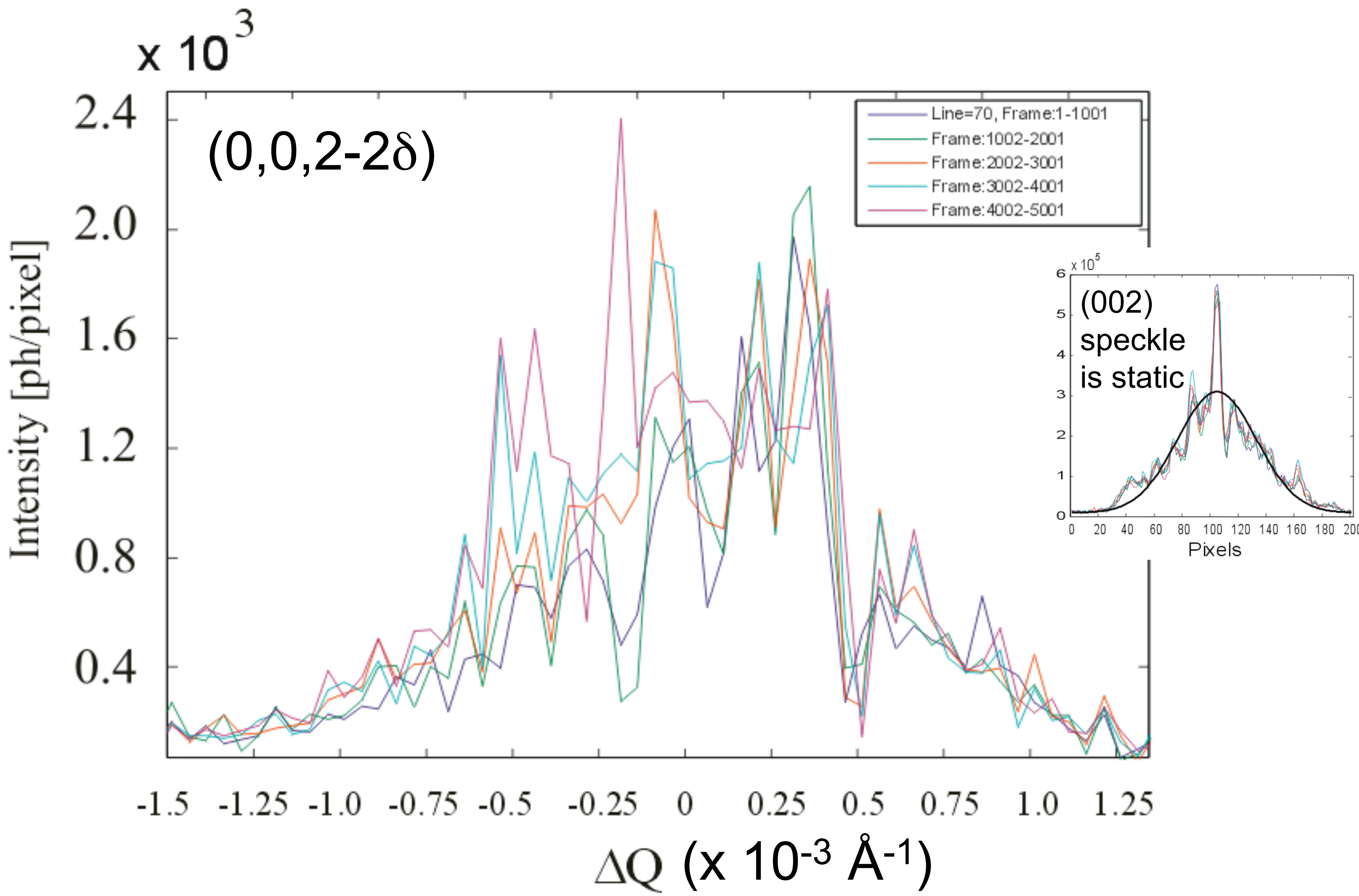


$$g_2(t) = 1 + \beta e^{-t/\tau_x}$$

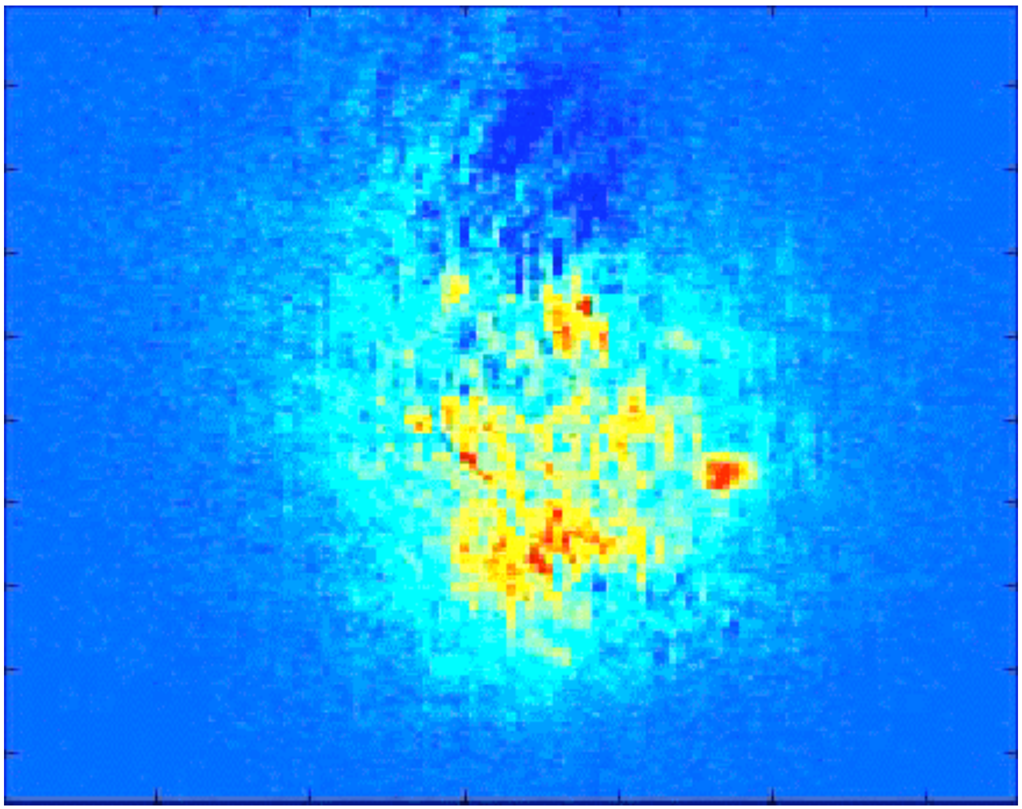
Dynamics with autocorrelation function $g_2(t)$



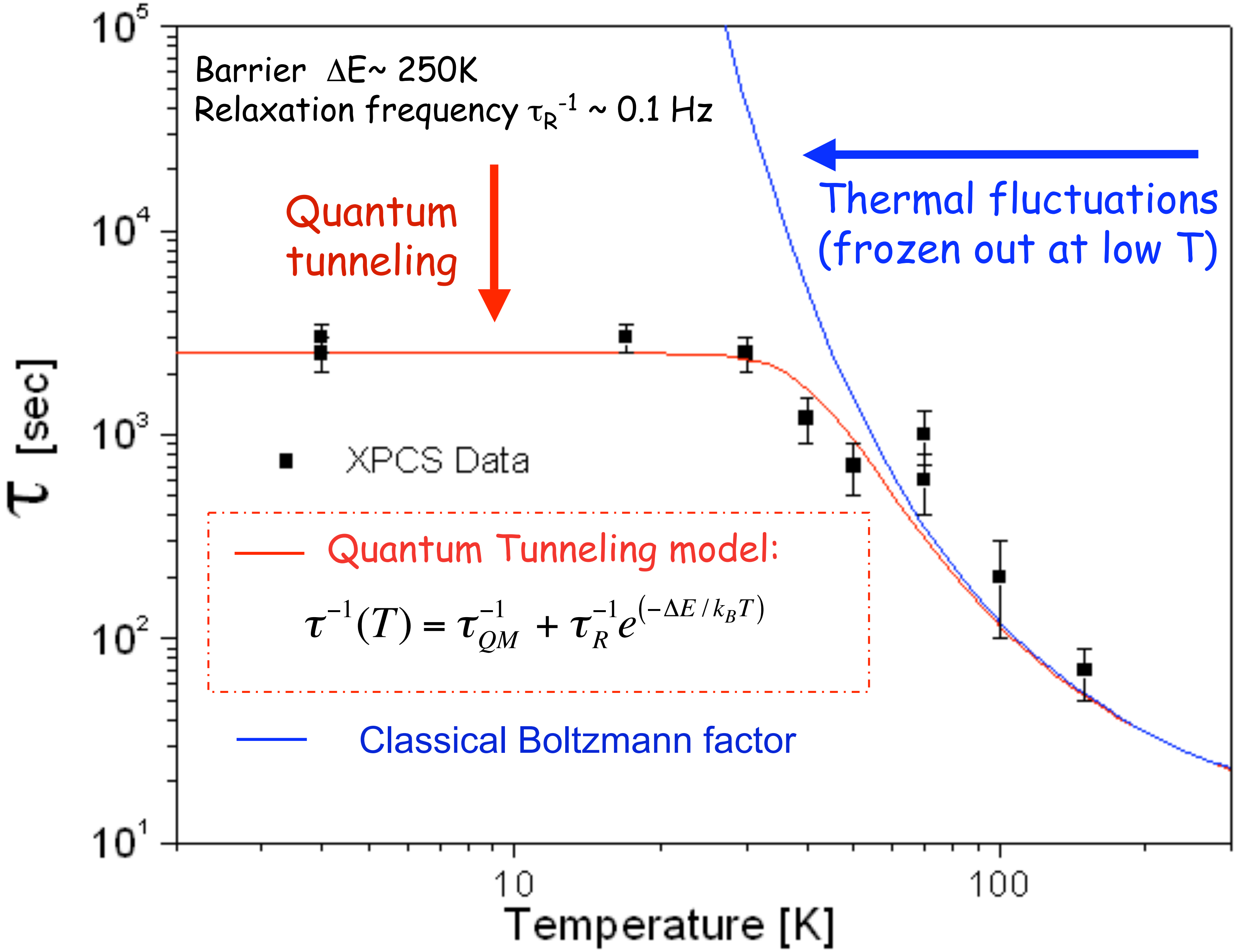
CDW is Dynamic at T=4 K !



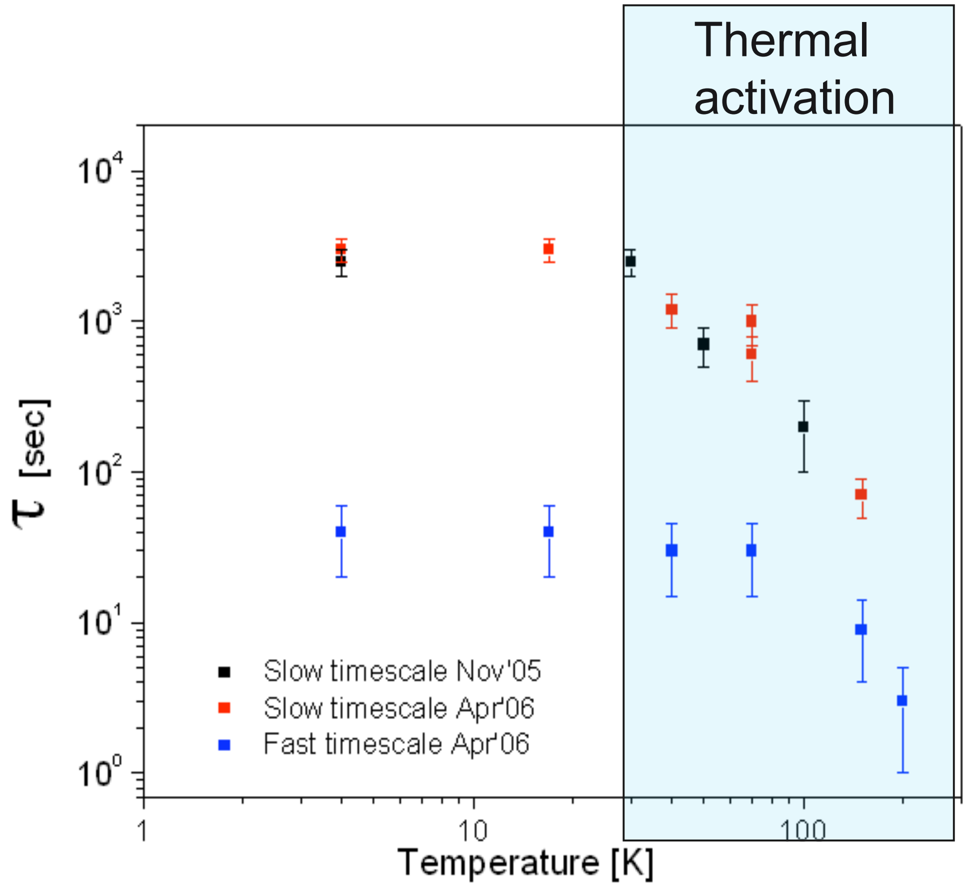
CDW speckle, the movie (17K)

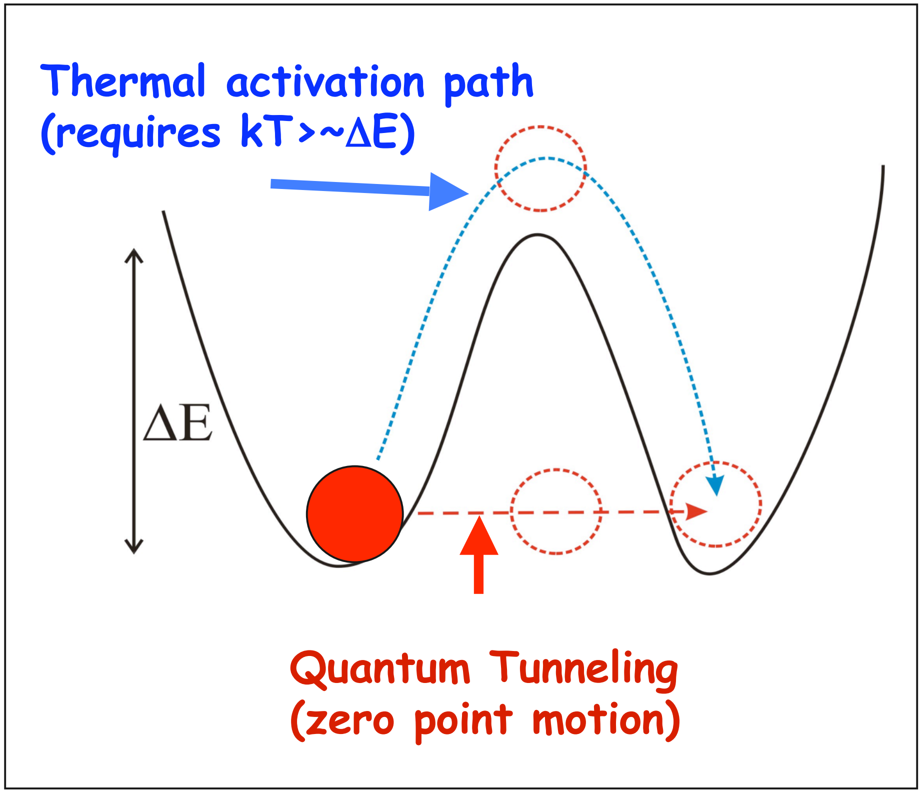


Temperature dependence of fluctuations

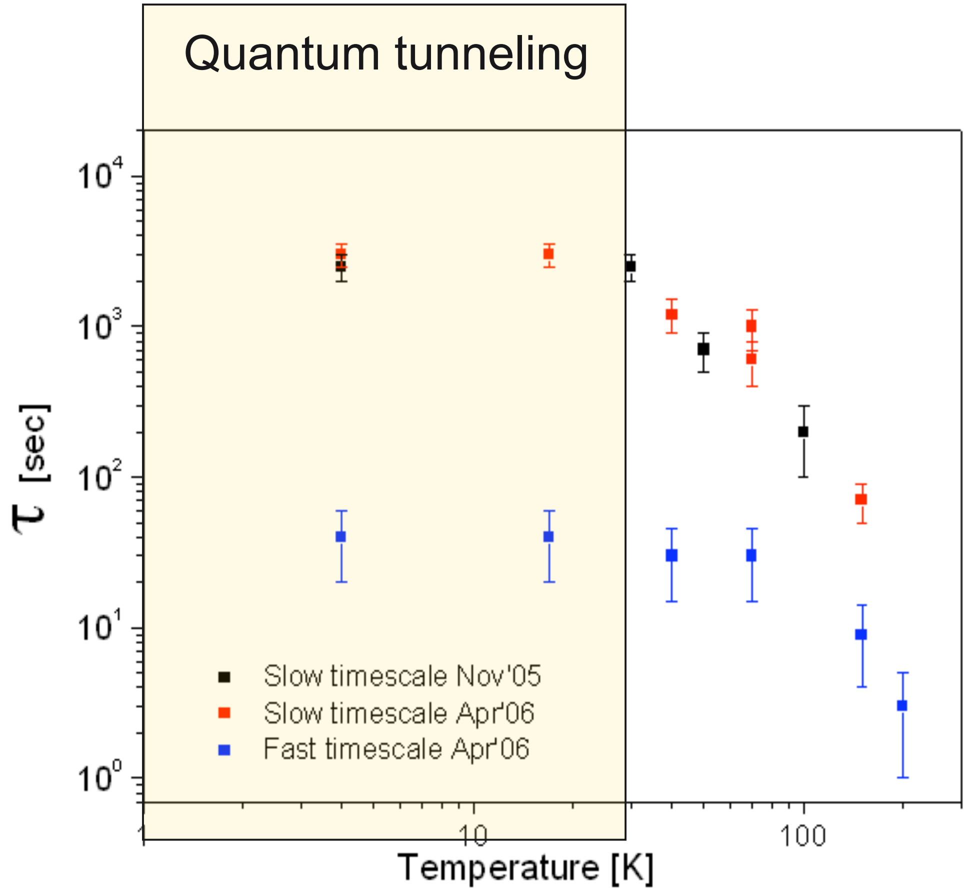


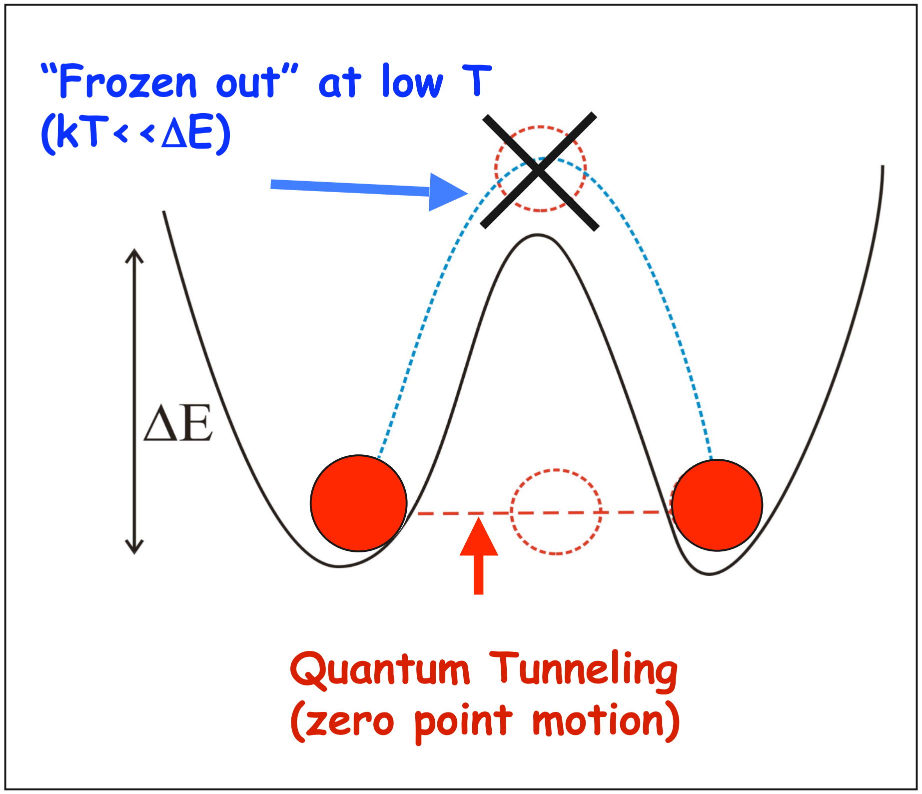
Domain Wall Motion:
Thermal activation model (high T)





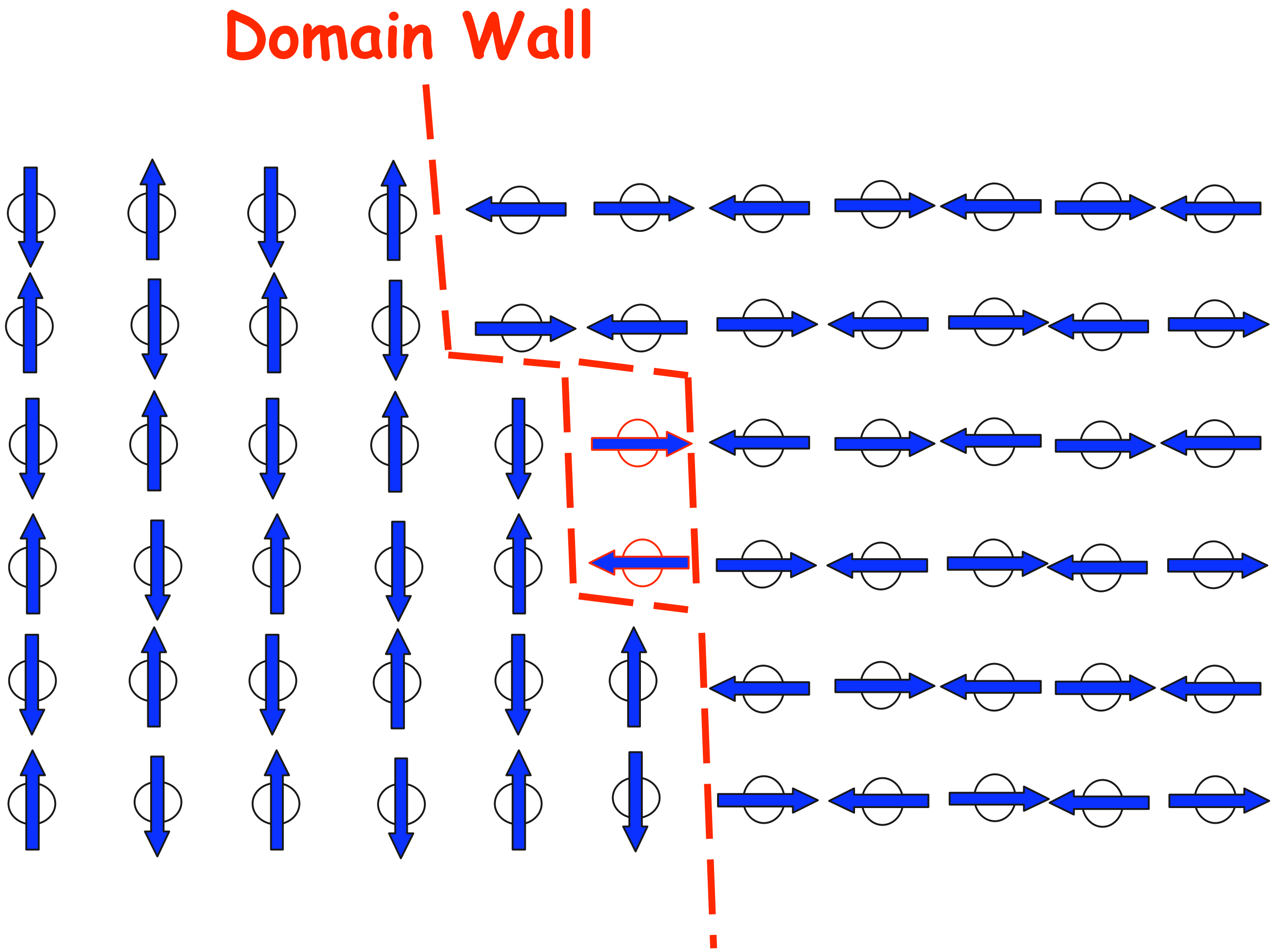
Domain Wall Motion: Quantum Tunneling model (low T)





Imaging Domain Wall Fluctuations at the Nanoscale

Is domain wall motion coherent?



Imaging Domain Wall Fluctuations at the Nanoscale

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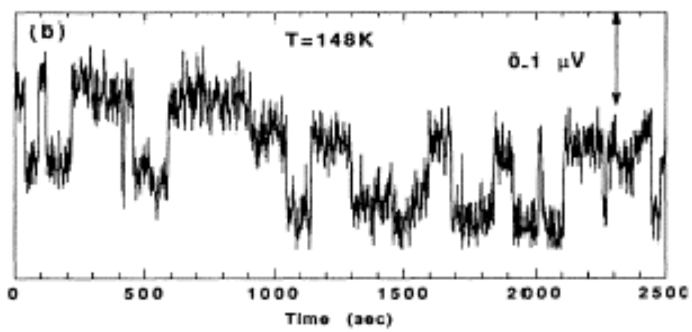
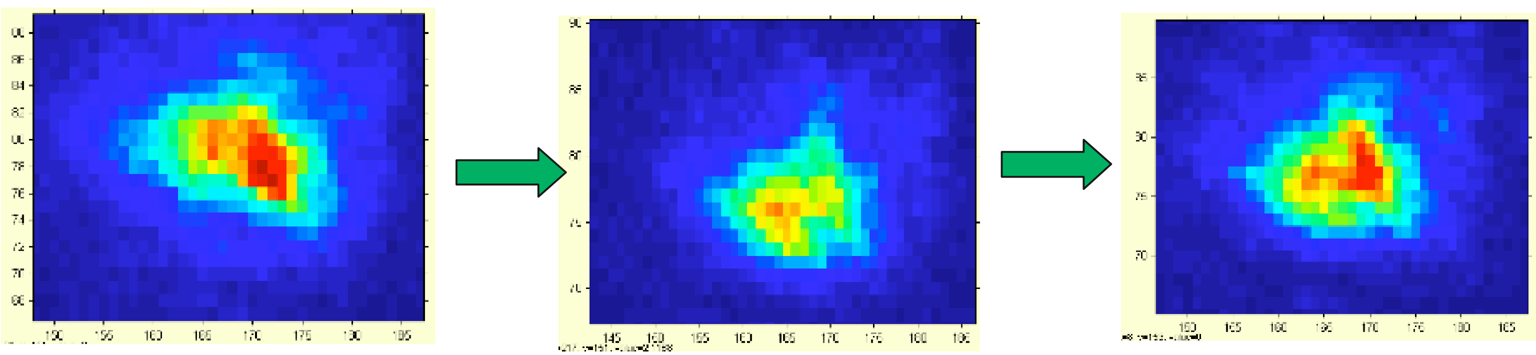


Transport in film.

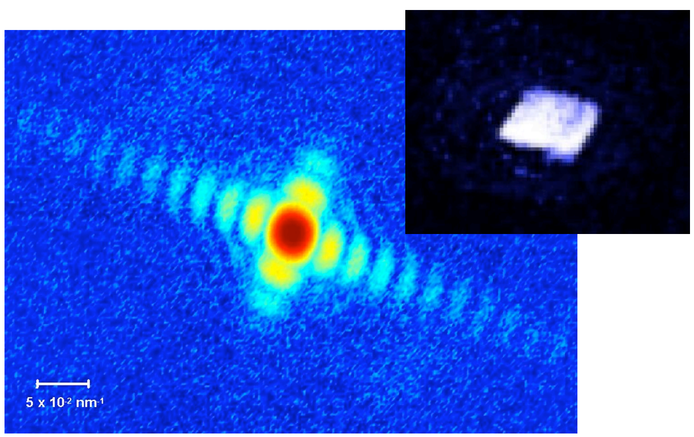
Smaller pinhole reveals switching between two states.



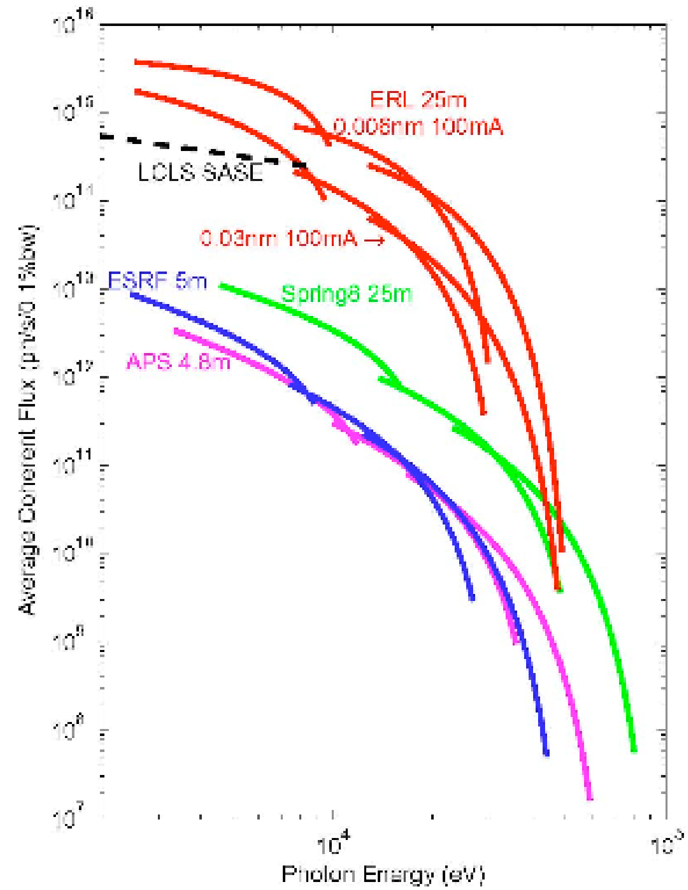
Combine coherent imaging and dynamics at an ERL

- ***3D movie of domain walls:*** nm resolution at μs.
  1) x 10^2 - 10^3 coherent flux (ERL).
  2) smart detectors.

- ***Coherent x-ray diffraction imaging****:* ~10^2 seconds per frame to get ~ 1 count per pixel.

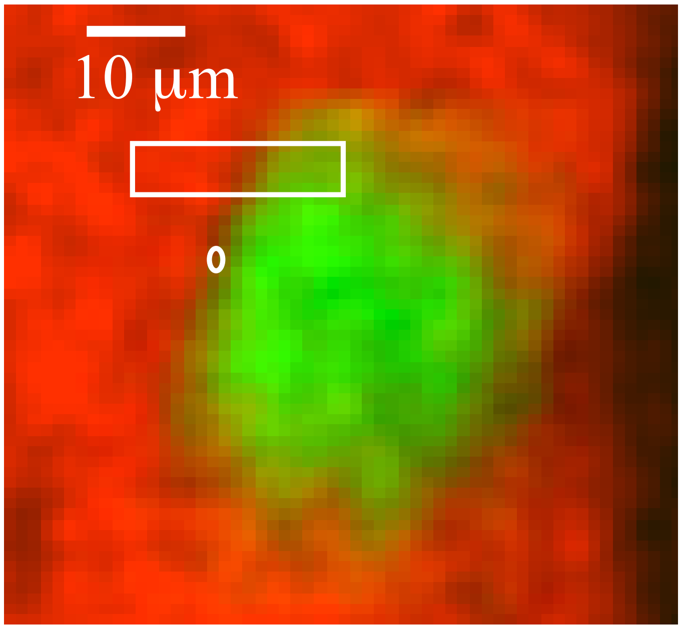


170 nm Ag cubes: I.K. Robinson, et. al.

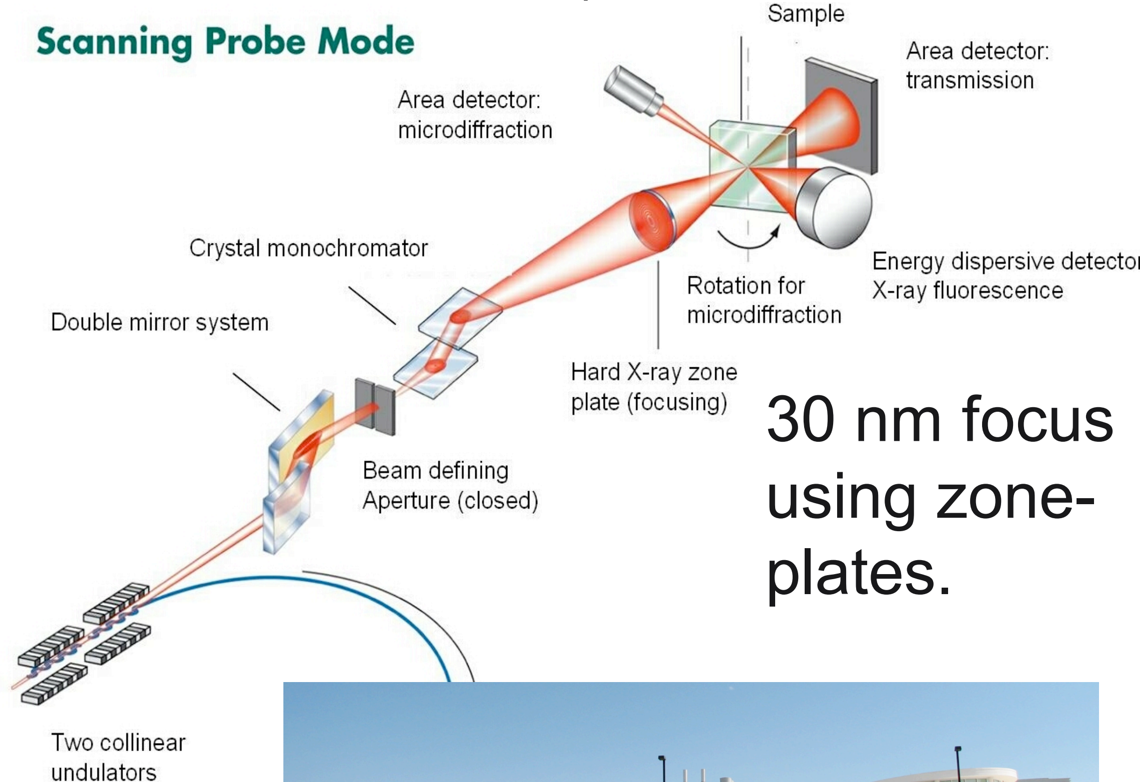


Center for Nanoscale Material's Hard X-ray Nanoprobe at the APS

G.B. Stephenson, J. Maser



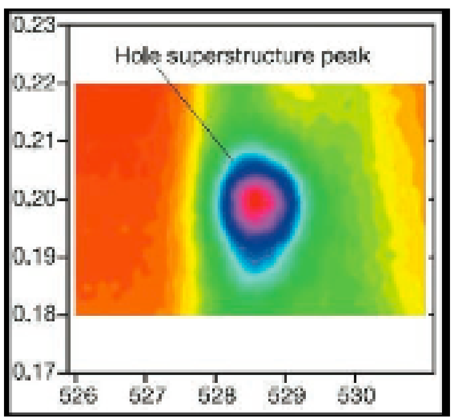
Zoom-in on domain wall using diffractive optics at 3rd-gen source



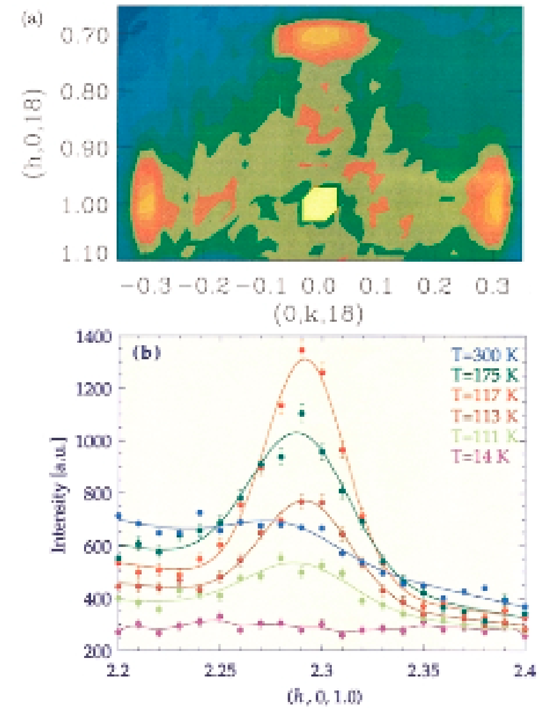
30 nm focus using zone-plates.

ERL Experiments: Melting of Charge & Spin Ordered States

- For example: dynamics of ‘striped’ phase melting in complex oxide compounds.
- Dynamics of striped domains may play a critical role in high-T_C superconductivity.
- Weak scattering at CDW superlattice peaks.
- Can we measure the SDW w/ERL?



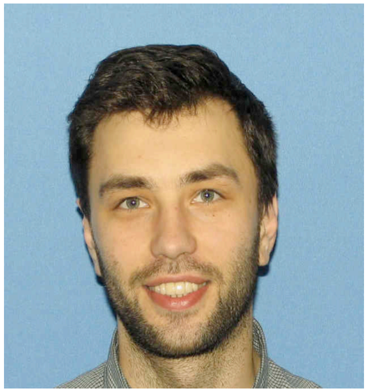
Hole ordering peak in $Sr_{14}Cu_{24}O_{41}$. (resonant)
Abbamonte, et al, Nature (2004)



Charge melting in $La_{1.2}Sr_{1.8}Mn_2O_7$,

L. Vasiliu-Doloc, et al., PRL (1999).

Quantum Speckle Collaborators



Dr. Oleg Shpyrko
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Jonathan Logan
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Clarisse Kim
Univ. of Chicago

Prof. Gabriel Aeppli
Univ. College, London

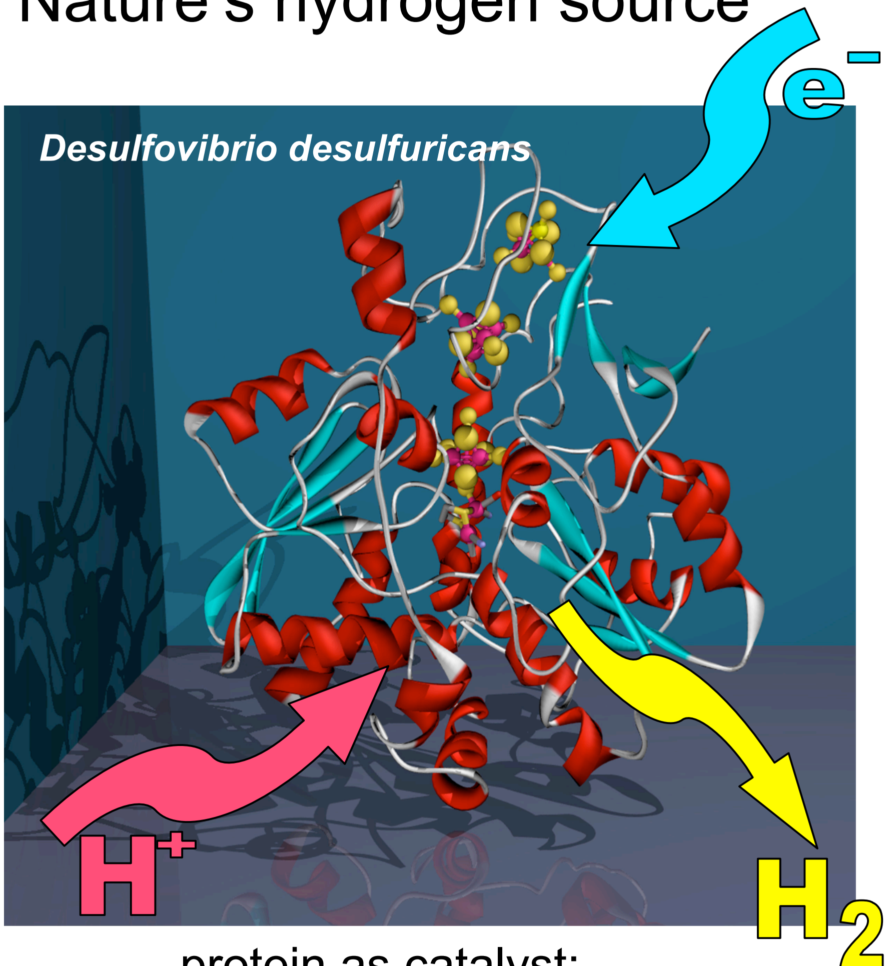
Prof. Tom
Rosenbaum
Univ. of Chicago

Dr. Yejun Feng
Univ. of Chicago

Rafael Jaramillo
Univ. of Chicago

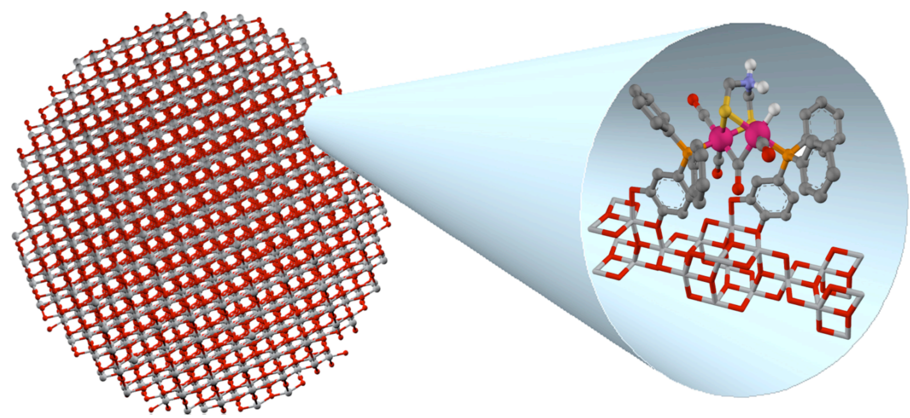
Many of nature's energy conversion processes occur on the nanoscale at fast time-scales

Nature's hydrogen source



protein as catalyst:
Fe-hydrogenase

synthetic hydrogen source
using solar energy

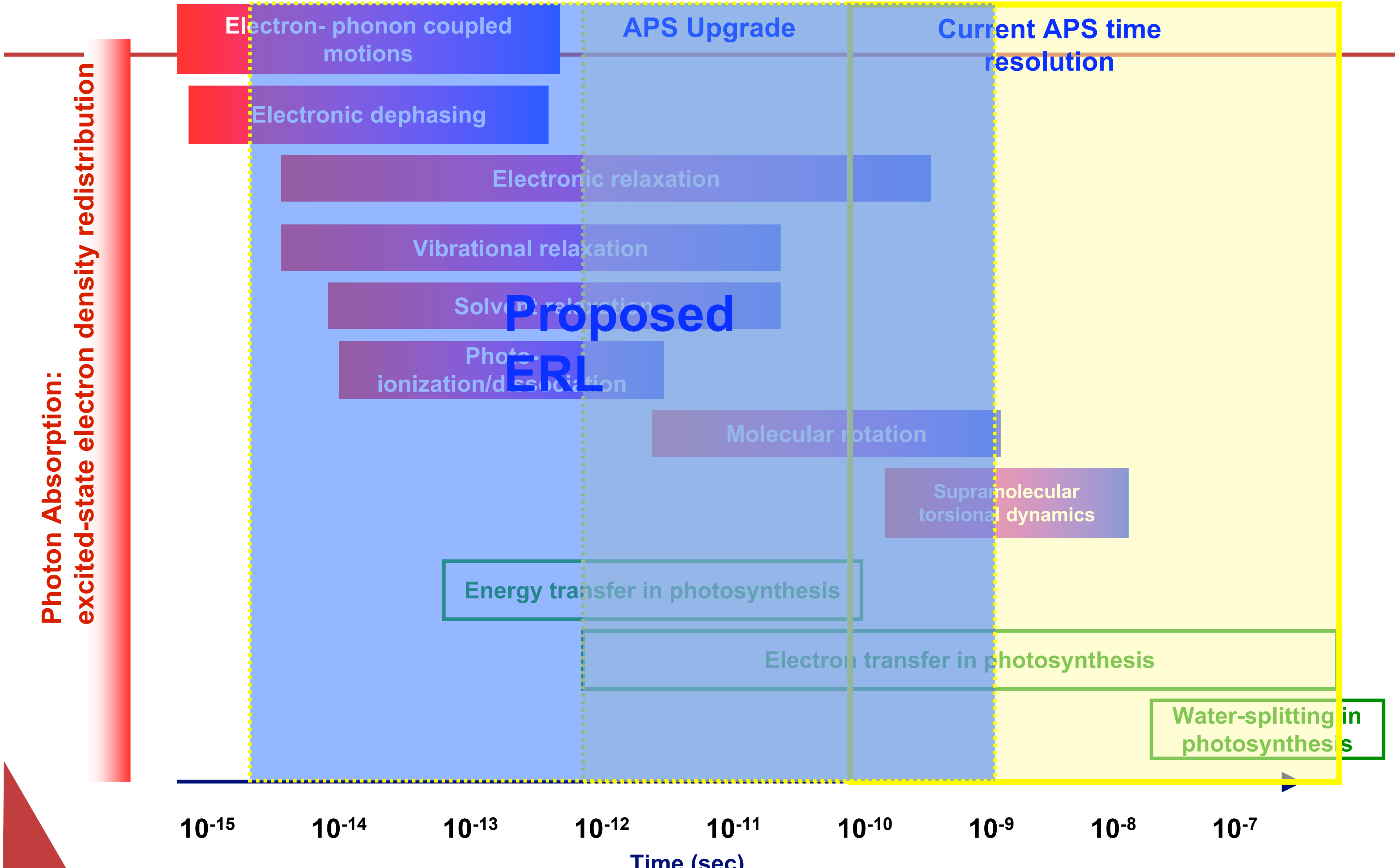


TiO_2 'e⁻-donor' + Fe_2S_2 active site mimic

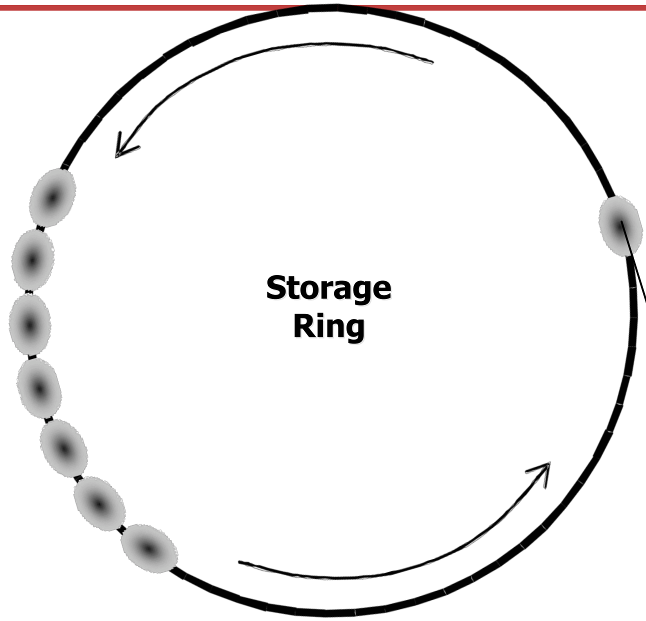
biomimetic hydrogen production on the surface of a metal-oxide nanoparticle.

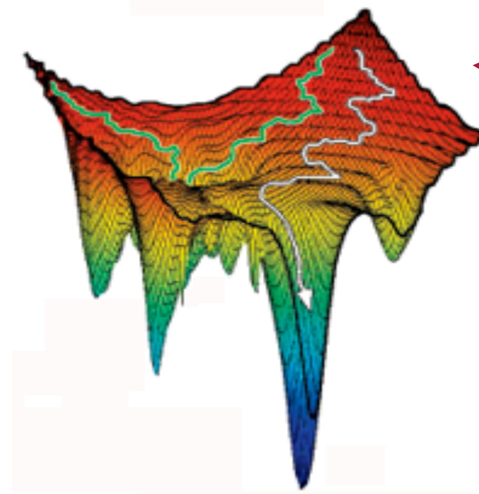
D. Tiede, X. Zuo, A. Goshe

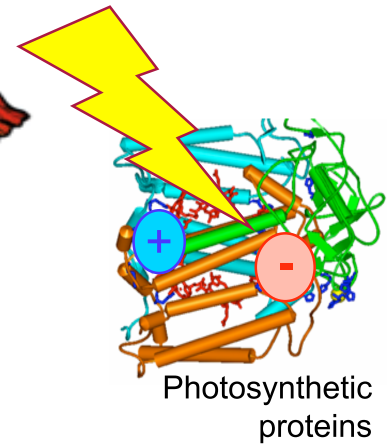
Time-scale for processes in solar energy conversion



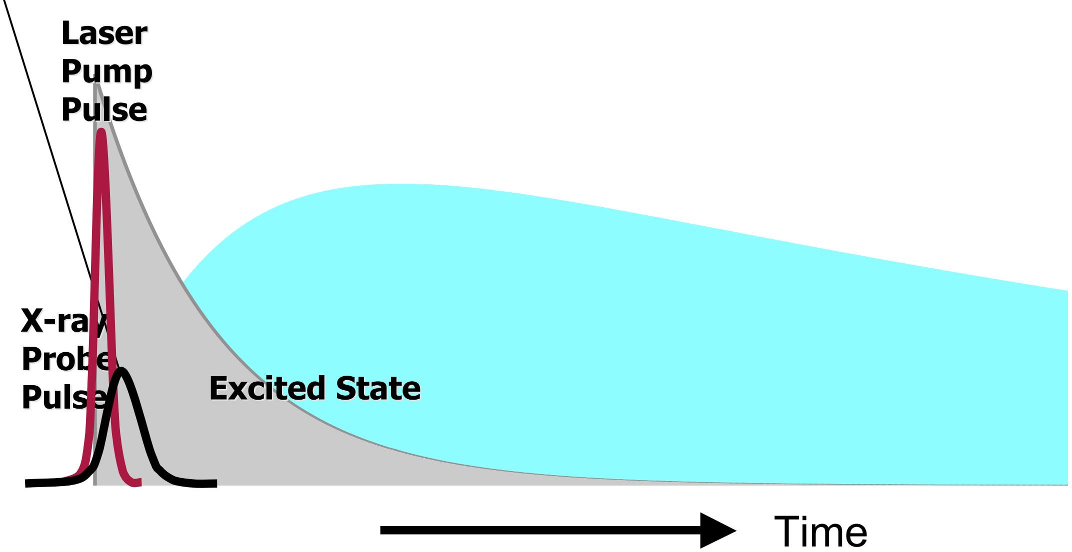
Structural Landscapes Underlying Function: Towards sub-picosecond time-resolved scattering.







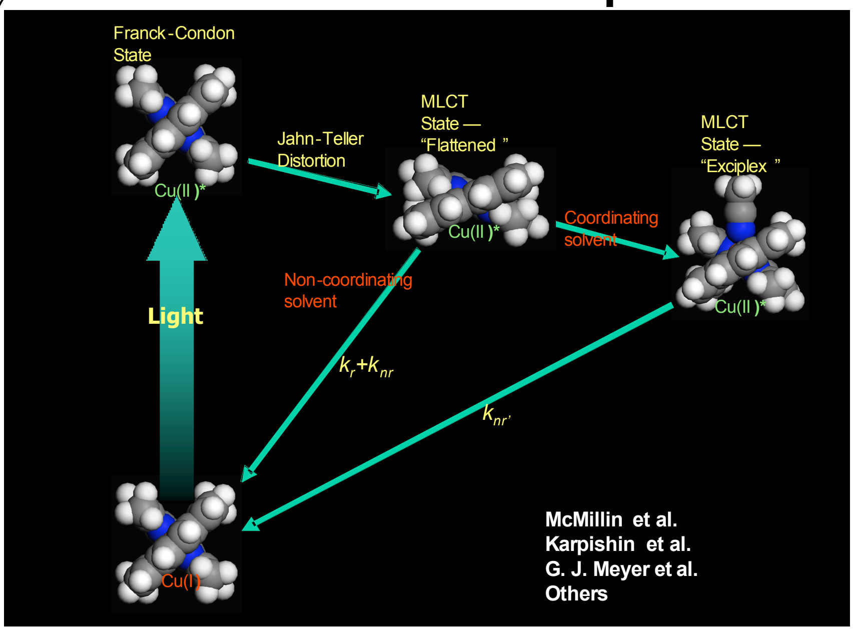
Laser/X-ray pump/probe
Intrinsic time resolution:
APS: ~100 ps fwhm (2005)
APS: ~ 1 ps (2012)
ERL: ~ 50 fs



Can we measure structural dynamics of individual nanoparticles?

D. Tiede, et al, structure
L. Chen, et al, spectroscopy

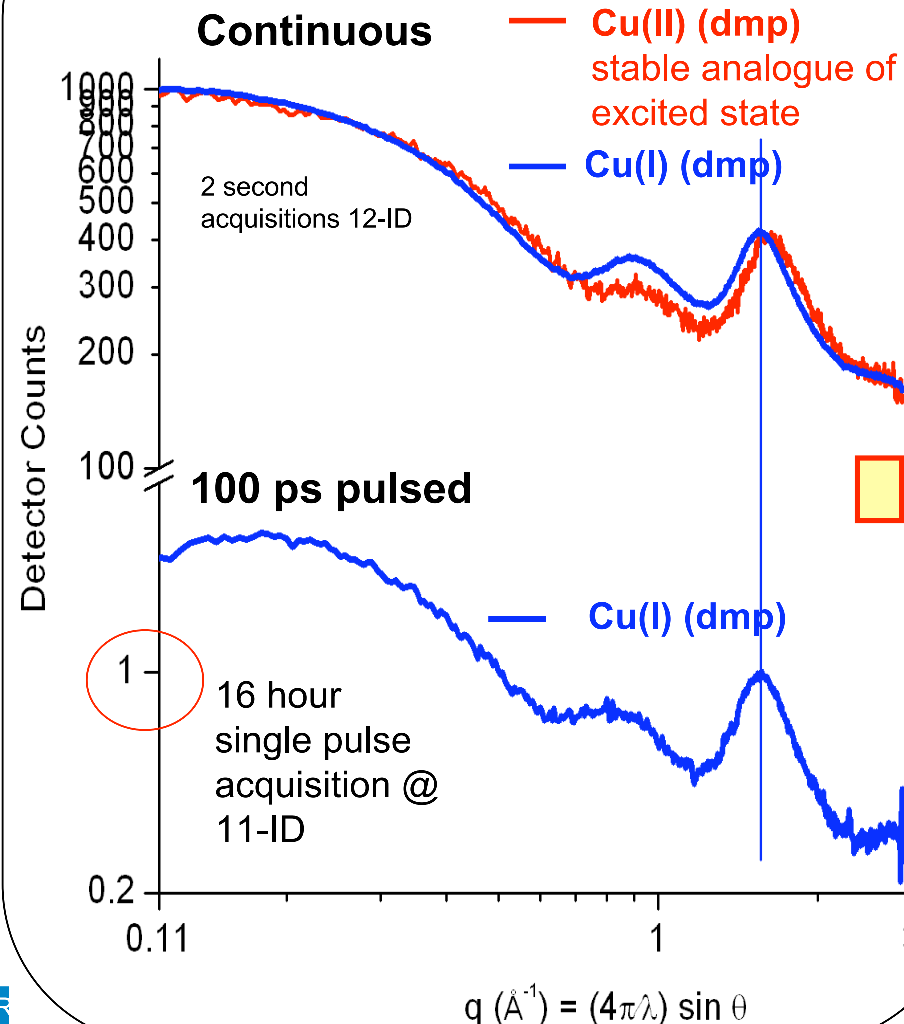
Model M-L CT complex



copper dimethyl-phenanthroline (dmp)

Chen, et al,J. Am. Chem. Soc. (2002) **124**,*10861;*(2003) **125**,7022.

Solution Scattering -
1st single bunch SAXS !!!



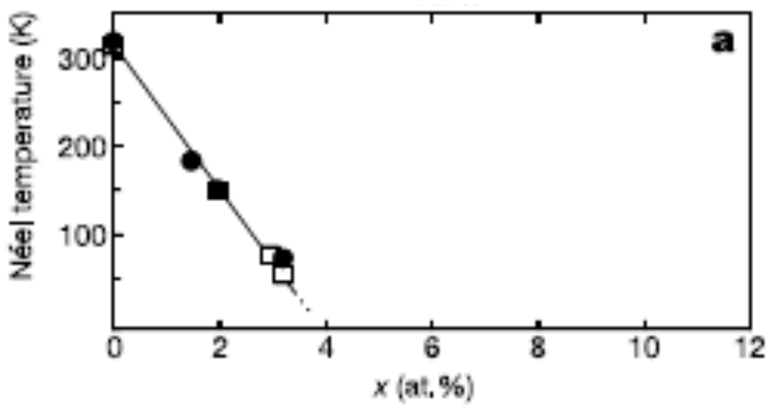
- Understanding the relationship between mesoscale phases (spin, charge, lattice) and physical properties is a grand challenge in condensed matter physics.
- X-ray photon correlation spectroscopy gives us a good way to measure their dynamics at relevant time- ***and*** length- scales.
  - ERL could enable 3D dynamic imaging of charge and spin dynamics (if we had the detectors).
  - Many condensed matter systems of interest.

Center for Nanoscale Materials

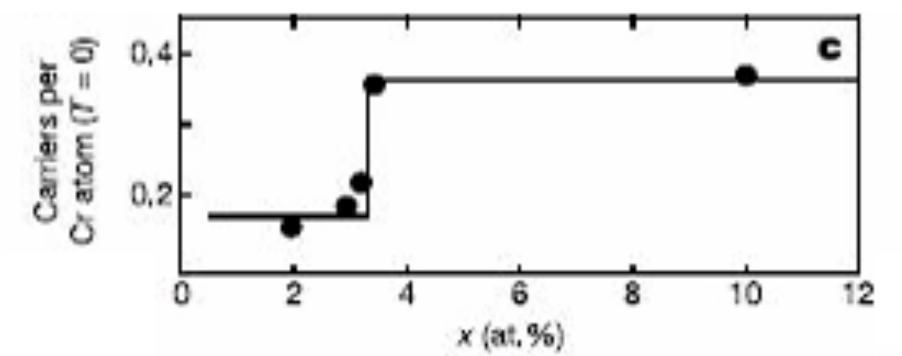
http://nano.anl.gov

Quantum Critical Behavior in $Cr_{1-x}V_x$

$T_N \rightarrow 0$, x_c=3.5%



Jump in carrier concentration



Generic phase diagram