Probing Materials' Behavior using Fast Electrons

Yimei Zhu

Dept. of Condensed Matter Physics
Brookhaven National Laboratory
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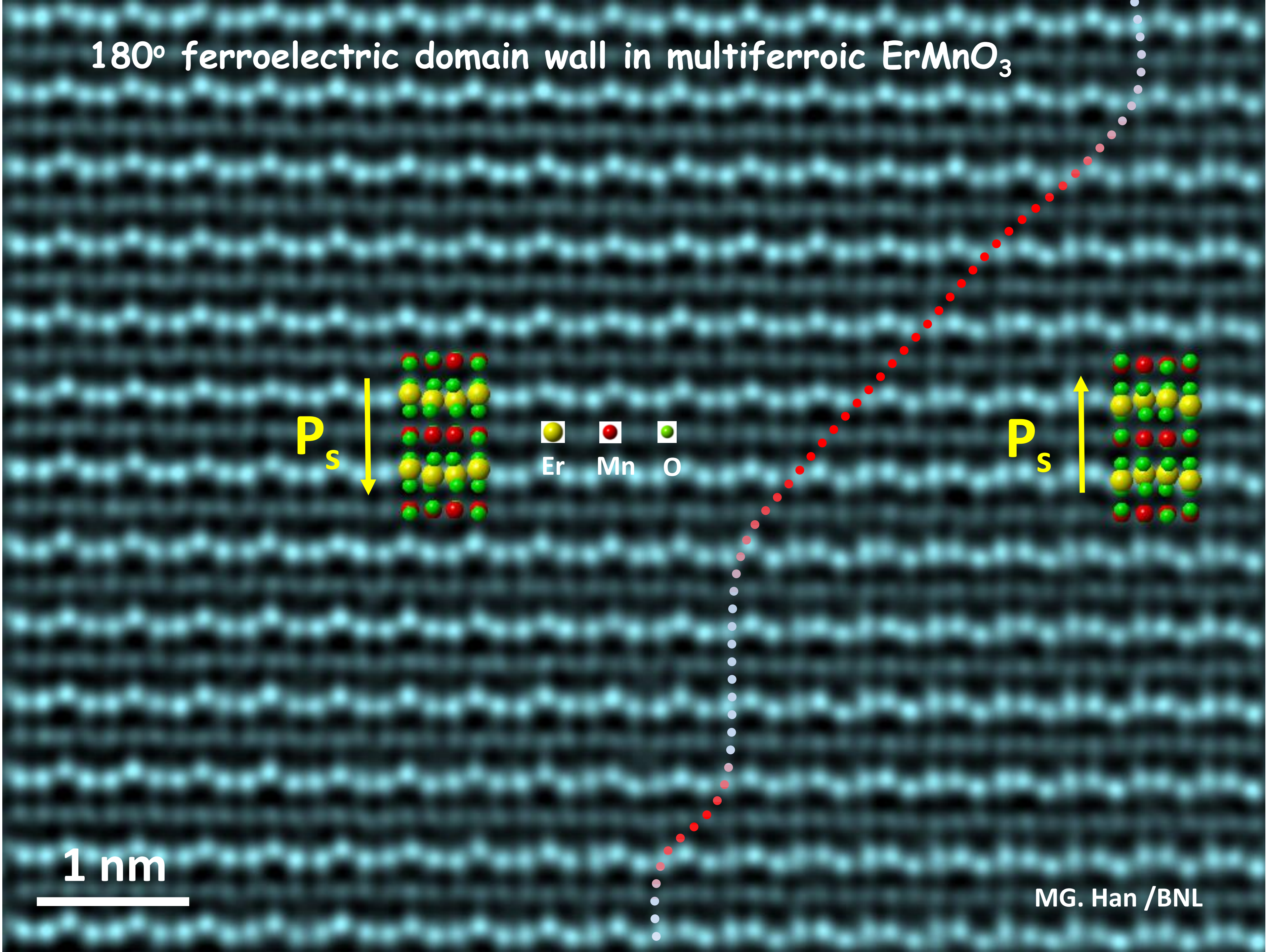
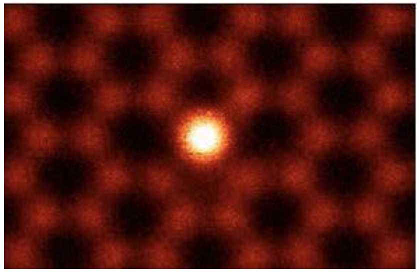
Workshop on Ultrafast
Dec 9-12, 2013, Key West, FL


180° ferroelectric domain wall in multiferroic $ErMnO_3$
P_s
Er
Mn
O
P_s
1 nm
MG. Han /BNL

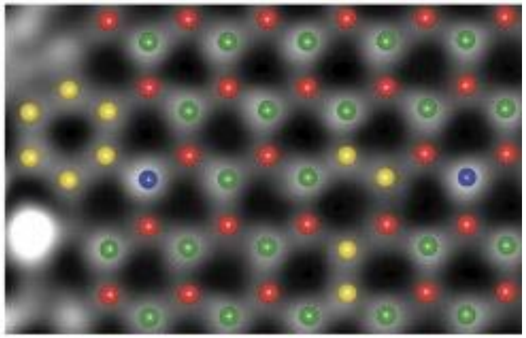
Imaging atoms

Krivanek et al, Nature (2010)

Single Si atom on graphene

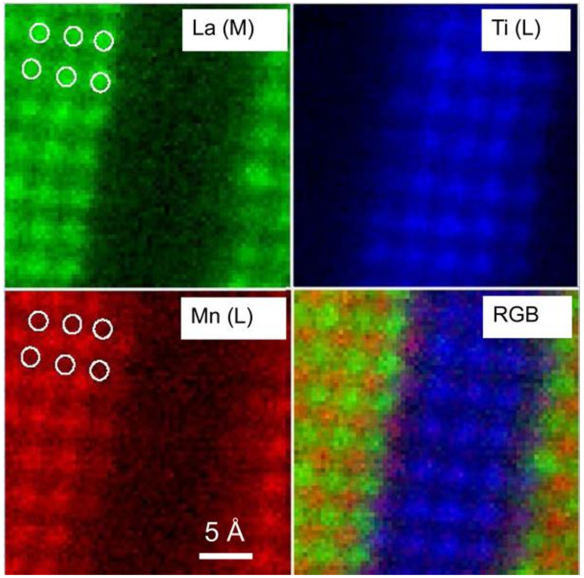


Single BN layer



red: B; yellow: C; green: N; blue: O

EELS mapping: $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3$

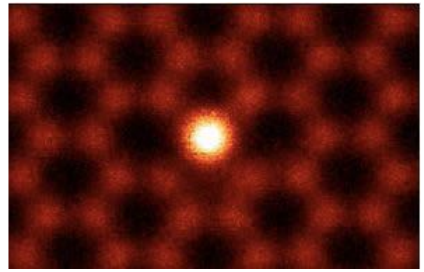


Muller et al Science 319, 1073 (2008)

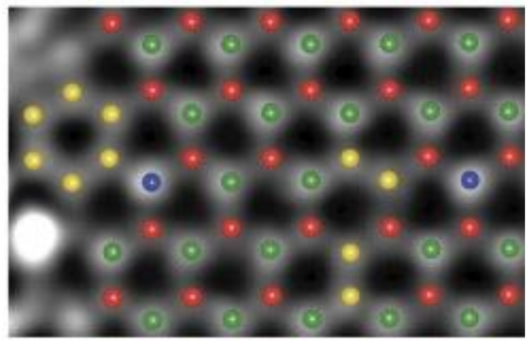
Imaging atoms

Krivanek et al, Nature (2010)

Single Si atom on graphene



Single BN layer



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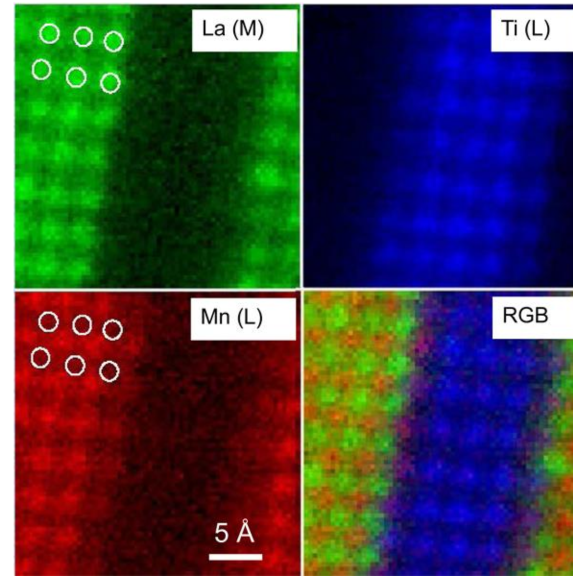


Muller et al Science 319, 1073 (2008)

Selected for a Viewpoint in *Physics*

PRL **111,** 046101 (2013) PHYSICAL REVIEW LETTERS week ending 26 JULY 2013

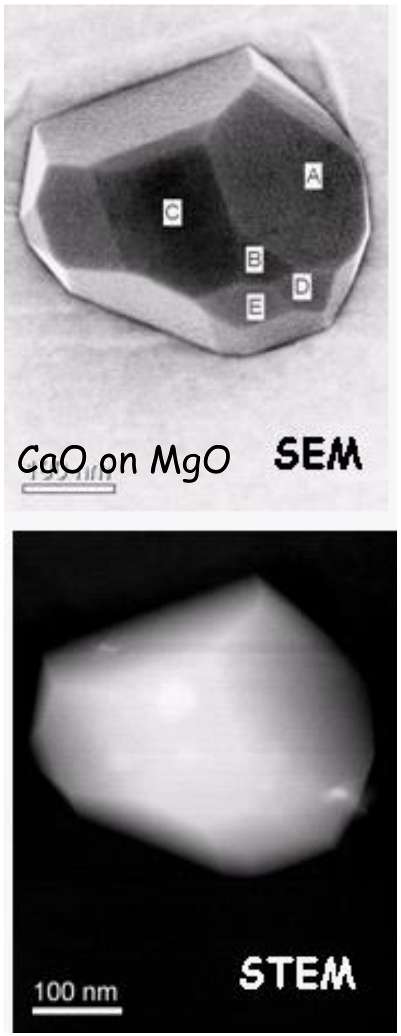
Thermal Magnetic Field Noise Limits Resolution in Transmission Electron Microscopy

Stephan Uhlemann,* Heiko Müller, Peter Hartel, Joachim Zach, and Max. Haider

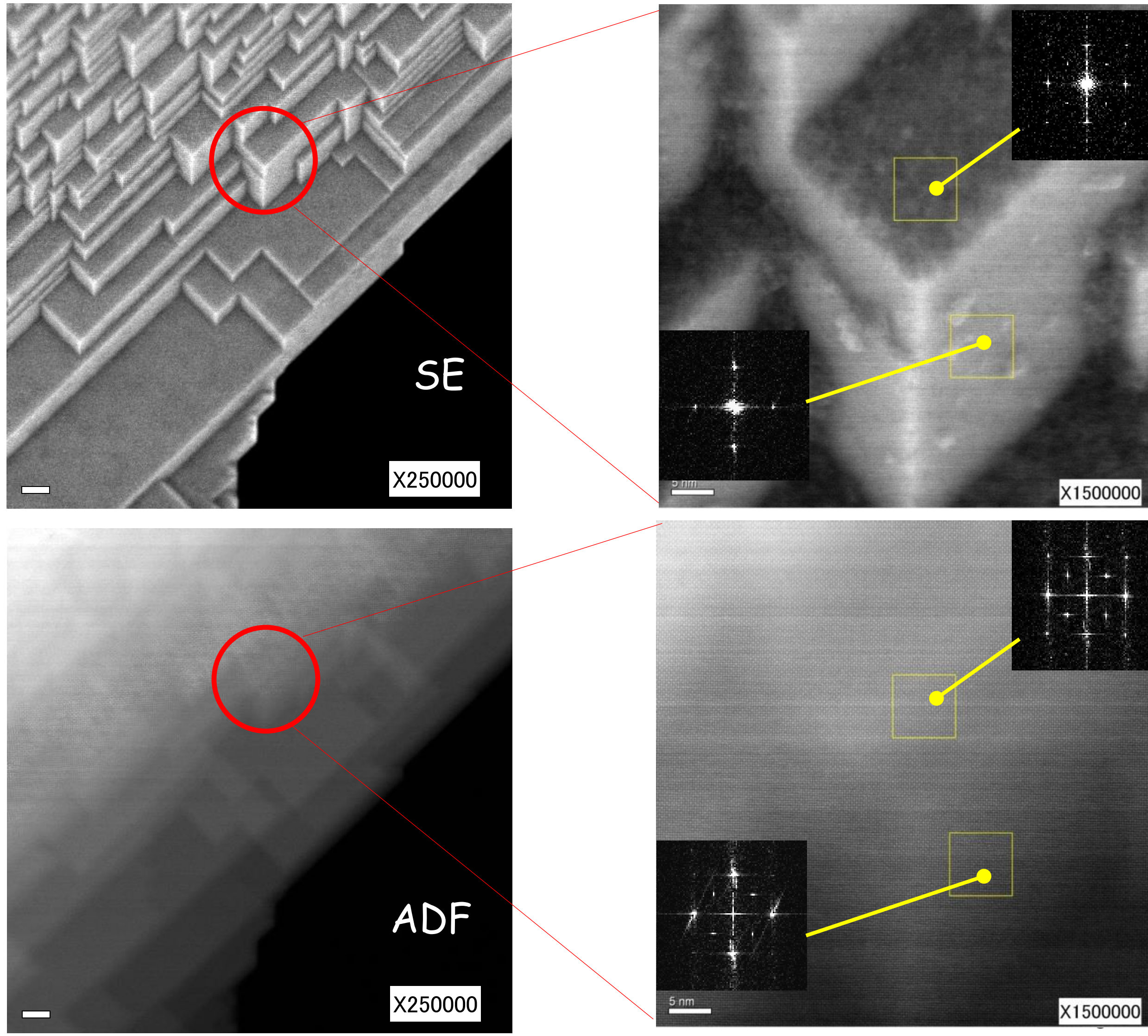
CEOS Corrected Electron Optical Systems GmbH, Englerstraße 28, 69126 Heidelberg, Germany

(Received 6 May 2013; published 22 July 2013)

Atomic surface imaging with secondary electrons

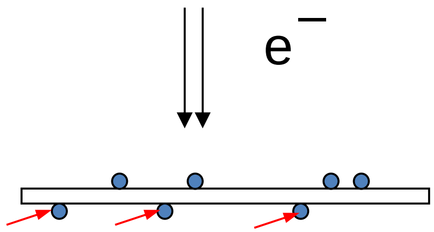


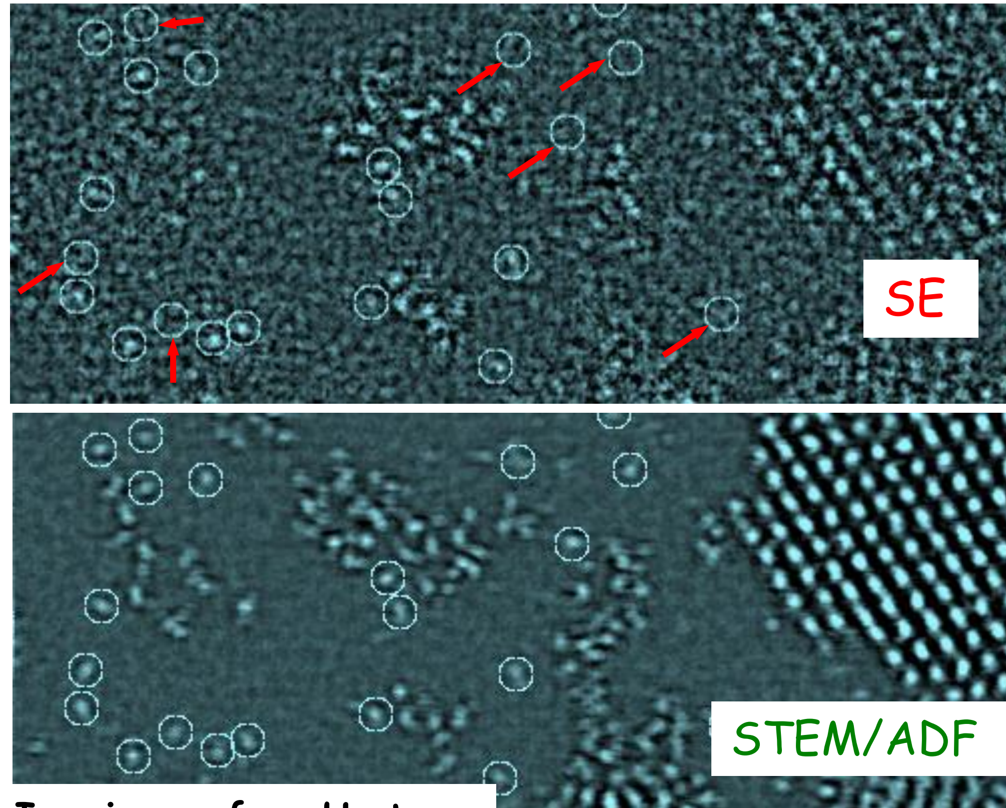
$SrTiO_3$



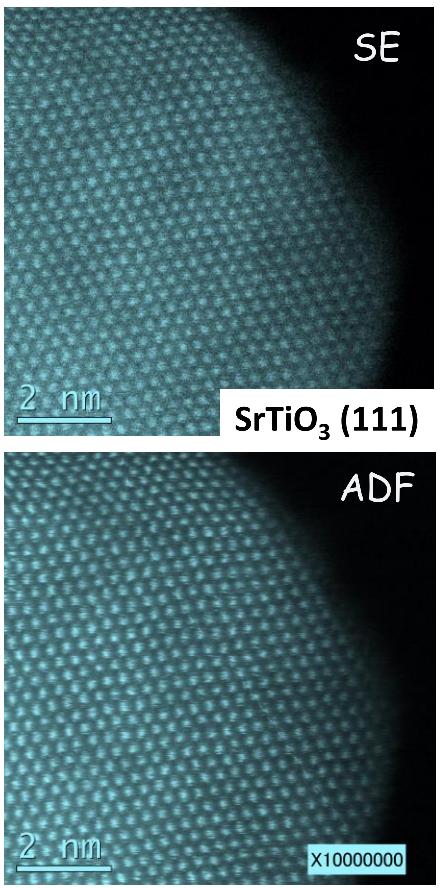
Atomic surface imaging with secondary electrons

Zhu, et al, Nature Materials, 8, 808 (2009)



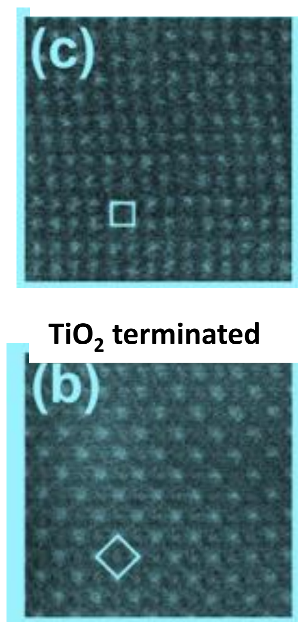


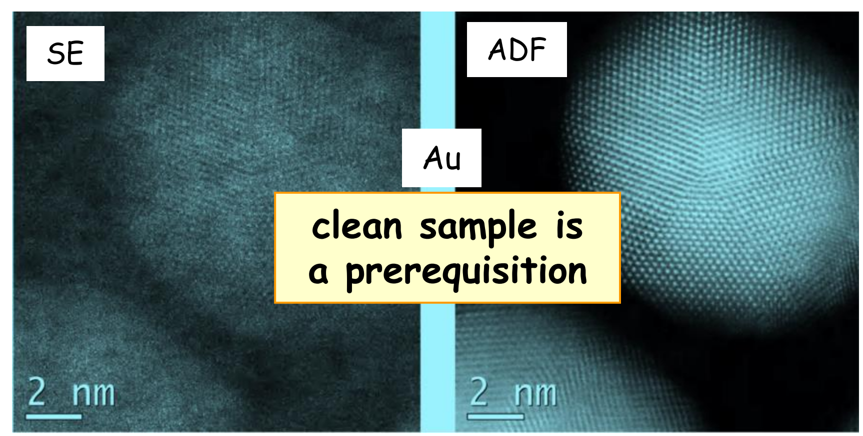
Imaging surface U atoms



$SrTiO_3$ (001)

SrO terminated





Uniqueness of electron scattering

Sensitive to valence electrons at small scattering angles

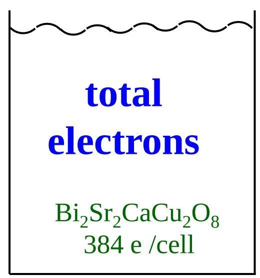
Mott formula

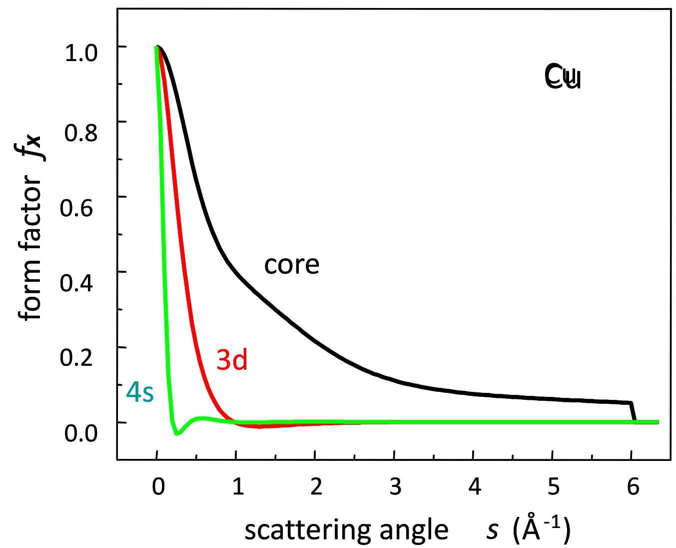
$$f^{e} = \frac{2\pi m_o e^2}{h^2 \varepsilon_0}\left(\frac{\lambda}{\sin\theta}\right)^2 (Z - f_x)$$

Electrons interact with electrostatic potential

X-rays interact with electron cloud

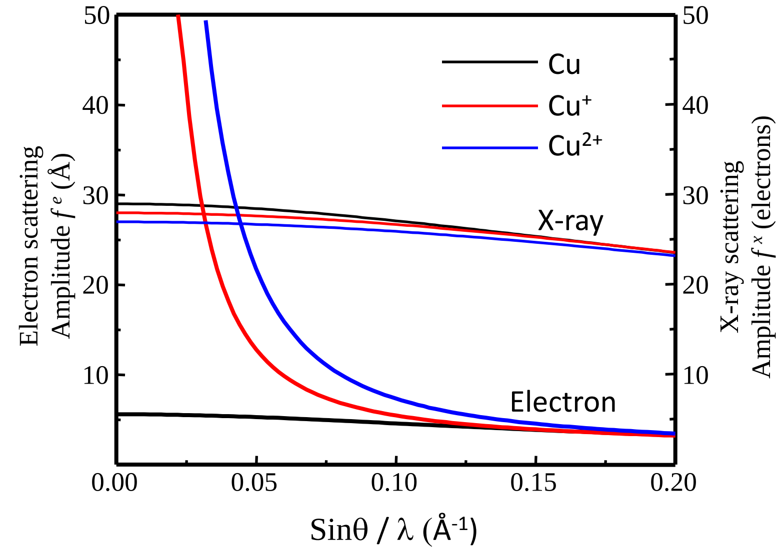
valence electrons





difficulties for x-ray suitability criterion

$$\alpha = \frac{V}{\sum n^2} >> 0.1 \sim 5$$



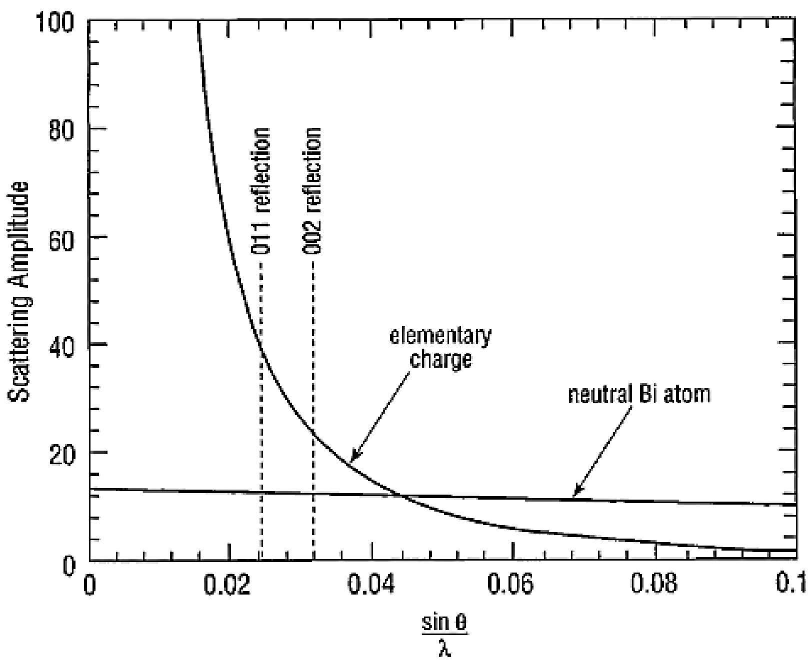
P. Coppens, (1997), "X-ray charge densities & chemical bonding"

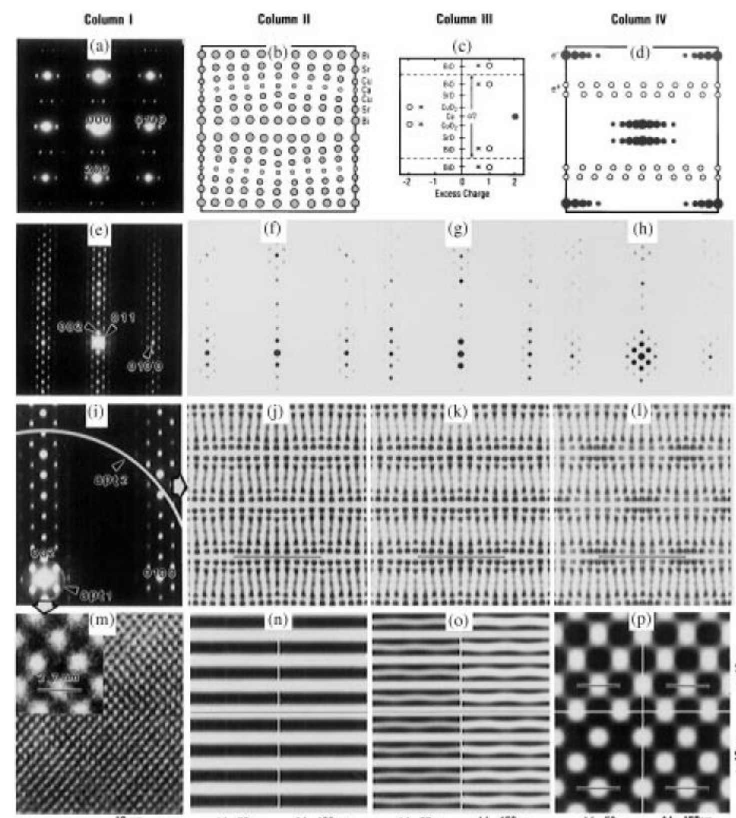
Direct Imaging of Charge Modulation

Yimei Zhu
Department of Applied Science, Brookhaven National Laboratory, Upton, New York 11973

J. Tafto
Department of Physics, University of Oslo, P.O. Box 1048, 0316 Blindern, Oslo 3, Norway
(Received 7 September 1995)

$Ba_2Sr_2CaCu_2O_8$

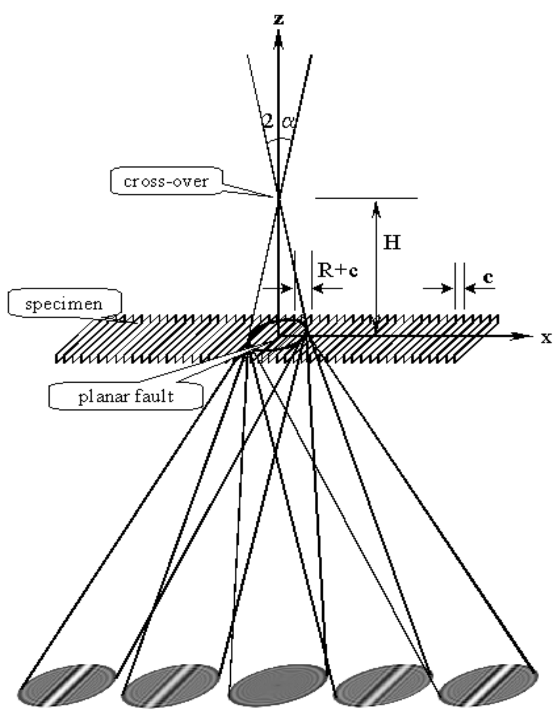




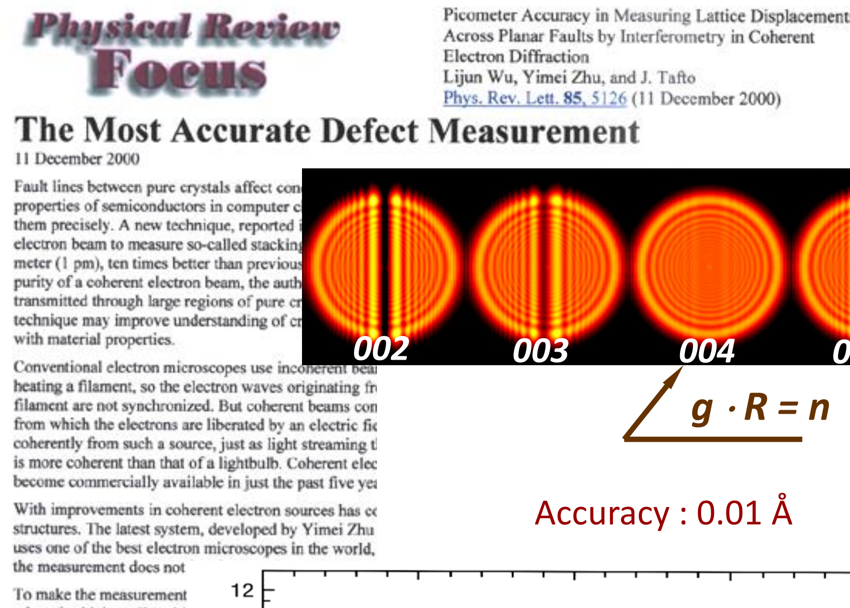
VOLUME 85, NUMBER 24 PHYSICAL REVIEW LETTERS 11 DECEMBER 2000

Picometer Accuracy in Measuring Lattice Displacements Across Planar Faults by Interferometry in Coherent Electron Diffraction

Lijun Wu, Yimei Zhu,* and J. Tafto†
Materials Science Division, Brookhaven National Laboratory, Upton, New York 11973
(Received 12 June 2000)



$$\varphi_g = \sum_{m,n} F_g \exp 2\pi i \left(\frac{\sqrt{r^2 + H^2}}{\lambda} - \vec{r} \cdot \vec{g} \right)$$


Physical Review Focus

Picometer Accuracy in Measuring Lattice Displacements Across Planar Faults by Interferometry in Coherent Electron Diffraction
Lijun Wu, Yimei Zhu, and J. Tafto
Phys. Rev. Lett. 85, 5126 (11 December 2000)

The Most Accurate Defect Measurement

11 December 2000

Fault lines between pure crystals affect con
properties of semiconductors in computer c
them precisely. A new technique, reported i
electron beam to measure so-called stacking
meter (1 pm), ten times better than previous
purity of a coherent electron beam, the auth
transmitted through large regions of pure cr
technique may improve understanding of cr
with material properties.

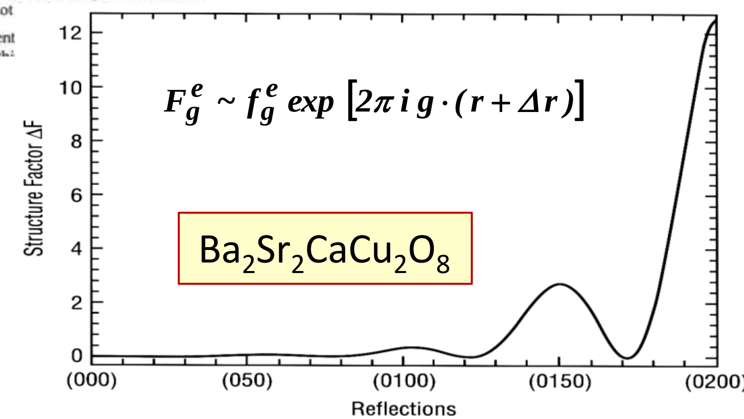
Conventional electron microscopes use incoherent bea
heating a filament, so the electron waves originating fr
filament are not synchronized. But coherent beams con
from which the electrons are liberated by an electric fi
coherently from such a source, just as light streaming t
is more coherent than that of a lightbulb. Coherent elec
become commercially available in just the past five yea

With improvements in coherent electron sources has c
structures. The latest system, developed by Yimei Zhu
uses one of the best electron microscopes in the world,
the measurement does not

To make the measurement

$g \cdot R = n$

Accuracy : 0.01 Å



Imaging electrons, spins, electromagnetic potentials

Valence electron distribution

charge density ρ :

$$\rho(r) = \frac{1}{\Omega} \sum_g F_g^x \exp(2\pi i\, g \cdot r)$$

structure factor

$$\rho = -\nabla^2 V \varepsilon_0$$

Electrostatic and magnetic potential distribution

electrostatic potential V
magnetic potential B :

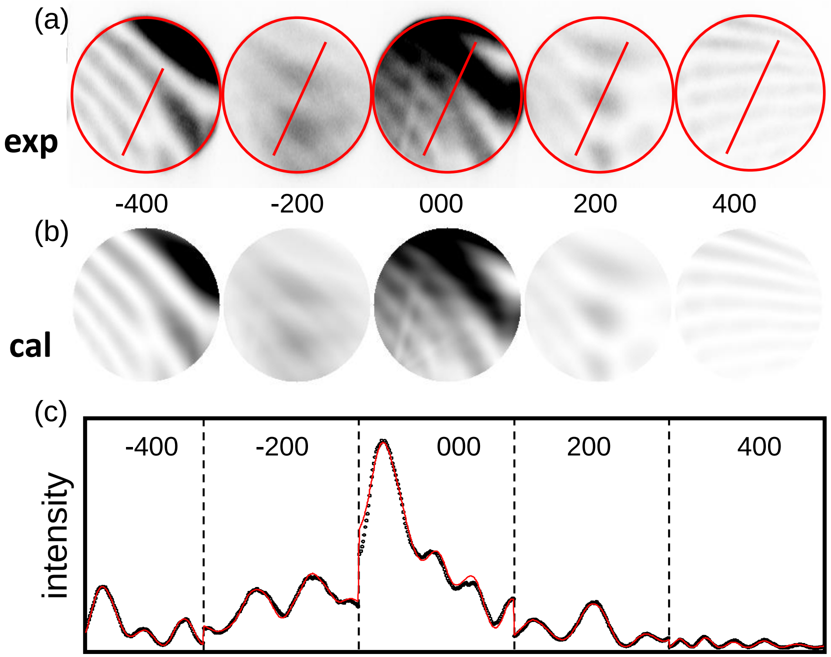
$$\phi(r) = C_E \int V(r,z)dz - \int \frac{e}{\hbar} \vec{A}(r,z) \cdot d\vec{r}$$

Aharonov-Bohm phase shift
of the wave function

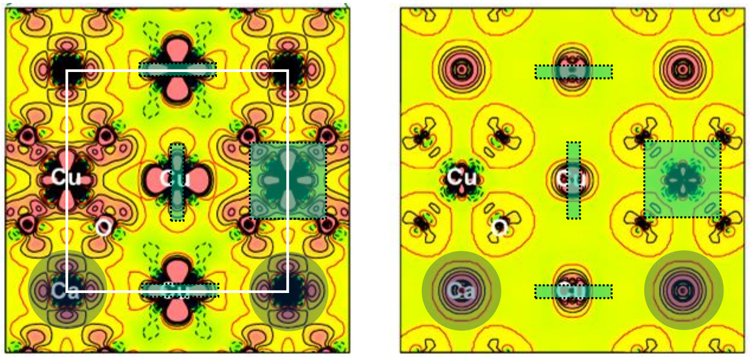
Mapping valence electron distribution with quantitative diffraction

$$\rho(r) = \frac{1}{\Omega}\sum_g F_g^x \exp(2\pi i g \cdot r)$$

$F - F_{IAM}$ → FFT

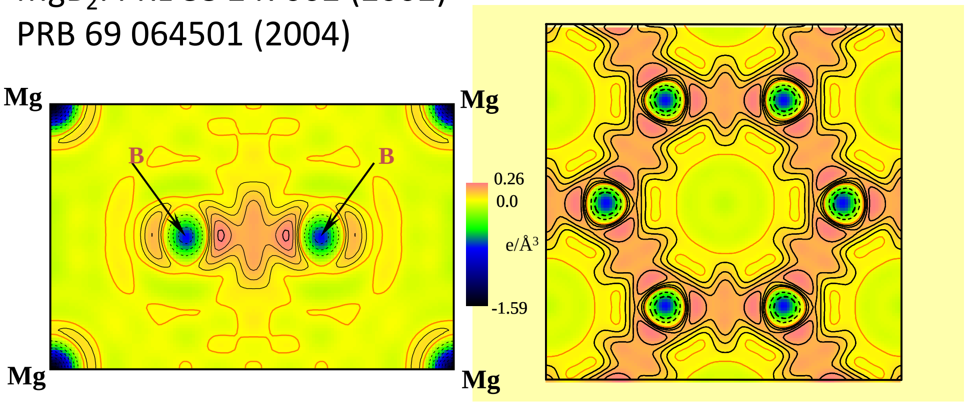


$CaCu_3Ti_4O_{12}$: PRL 99 037602 (2007)



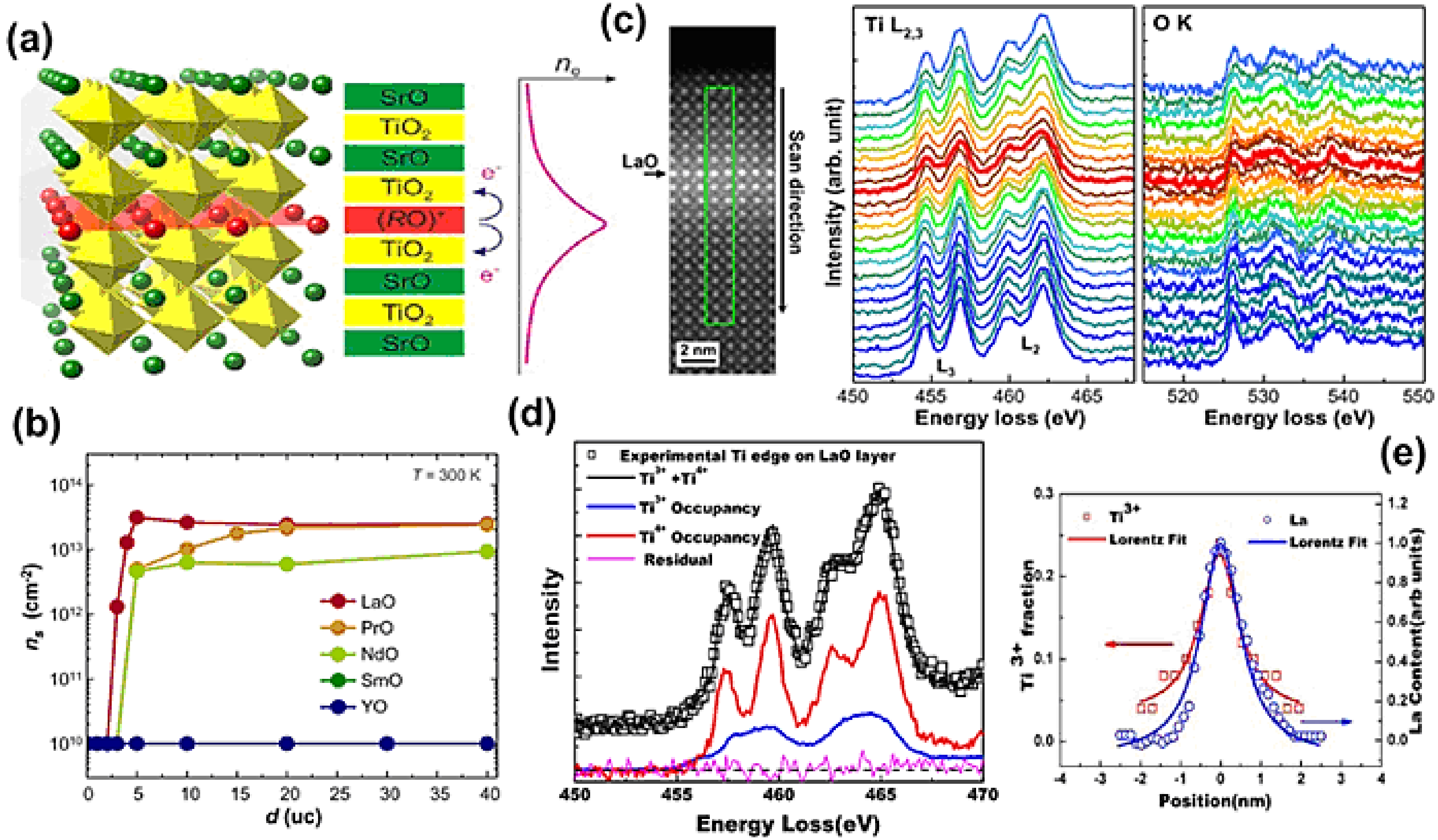
Experiment : ED + X-ray, 90K   DFT, w/o disorder

MgB_2: PRL 88 247002 (2002)
PRB 69 064501 (2004)



Probing bonding states and charge transfer with EELS

2-D Electron Gas: $SrTiO_3/RO/SrTiO_3$ (R=La, Pr, Nd, Sm, Y)



In collaboration with Prof. C.B Eom

Jang et al Science, 311, 886 (2011)

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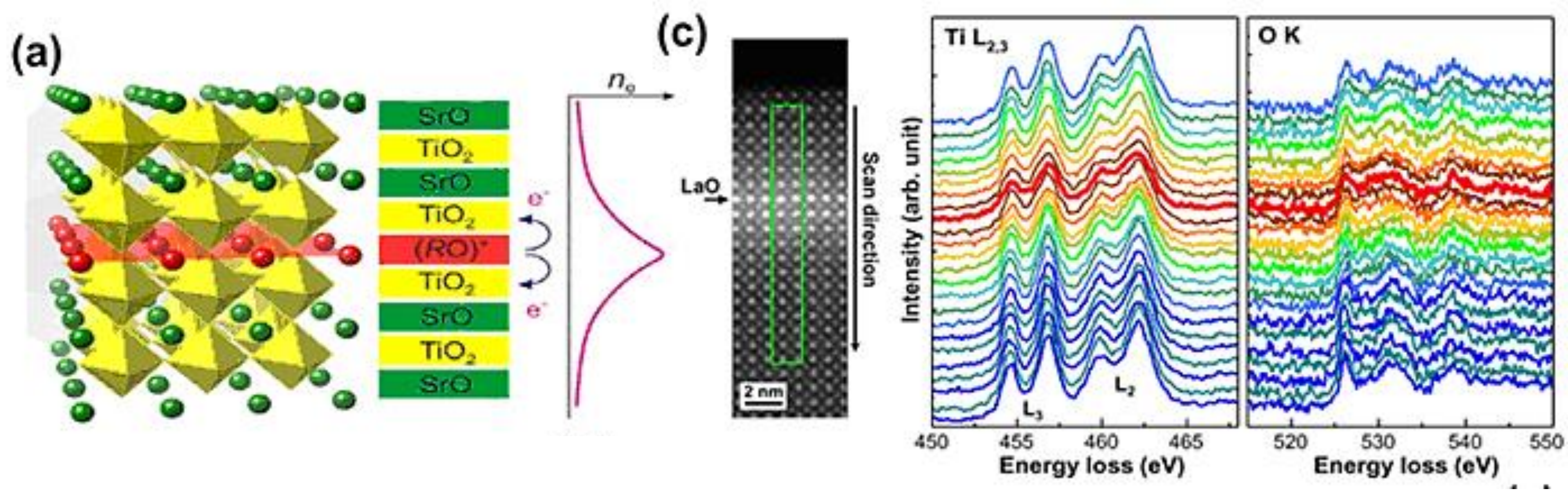
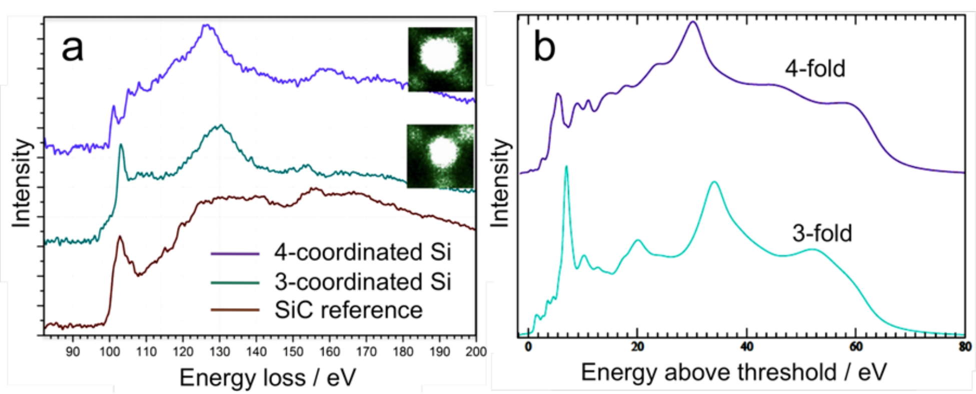
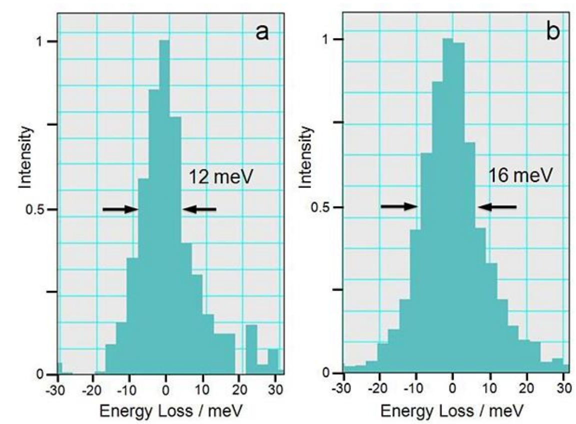


individual Si substitutional atoms on SiC



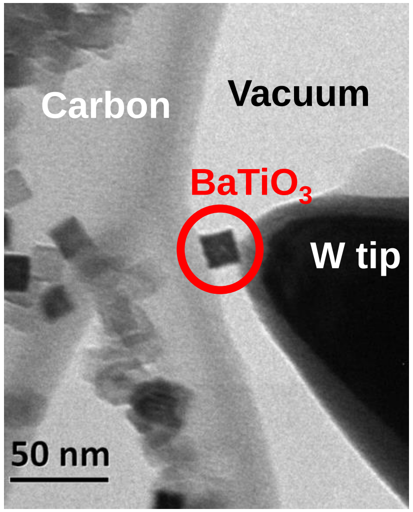


Ondrej Krivanek

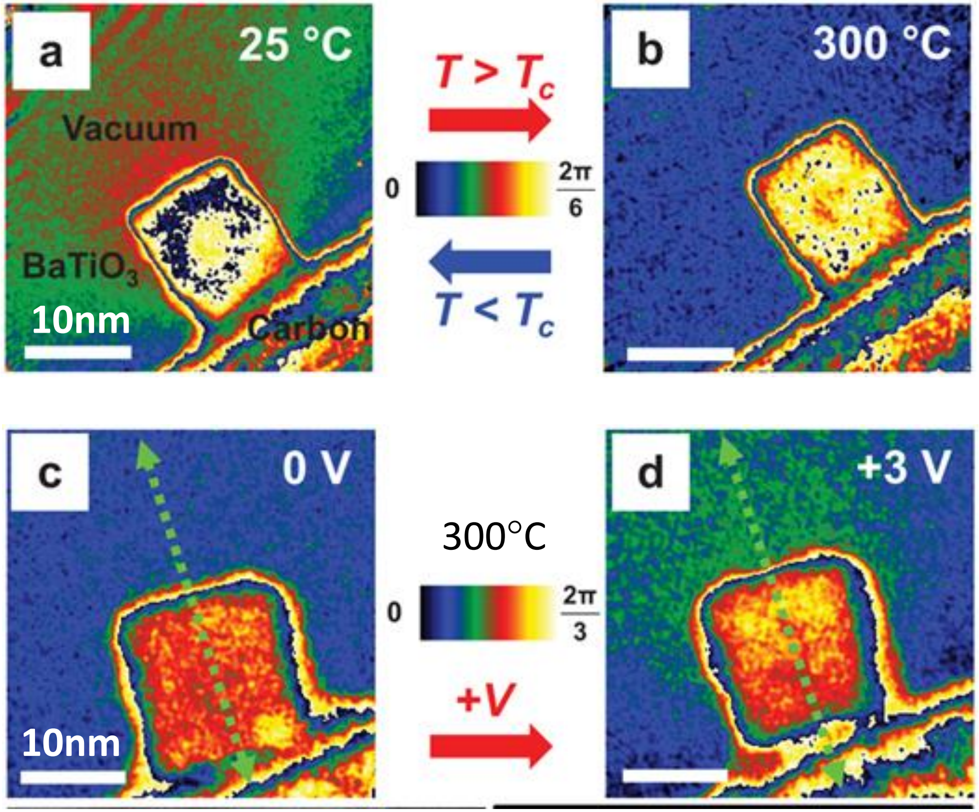
Direct imaging of ferroelectric order using holography

- RT ferroelectric order can be down to 10-15nm, below 10nm superparaelectric.

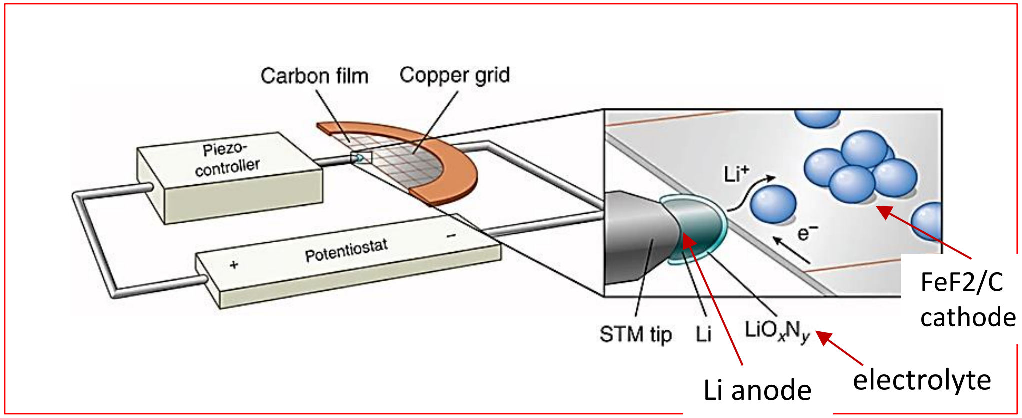
$BaTiO_3$
Curie temperature 130°C

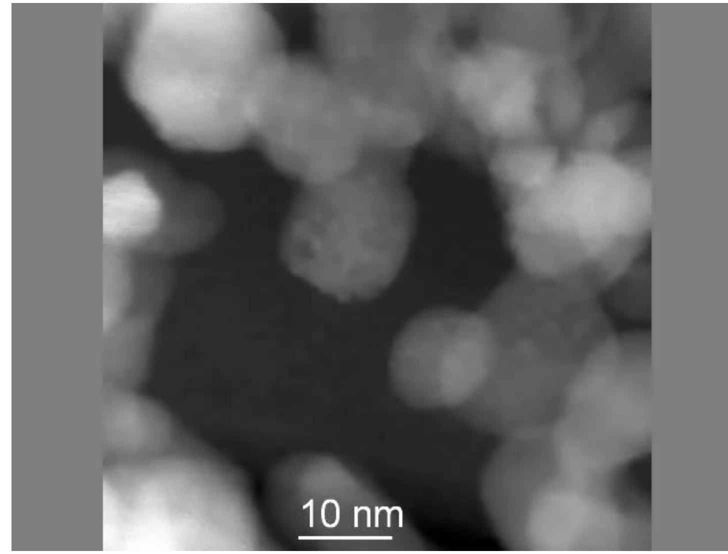


Polking, Han, et al,
Nat. Mat., **11** 700 (2012)

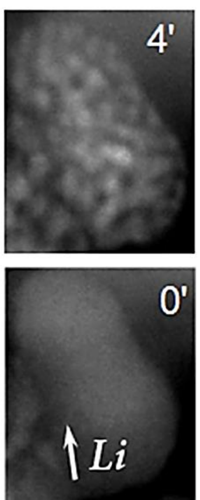


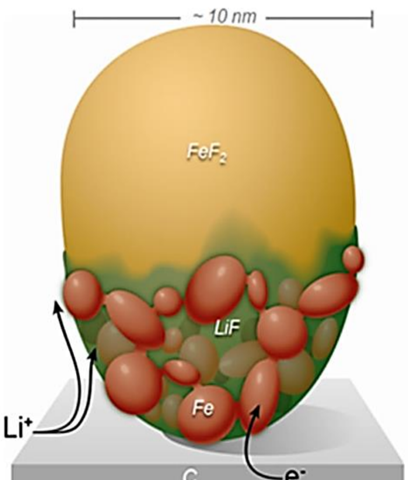
Direct observation of the Lithiation process in Li-ion-batteries

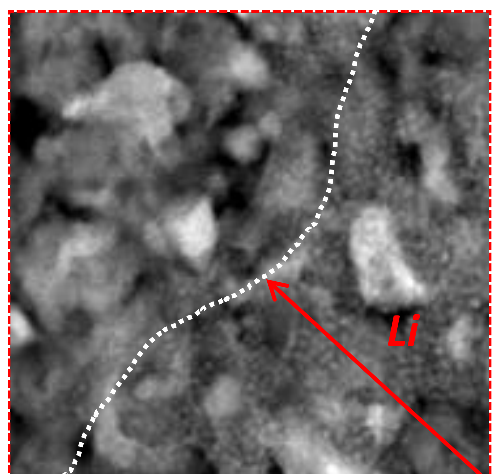


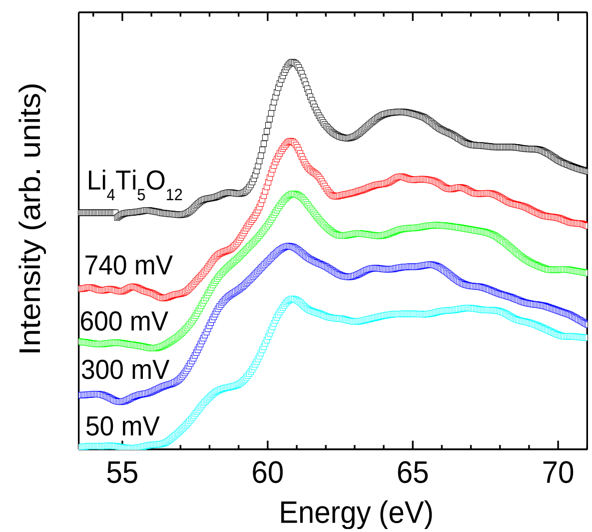


$FeF_2 + 2Li^+ + 2e^- \rightarrow 2LiF + Fe$





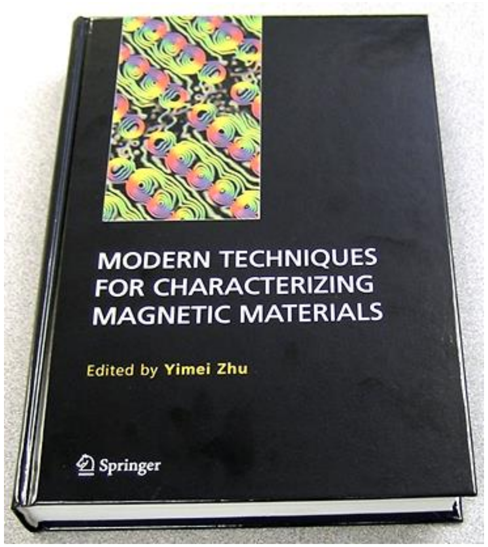




Wang et al Nature Comm. 3, 1201 (2012)

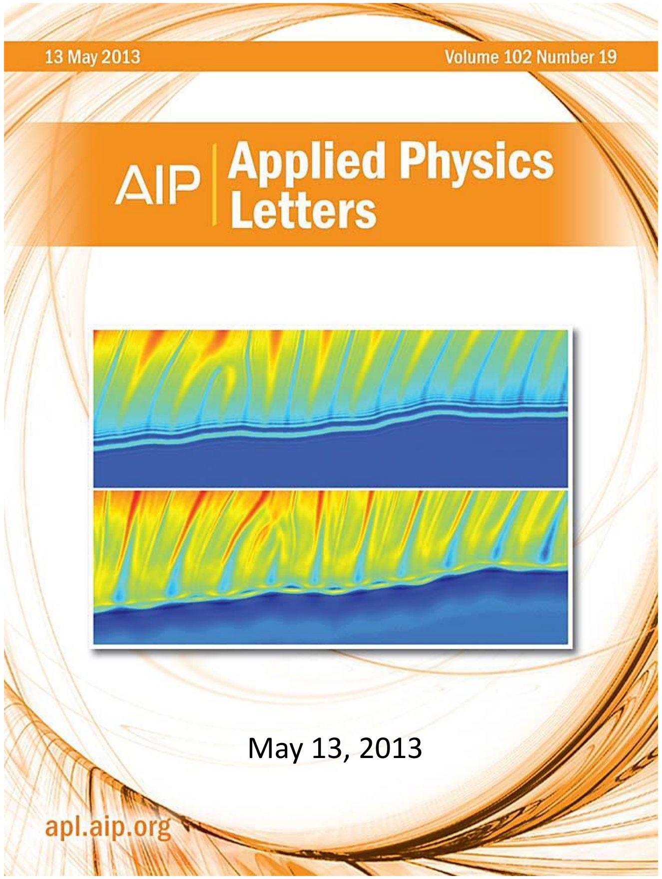
Can we image individual spins ?

For BCC Fe NP, the max phase shift is ~50urad, 10^{-3} of $2\pi/100$



	EH	LM	MFM	SP-STM	n	MOKE	X-rays
Strength	Mapping fields, measuring moments	Quick and easy, large field of view	Easy, cheap	Single-spin sensitivity, manipulation	Resolving unknown spin structures, spin wave Q(w)	Super fast dynamics	Super fast dynamics
Weakness	Requires practice	Qualitative	Perturbs sample	Surface-only	Bulk/crystal-only, requires a neutron source	Surface-only	Requires a synchrotron
Time resolution	100 ms	10 ms	1 s	1 s	n/a	1 fs	1 fs
Spatial resolution	3 nm	10 nm	50 nm	0.1 nm	n/a	100 nm	20 nm
Sensitivity	$\pi/100$ rad phase shift	1 μrad deflection	100 pN force	<1 μ_B moment	<1 μ_B moment	20 μrad rotation	?

Phase plate for magnetic imaging

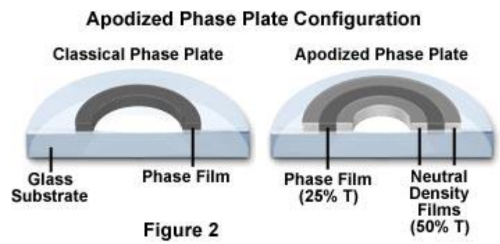


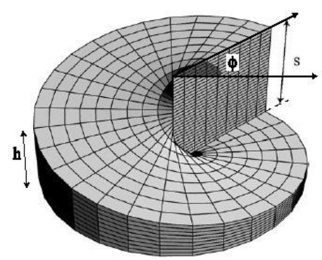
May 13, 2013

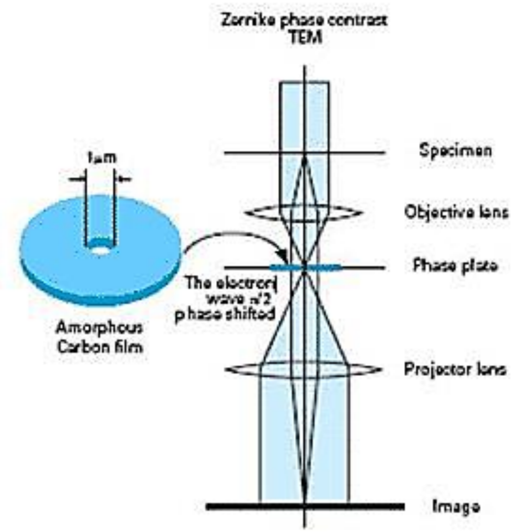
Pollard, Malac, Belleggia, Kawasaki, Zhu
APL 102, 192401 (2013)

Hole free Phase Plate

Zernike phase plate:
convert phase contrast to
amplitude contrast



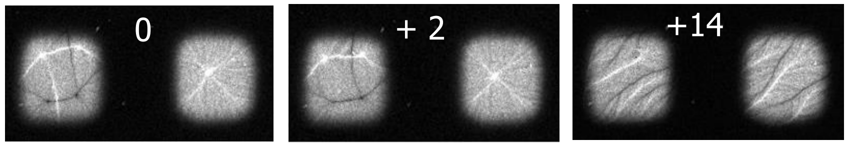




www.Jeolusa.com

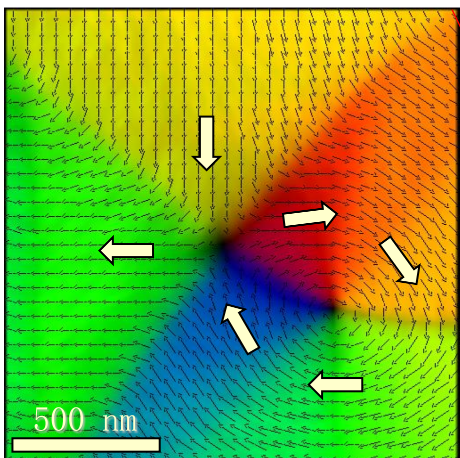
Imaging magnetic moments

in-situ

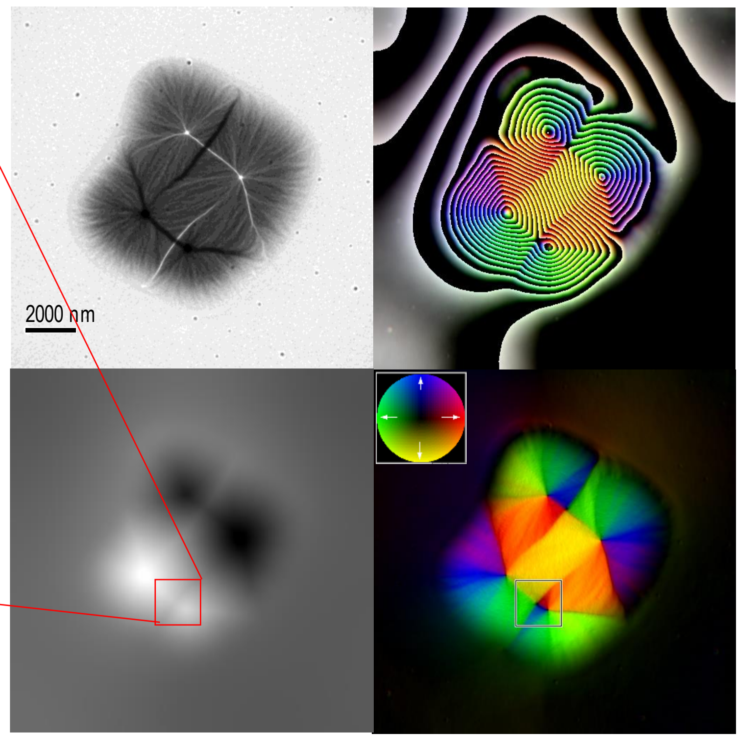


Lorentz contrast

vortex & antivortex

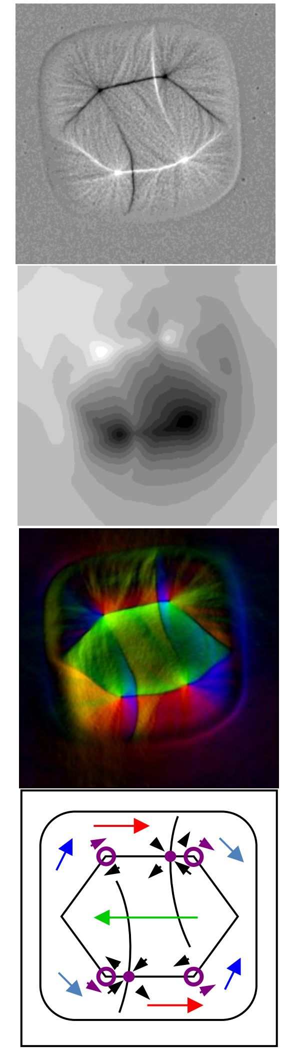


magnetization
vector map



phase image

V.Volkov and Y. Zhu
PRL. 91 043904(2003)

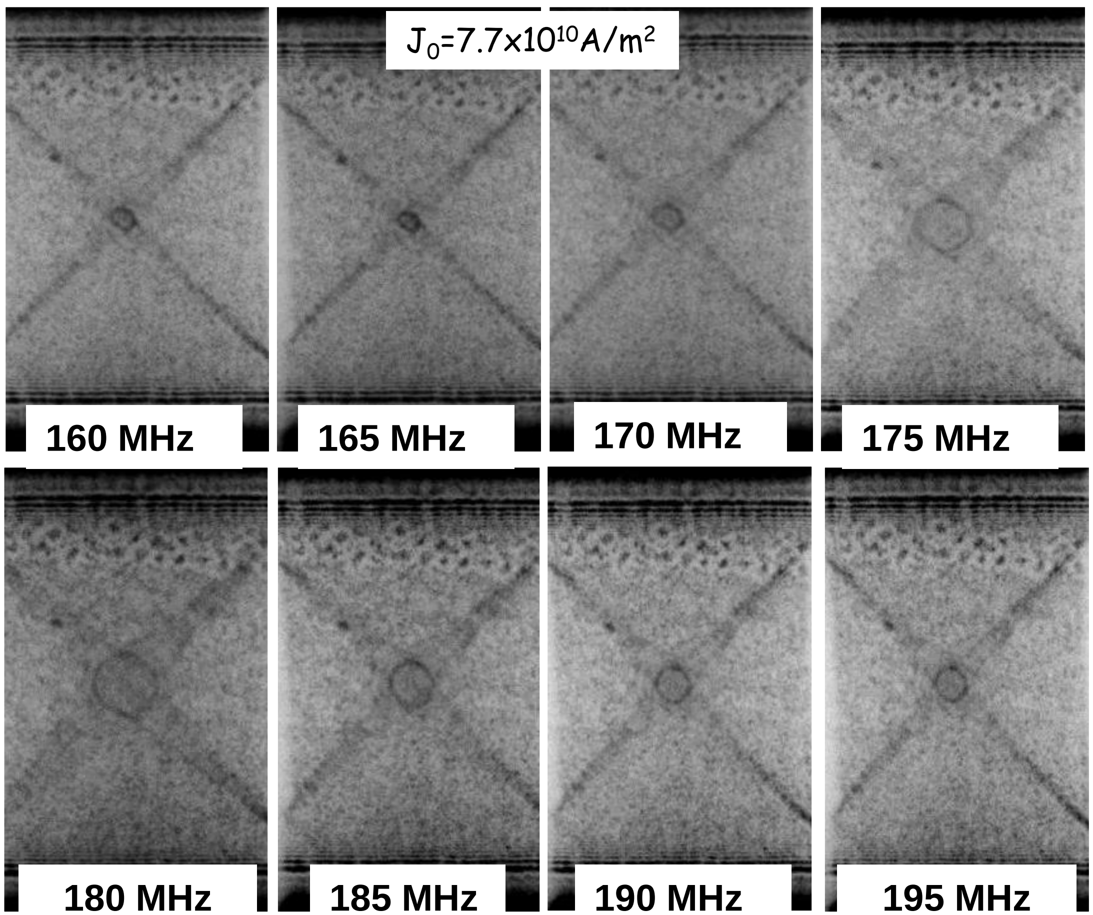


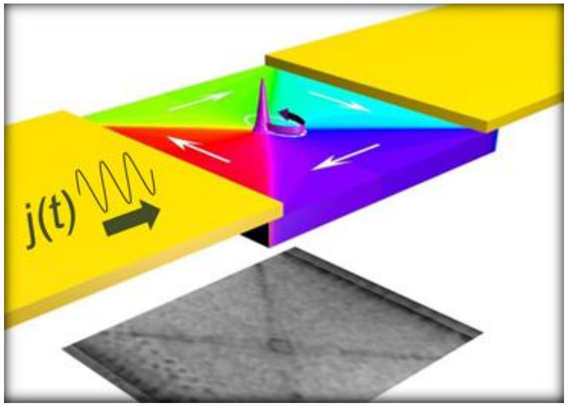
image

t B map

structure

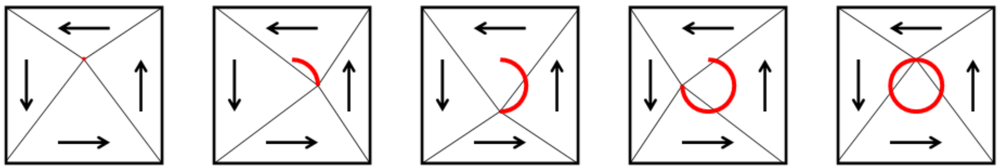
Imaging vortex-precession orbit via resonance excitation

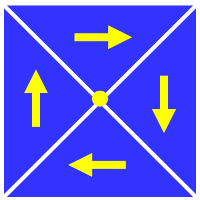


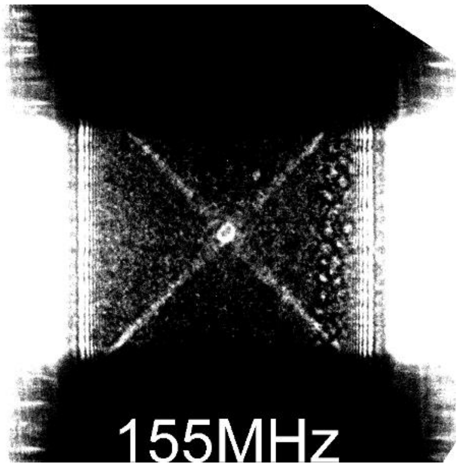


Vortex dynamics in nanomagnet

Direct observations of vortex precession orbit



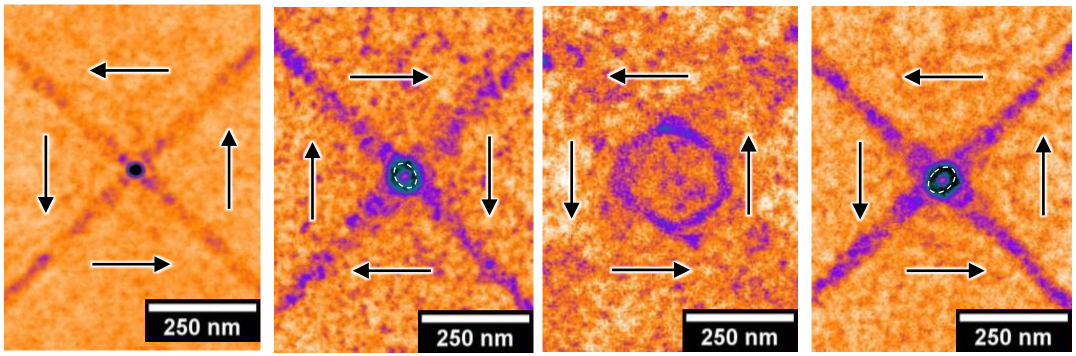




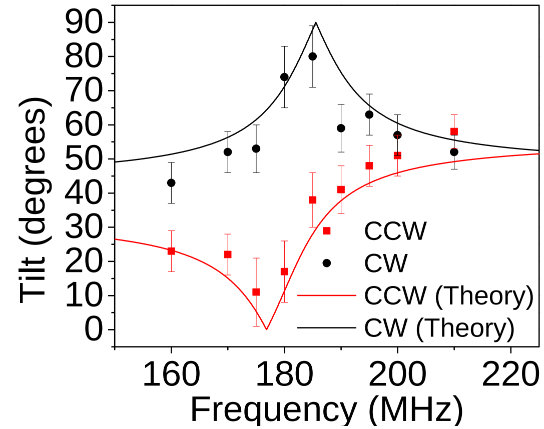
2um Py square

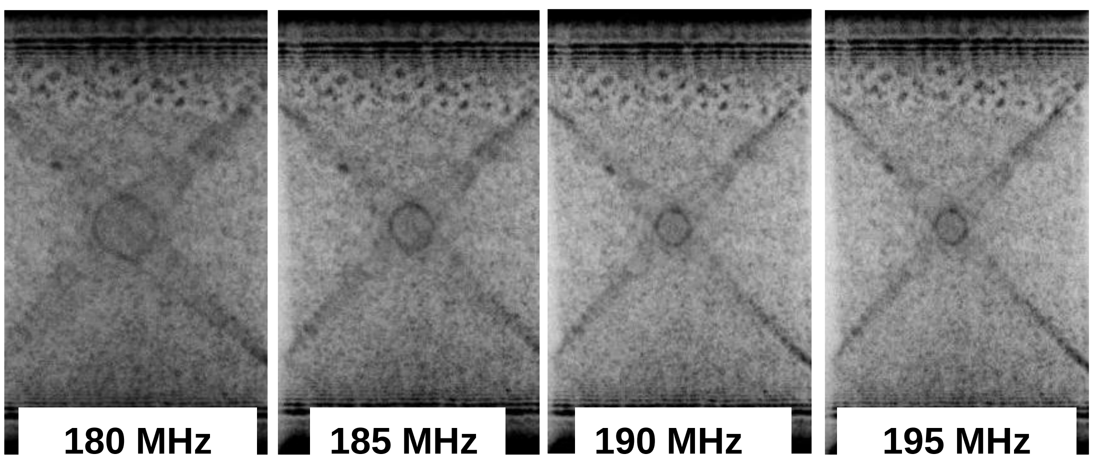
Pollard et al Nature Comm. **3** 1028 (2012)

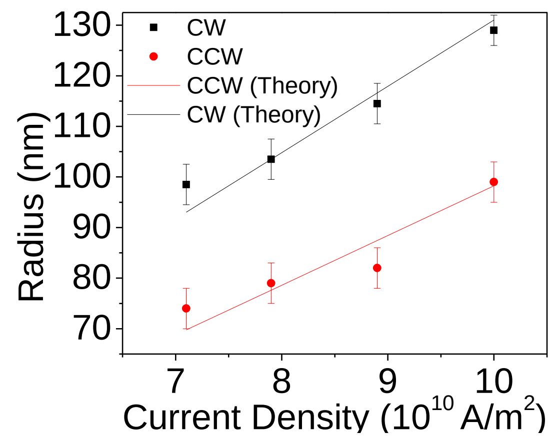
Imaging vortex-precession orbit via resonance excitation



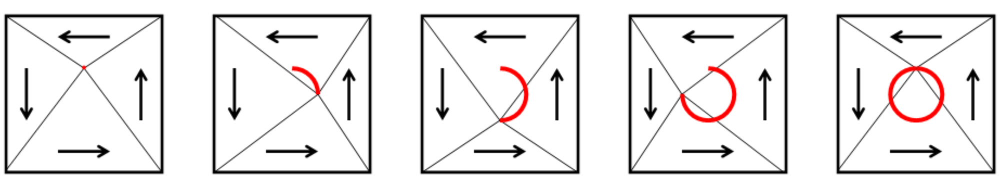
No excitation, CCW | 200 MHz, CW | 180 MHz, CCW | 200 MHz, CCW

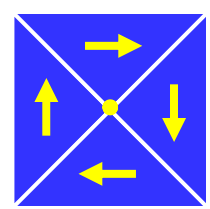






Direct observations of vortex precession orbit





Pollard et al Nature Comm. **3** 1028 (2012)

Our goals: understanding strongly correlated materials

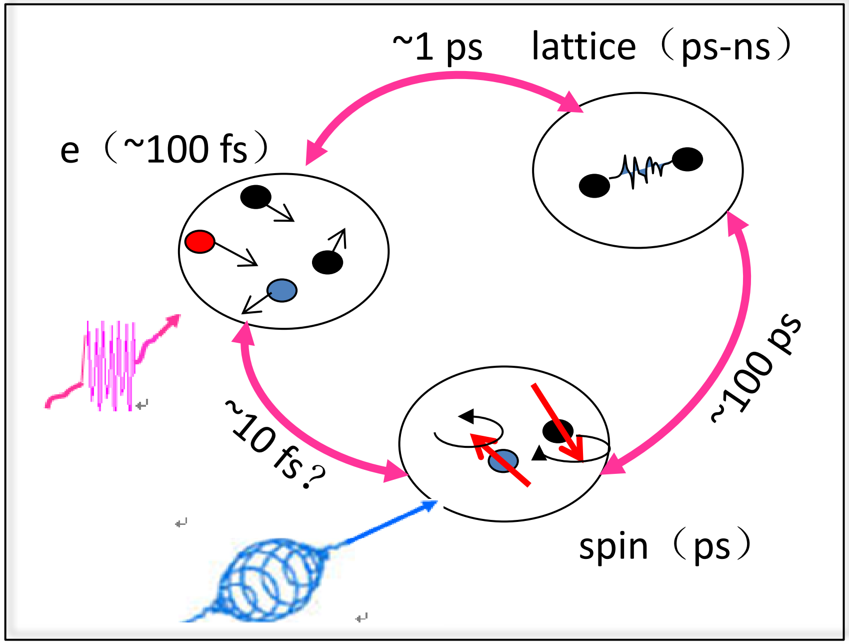
Challenges:

Coupling electronic-lattice system

→charge, orbital and spin order

Strong interplay between charge, spins, orbital and lattice

→ complex phase diagrams, exotic material properties



One solution:

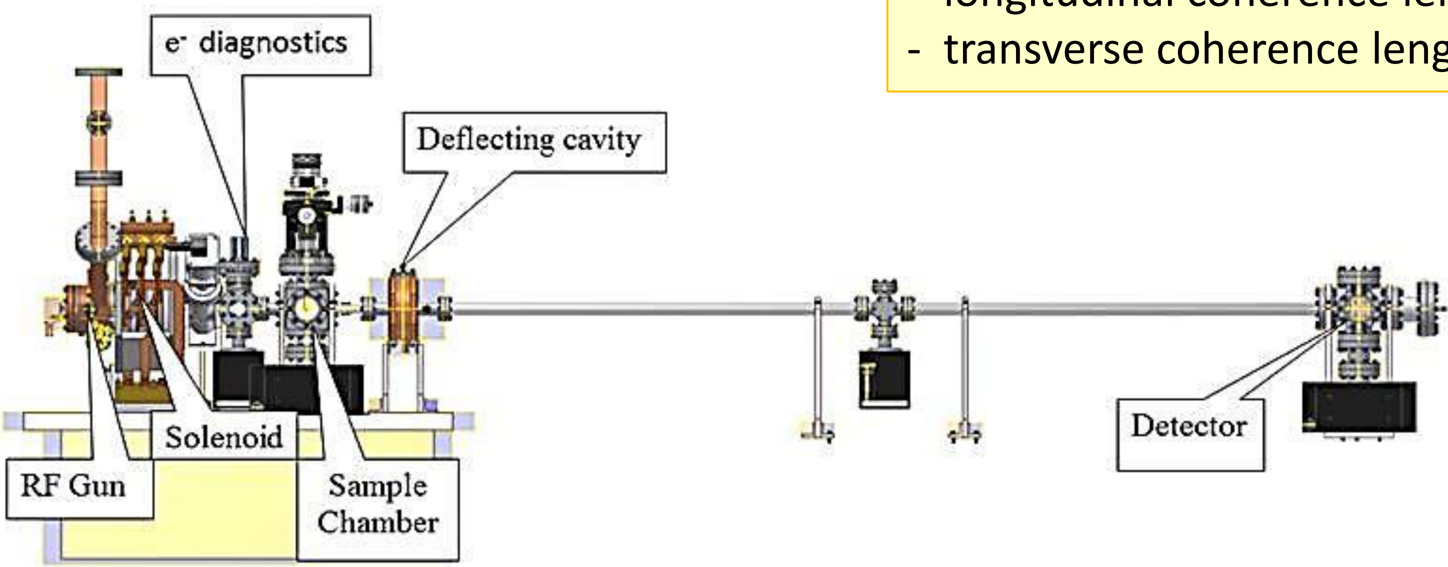
Decouple the subsystems in the time domain and then observe the dynamics of subsystems separately.

<u>Ultrafast ED</u>: better time resolution & simultaneously observe diverse degrees of freedom.

Ultrafast electron diffraction at BNL

Currently optimized at
2.8MeV with 120fs resolution

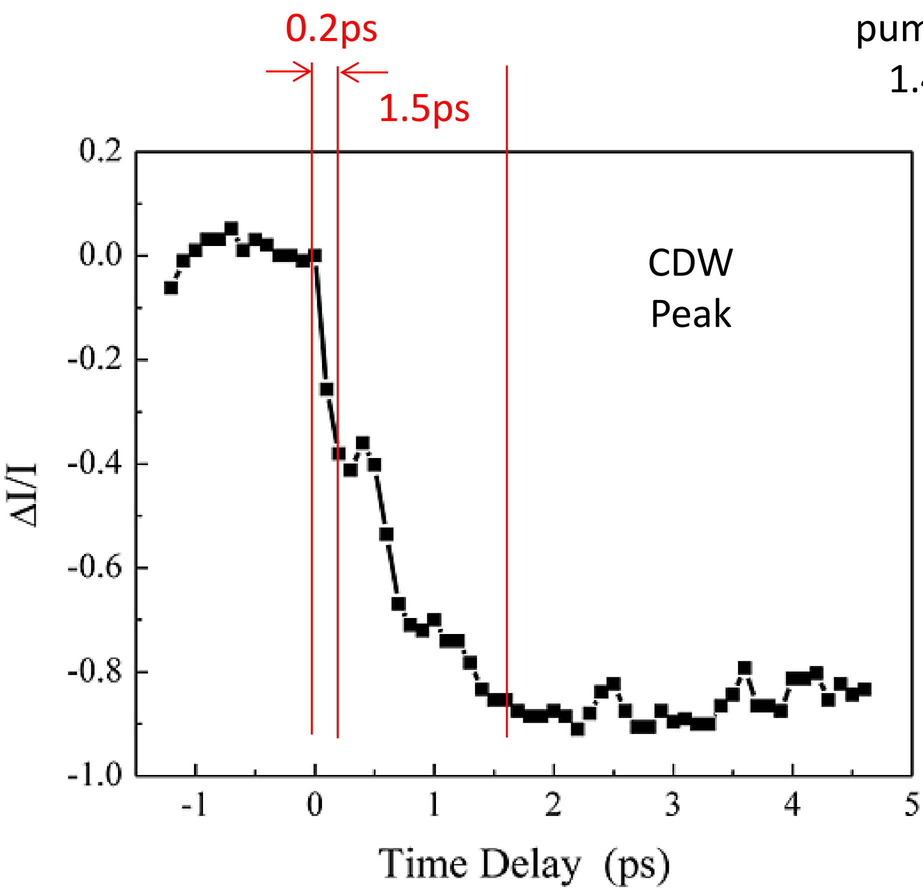
- 2-4 MeV electron energy
- 100 fs pulse, Hz repetition rate
- 10^6 electrons in a single pulse
- energy spread <0.01%
- 30urad divergence
- beam size on detector 200um
- Synchronization of RF & laser <50fs
- cryogenic capability
- longitudinal coherence length 1-2nm
- transverse coherence length ~10nm

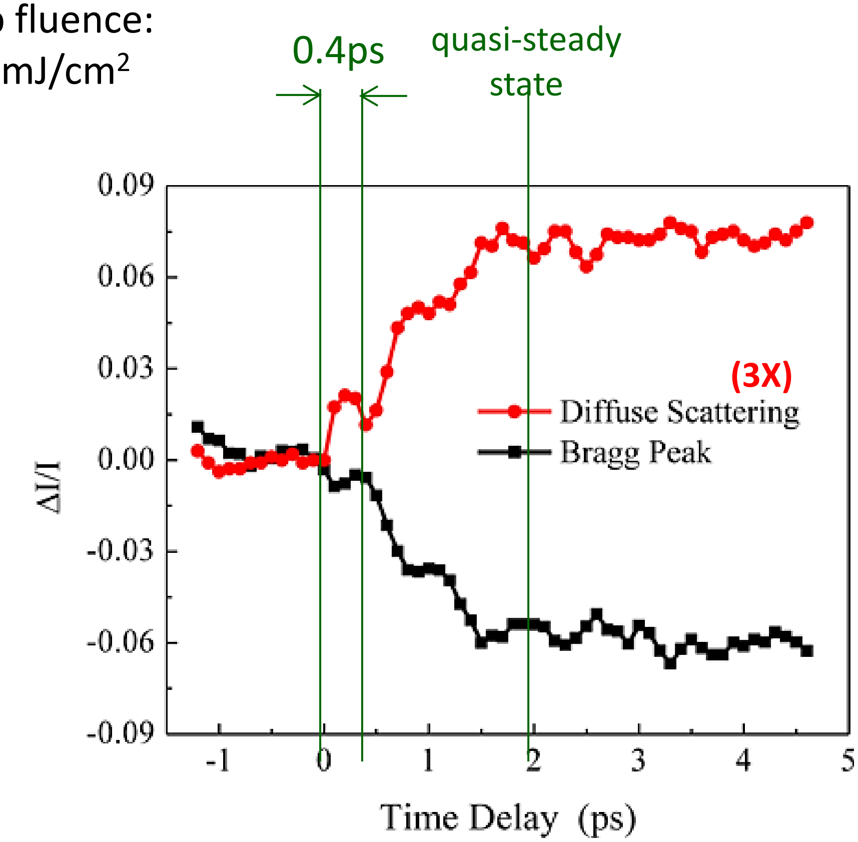


XJ Wang's talk Monday morning

2H-$TaSe_2$

pump fluence: 1.4 mJ/cm^2



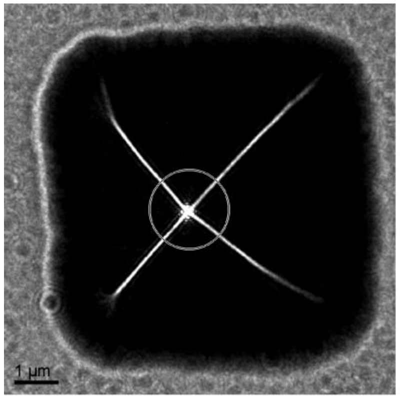


SP intensity reduced to ~ 0, without obvious recovery in the following 50 ps

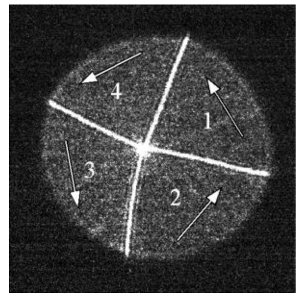
Coherent phonon: 2.5THz (~ 400fs)

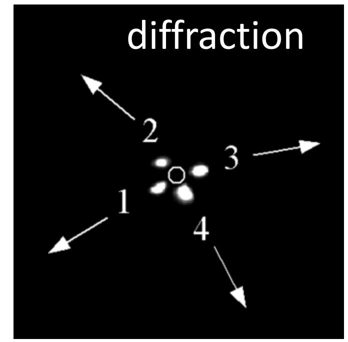
P. Zhu et al, APL 2013

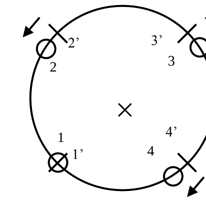
Ultrafast electron diffraction for spin dynamics ?

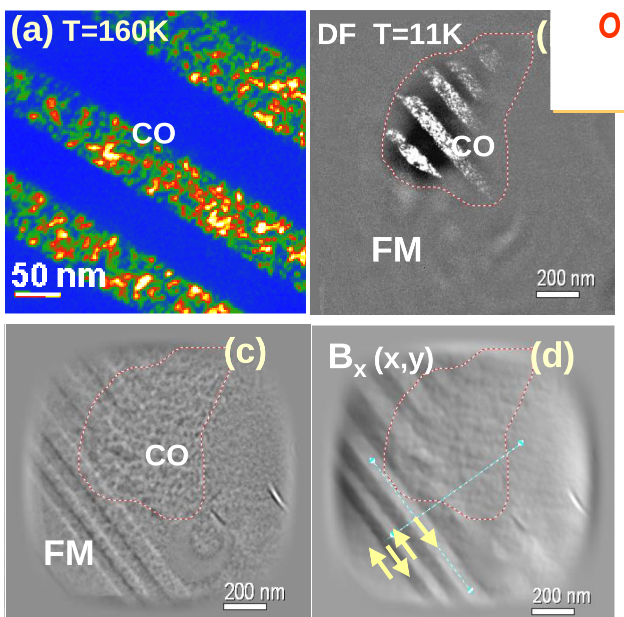


Split of transmitted and diffraction spots
due to magnetic domains in permalloy



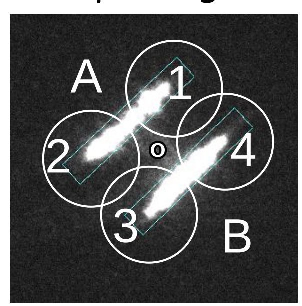


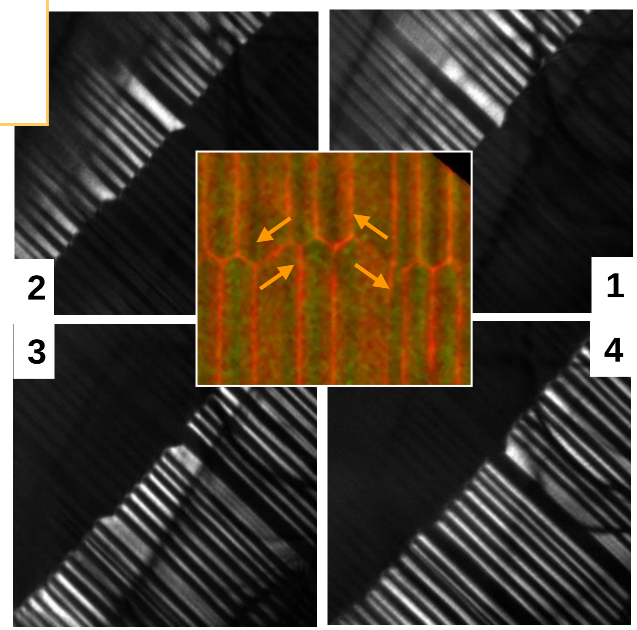




orbital-spin ordering
$La_{1/4}Pr_{3/8}Ca_{3/8}MnO_3$

(000) beam
splitting





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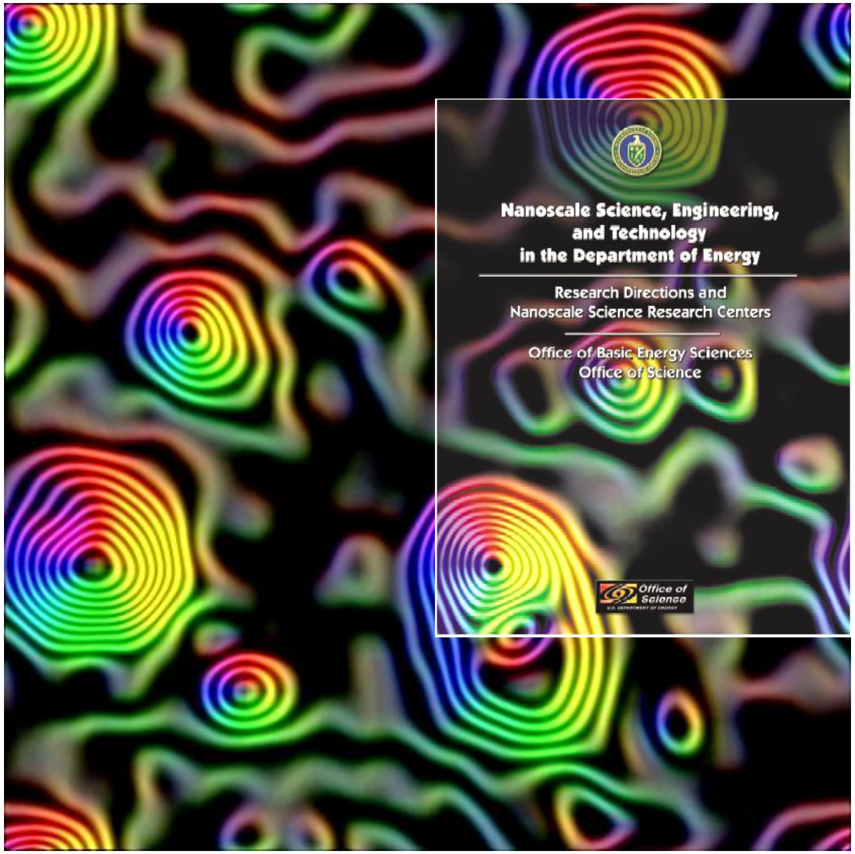

Thank you !