Resonant X-ray scattering at the nanoscale

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NSLS-II & CFN 2018 Users' Meeting
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Outline

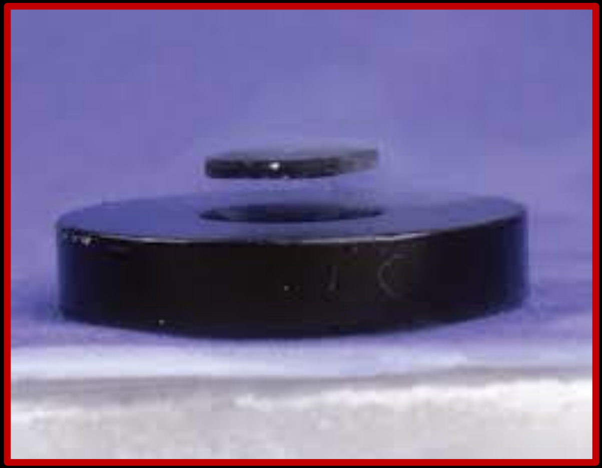
- Strongly-correlated quantum solids
- Competing orders and nanoscale granularity
- Resonant Soft X-ray Scattering: in a nutshell
- Soft X-ray nanodiffraction at beamline CSX
- Scale-invariant spin textures in nickelates

Strongly-interacting quantum matter

Fundamental building blocks

Interactions U

Many-body phenomena



Strongly-interacting quantum matter

Property

Superconductivity

Magnetoelectrics

Colossal magneto-resistance

Magnetic skyrmions

Ferroelectricity

Mott insulators

Application

Quantum computing

Data storage

Spintronics

Medical imaging

Energy transport

Quantum sensing

Energy storage

Technology

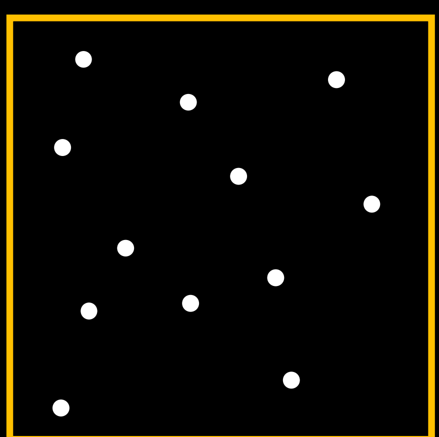
Strongly-interacting quantum matter

$\ll 1$ U/W $\gg 1$

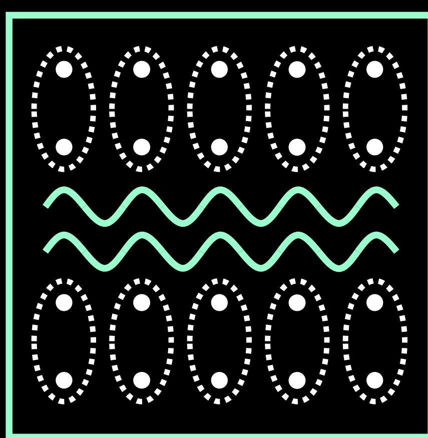
Fermi Gas

Emergent phenomena

Superconductivity
Mott insulator
Kondo insulator
Nematic fluid
Charge/spin-density-wave



Symmetry Breaking



Incoherent superposition
of single-particle w.f.

Many-body quantum order
(macroscale phase coherence)

Strongly-interacting quantum matter

Reduce kinetic energy (*W*)

High magnetic fields

Lattice engineering

Doping

Confining potentials (e.g., optical traps)

Increase interactions (*U*)

Chemistry

Electrostatics (dielectric screening)

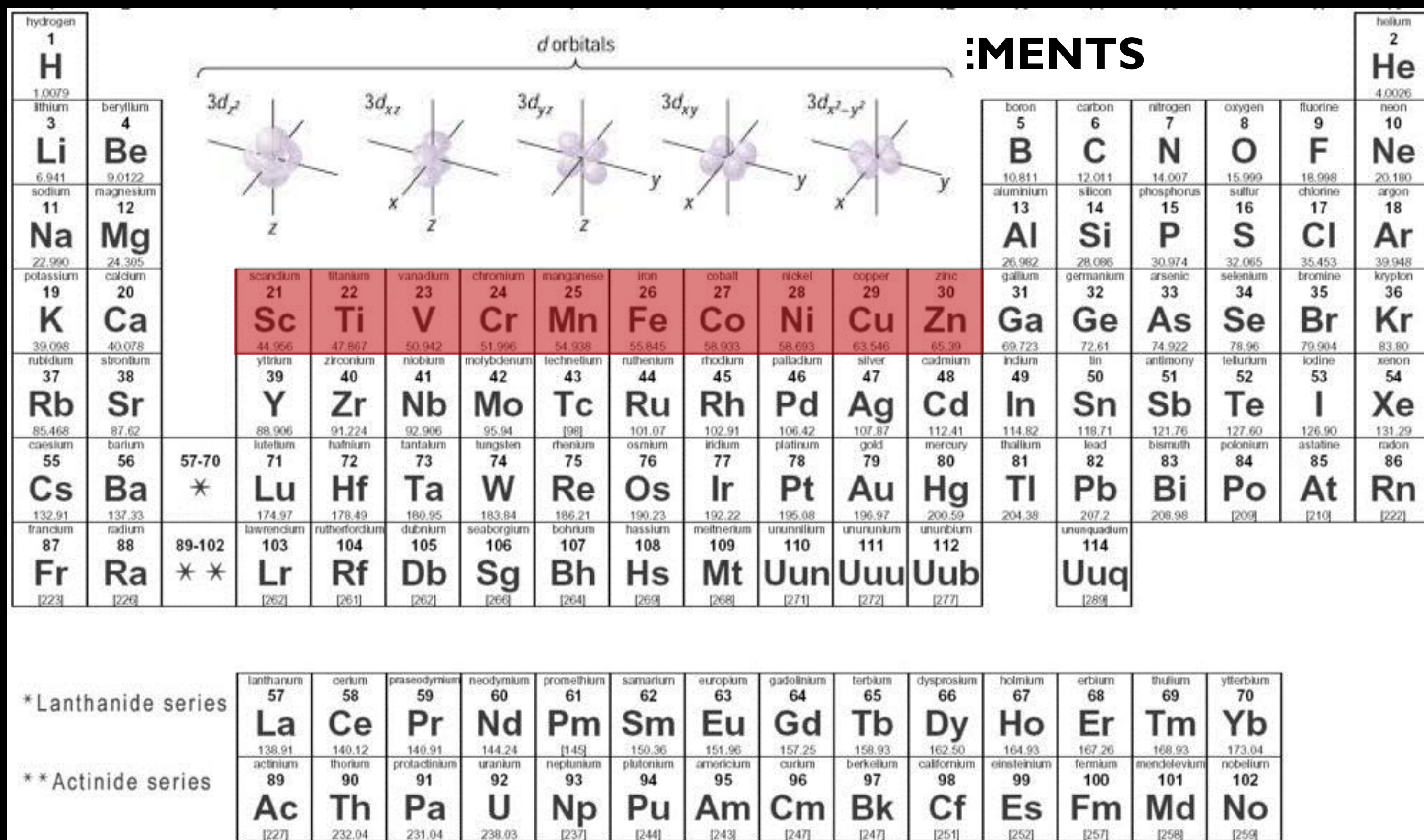
Doping

Dimensionality

Strongly-interacting quantum matter

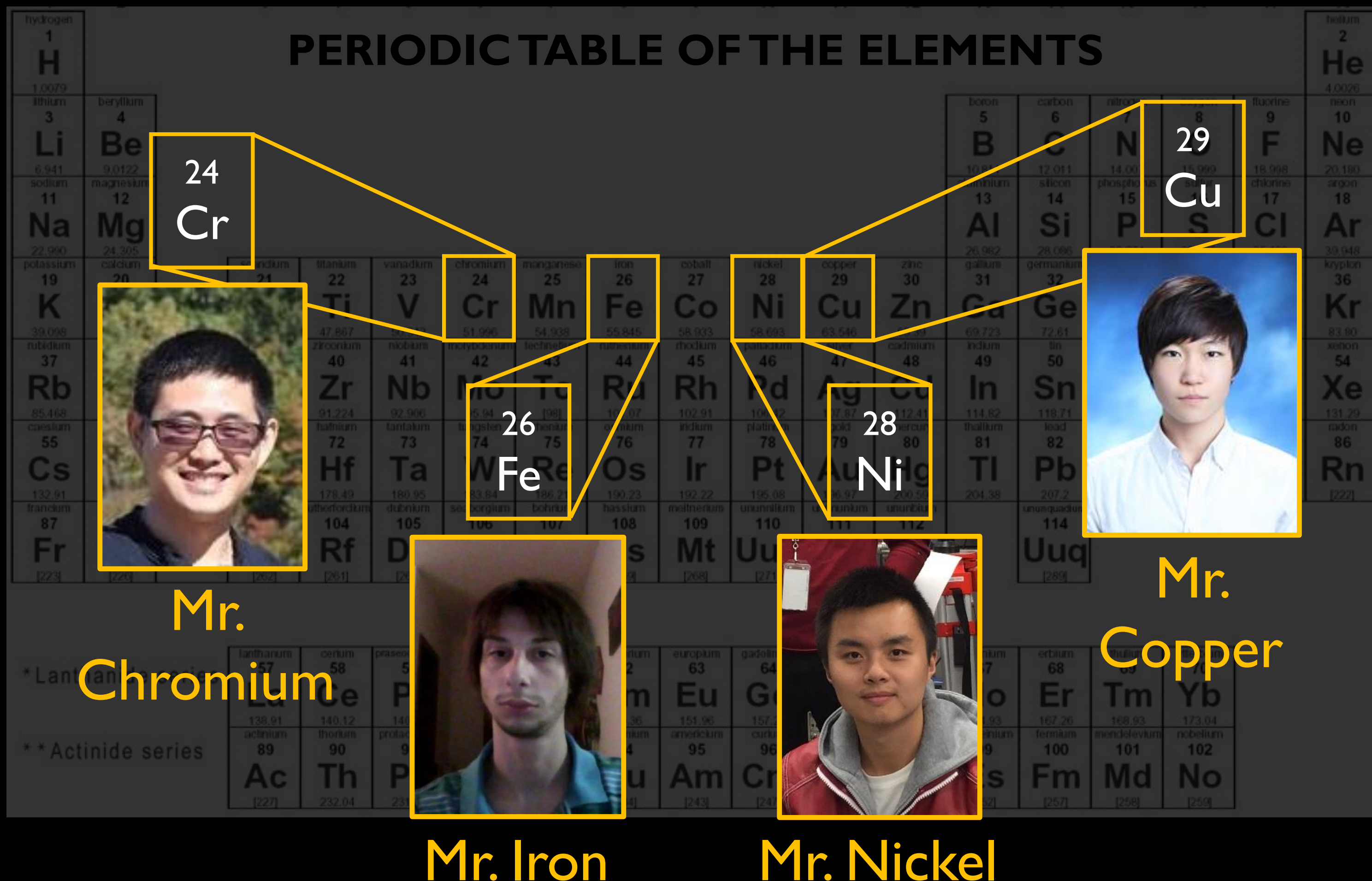
Reduce kinetic energy (*W*)

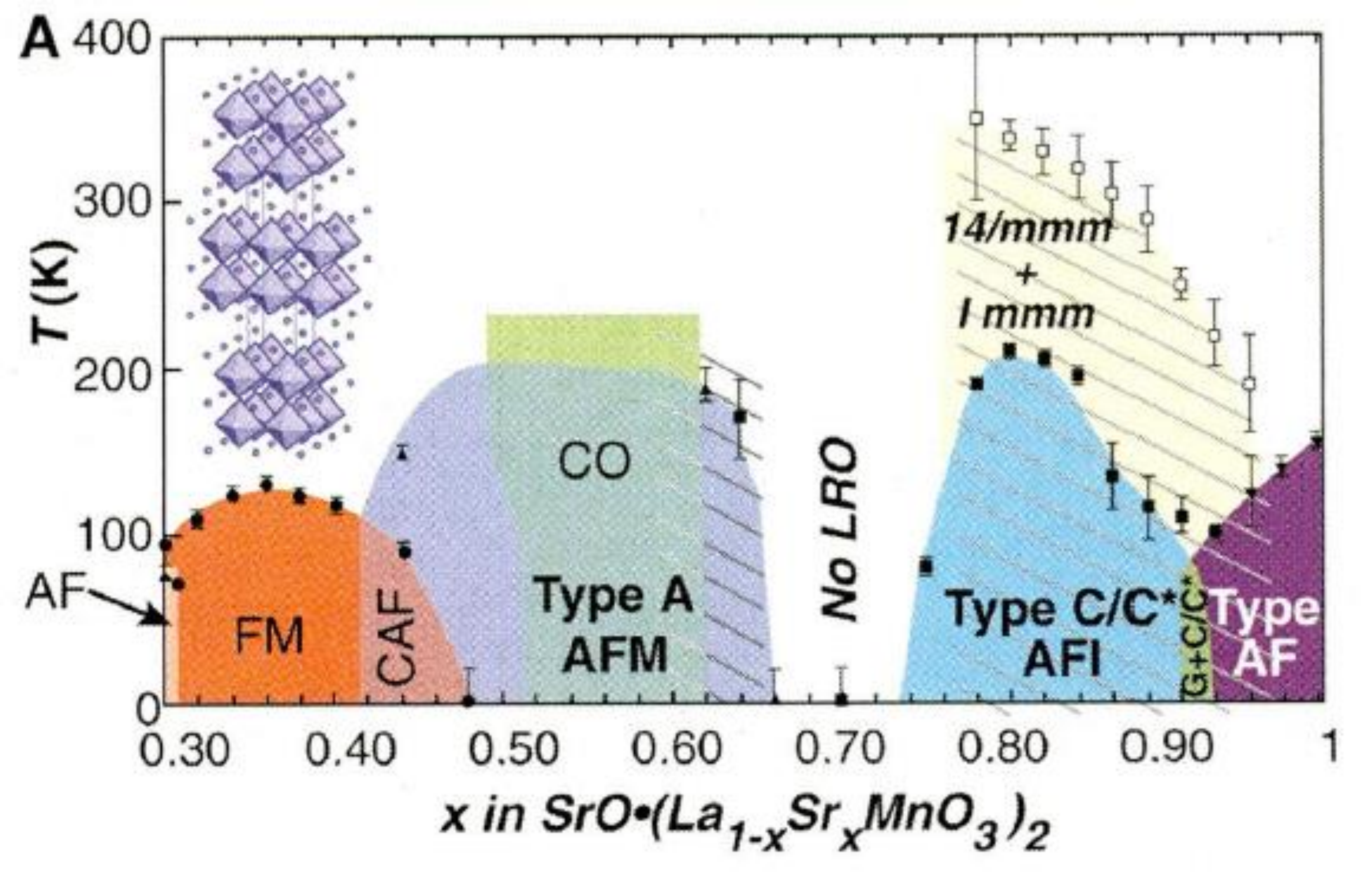
Increase interactions (*U*)



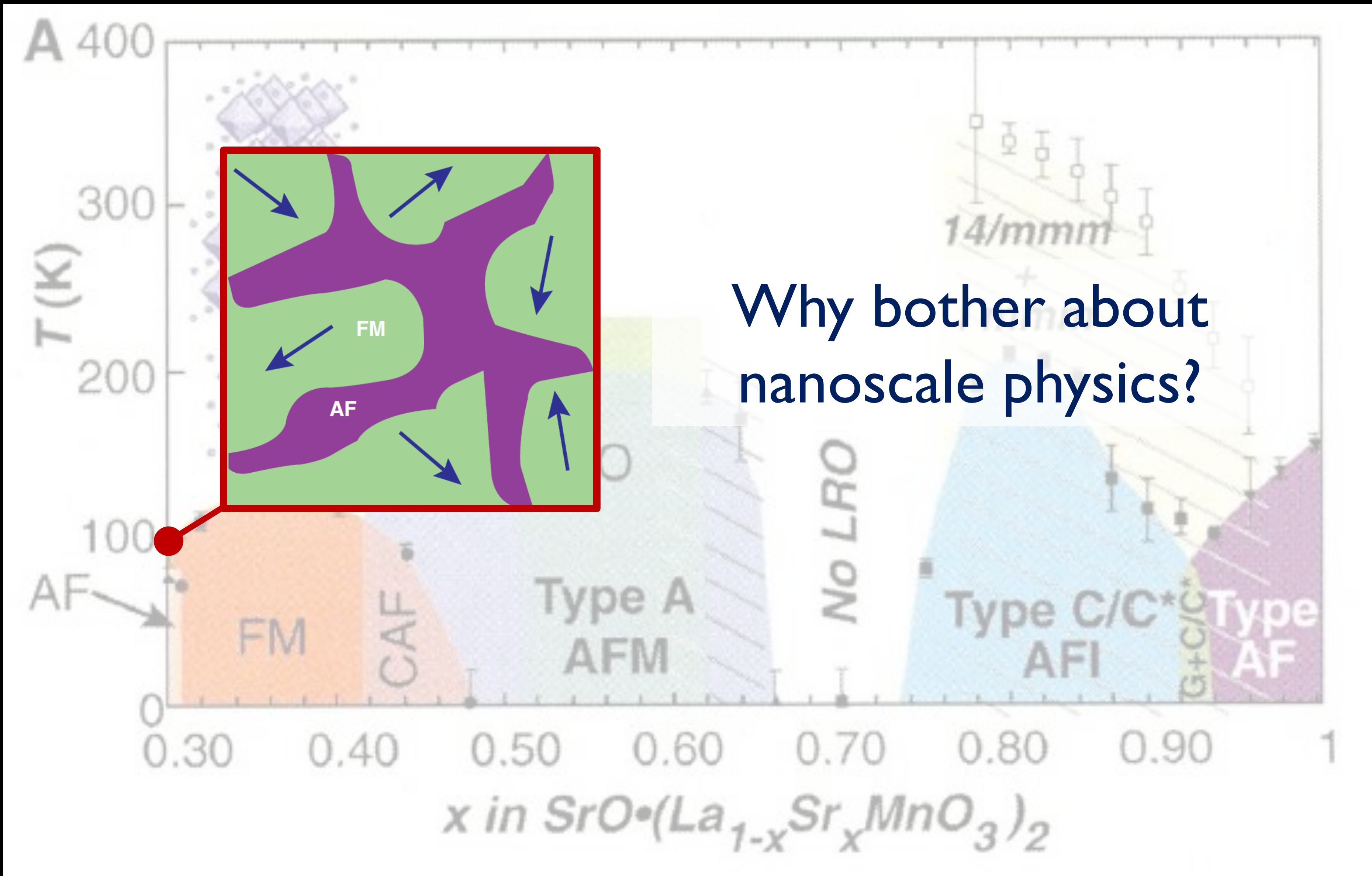
Strongly interacting systems

Strong interactions (*U*) ⟷ Localized orbitals





E. Dagotto, *Science* **309**, 257 (2005)

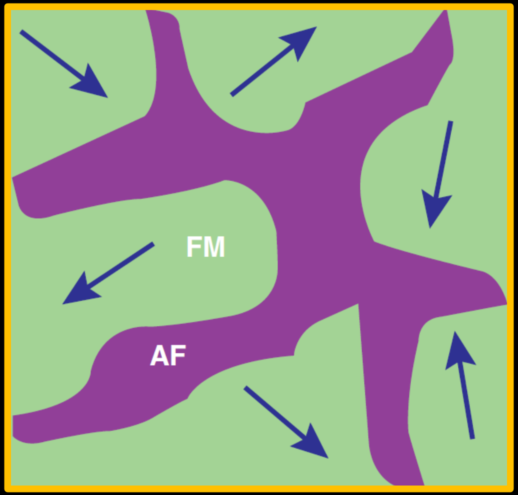


Why bother about nanoscale physics?

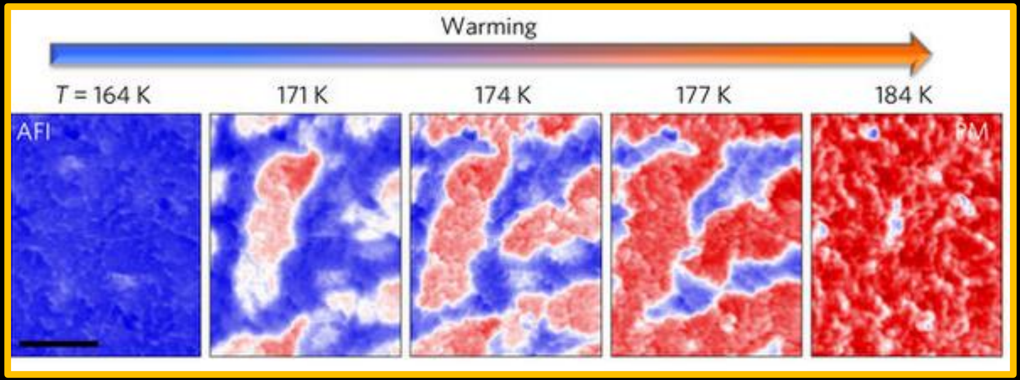
E. Dagotto, *Science* **309**, 257 (2005)

Strongly-interacting quantum matter

Phase segregation



Percolation phenomena

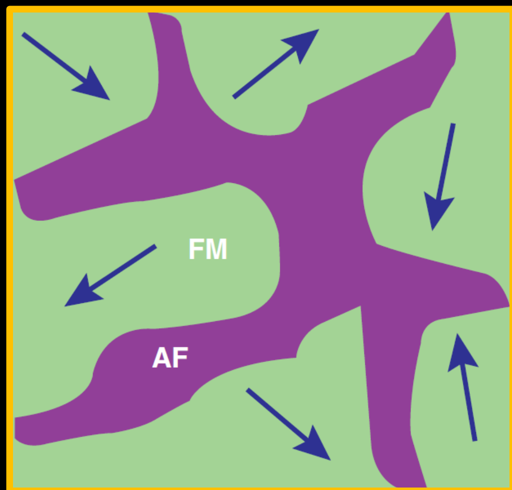


A. S. McLeod, *Nat. Physics* **13**, 80 (2017)

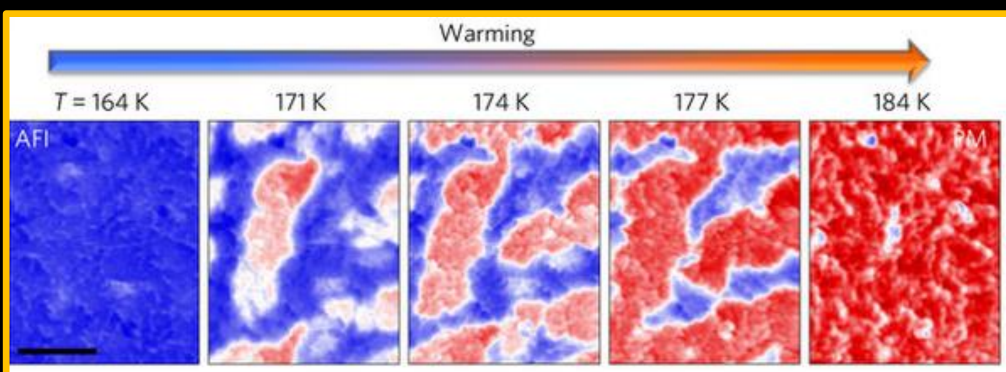
Quantum matter is almost inevitably inhomogeneous at the nanoscale

Strongly-interacting quantum matter

Phase segregation

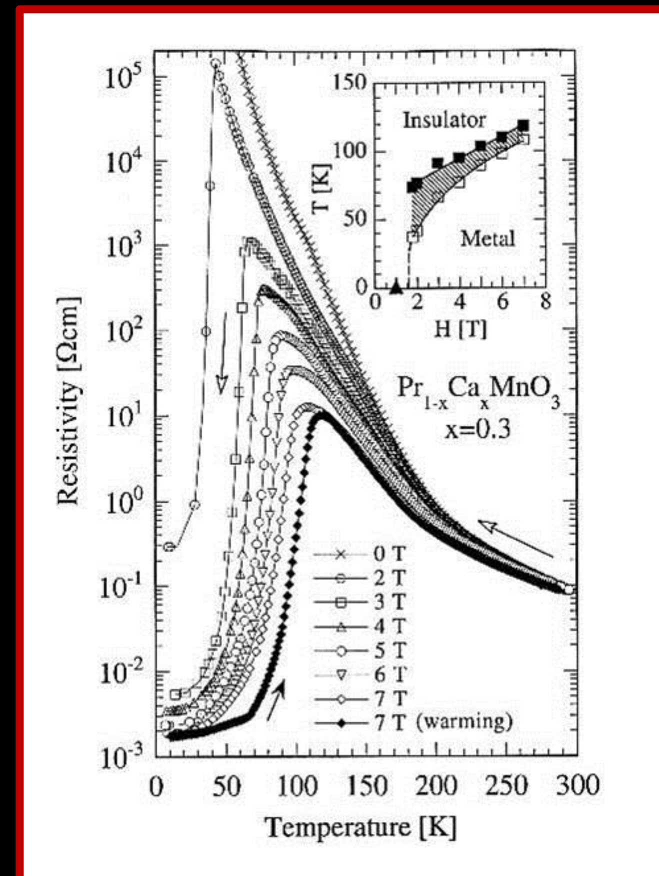


Percolation phenomena



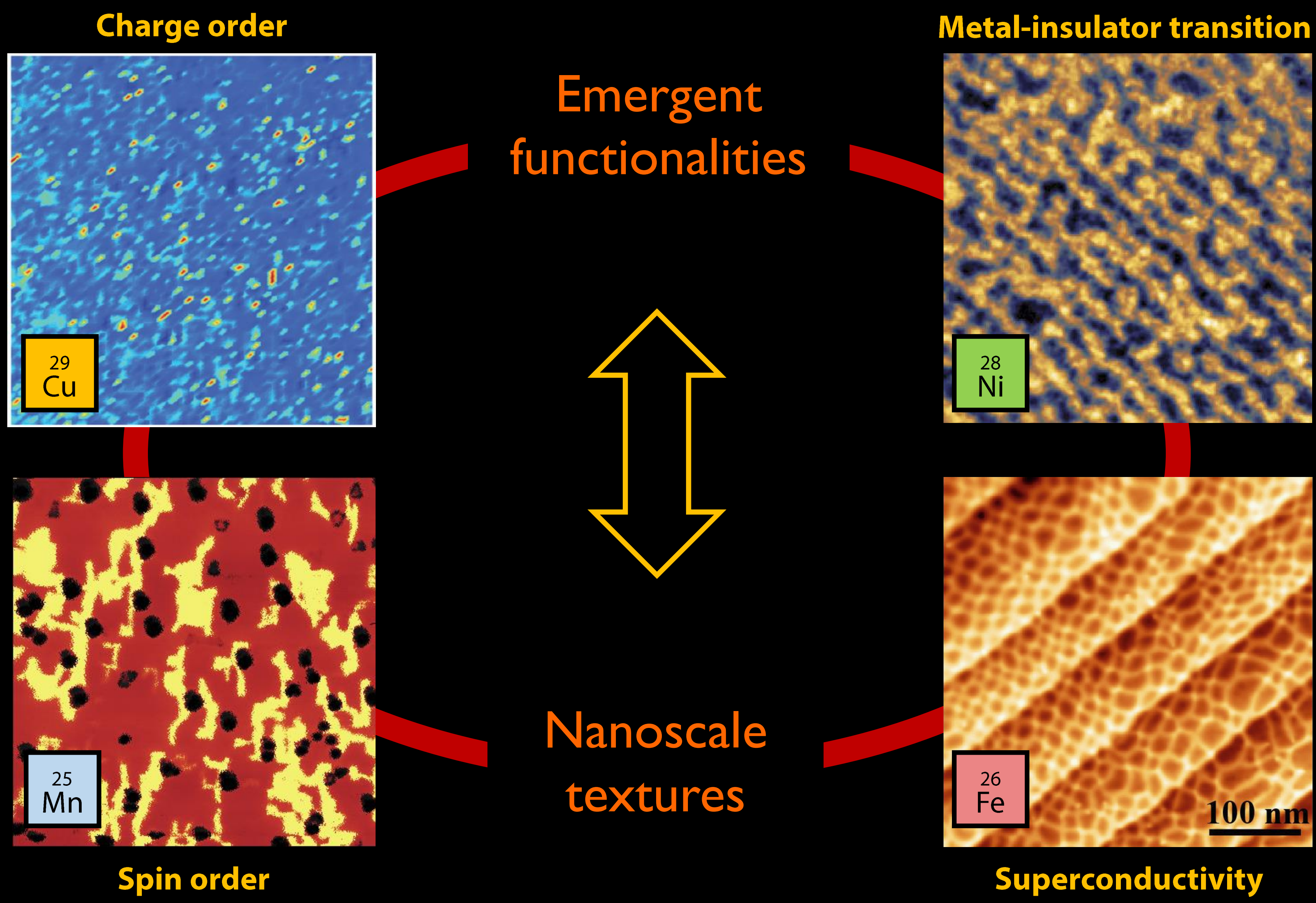
A. S. McLeod, *Nat. Physics* **13**, 80 (2017)

Colossal resistive switching



Y. Tomioka, *et al.*, *Physics of Manganites* (1999)

Strongly-interacting quantum matter



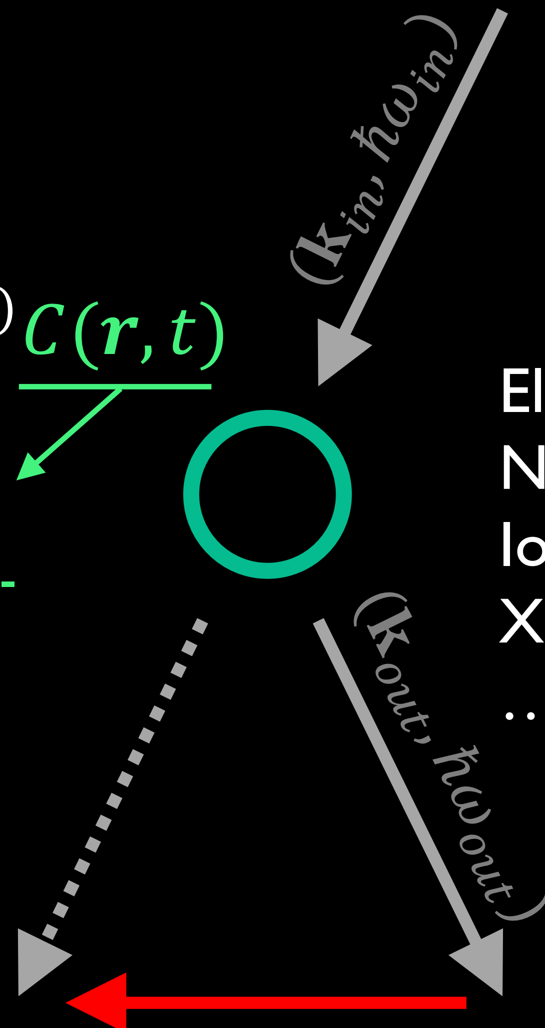
Scattering probes

Structure factor

$$S(\boldsymbol{Q},\omega) =$$

$$= \int dt\, d\boldsymbol{r}\, e^{i(\boldsymbol{Q}\cdot\boldsymbol{r}-\omega t)} C(\boldsymbol{r},t)$$

Correlation function (density-density, spin-spin, …)

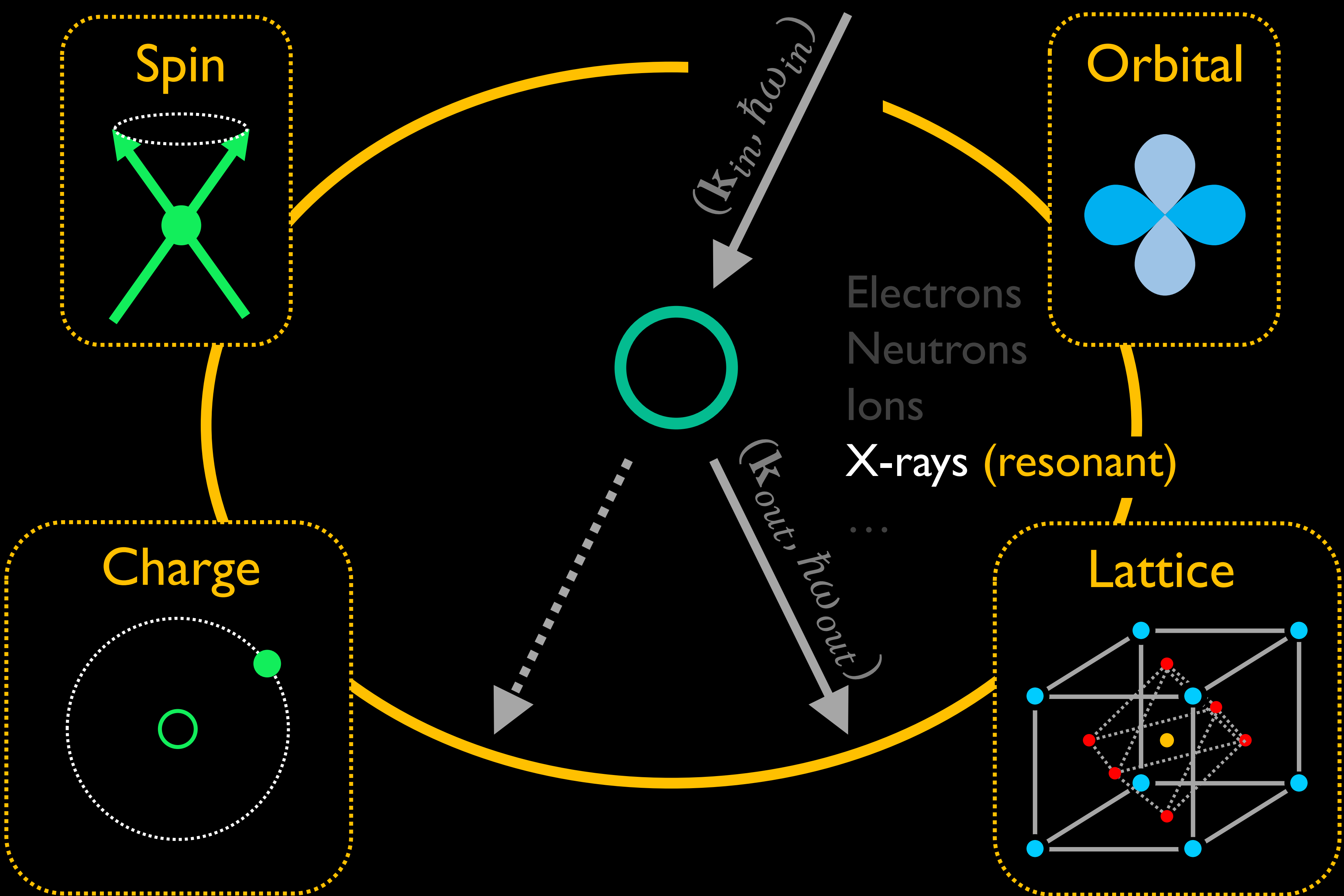


Electrons
Neutrons
Ions
X-rays
…

Momentum $\mathbf{Q} = \mathbf{k}_{in} - \mathbf{k}_{out}$

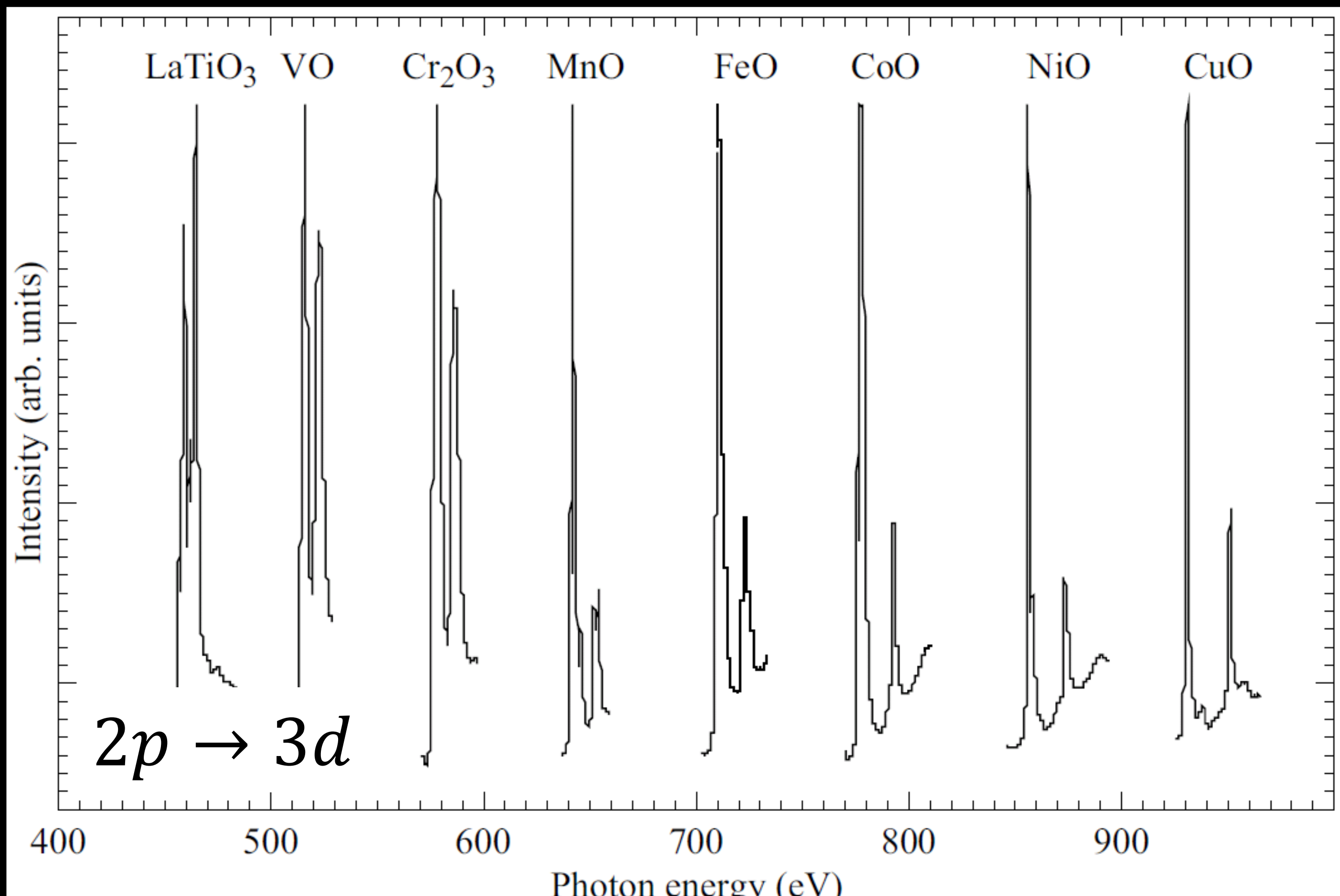
Energy $\omega = \omega_{in} - \omega_{out}$

Scattering probes



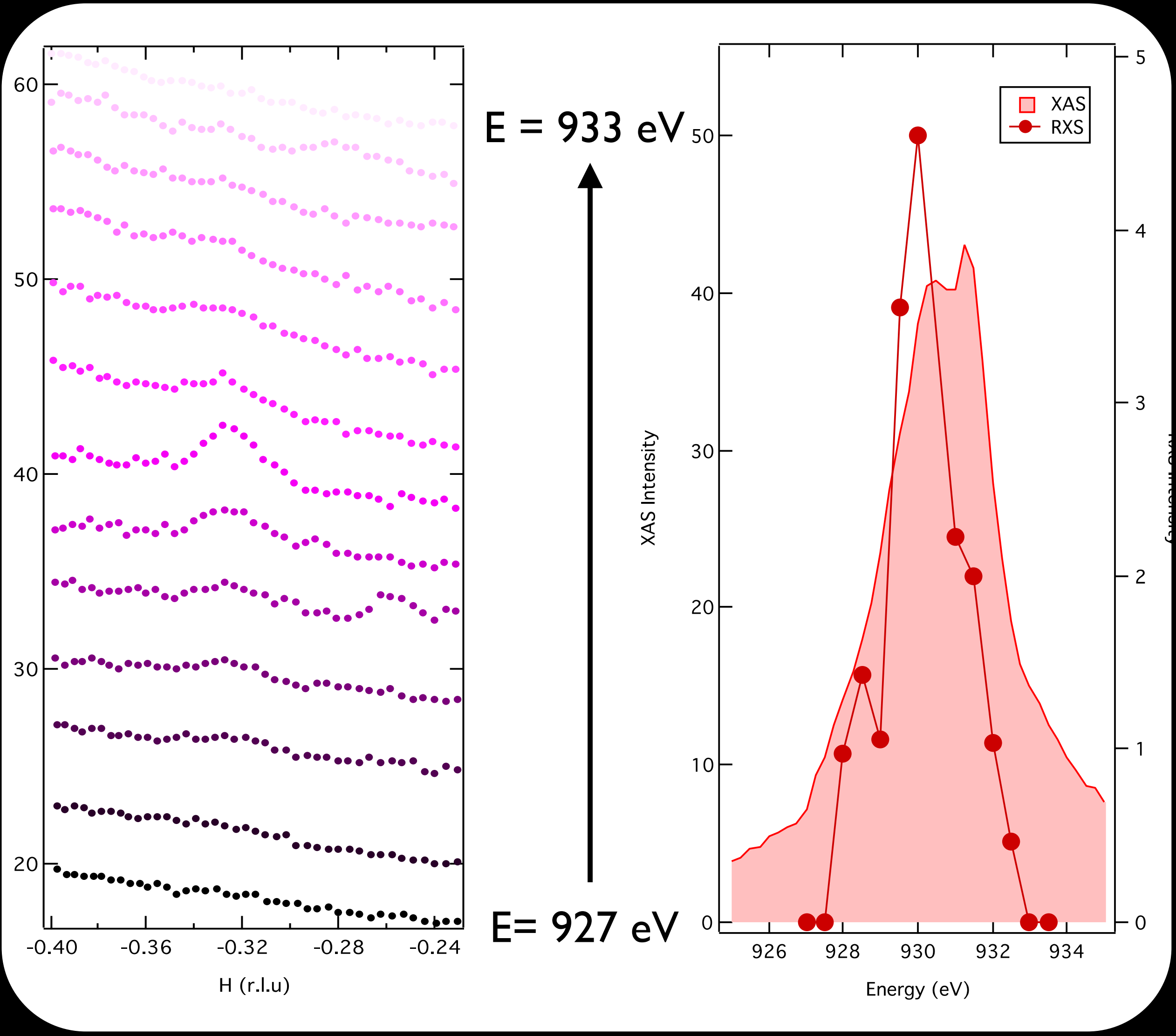
Resonant scattering

22	23	24	25	26	27	28	29
Ti	V	Cr	Mn	Fe	Co	Ni	Cu



Resonant scattering

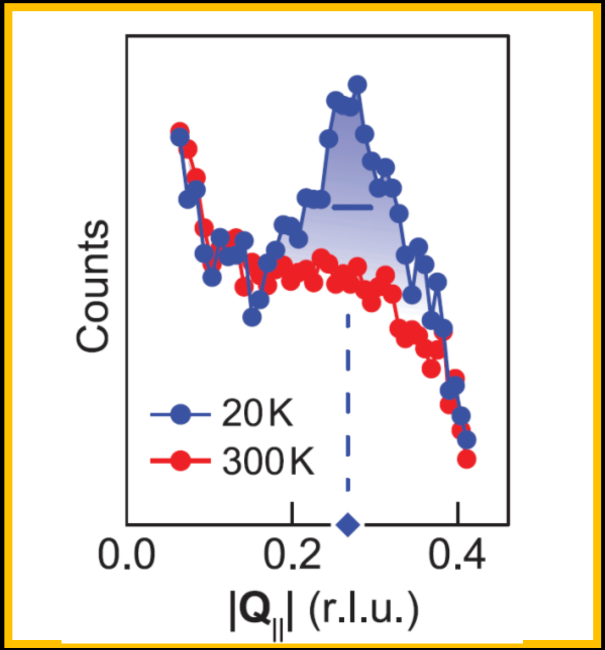
Charge-density-wave (Nd_2CuO_4)



Kang *et al*, in preparation (2018)

Electronic orders at the nanoscale

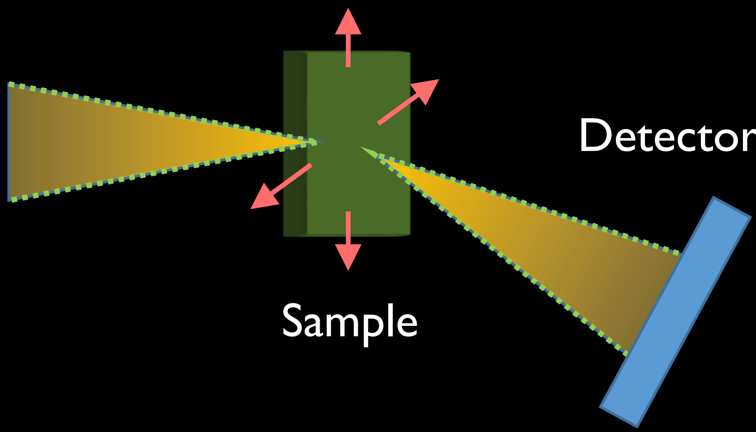
Reciprocal space (scattering)



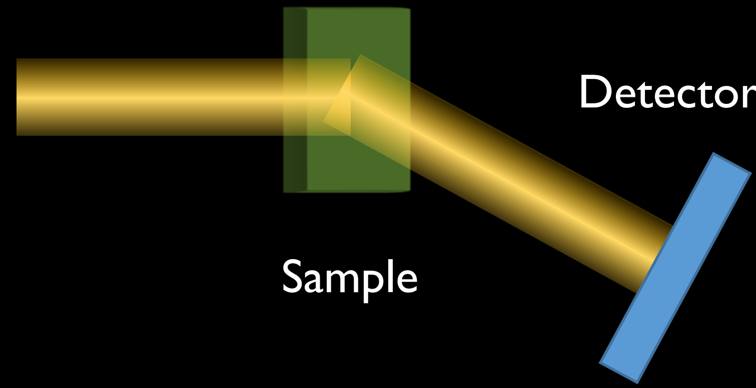
Recover real-space information

HOW

Scanning nanodiffraction (50 nm)



Full-field lensless imaging (10 nm)



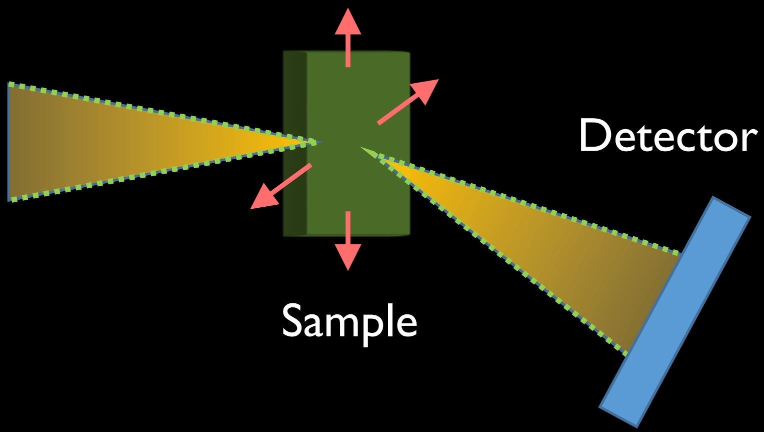
Electronic orders at the nanoscale

WHY

- Nanoscale granularity:
  - Intrinsic (phase competition & segregation)
  - Extrinsic (disorder, defects, doping, …)
- Scale-invariant phenomena:
  - Extended range of length scales (10 nm to 10 μm)
- Emergent physics at the edge or boundary:
  - Domain walls; lateral interfaces; nanoengineered structures

HOW

Scanning nanodiffraction (50 nm)



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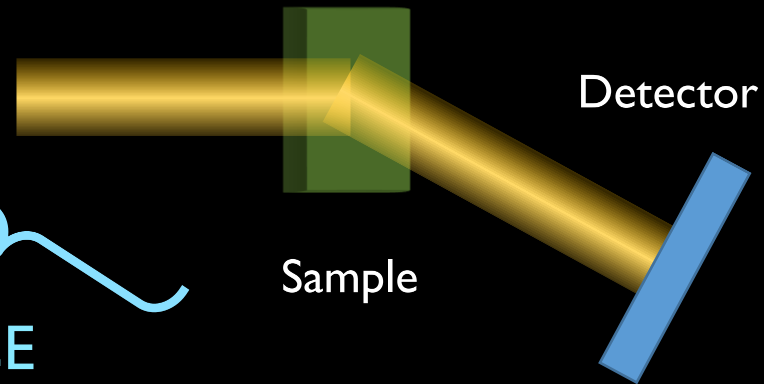
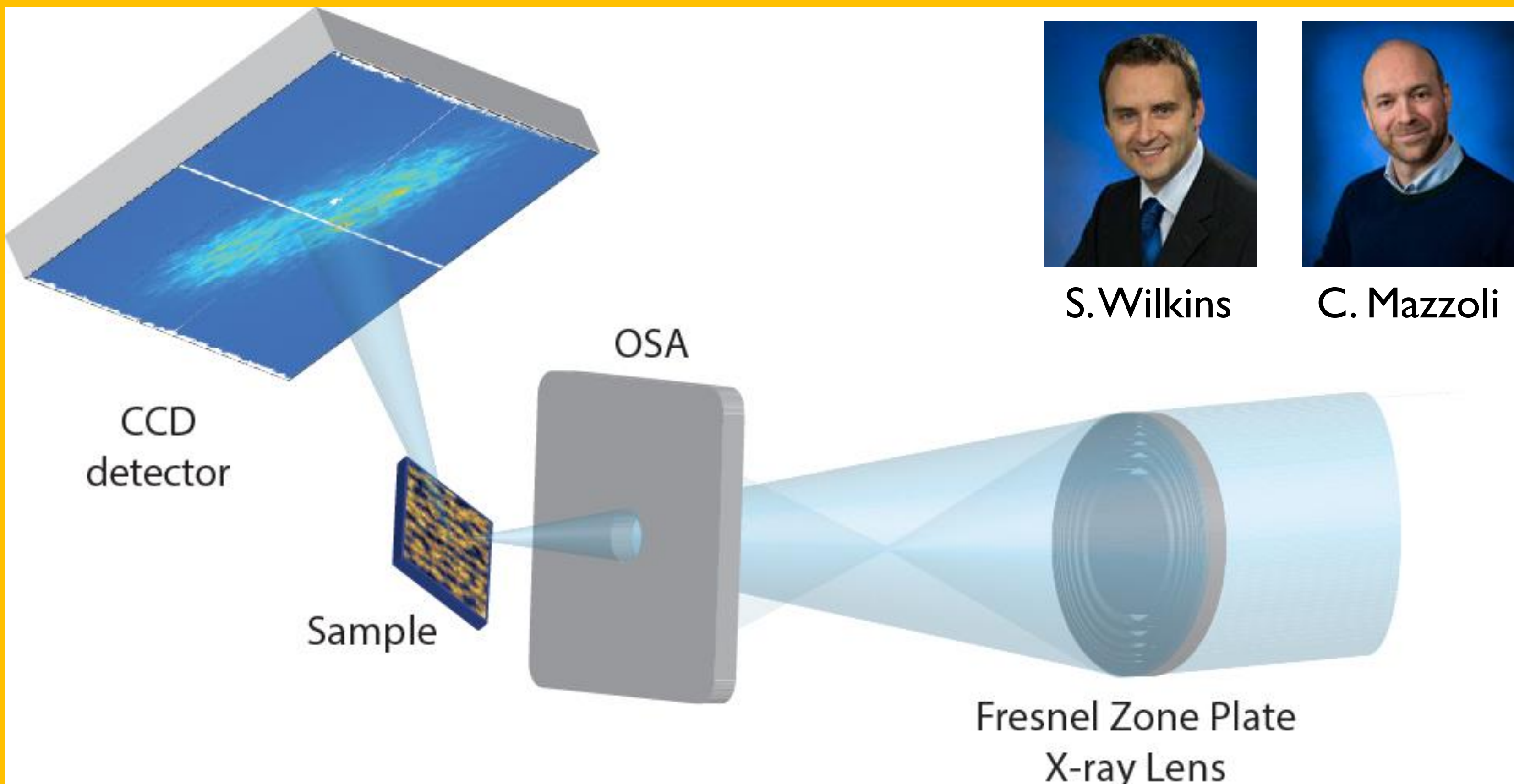


Need COHERENCE

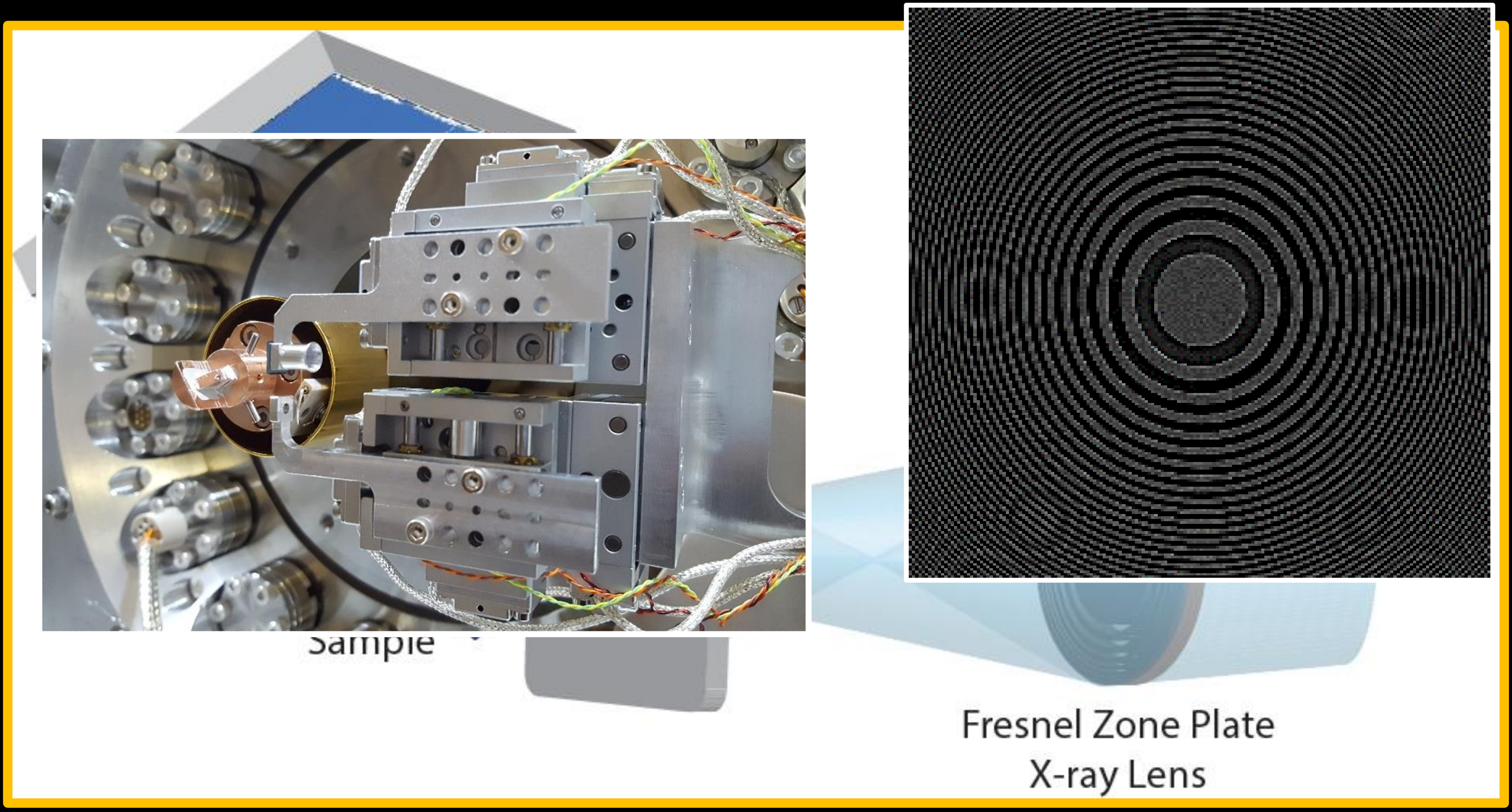
Soft X-ray nanodiffraction

Scanning resonant nanodiffraction



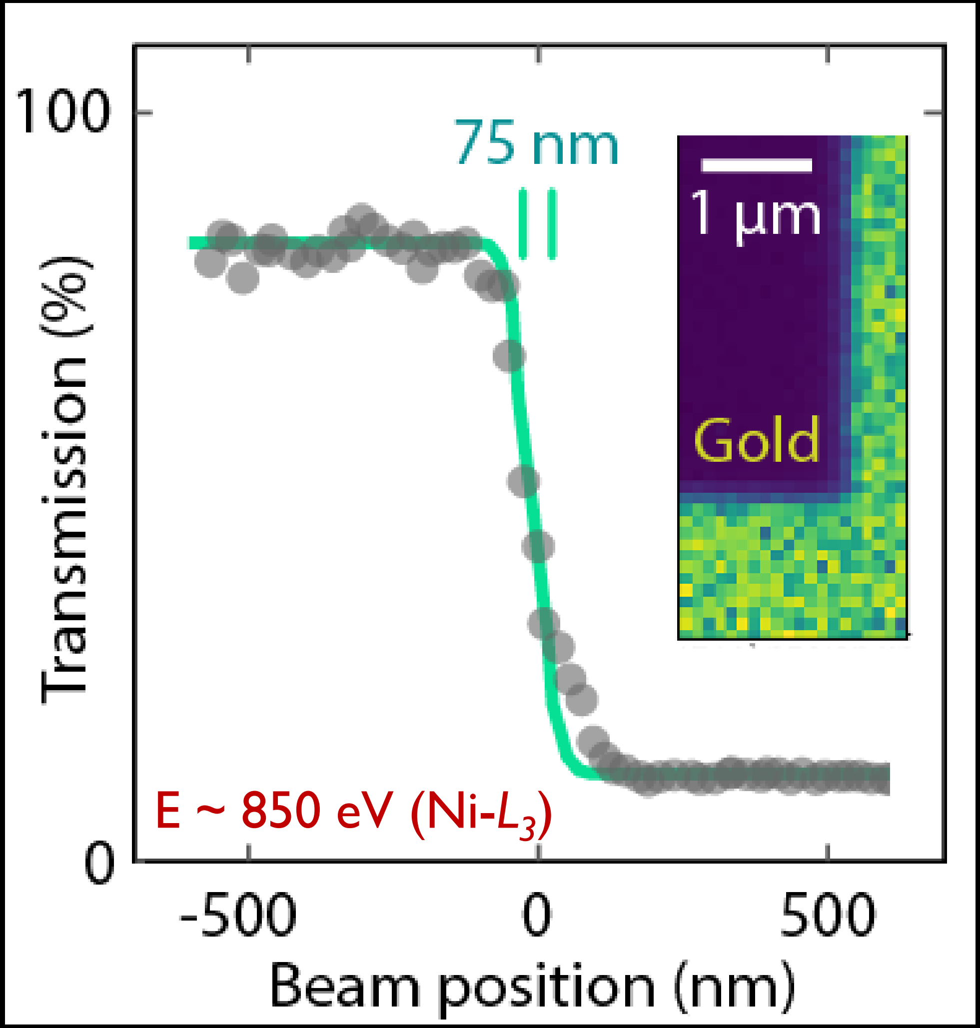
Soft X-ray nanodiffraction

Scanning resonant nanodiffraction



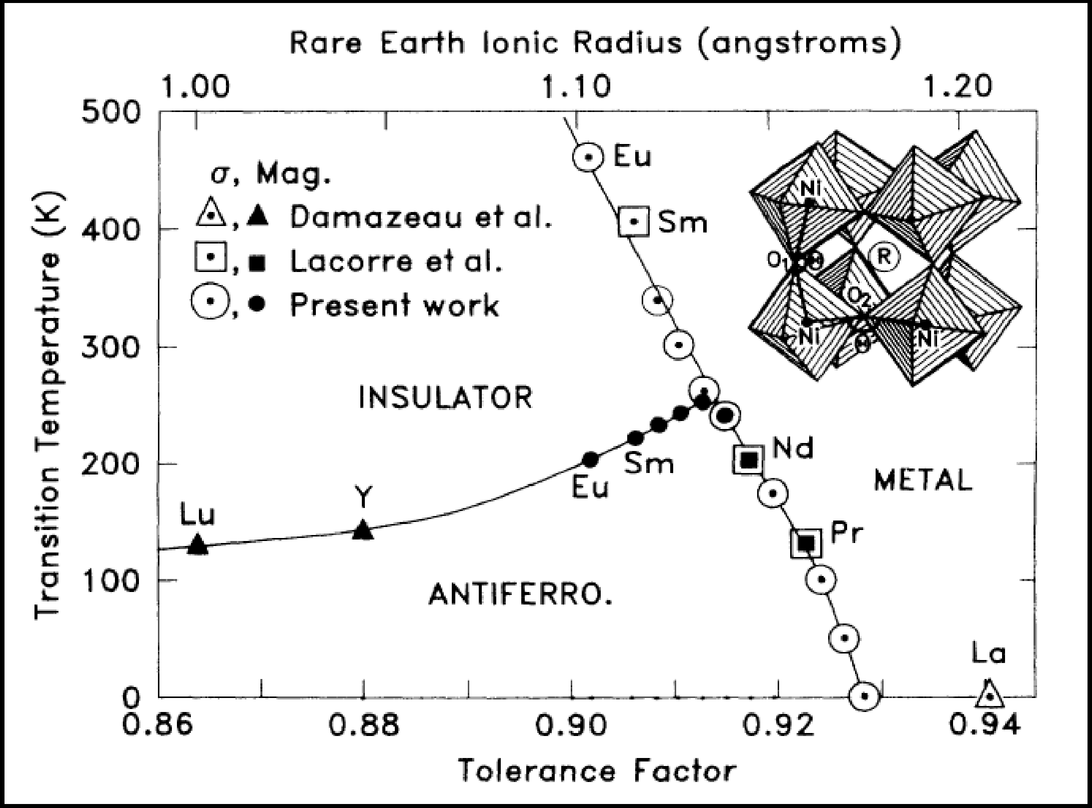
Soft X-ray nanodiffraction

Zone-plate focusing optics: 75 nm spot size



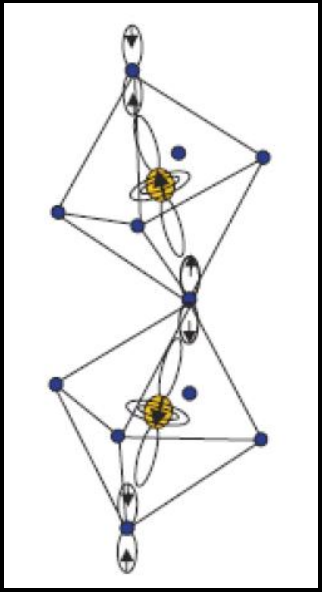
Nickelates

Cubic nickelate perovskites ($RNiO_3$)

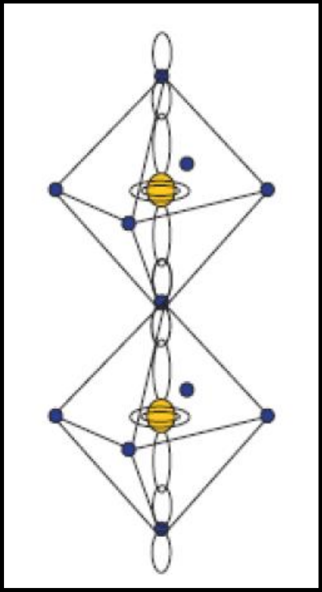


Torrance *et al.* PRB 1992

Low-T (insulator)



High-T (metal)



Nickelates

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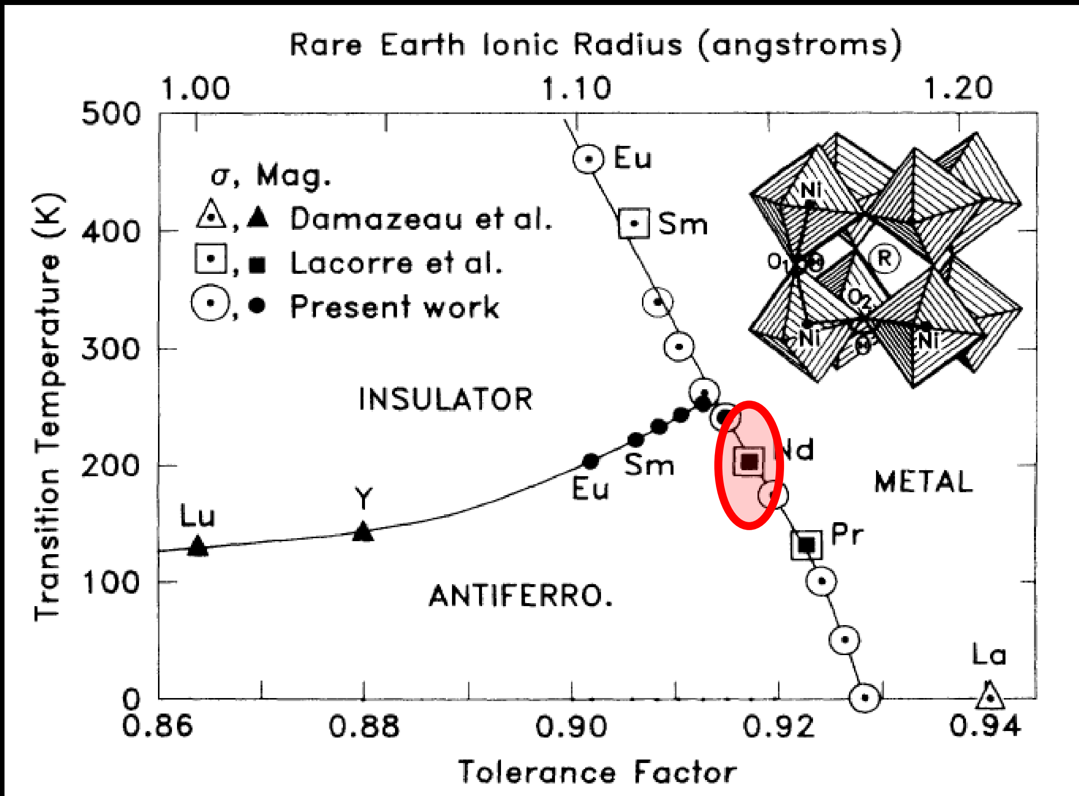
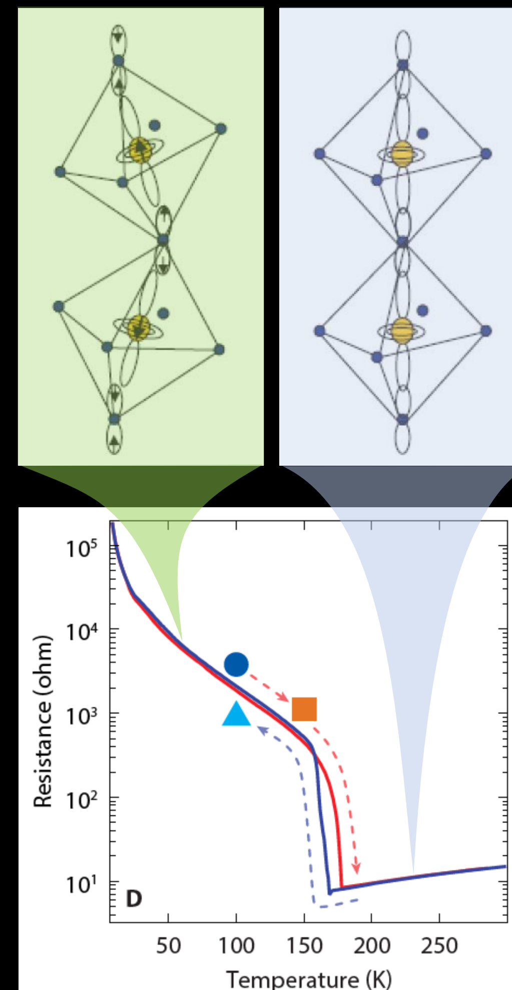


Torrance *et al.* PRB 1992

$NdNiO_3$

Low-T (insulator)

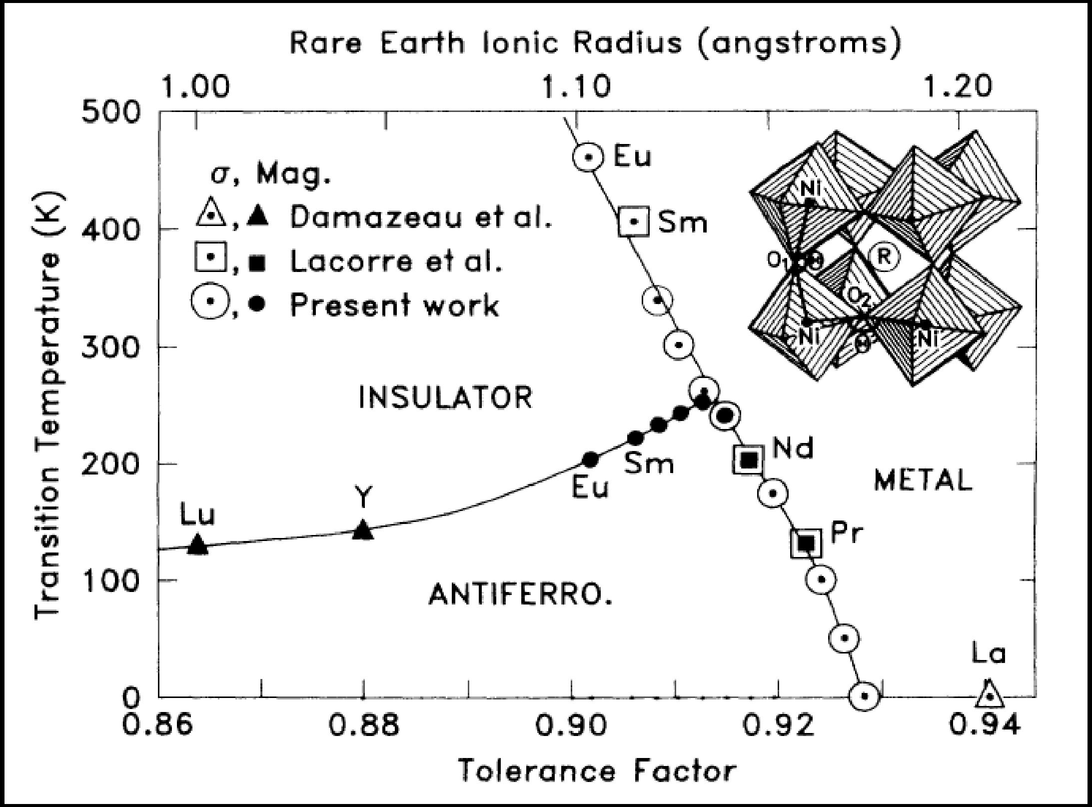
High-T (metal)



Metal insulator transition

Nickelates

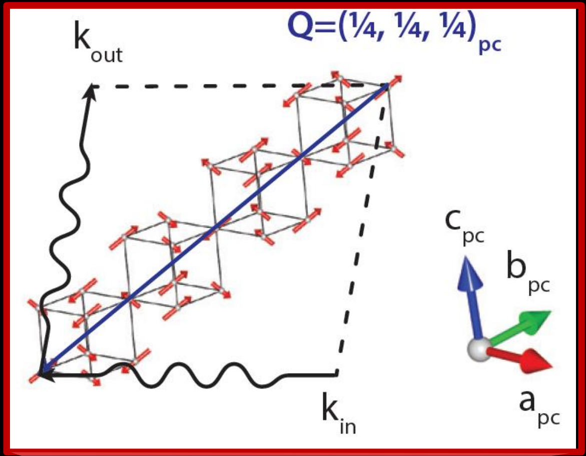
Cubic nickelate perovskites ($RNiO_3$)

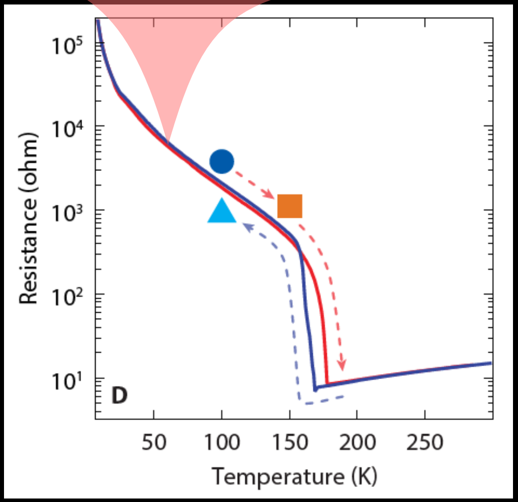


Torrance *et al.* PRB 1992

Goal: map antiferromagnetic order across metal-insulator transition

Magnetic order

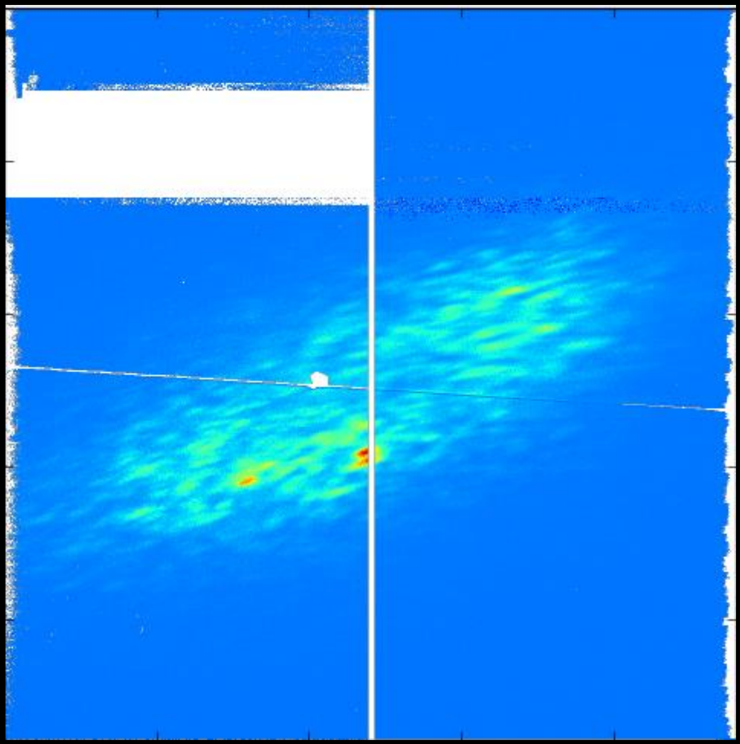




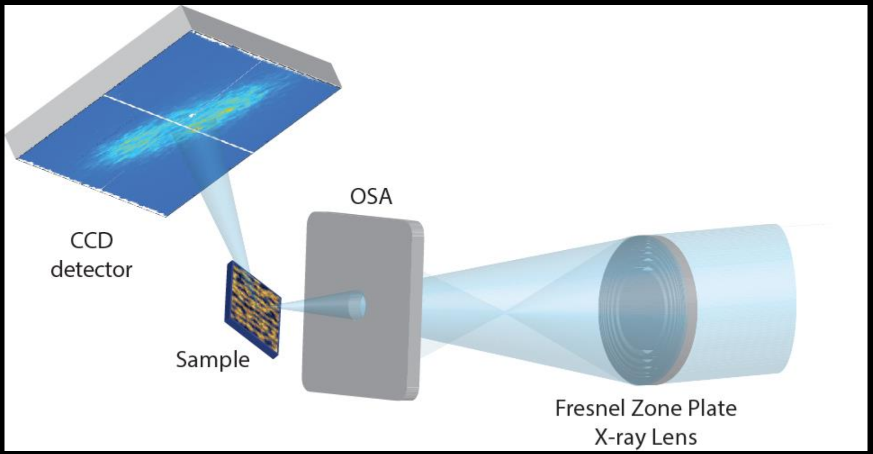
Metal insulator transition

Nickelates

RECIPROCAL SPACE

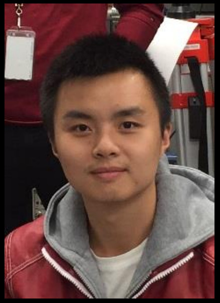


Coherent magnetic scattering from spin-density wave in $NdNiO_3$



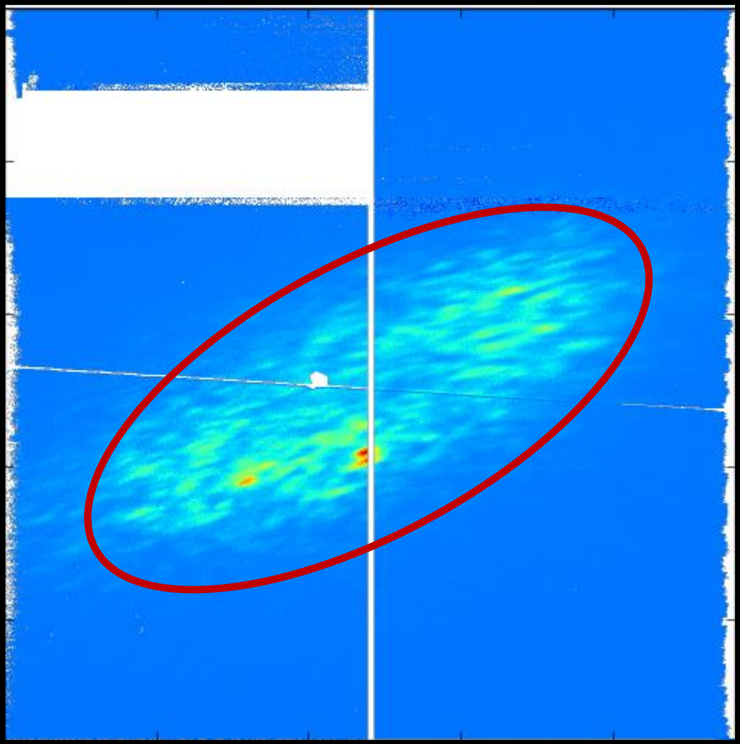
Johnny Pelliciari

Jiarui Li



Nickelates

RECIPROCAL SPACE

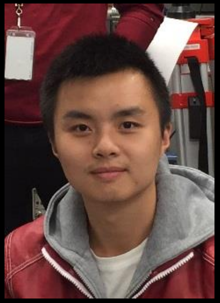


Speckle pattern: coherent interference between magnetic domains

Coherent magnetic scattering from spin-density wave in $NdNiO_3$

Johnny Pelliciari

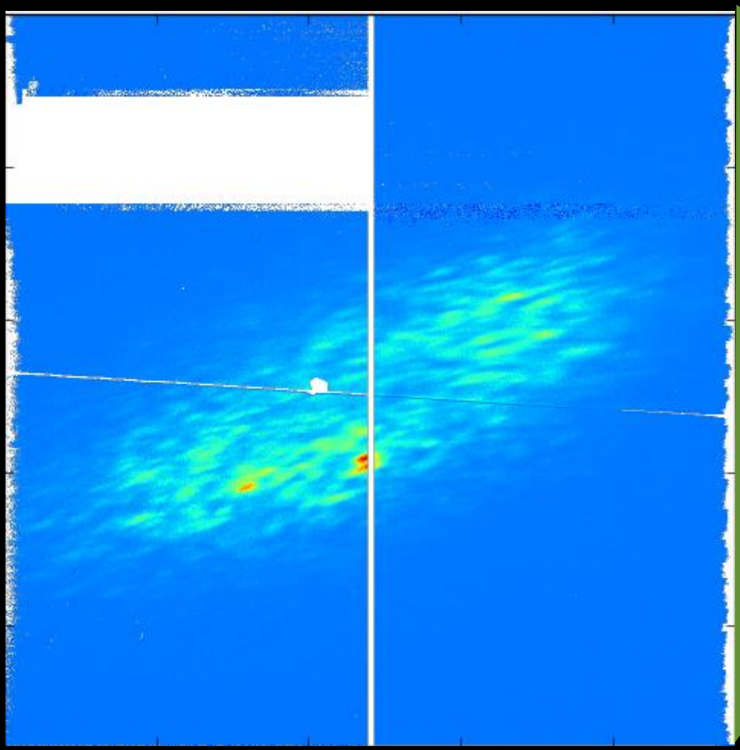
Jiarui Li



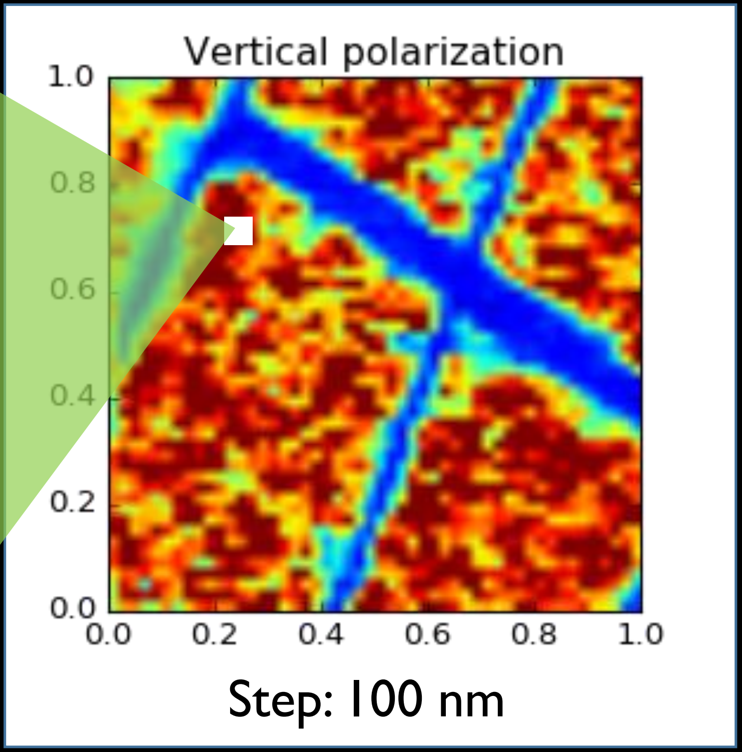
Nickelates

First case study: spin-density-wave in $NdNiO_3$ thin films

RECIPROCAL SPACE → REAL SPACE (mapping)

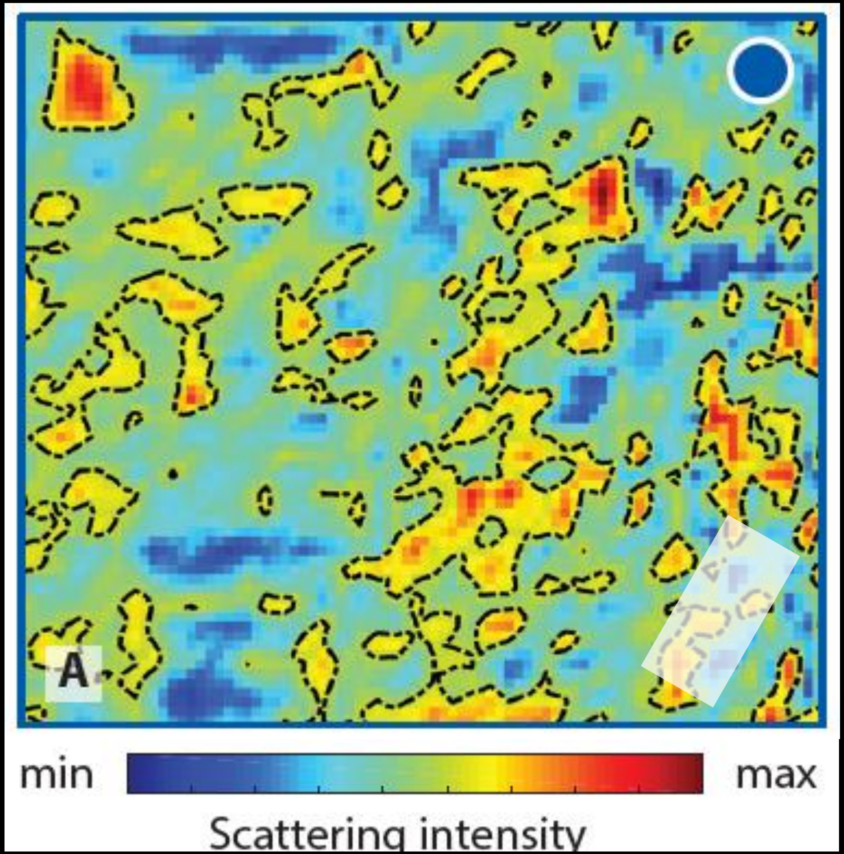


Coherent magnetic scattering from spin-density wave in $NdNiO_3$



Nano-mapping of order parameter

Nickelates

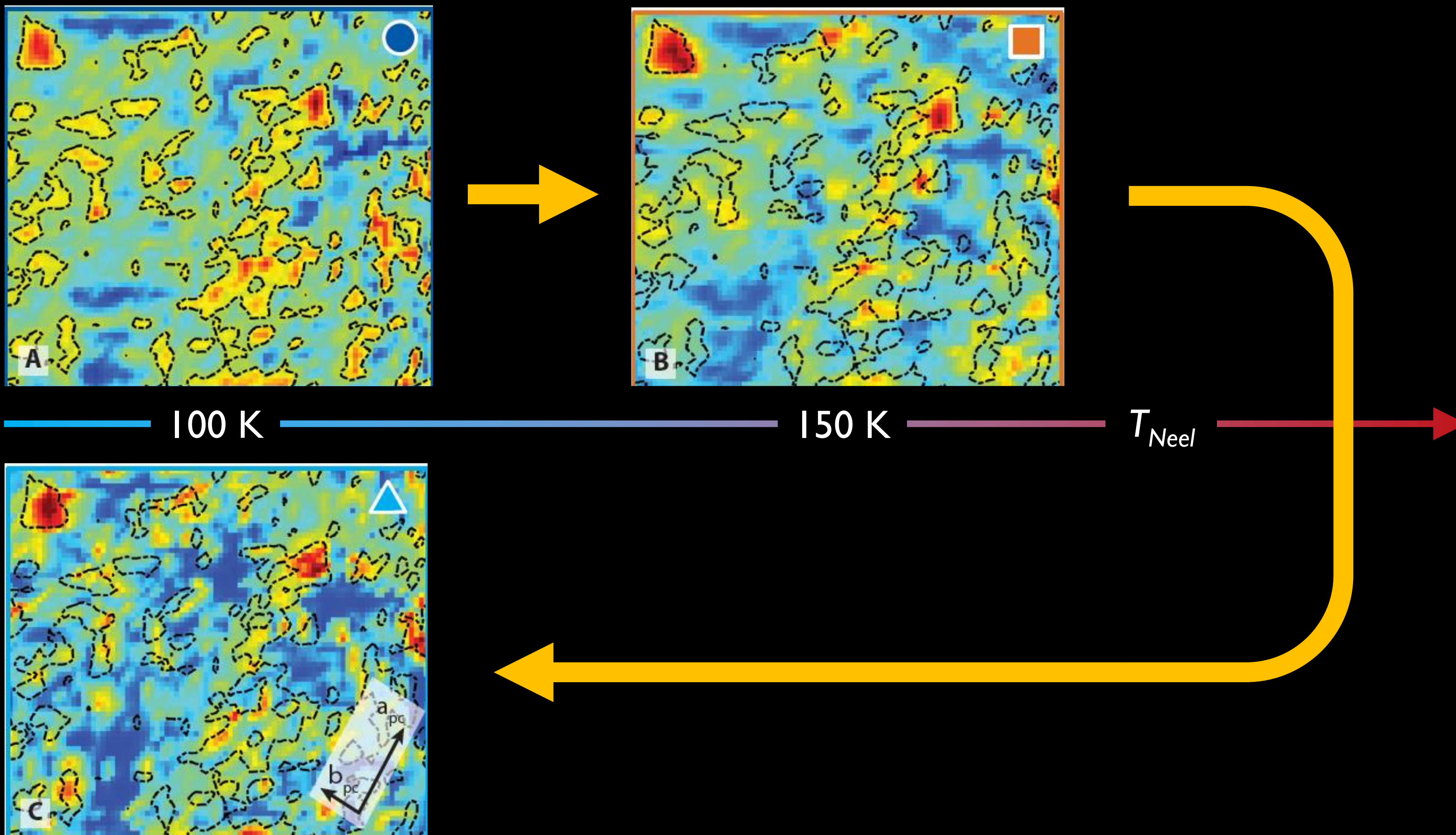


SDW order parameter

1x1 μm^2 square

Nanoscale inhomogeneity on length scales 0.1 – 10 μm

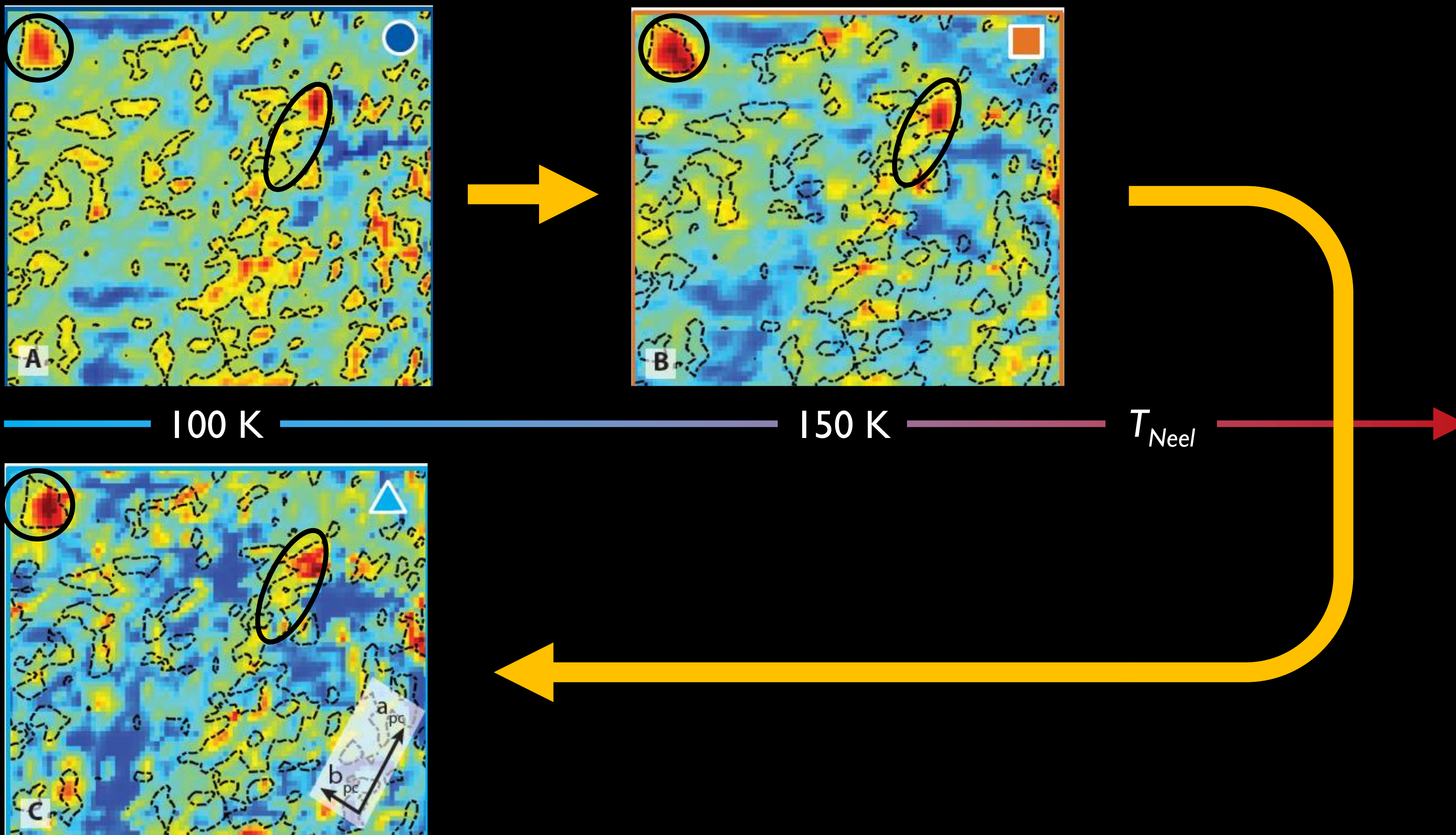
Nickelates



Domain pinning = memory effect

Possibly a hidden local parameter controlling domain nucleation

Nickelates



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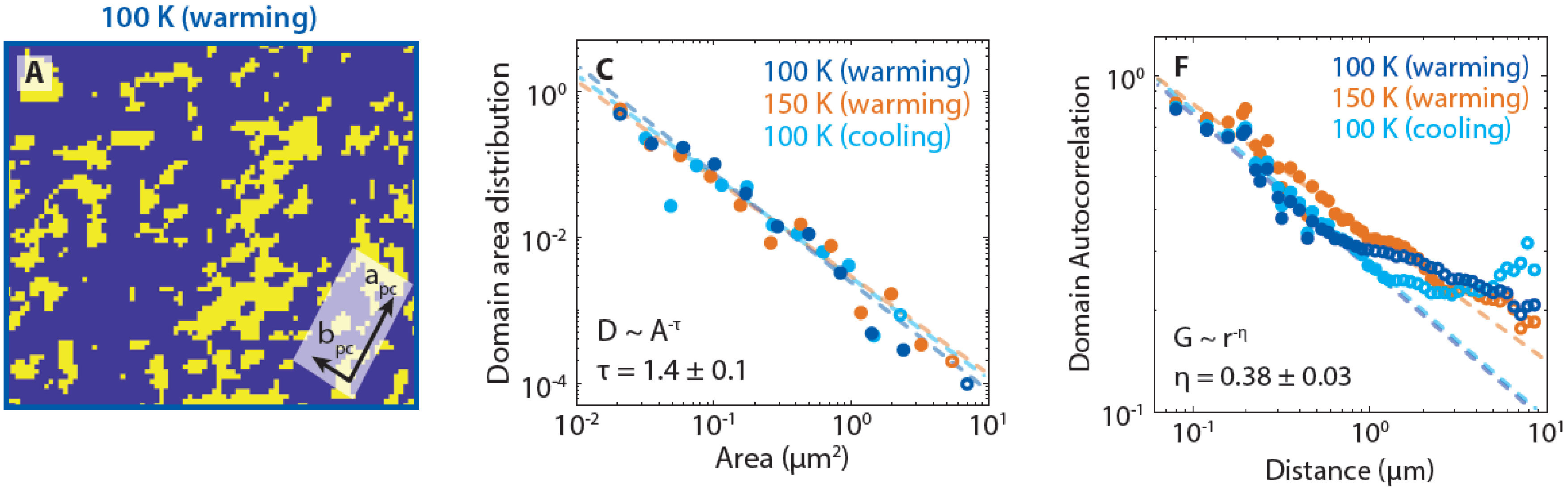
Nickelates

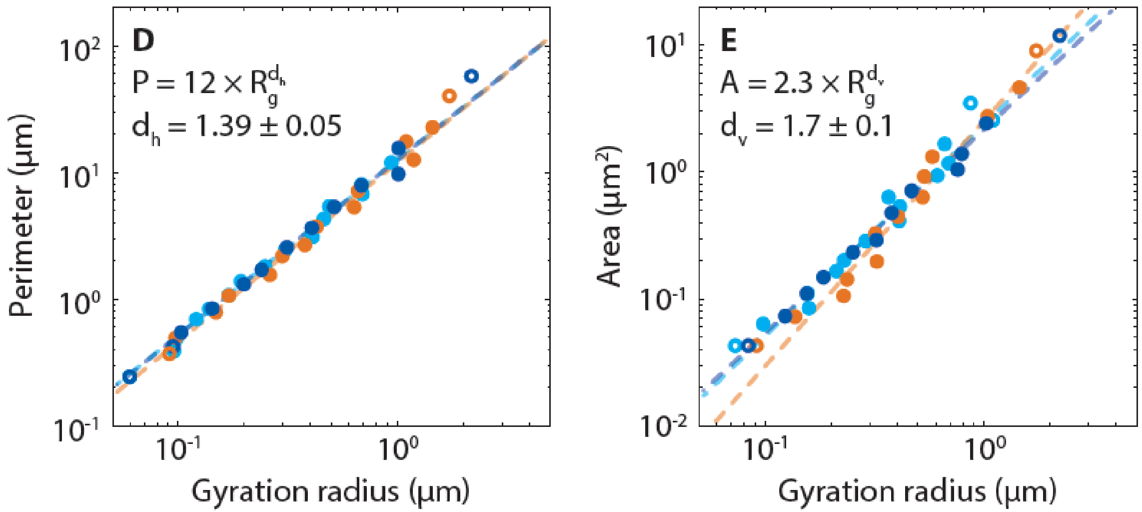
Scale-invariant textures – a fractal magnetic landscape



NdNiO3 manifests near-critical behavior – static (quenched) spatial fluctuations appear at all length scales

Nickelates

Scale-invariant textures – a fractal magnetic landscape



NdNiO3 manifests near-critical behavior – static (quenched) spatial fluctuations appear at all length scales

Soft X-ray nanodiffraction

Scanning resonant nanodiffraction

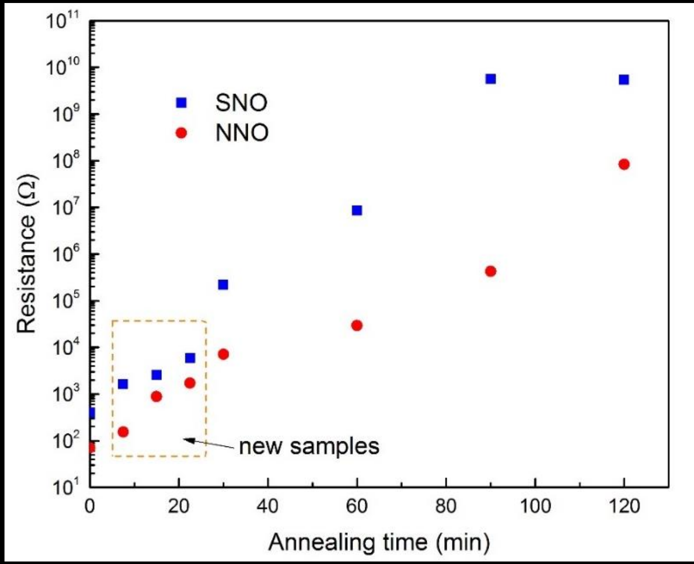
PROS

- Direct visualization of order parameter
- No phase retrieval needed
- Versatile (applies to any material)

CONS

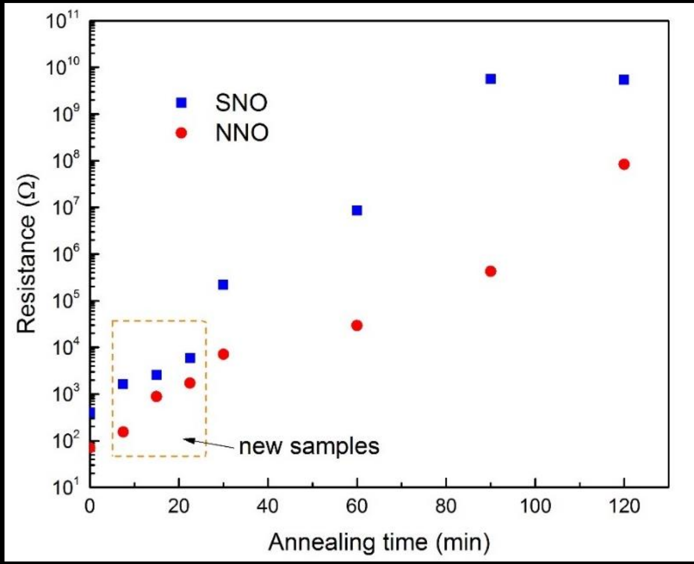
- Slow, point-by-point scanning required (1-4 hrs)
- Spatial resolution limited by NA of optics (not detector)
- 10-15% throughput across ZP lenses

Doped nickelates



Electron-doping induced a colossal metal-insulator transition

Doped nickelates



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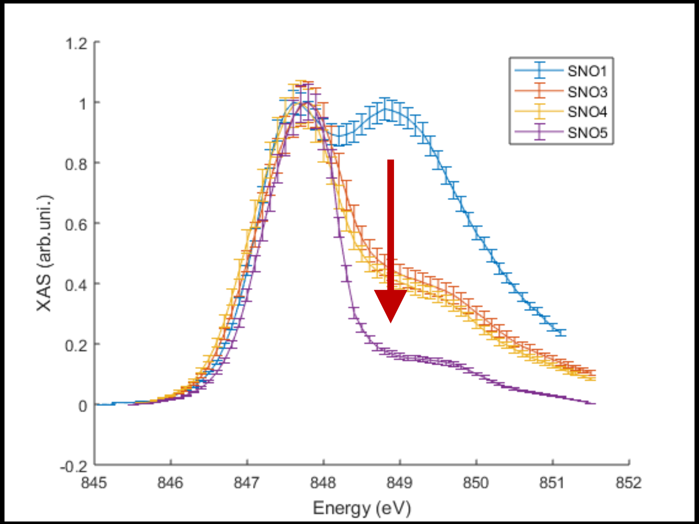
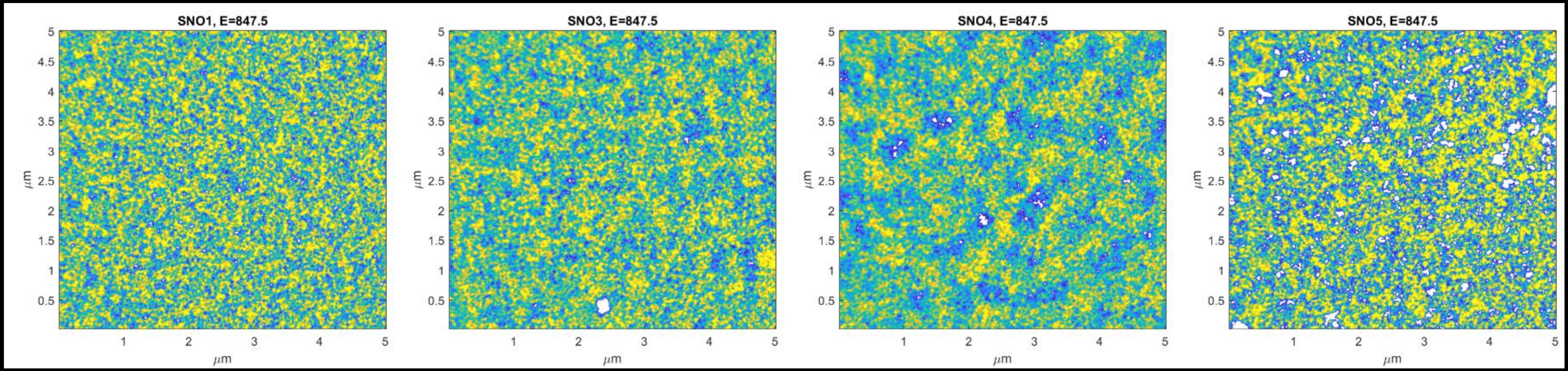
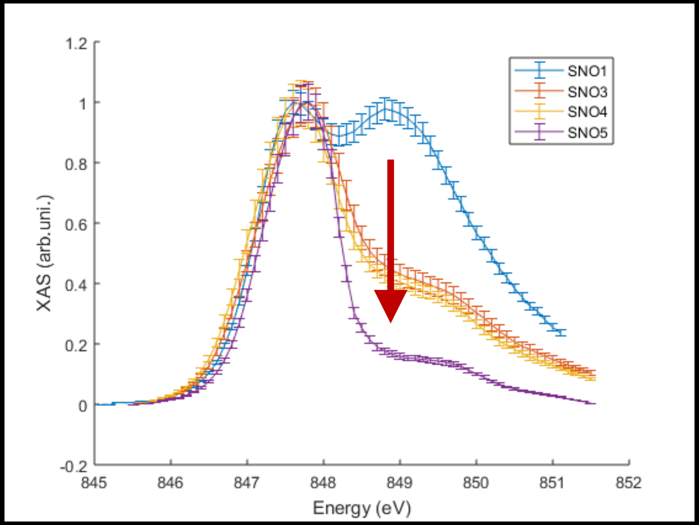


XAS shows doping induced crossover in electronic ground state

Doped nickelates

XPEEM mapping across Ni-L_3 edge (@ESM beamline)





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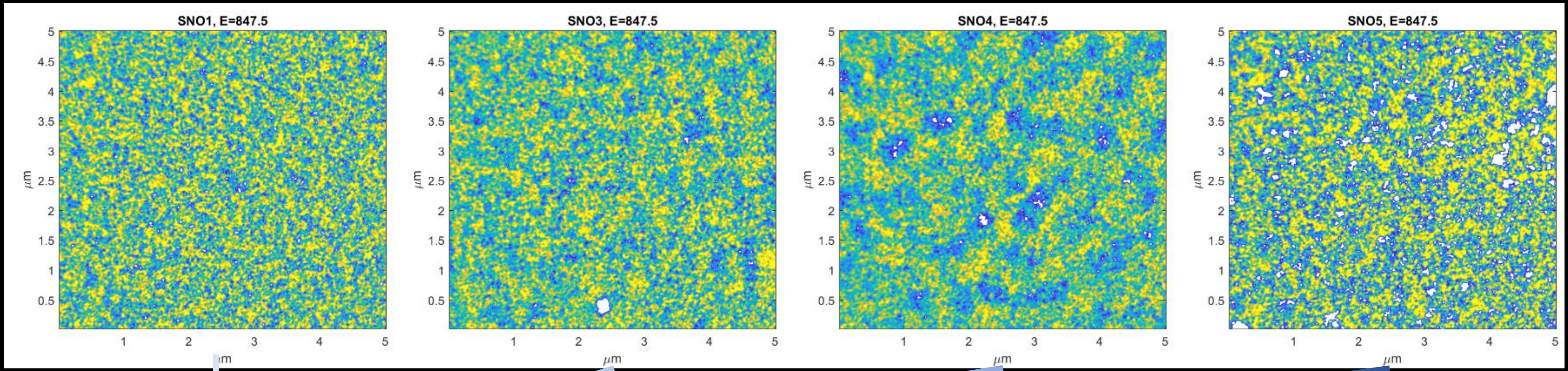
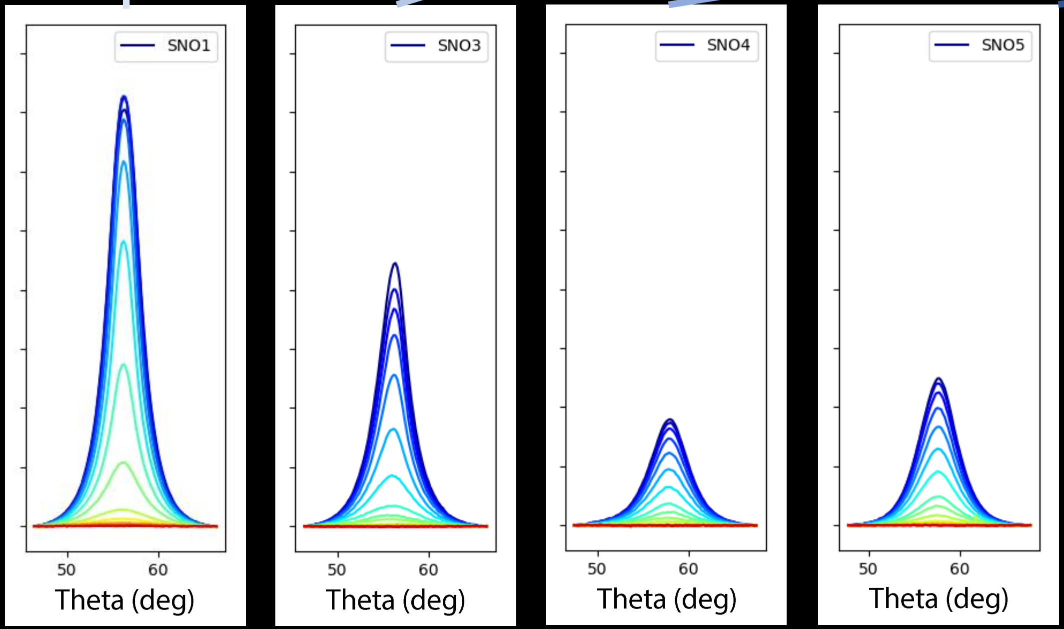


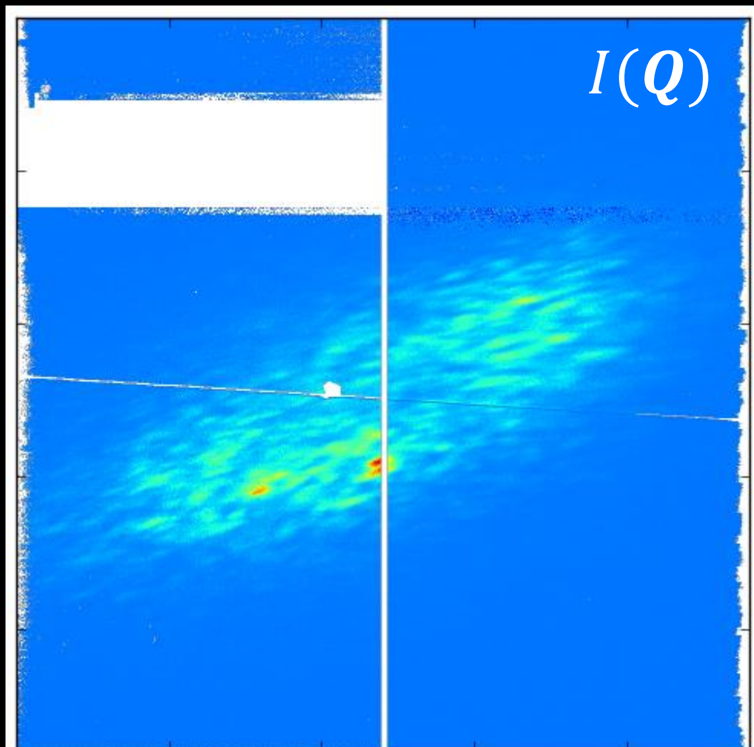


Doping induced phase separation as AFM order is suppressed.

The length scale of inhomogeneity is maximal when AFM order is weakest

Coherent Diffractive Imaging

RECIPROCAL SPACE → REAL SPACE



Coherent magnetic Bragg diffraction

Probe Function: E-field profile at the sample

$$F(\boldsymbol{Q}) = \mathcal{F}[O(\boldsymbol{r}) \cdot P(\boldsymbol{r})]$$

Object Function: what we are interested in

$$F(\boldsymbol{Q}) = \sqrt{I(\boldsymbol{Q})} \cdot \exp[i\phi(\boldsymbol{Q})]$$

From measurement

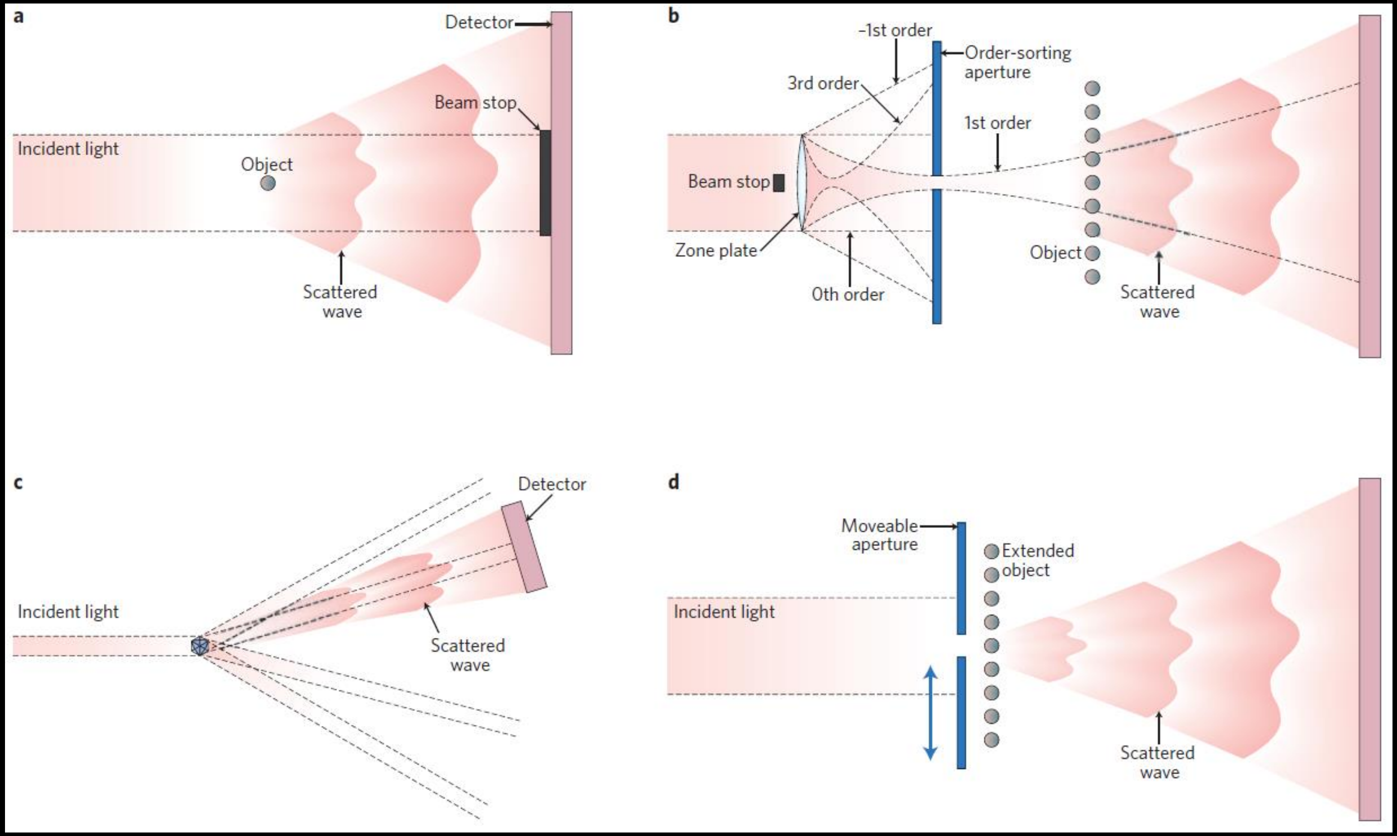
We need to recover the phases of the exit waves

COMPUTATIONAL IMAGING ←

Coherent Diffractive Imaging

Plane-wave CDI

Fresnel CDI



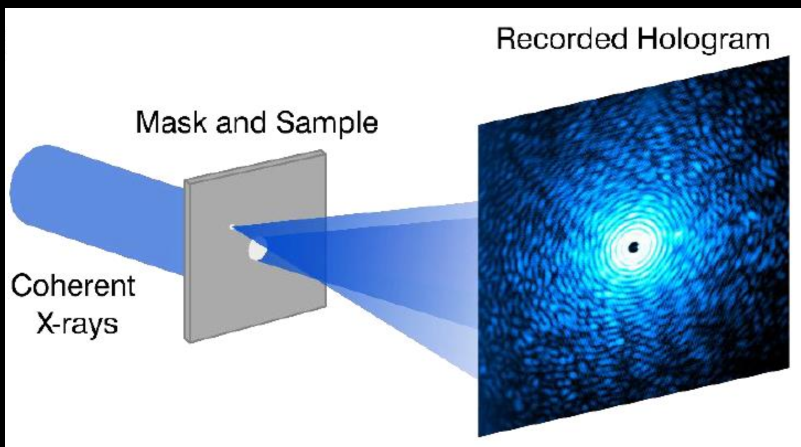
Bragg CDI

Ptychographic CDI

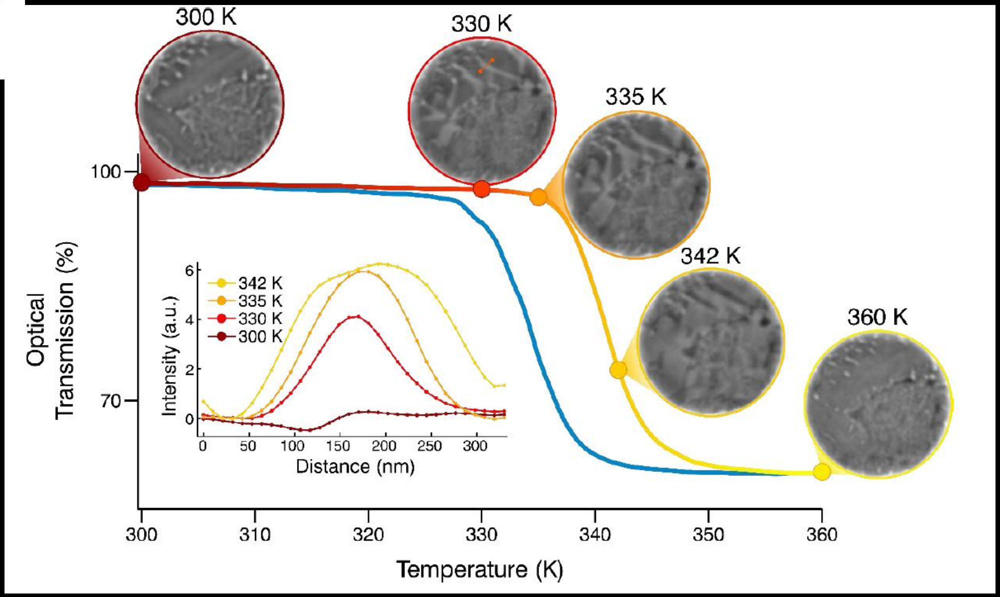
Chapman & Nugent, *Nature Photonics* 2010

Coherent Imaging

Coherent Diffractive Imaging



Metal-insulator transition in VO_2



Resonant holography

(few sec for a full hologram)

Vidas *et al.*, arXiv:1612.07998 (2016)

Coherent Imaging

Coherent Diffractive Imaging

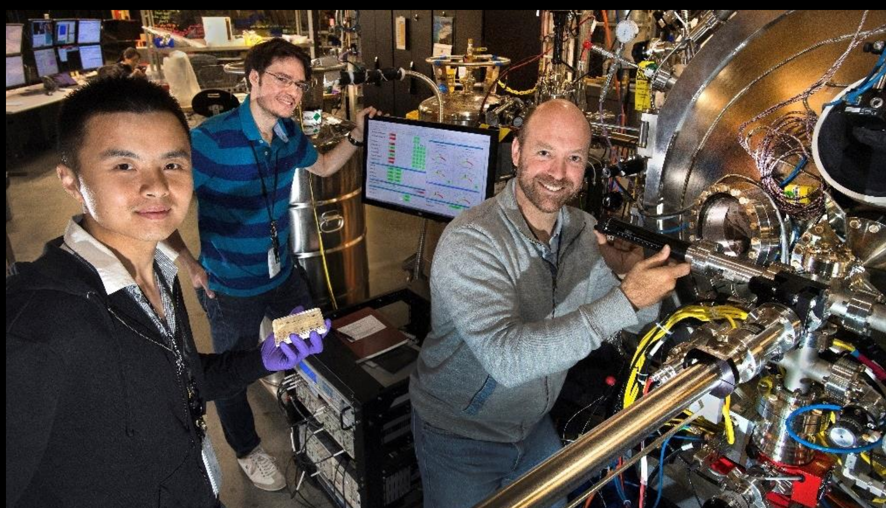
PROS

- Spatial resolution limited by NA of detector (10 nm)
- Fast acquisition (1-100 sec)
- No need for diffractive optics
- Depth resolution (3D Bragg CDI)

CONS

- Intensive computational effort (phase retrieval)
- Sometimes requires sample pre-patterning
- Beware dynamical scattering effects

Acknowledgments



MIT Photon Scattering Lab

Jiarui Li
Jonathan Pelliciari
Min Gu Kang
Zhihai Zhu
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