



Massachusetts
Institute of
Technology

Resonant X-ray scattering at the nanoscale

Riccardo Comin

Massachusetts Institute of Technology

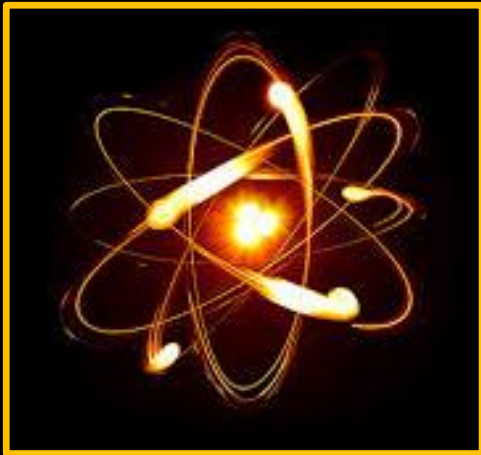
NSLS-II & CFN 2018 Users' Meeting
Brookhaven, 23 May 2018

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

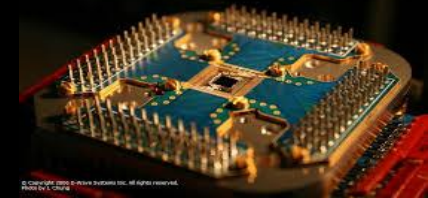
Medical imaging

Energy transport

Quantum sensing

Energy storage

Technology



Strongly-interacting quantum matter

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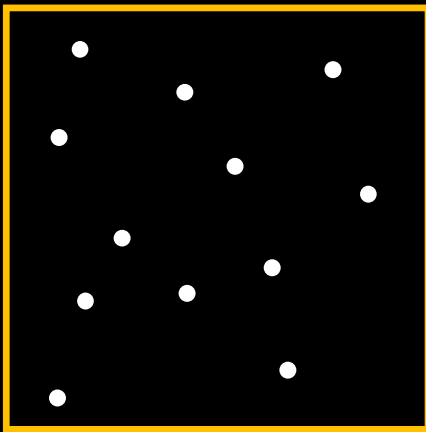
U/W

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Fermi Gas

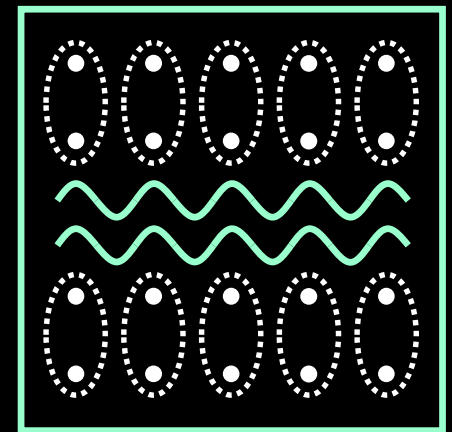
Emergent phenomena

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



Incoherent superposition
of single-particle w.f.

Symmetry Breaking

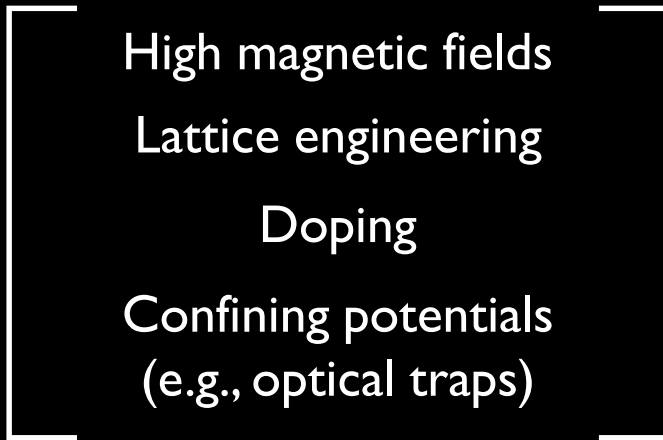


Many-body quantum order
(macroscale phase coherence)

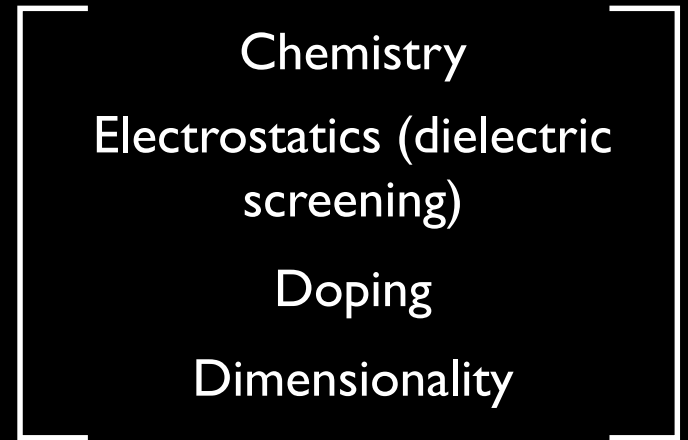
Strongly-interacting quantum matter



Reduce kinetic energy (W)



Increase interactions (U)



Strongly-interacting quantum matter



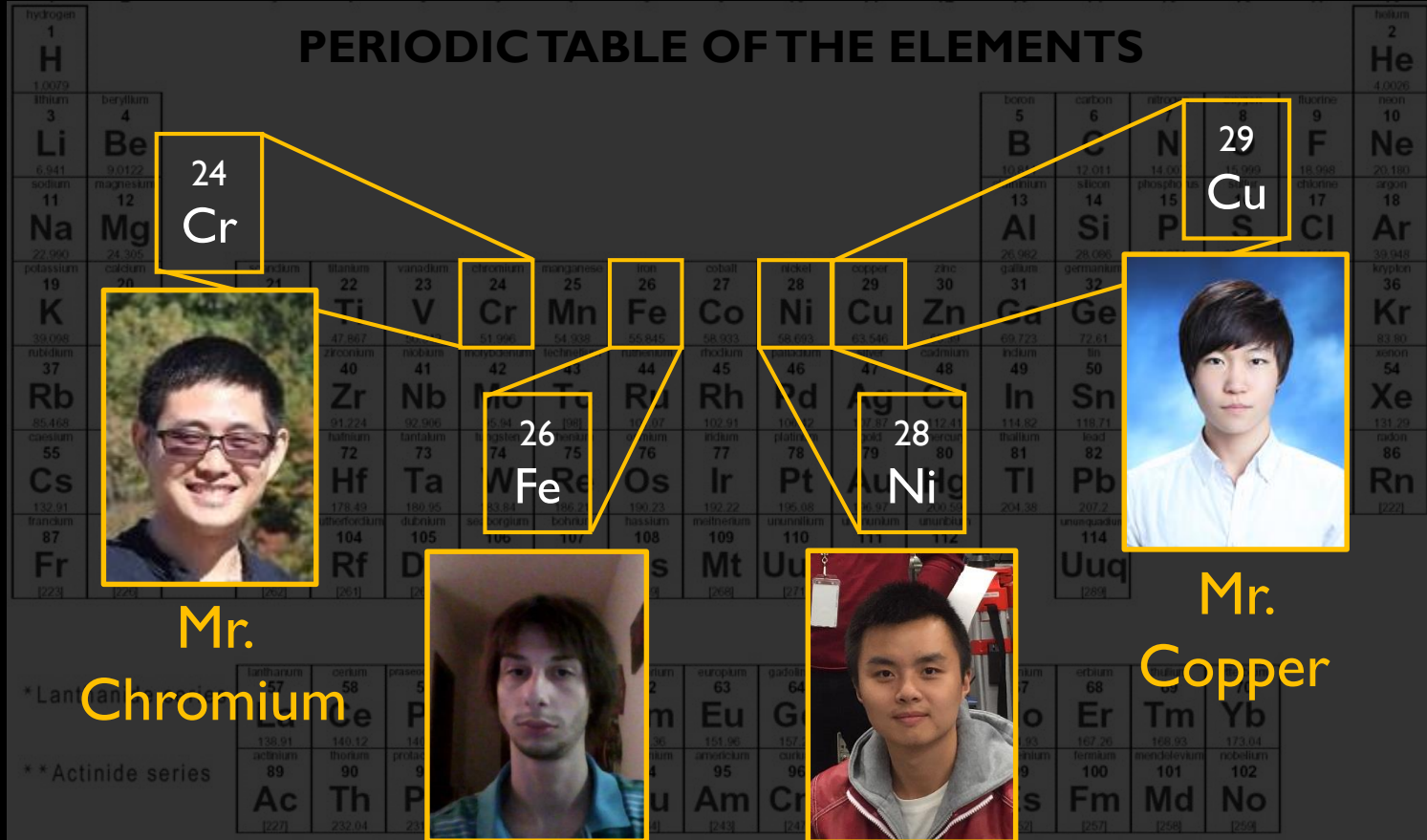
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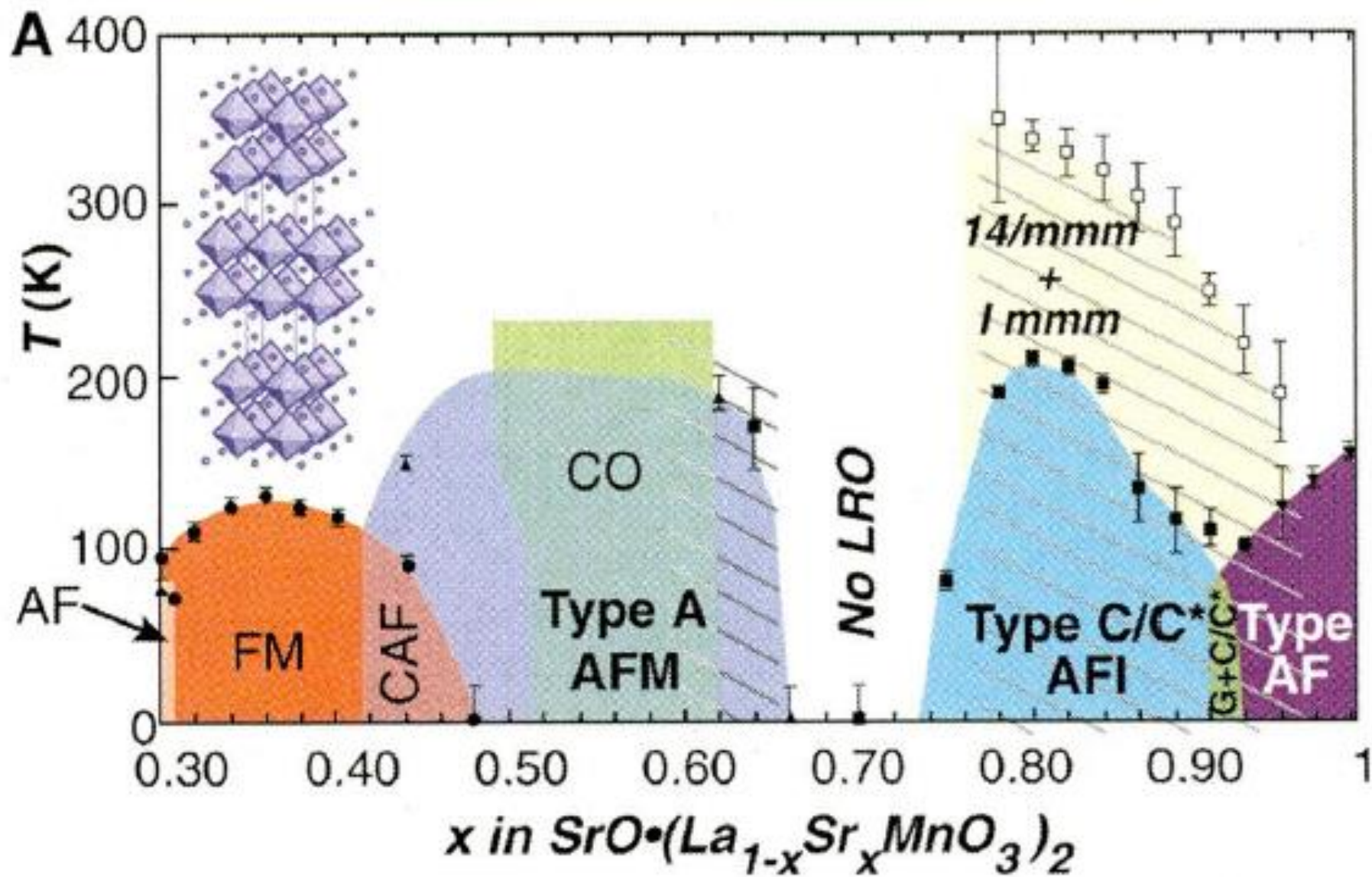
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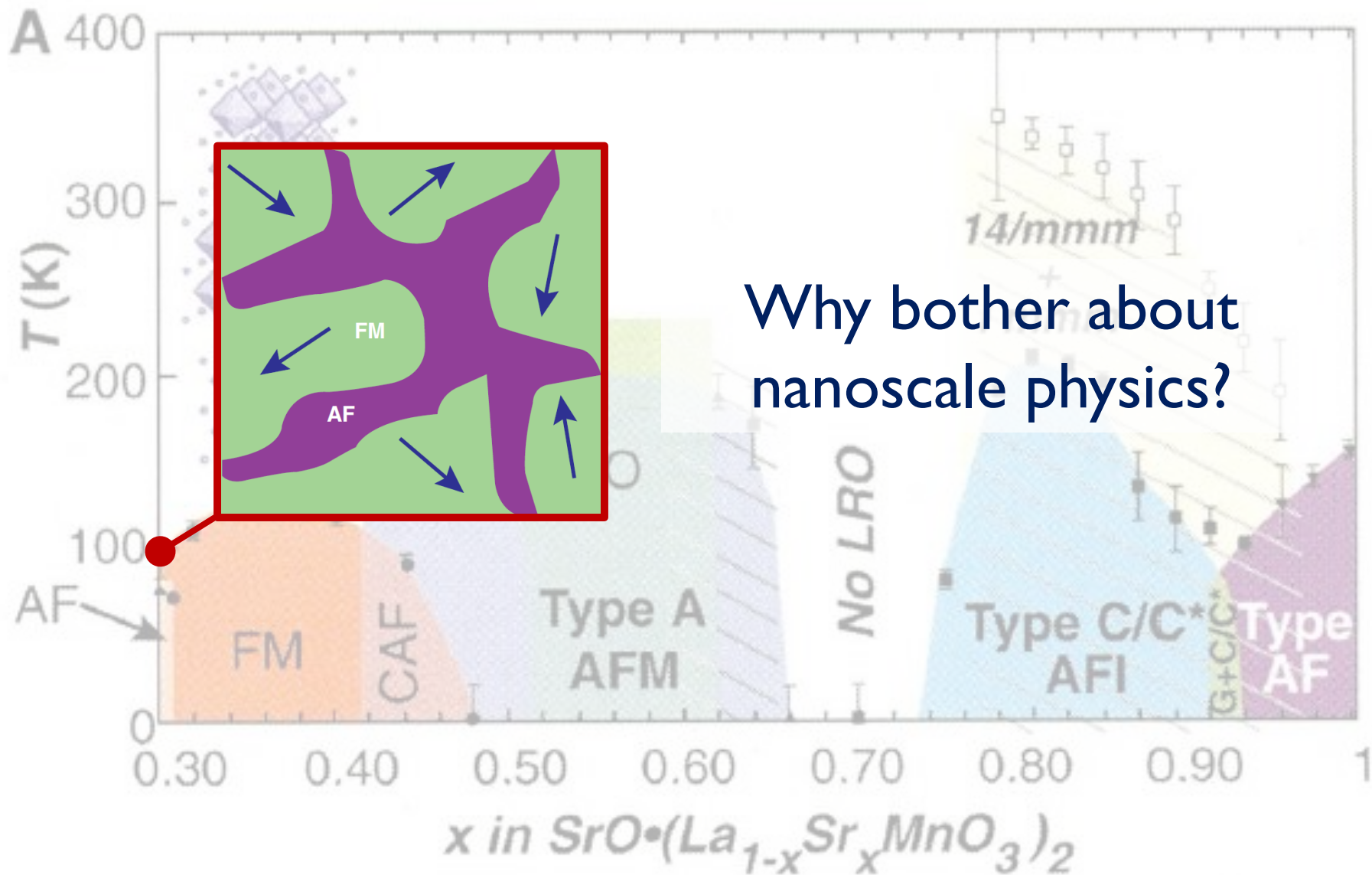
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hydrogen 1 H 1.0079																	helium 2 He 4.0026																																																																																																																																				
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sodium 11 Na 22.990	magnesium 12 Mg 24.305																																																																																																																																																				
potassium 19 K 39.098	calcium 20 Ca 40.078																																																																																																																																																				
rubidium 37 Rb 85.468	strontium 38 Sr 87.62																																																																																																																																																				
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Strongly interacting systems

Strong interactions (U) \longleftrightarrow Localized orbitals



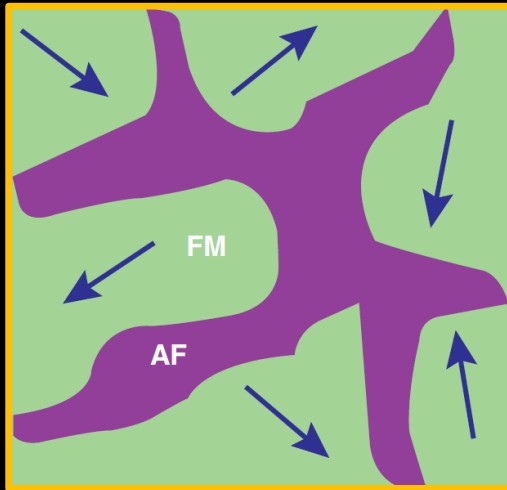




Why bother about
nanoscale physics?

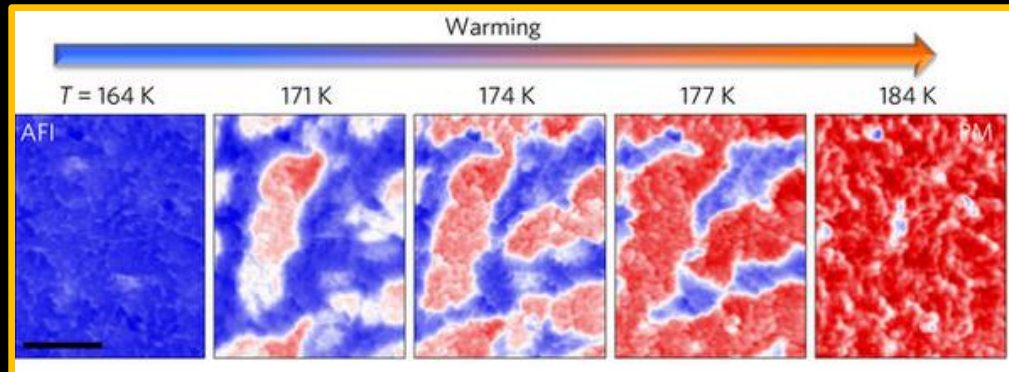
Strongly-interacting quantum matter

Phase segregation



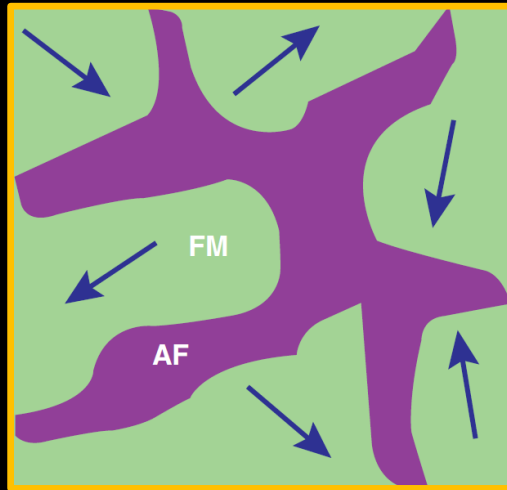
Quantum matter is almost inevitably inhomogeneous at the nanoscale

Percolation phenomena

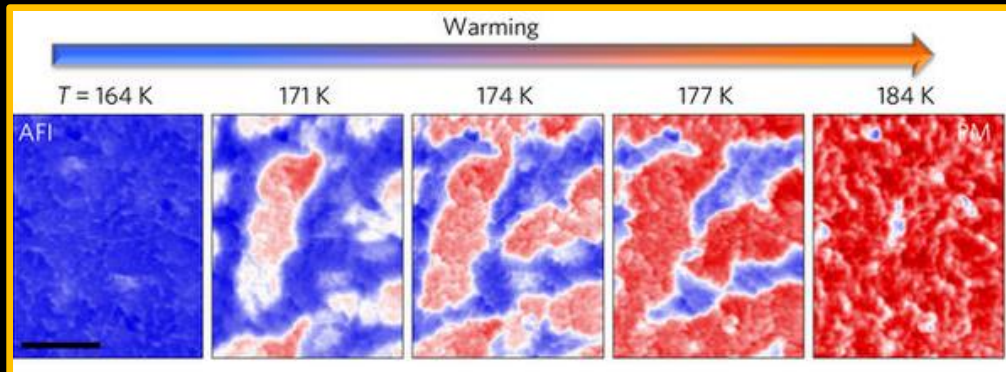


Strongly-interacting quantum matter

Phase segregation

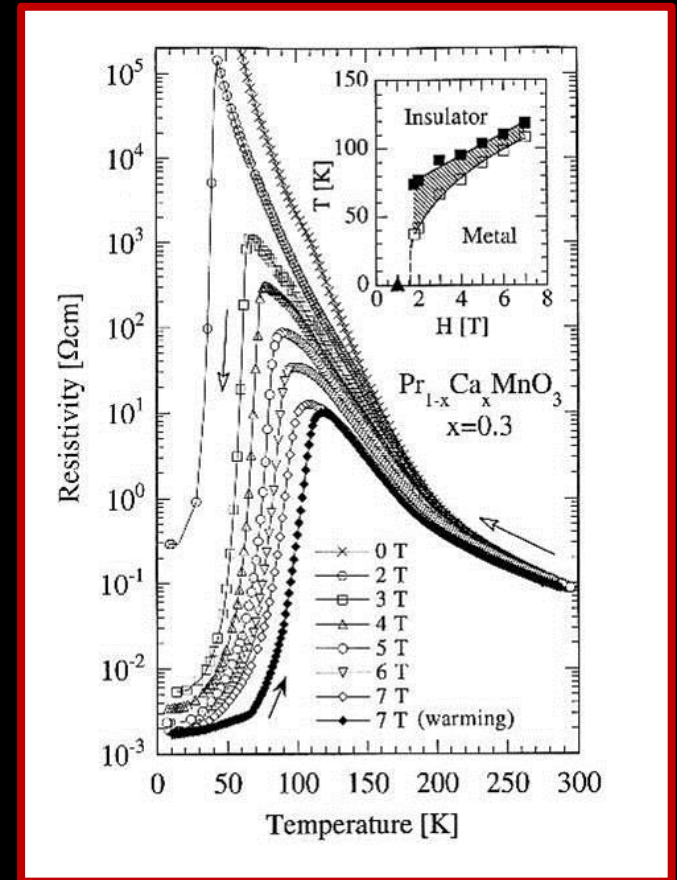


Percolation phenomena



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

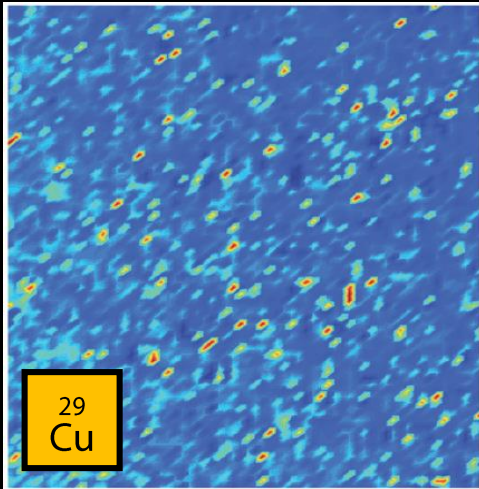
Colossal resistive switching



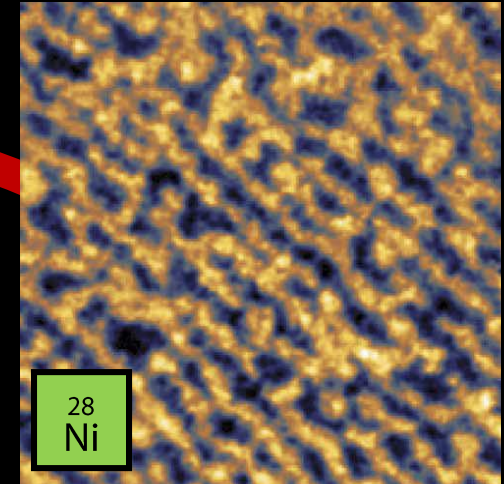
Y. Tomioka, et al., *Physics of Manganites* (1999)

Strongly-interacting quantum matter

Charge order



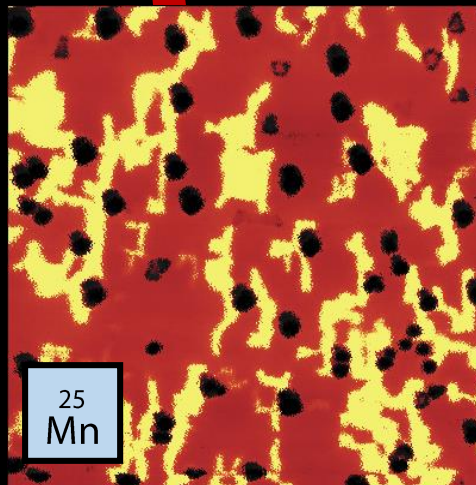
Metal-insulator transition



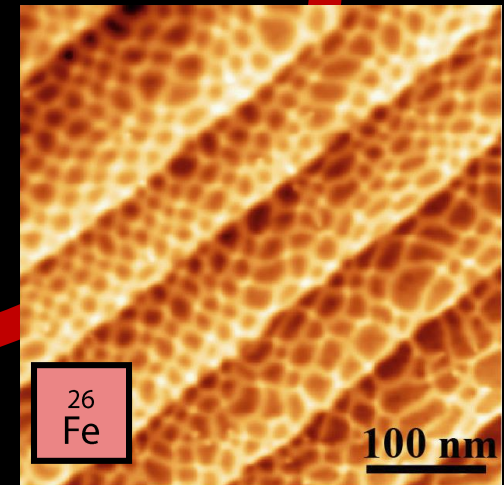
Emergent
functionalities



Nanoscale
textures



Spin order



Superconductivity

Scattering probes

Structure factor

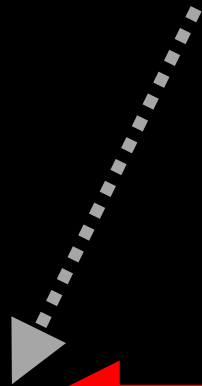
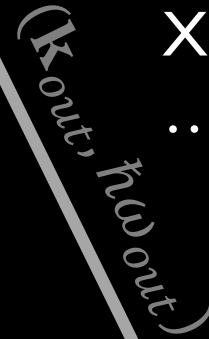
$$S(\mathbf{Q}, \omega) =$$

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

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

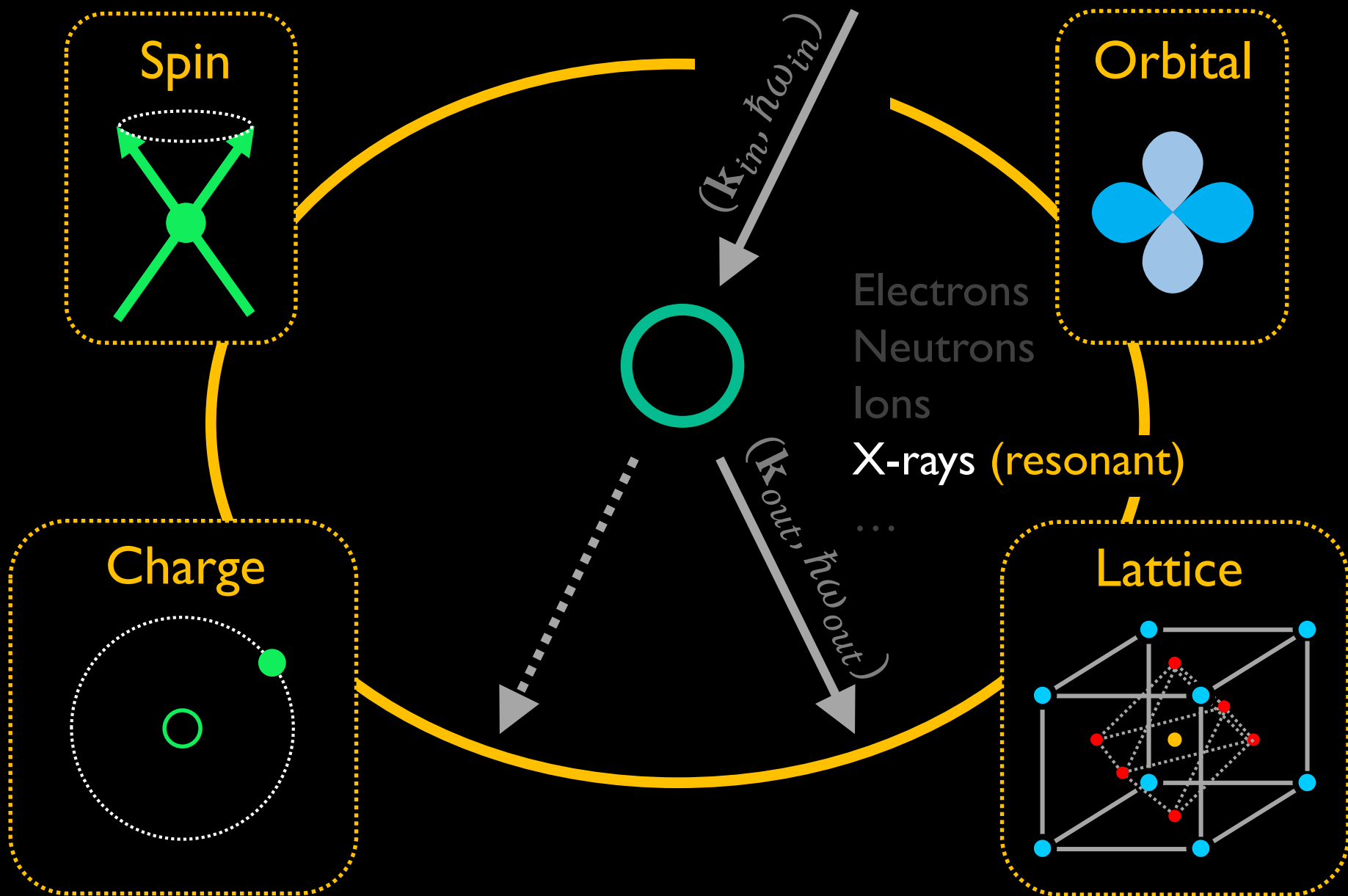


Electrons
Neutrons
Ions
X-rays
...



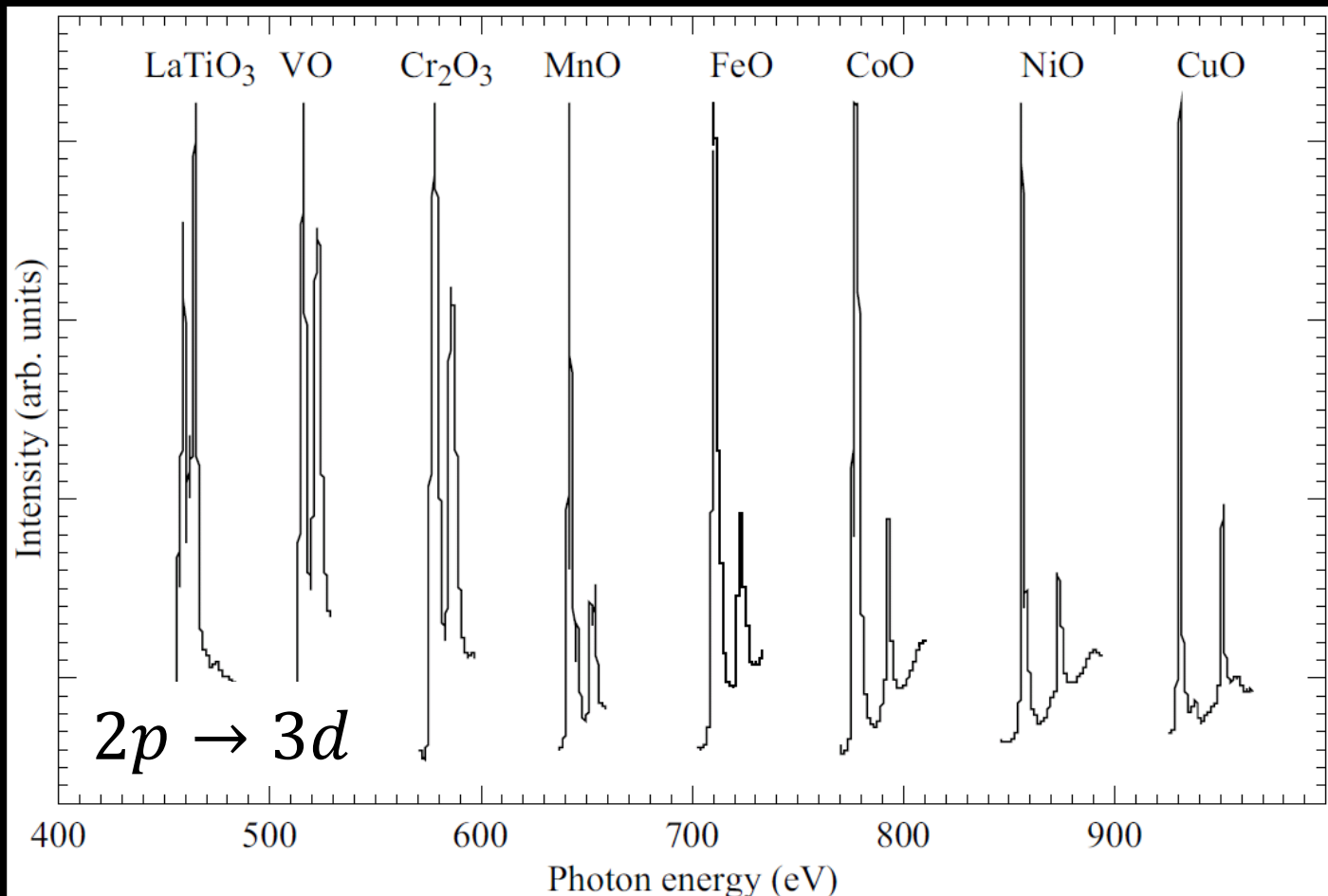
Momentum $\mathbf{Q} = \mathbf{k}_{in} - \mathbf{k}_{out}$
Energy $\omega = \omega_{in} - \omega_{out}$

Scattering probes



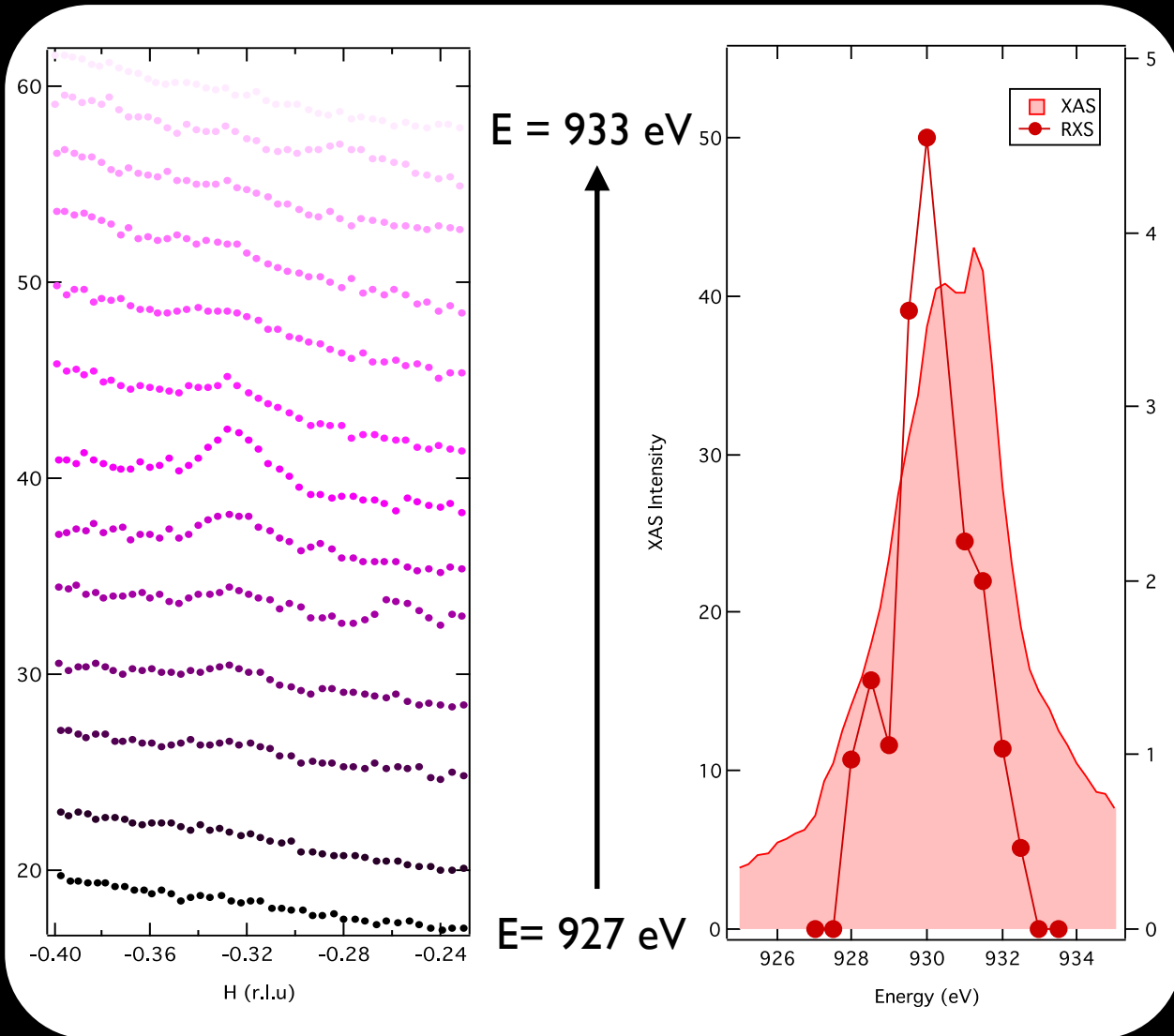
Resonant scattering

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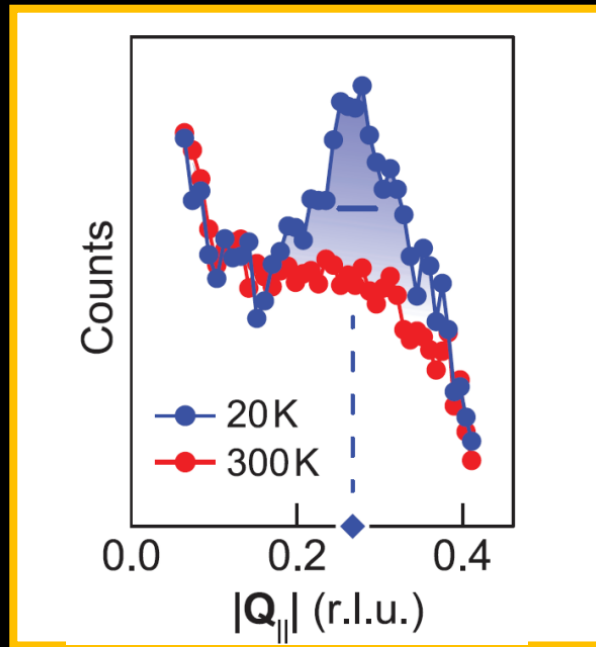
Resonant scattering

Charge-
density-
wave
(Nd_2CuO_4)



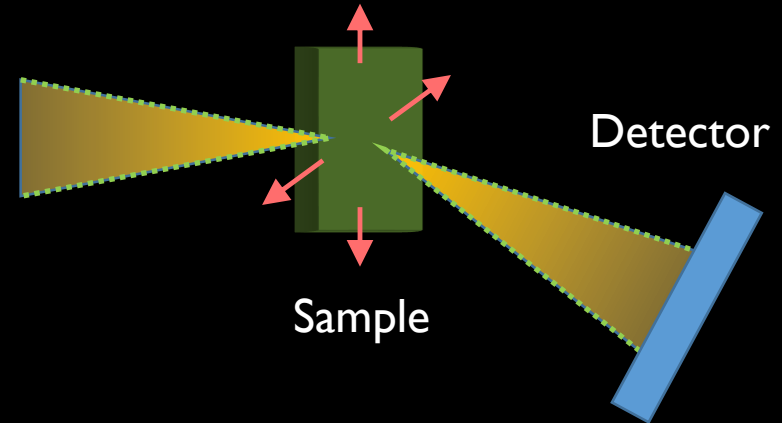
Electronic orders at the nanoscale

Reciprocal space (scattering)

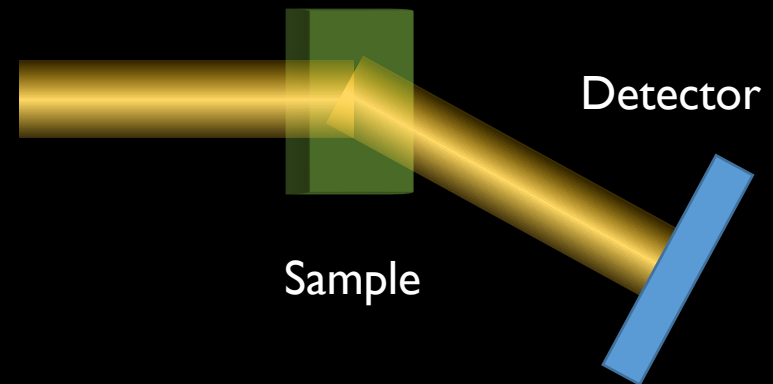


HOW

Scanning nanodiffraction (50 nm)



Full-field lensless imaging (10 nm)



Recover real-space information

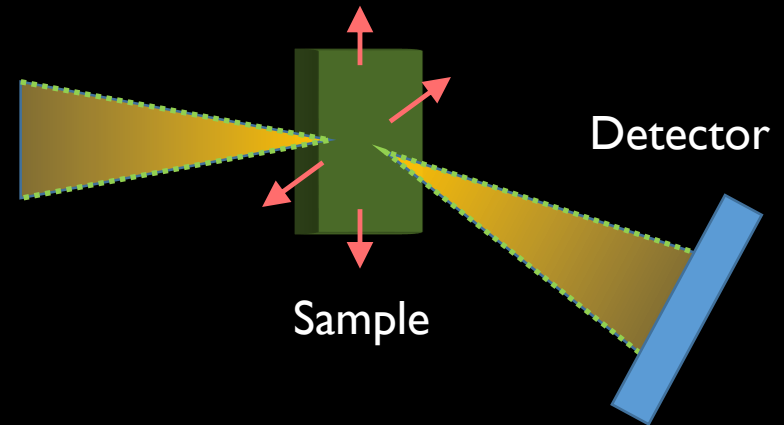
Electronic orders at the nanoscale

WHY

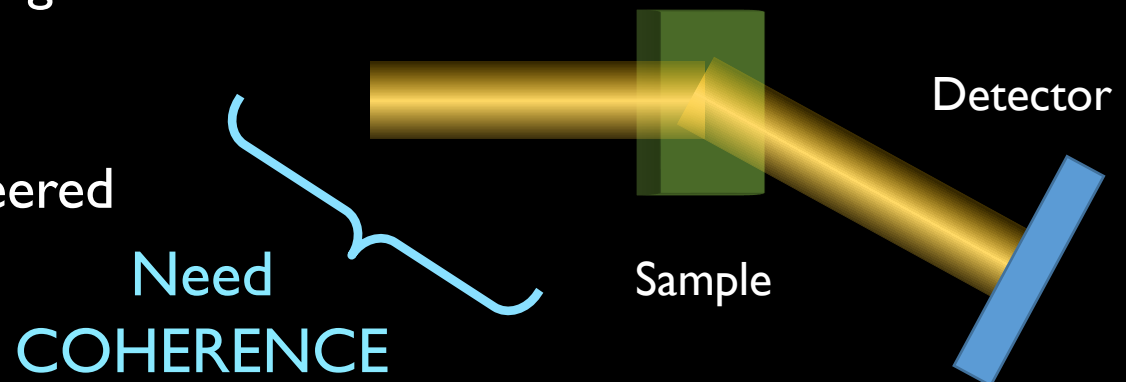
- Nanoscale granularity:
 - Intrinsic (phase competition & segregation)
 - Extrinsic (disorder, defects, doping, ...)
- Scale-invariant phenomena:
 - Extended range of length scales (10 nm to 10 μm)
- Emergent physics at the edge or boundary:
 - Domain walls; lateral interfaces; nanoengineered structures

HOW

Scanning nanodiffraction (50 nm)

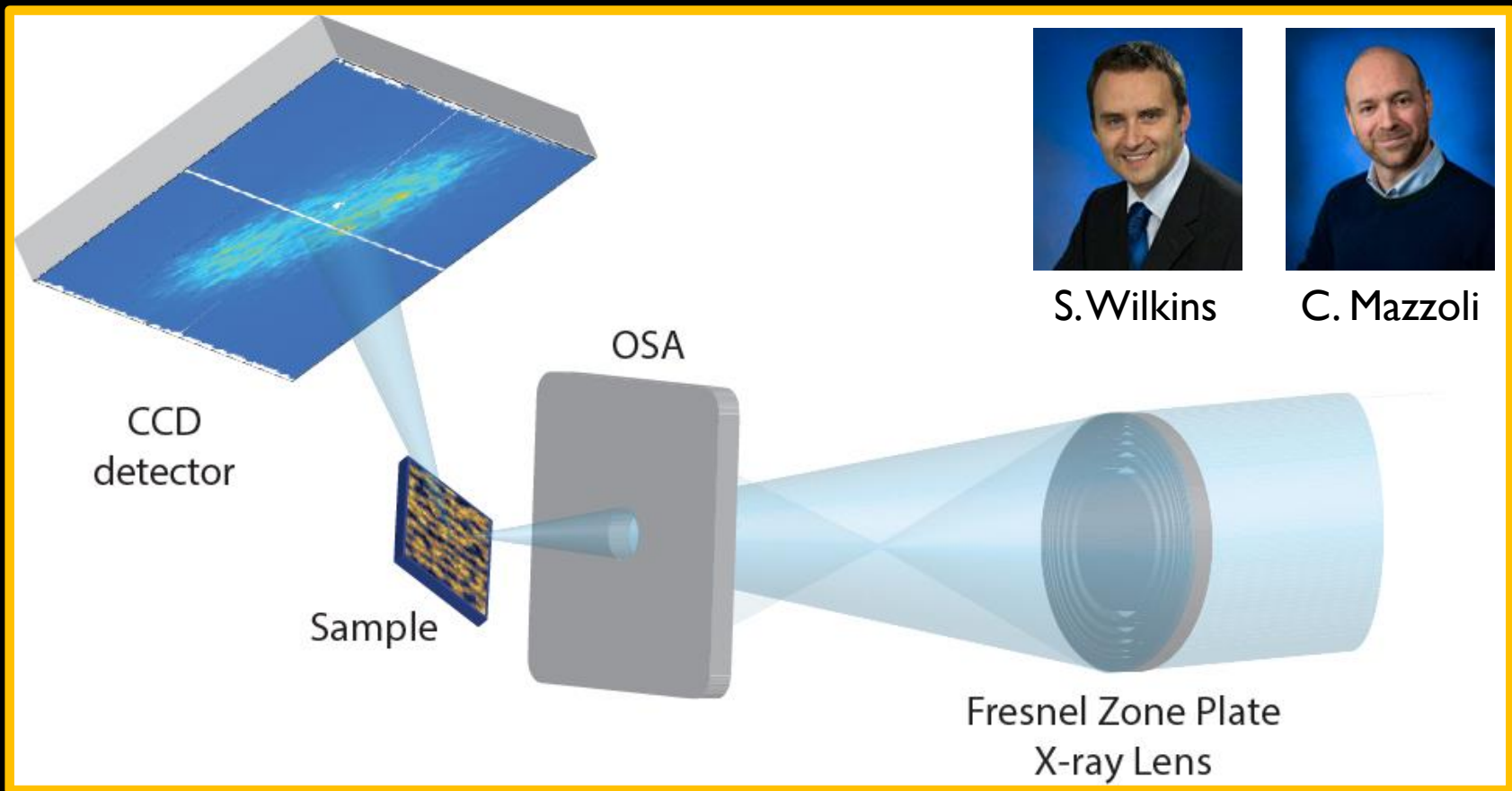


Full-field lensless imaging (10 nm)



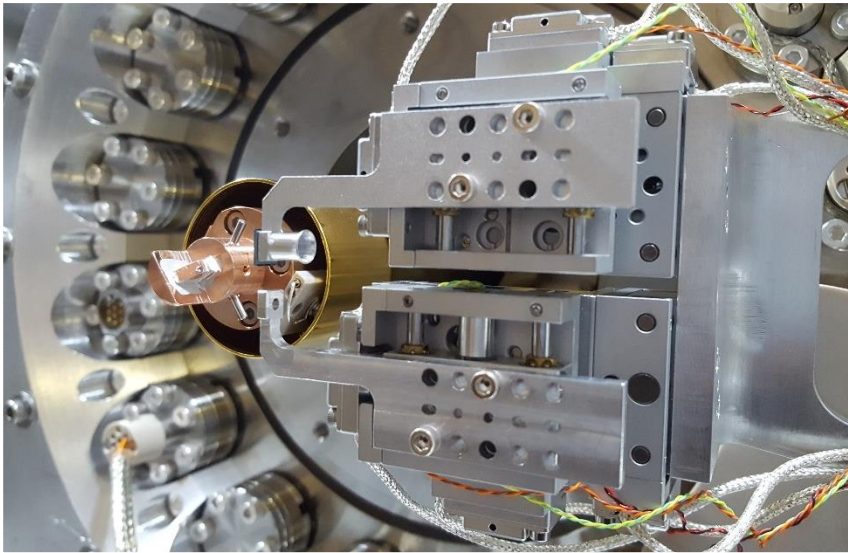
Soft X-ray nanodiffraction

Scanning resonant nanodiffraction

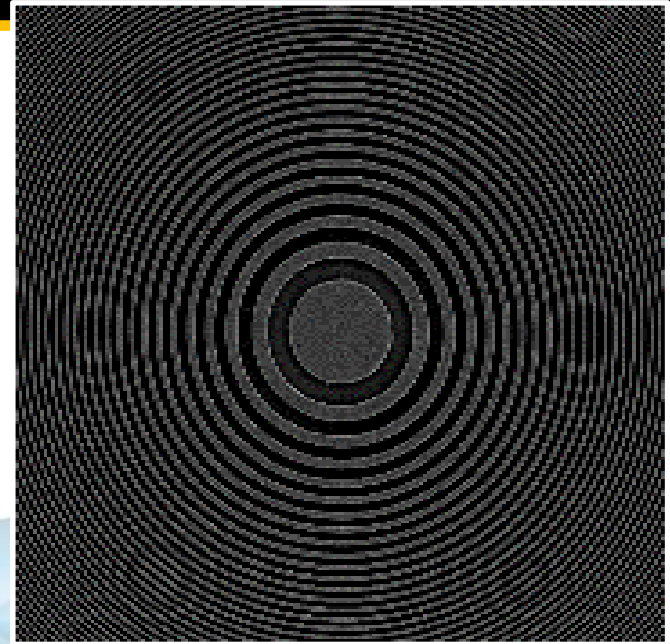


Soft X-ray nanodiffraction

Scanning resonant nanodiffraction



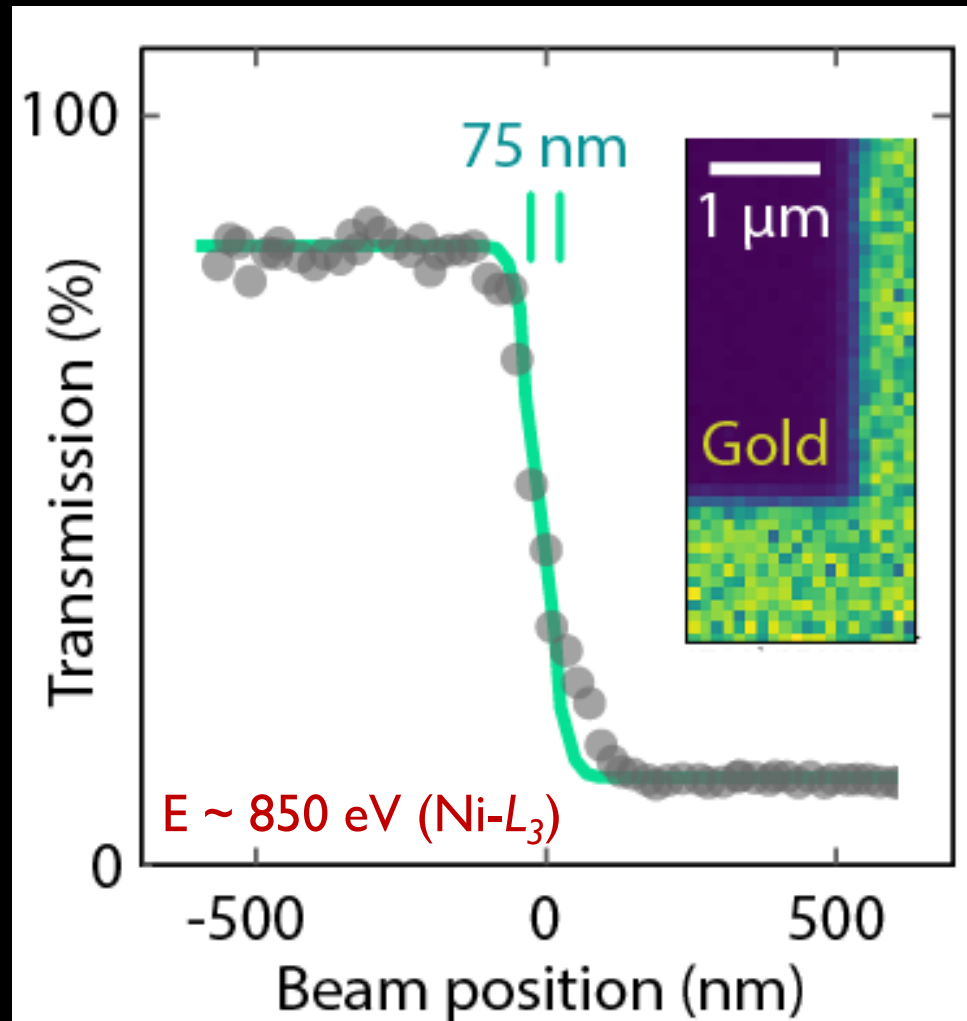
sample



Fresnel Zone Plate
X-ray Lens

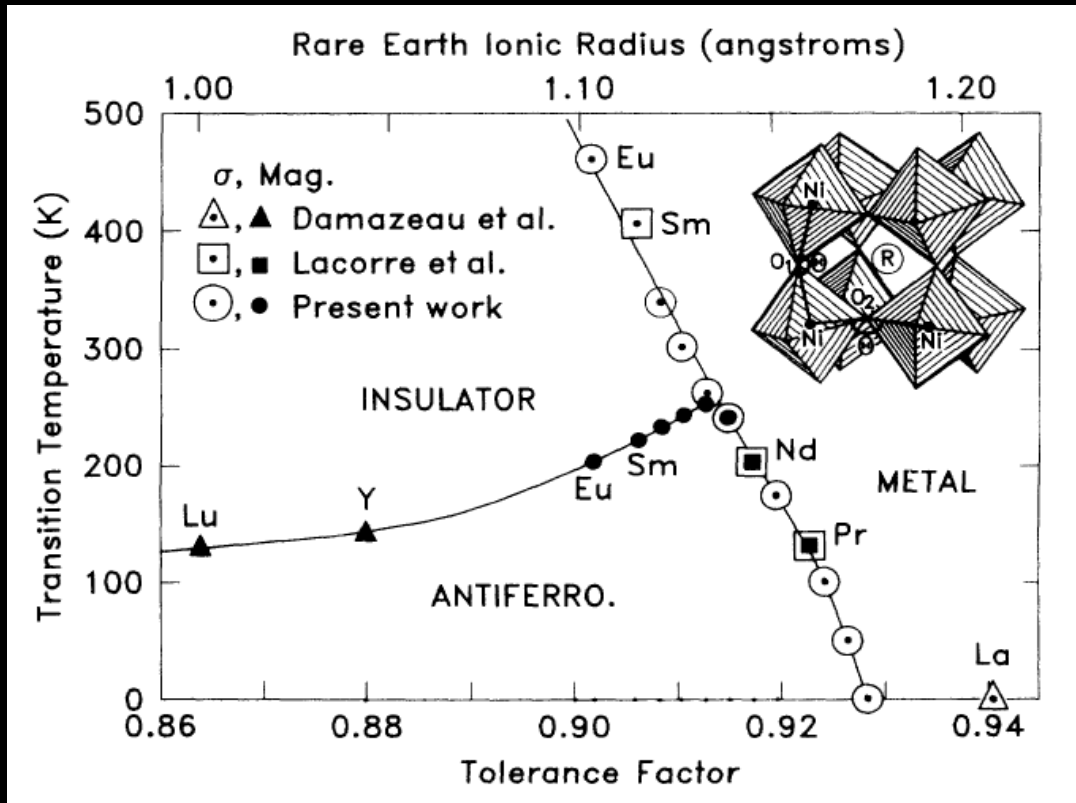
Soft X-ray nanodiffraction

Zone-plate focusing optics: 75 nm spot size

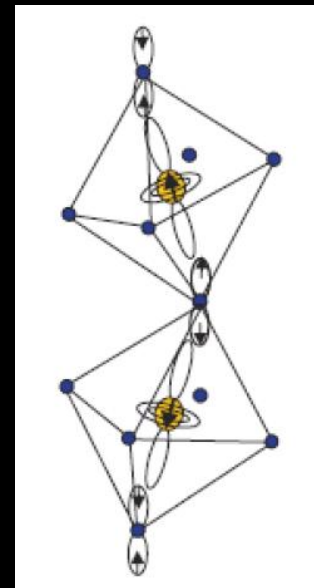


Nickelates

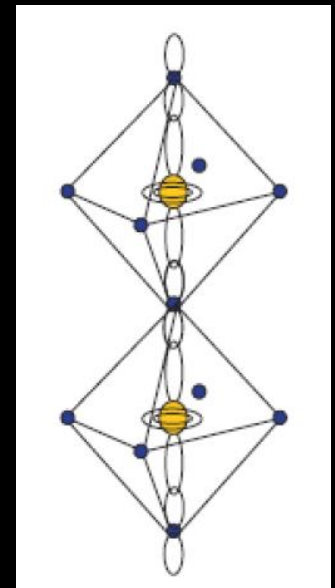
Cubic nickelate perovskites ($RNiO_3$)



Low-T
(insulator)



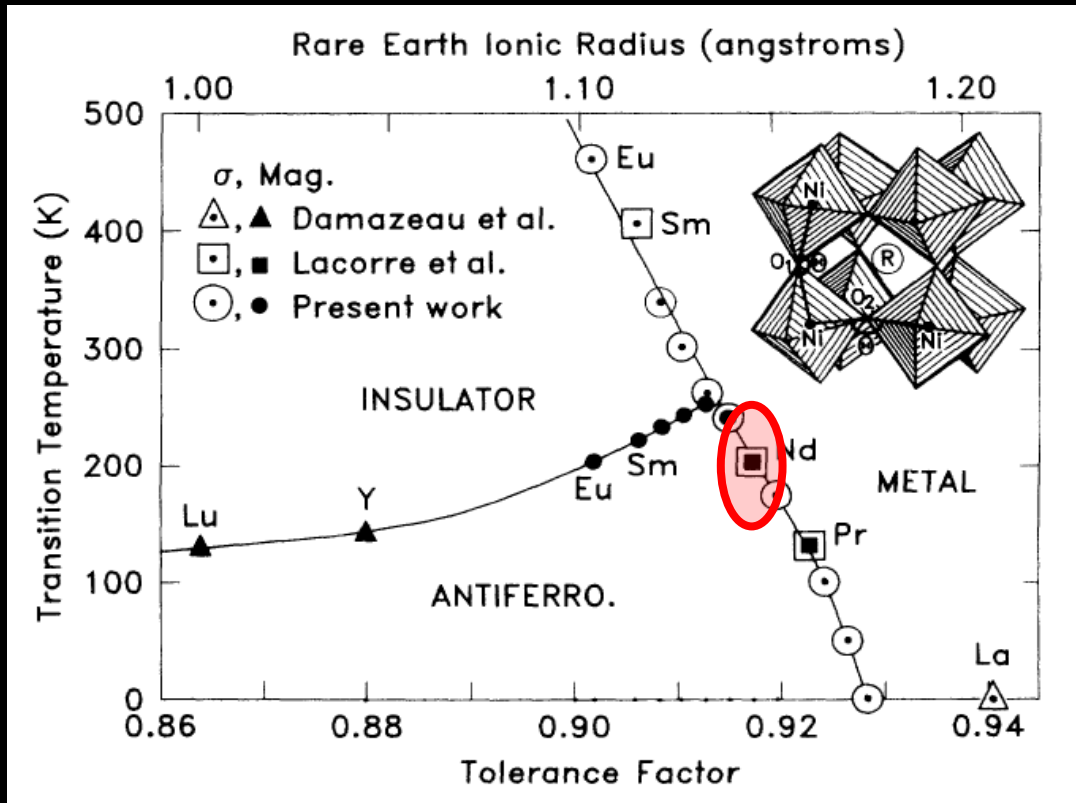
High-T
(metal)



Torrance et al. PRB 1992

Nickelates

Cubic nickelate perovskites ($RNiO_3$)

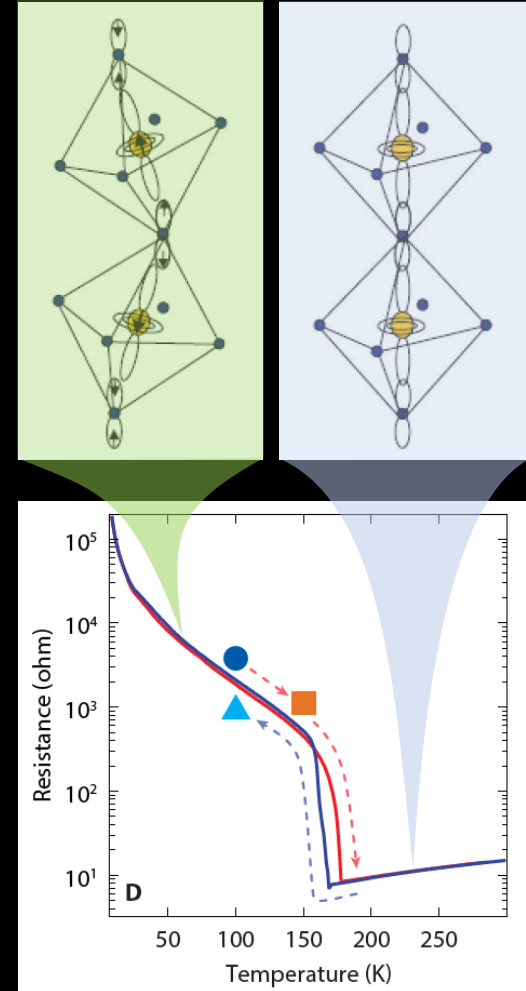


Torrance et al. PRB 1992

$NdNiO_3$

Low-T
(insulator)

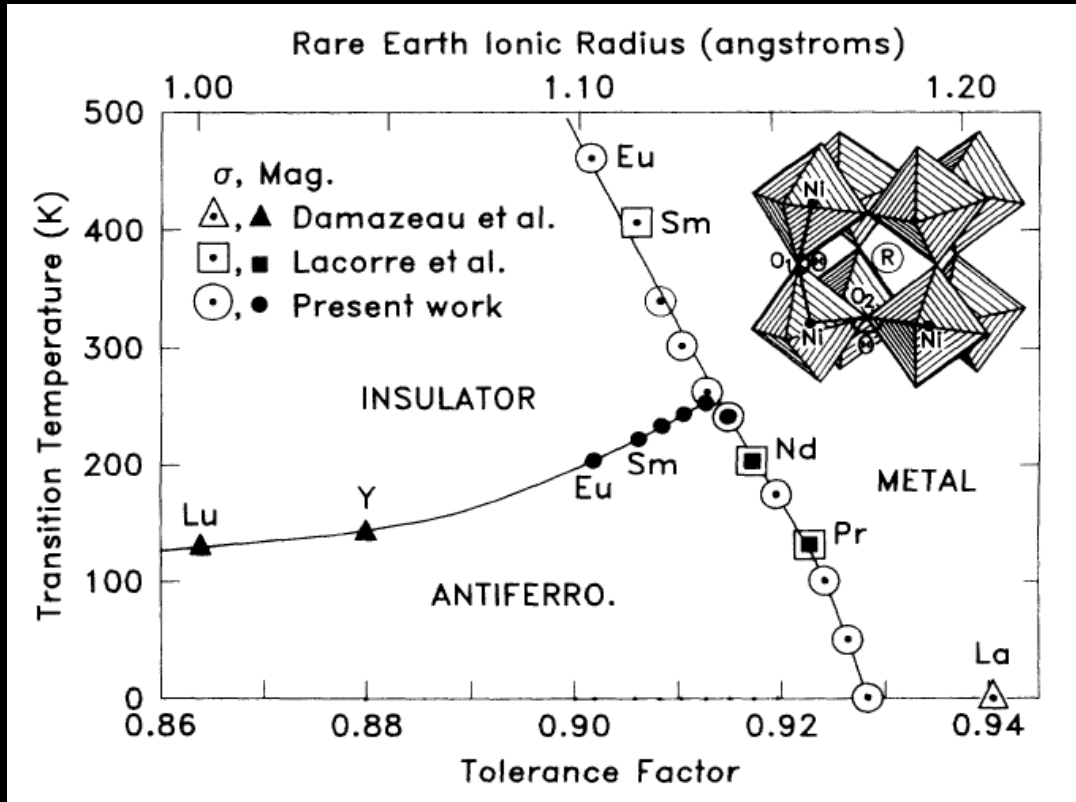
High-T
(metal)



Metal insulator transition

Nickelates

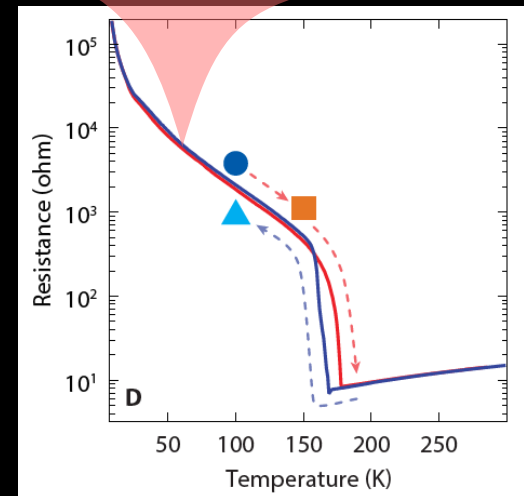
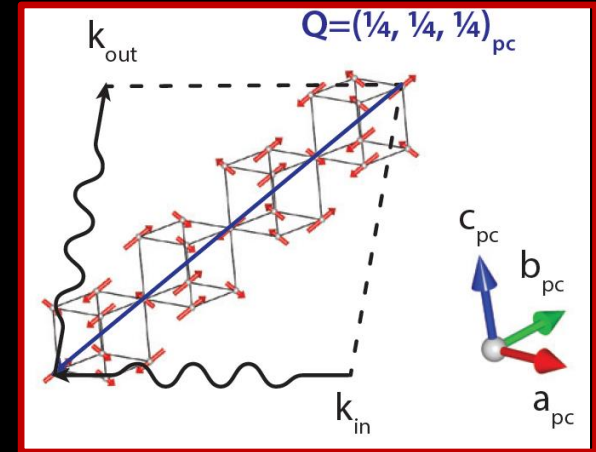
Cubic nickelate perovskites ($RNiO_3$)



Torrance *et al.* PRB 1992

Goal: map antiferromagnetic order across metal-insulator transition

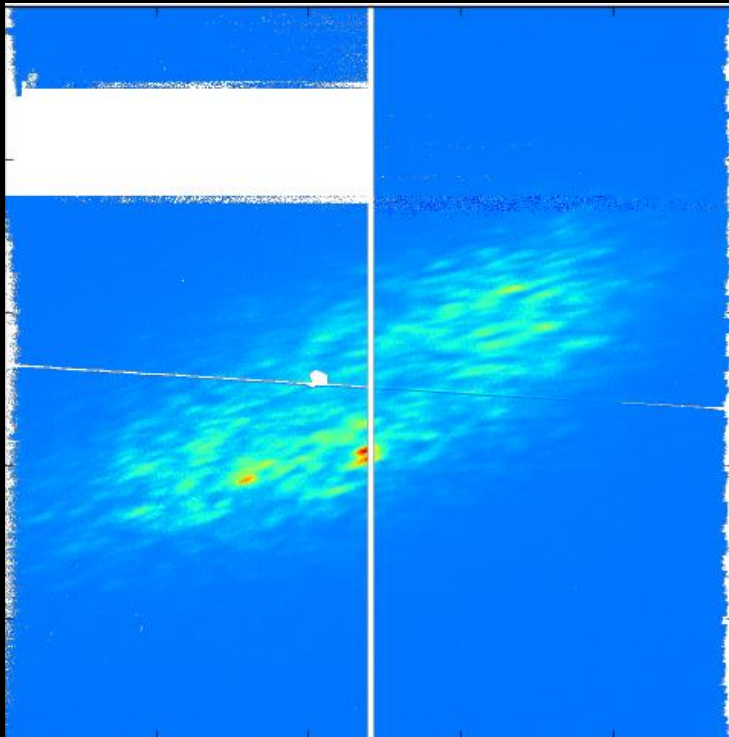
Magnetic order



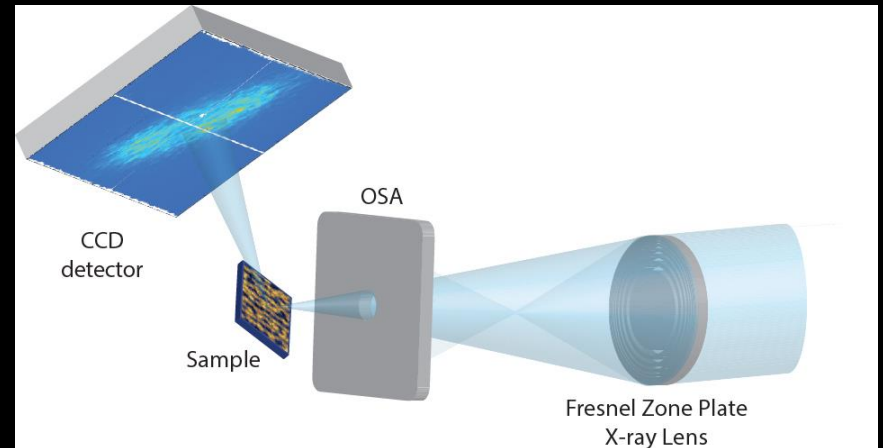
Metal insulator transition

Nickelates

RECIPROCAL SPACE



Coherent magnetic scattering from
spin-density wave in NdNiO_3



Johnny Pelliciarì

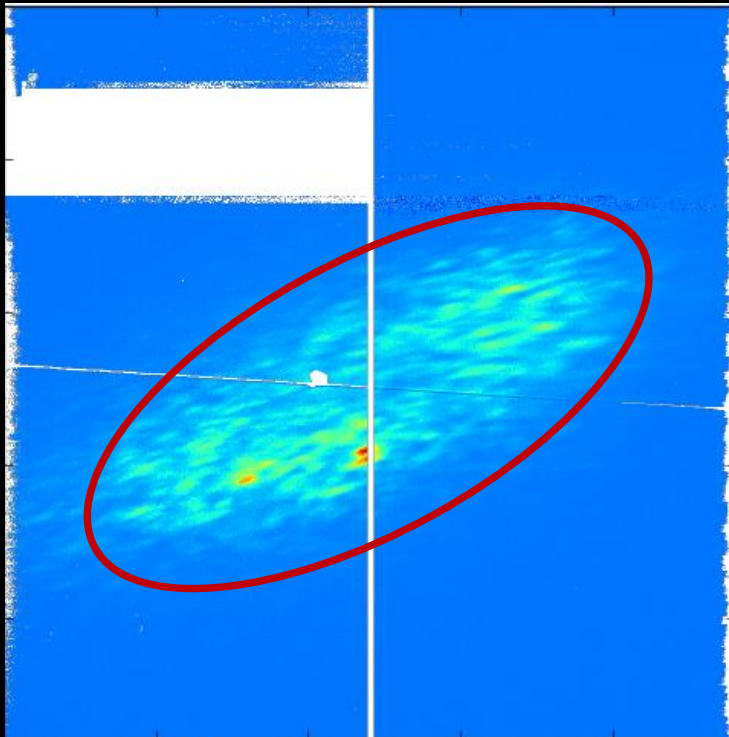


Jiarui Li



Nickelates

RECIPROCAL SPACE



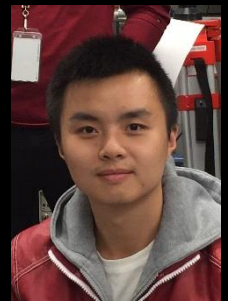
Speckle pattern: coherent interference between magnetic domains

Coherent magnetic scattering from spin-density wave in NdNiO_3

Johnny Pelliciani



Jiarui Li



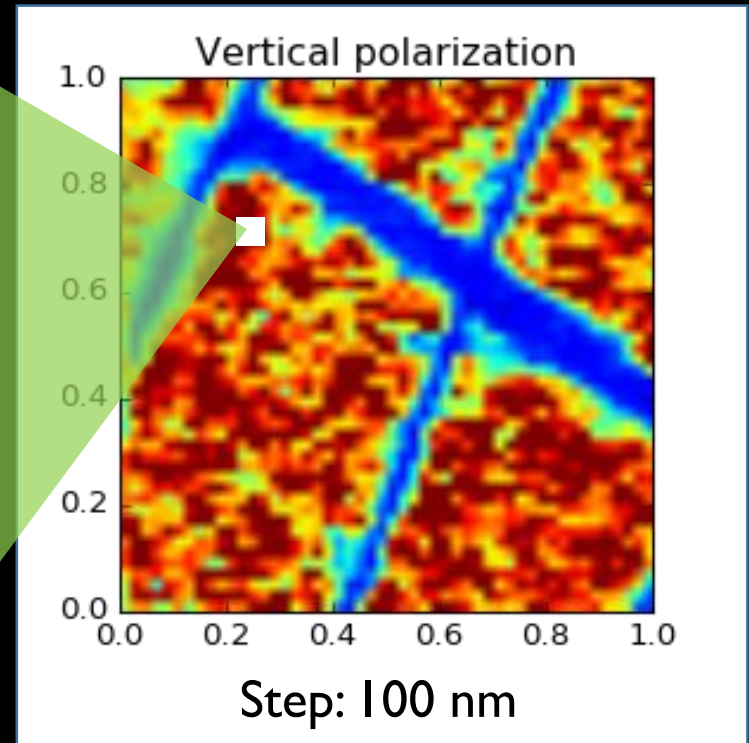
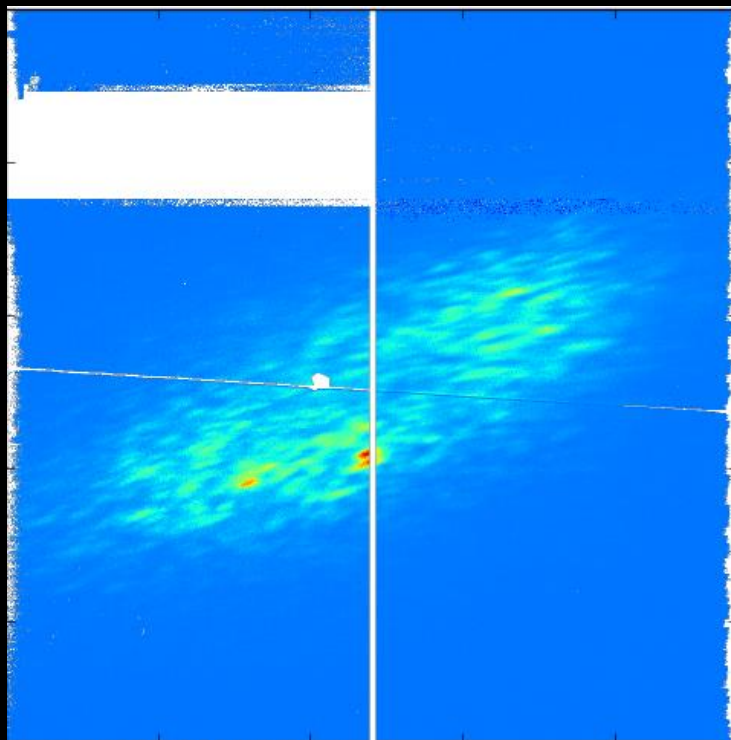
Nickelates

First case study: spin-density-wave in NdNiO₃ thin films

RECIPROCAL SPACE



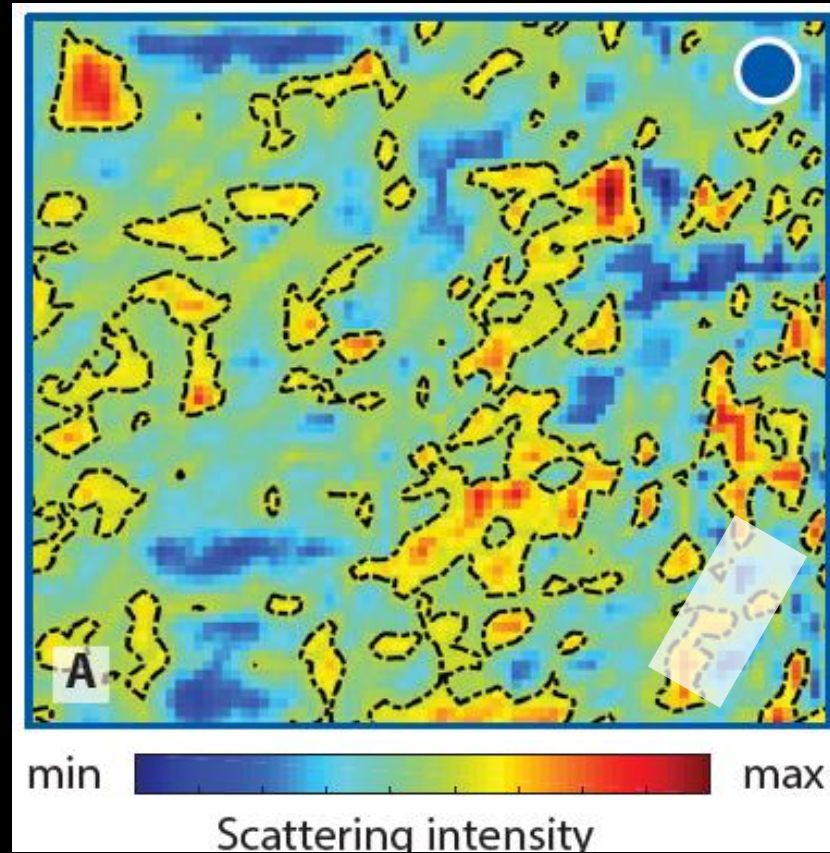
REAL SPACE (mapping)



Coherent magnetic scattering from spin-density wave in NdNiO₃

Nano-mapping of order parameter

Nickelates

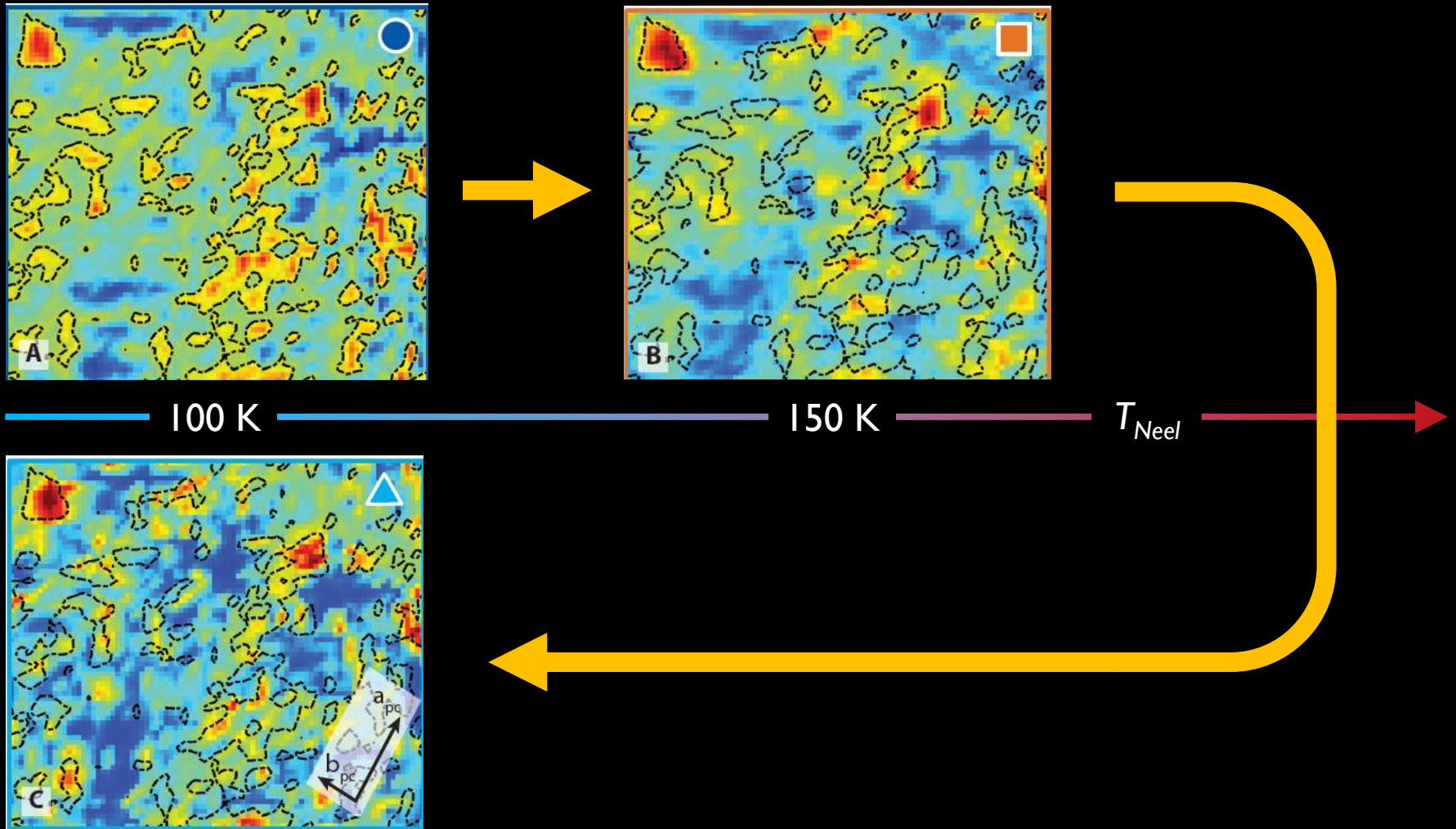


SDW order
parameter

$1 \times 1 \mu\text{m}^2$
square

Nanoscale inhomogeneity on length scales $0.1 - 10 \mu\text{m}$

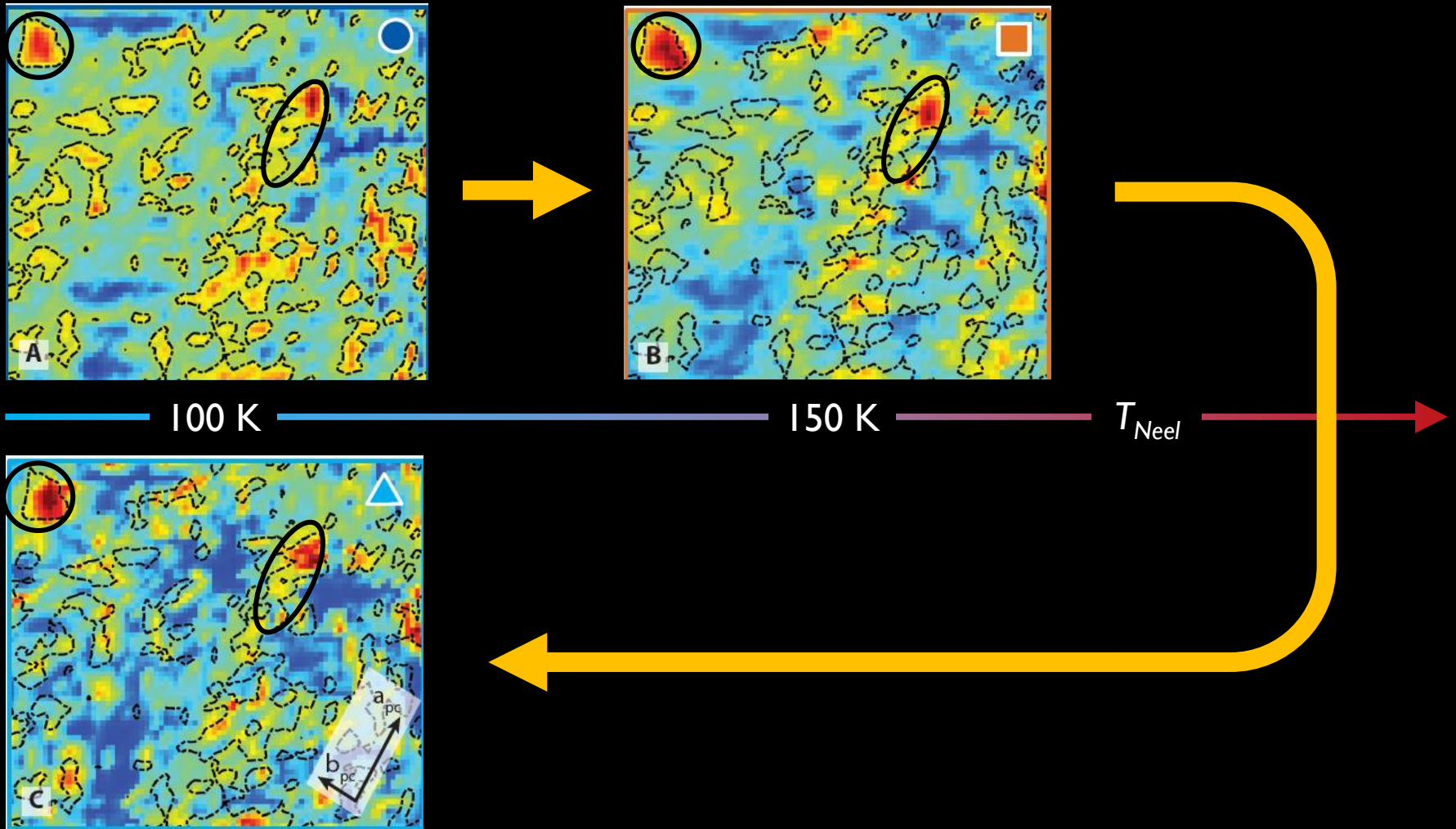
Nickelates



Domain pinning = memory effect

Possibly a hidden local parameter controlling domain nucleation

Nickelates



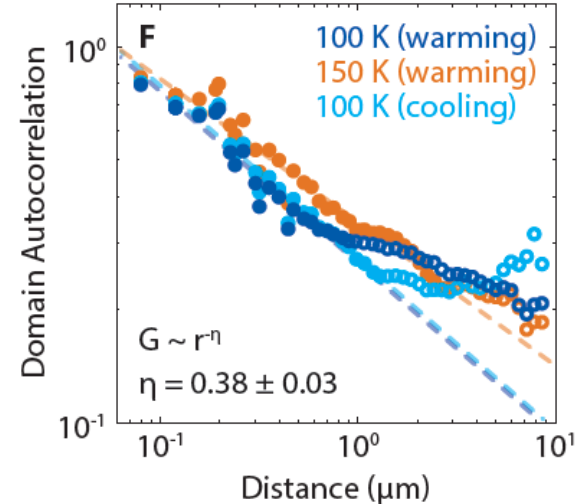
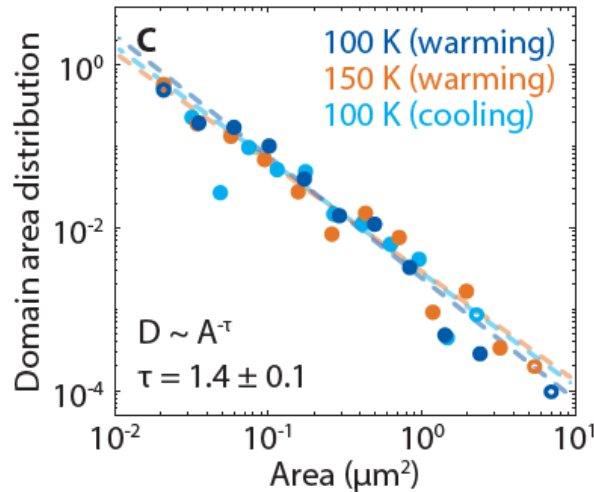
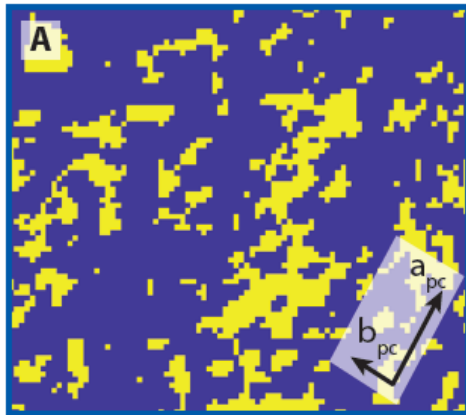
Domain pinning = memory effect

Possibly a hidden local parameter controlling domain nucleation

Nickelates

Scale-invariant textures – a **fractal** magnetic landscape

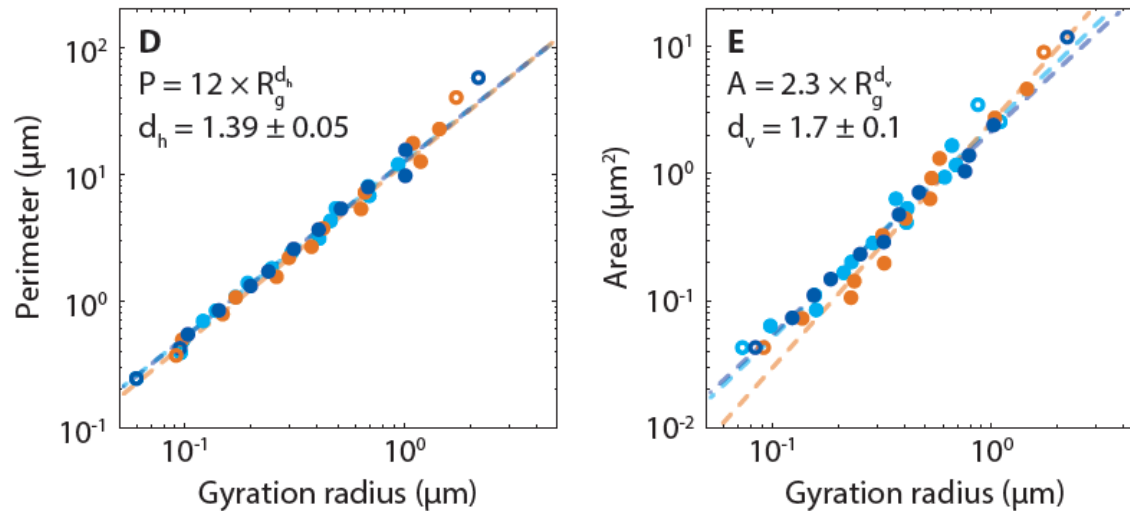
100 K (warming)



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

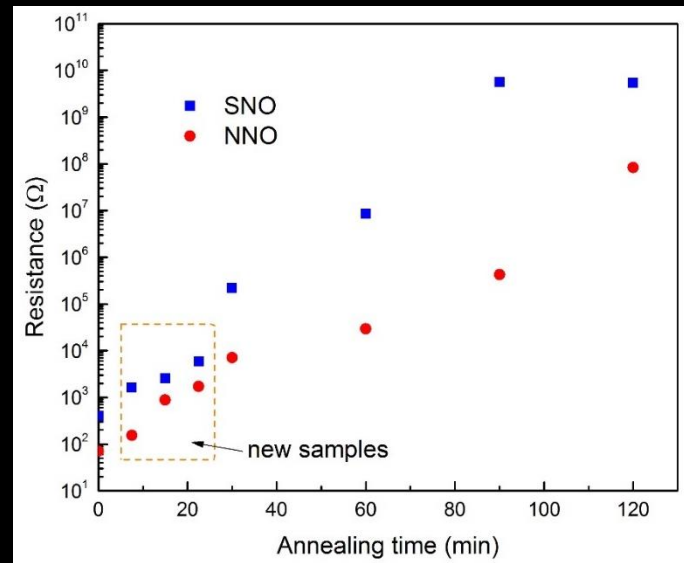
PROS

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

CONS

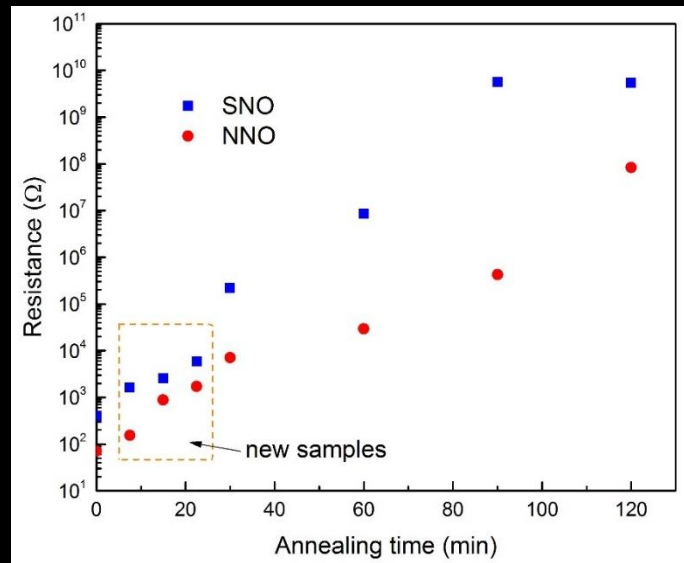
- Slow, point-by-point scanning required (1-4 hrs)
- Spatial resolution limited by NA of optics (not detector)
- 10-15% throughput across ZP lenses

Doped nickelates

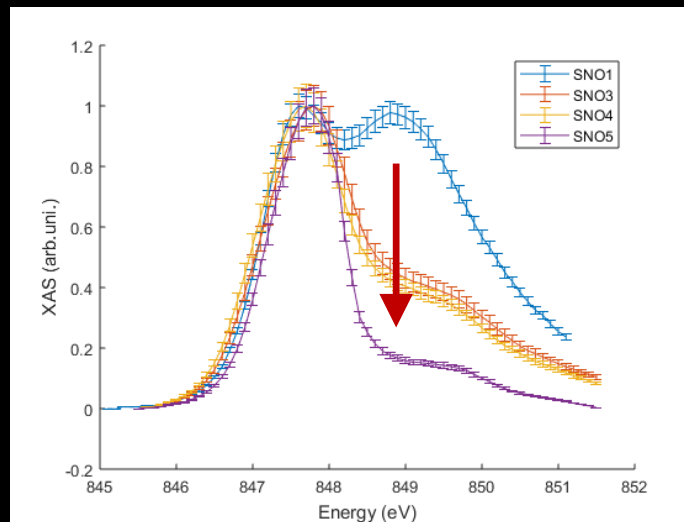


Electron-doping induced a colossal metal-insulator transition

Doped nickelates



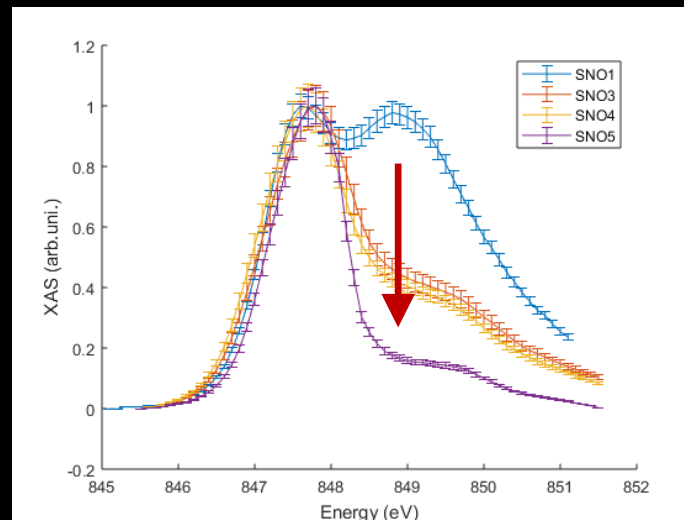
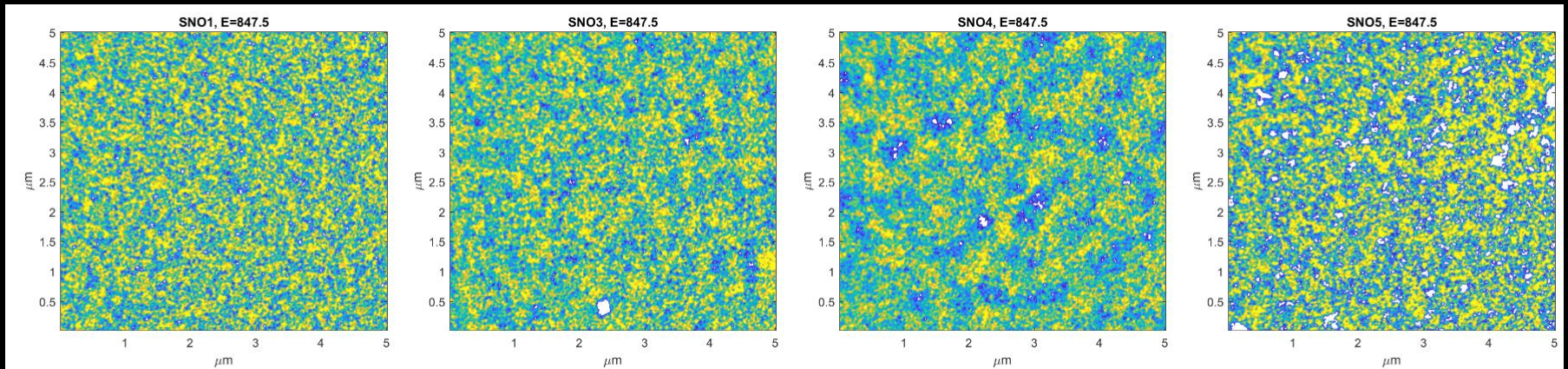
Electron-doping induced a colossal metal-insulator transition



XAS shows doping induced crossover in electronic ground state

Doped nickelates

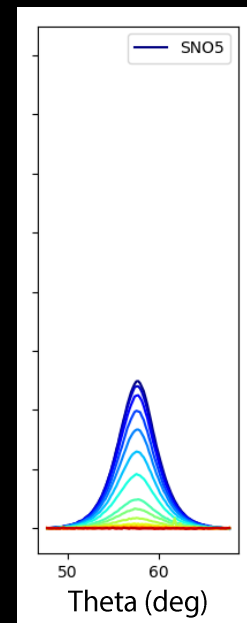
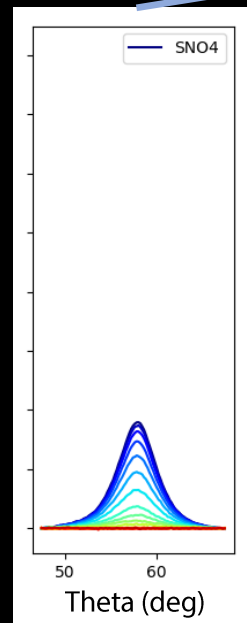
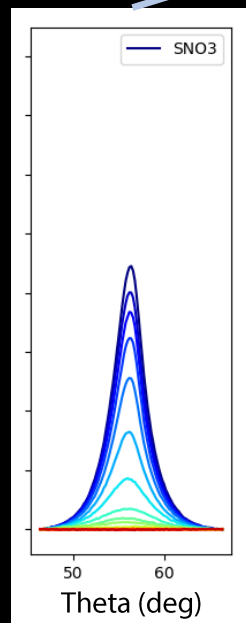
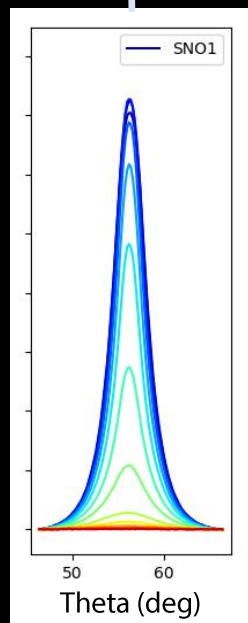
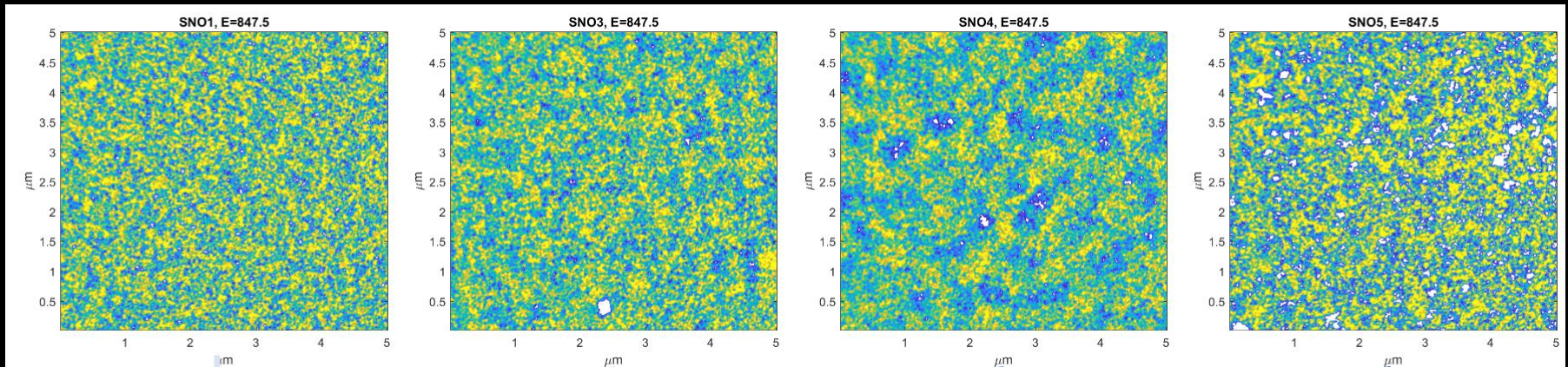
XPEEM mapping across Ni-L₃ edge (@ESM beamline)



XAS shows doping induced crossover in electronic ground state

Doped nickelates

XPEEM mapping across Ni-L₃ edge (@ESM beamline)



Doping induced phase separation as AFM order is suppressed.

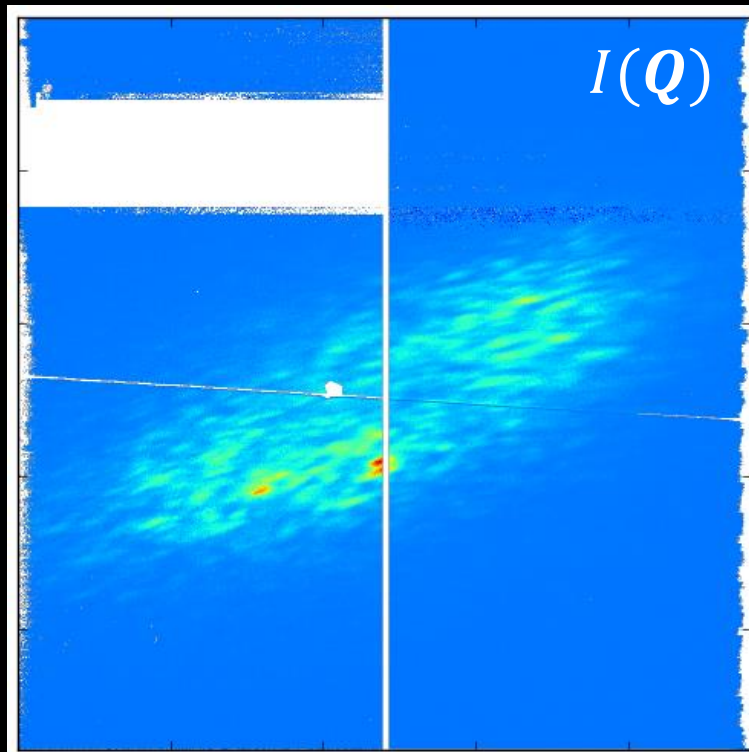
The length scale of inhomogeneity is maximal when AFM order is weakest

Coherent Diffractive Imaging

RECIPROCAL SPACE



REAL SPACE



Coherent magnetic Bragg diffraction

Probe Function: E-field profile at the sample

$$F(\mathbf{Q}) = \mathcal{F}[\underbrace{O(\mathbf{r})}_{\text{Object Function}} \cdot \underbrace{P(\mathbf{r})}_{\text{Probe Function}}]$$

Object Function: what we are interested in

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

From measurement

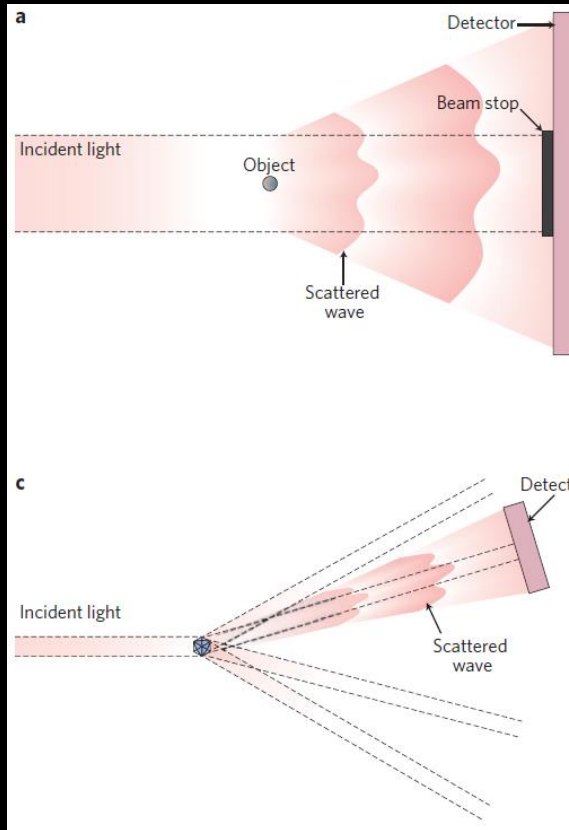
We need to recover the phases of the exit waves

COMPUTATIONAL IMAGING

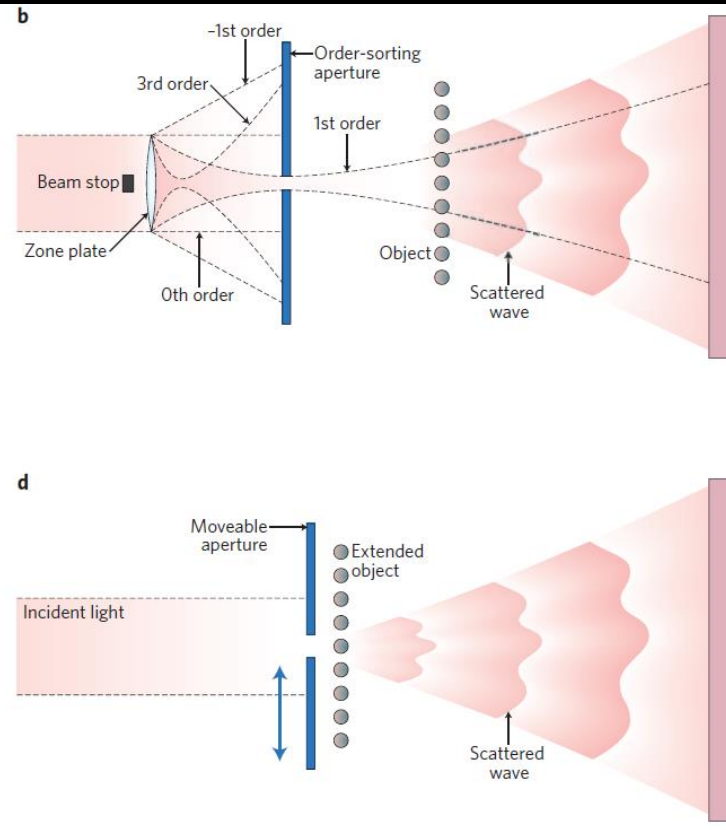


Coherent Diffractive Imaging

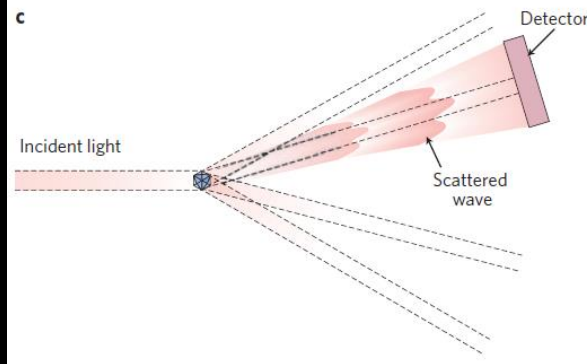
Plane-wave CDI



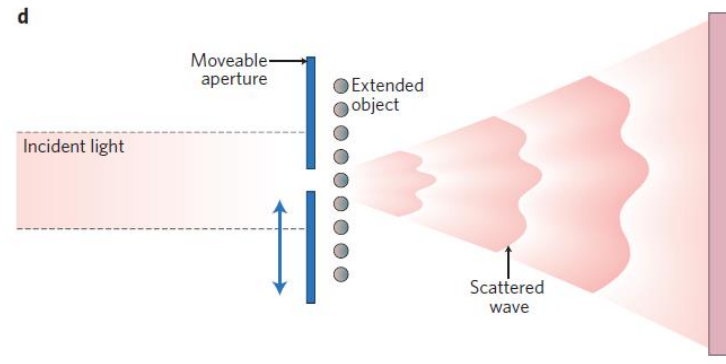
Fresnel CDI



Bragg CDI

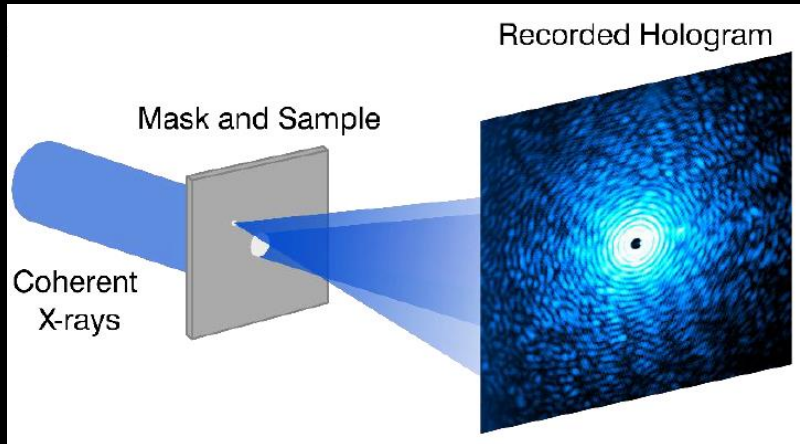


Ptychographic CDI



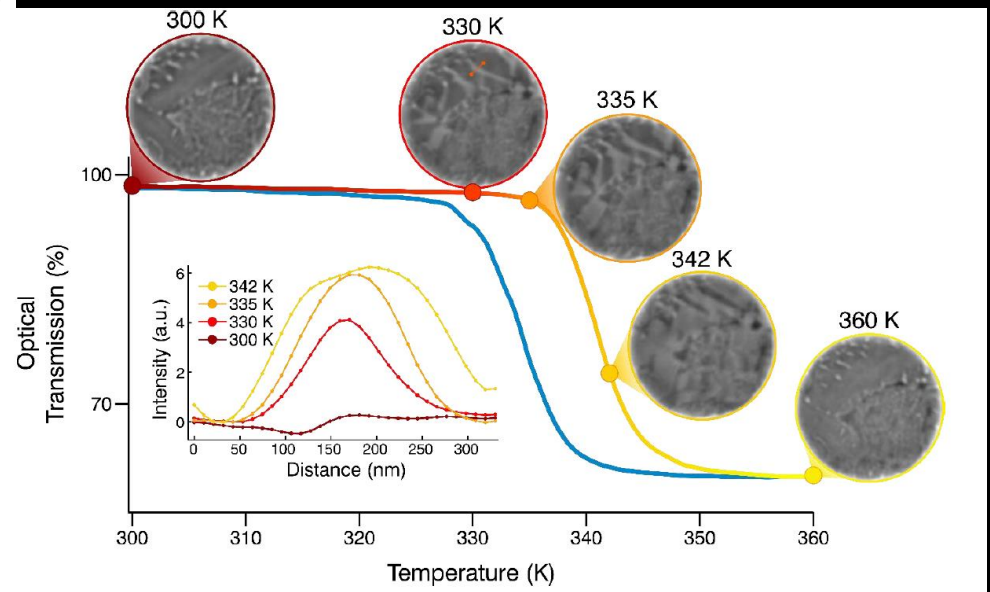
Coherent Imaging

Coherent Diffractive Imaging



Metal-insulator transition in VO_2

Resonant holography
(few sec for a full hologram)



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

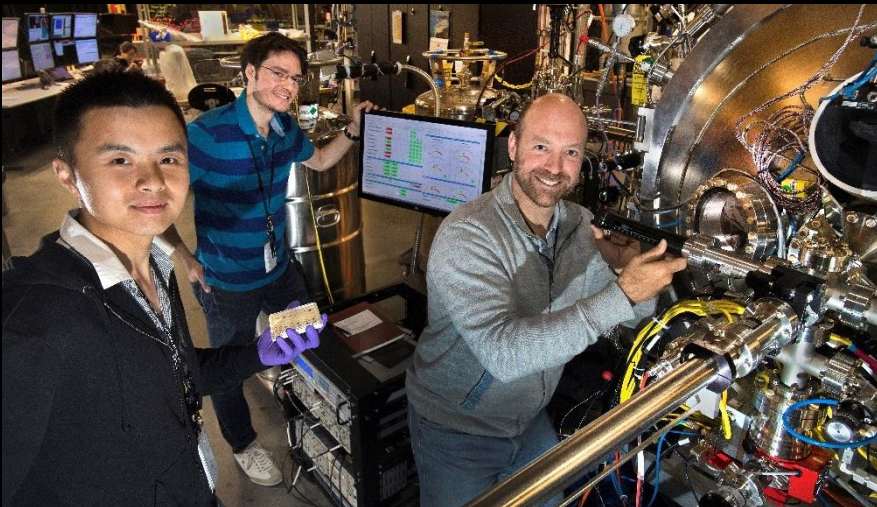
- Intensive computational effort (phase retrieval)
- Sometimes requires sample pre-patterning
 - Beware dynamical scattering effects

Acknowledgments



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