



Massachusetts  
Institute of  
Technology

# Soft X-ray scattering and imaging of quantum electronic solids

Riccardo Comin

Massachusetts Institute of Technology

FUSEE workshop  
Trieste, 11 Dec 2019

# Outline

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- **Intro:**
  - Density-wave phenomena
  - Resonant X-ray scattering
- **Coherent soft X-ray imaging:**
  - Resonant scanning nanodiffraction: **scale-invariant nanoscale magnetic textures** in rare earth nickelates
  - **Coherent diffractive phase contrast imaging** of antiferromagnetic domain textures

# MIT Photon Scattering Lab



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Mingu Kang

Jiarui Li

Abe Levitan

Qian Song

Connor Occhialini

Luiz Martins

David Rower



# Acknowledgments

## Collaborators



C. Mazzoli  
S. Wilkins



F. Simmons  
E. Carlson  
S. Ramanathan



S. Catalano  
M. Gibert  
J.-M. Triscone

## Funding



# Density-wave phenomena in strongly-correlated matter

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# Strongly-interacting quantum matter

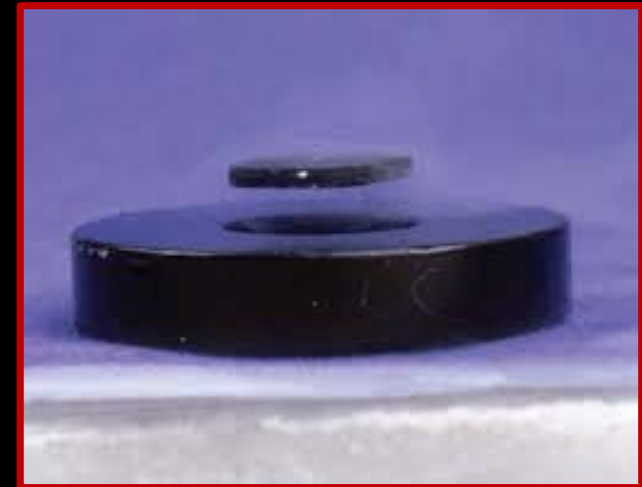
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Fundamental  
building blocks



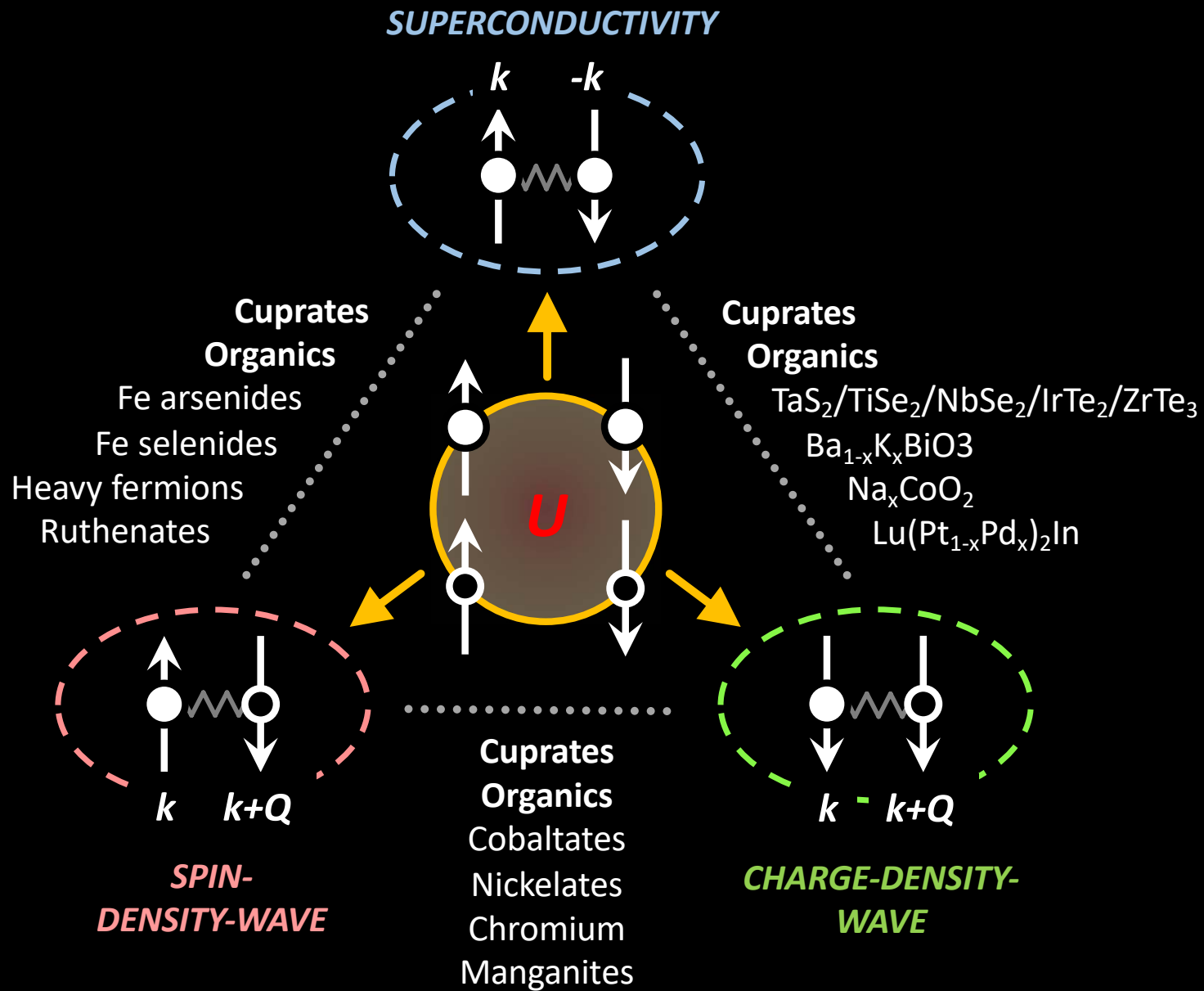
Interactions

Many-body  
phenomena



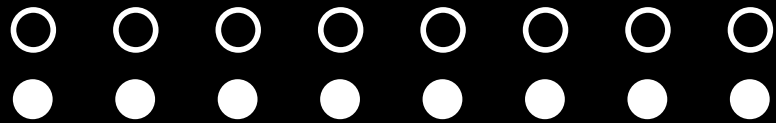
*“More is different”* (P.W.Anderson, 1972):  
Interactions foster new organizing principles and collective  
behavior in many-body systems

# Density-wave phases

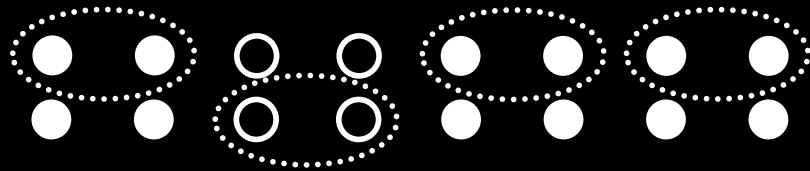


# Instabilities of a strongly-interacting electron system

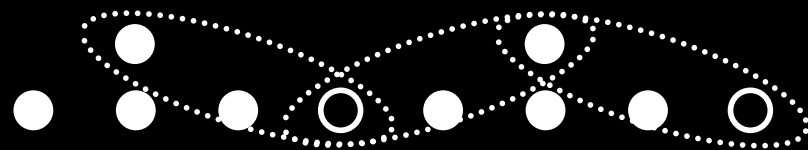
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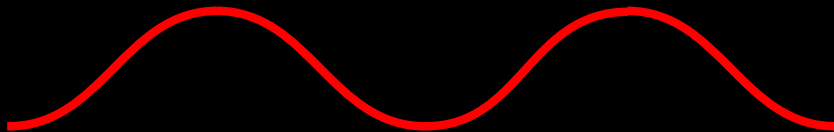
Mott-Hubbard ground state



Superconductivity (Cooper pairing)



Density-wave (particle-hole pairing)

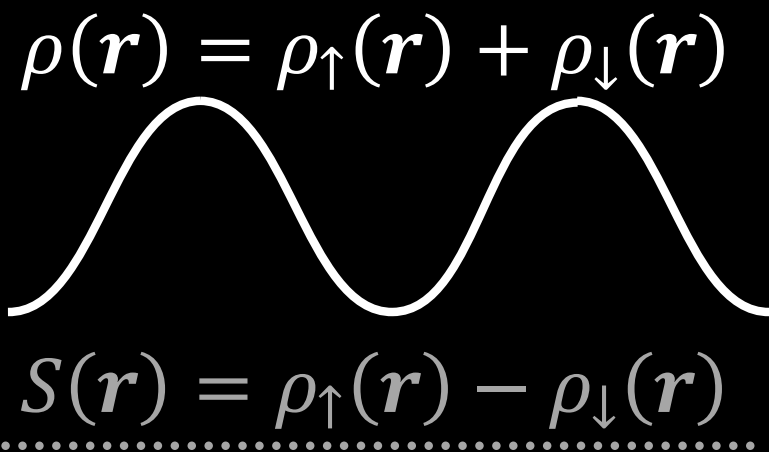
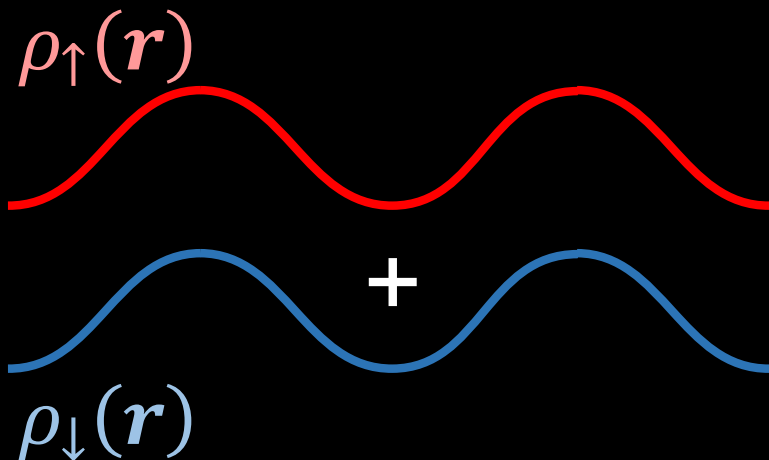


$\rho(\mathbf{r})$



# Density-wave phases

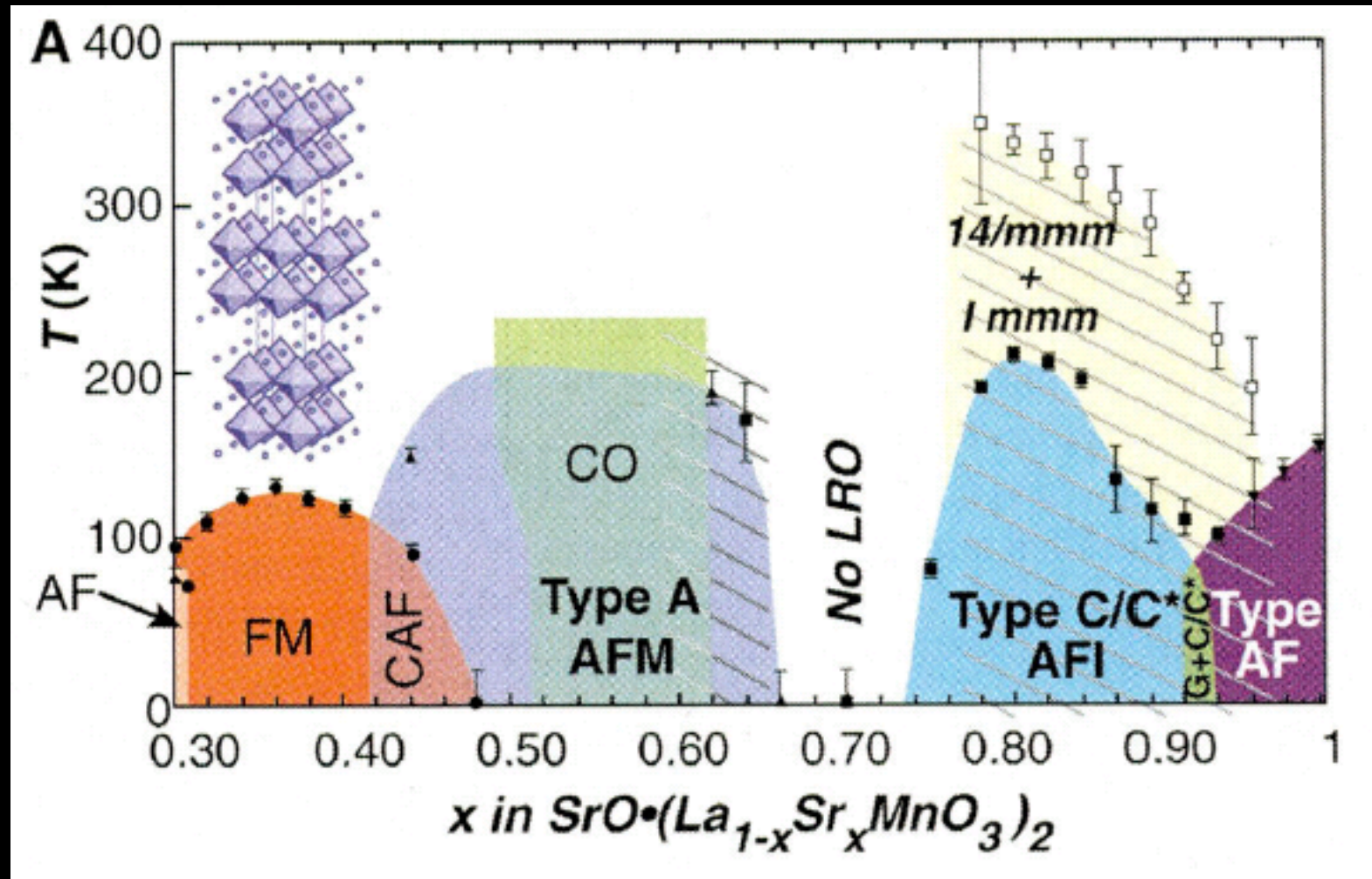
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Charge-density-wave

Spin-density-wave

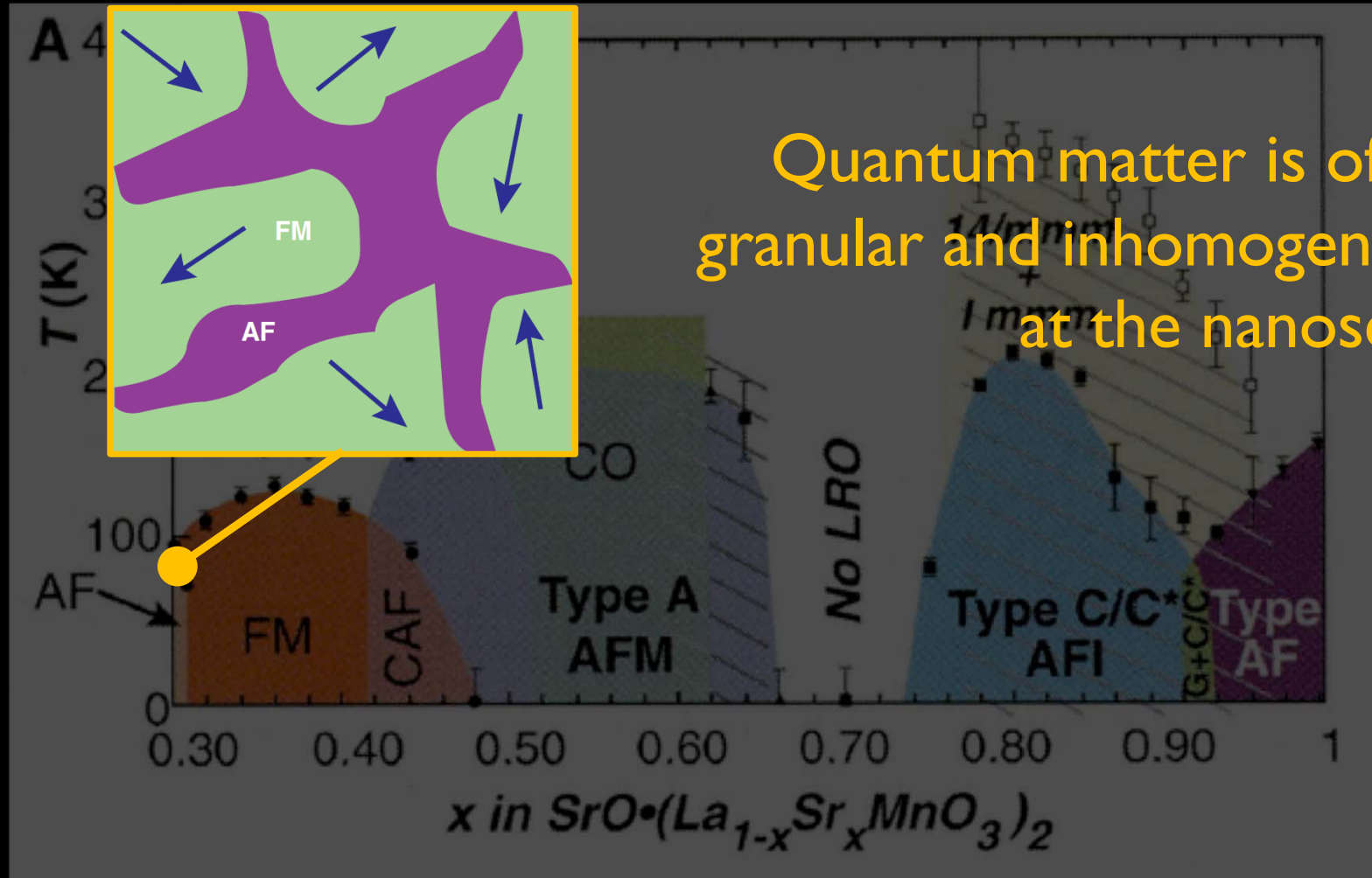
# Strongly-interacting quantum matter



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

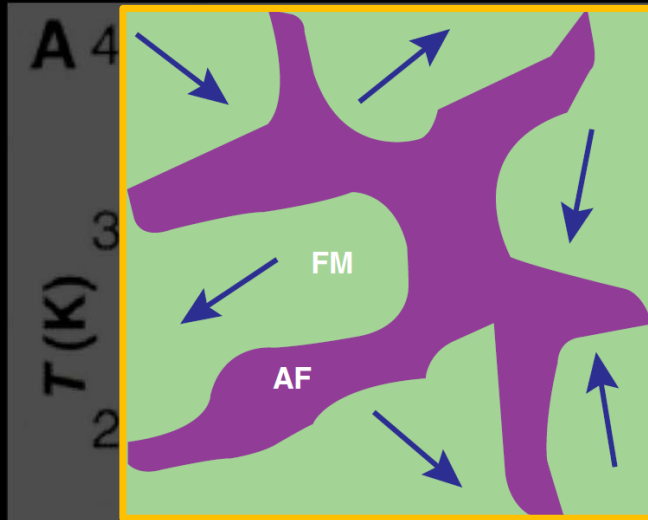
# Strongly-interacting quantum matter

Phase segregation

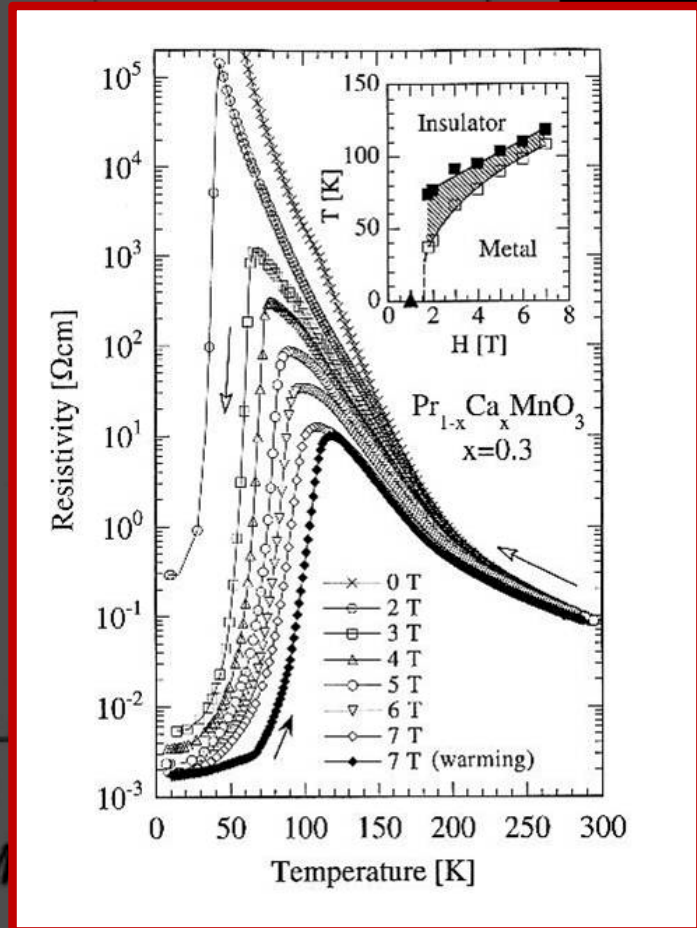


# Strongly-interacting quantum matter

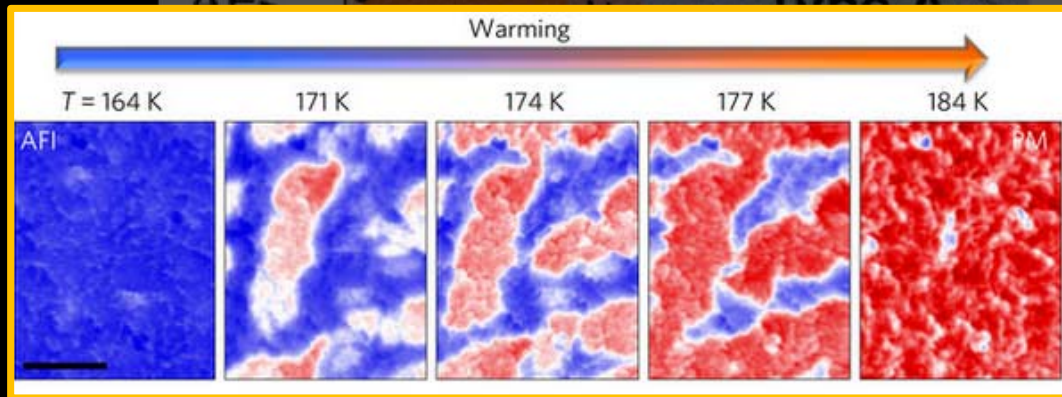
Phase segregation



Colossal resistive switching



Percolation phenomena

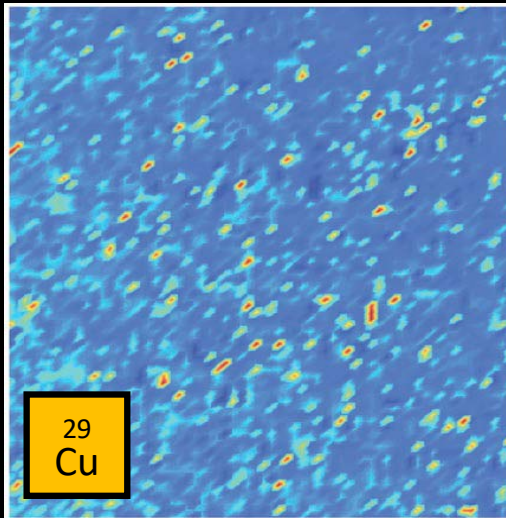


A. S. McLeod, *Nat. Physics* **13**, 80 (2017)

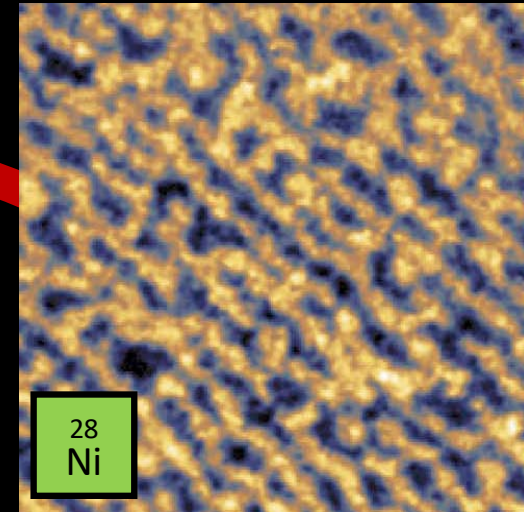
Y. Tomioka, et al., *Physics of Manganites* (1999)

# Strongly-interacting quantum matter

Charge order



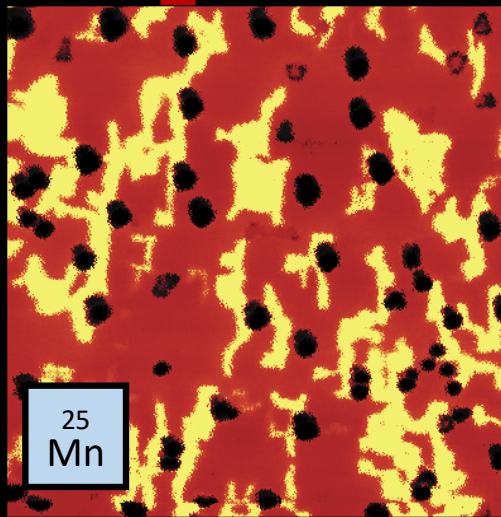
Metal-insulator transition



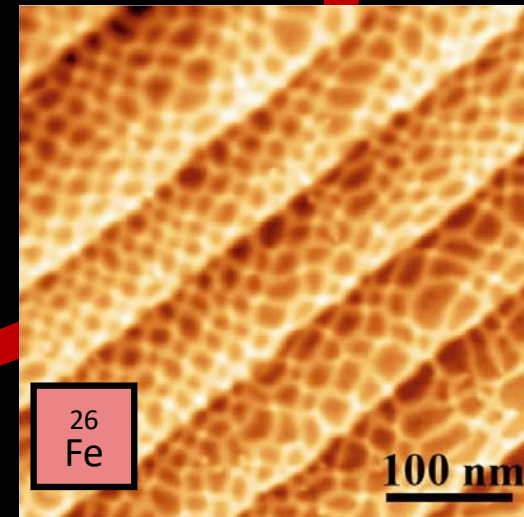
Emergent  
nanoscale  
textures



Macroscale  
quantum  
phenomena



Spin order



Superconductivity

# Soft X-ray scattering probes of density waves

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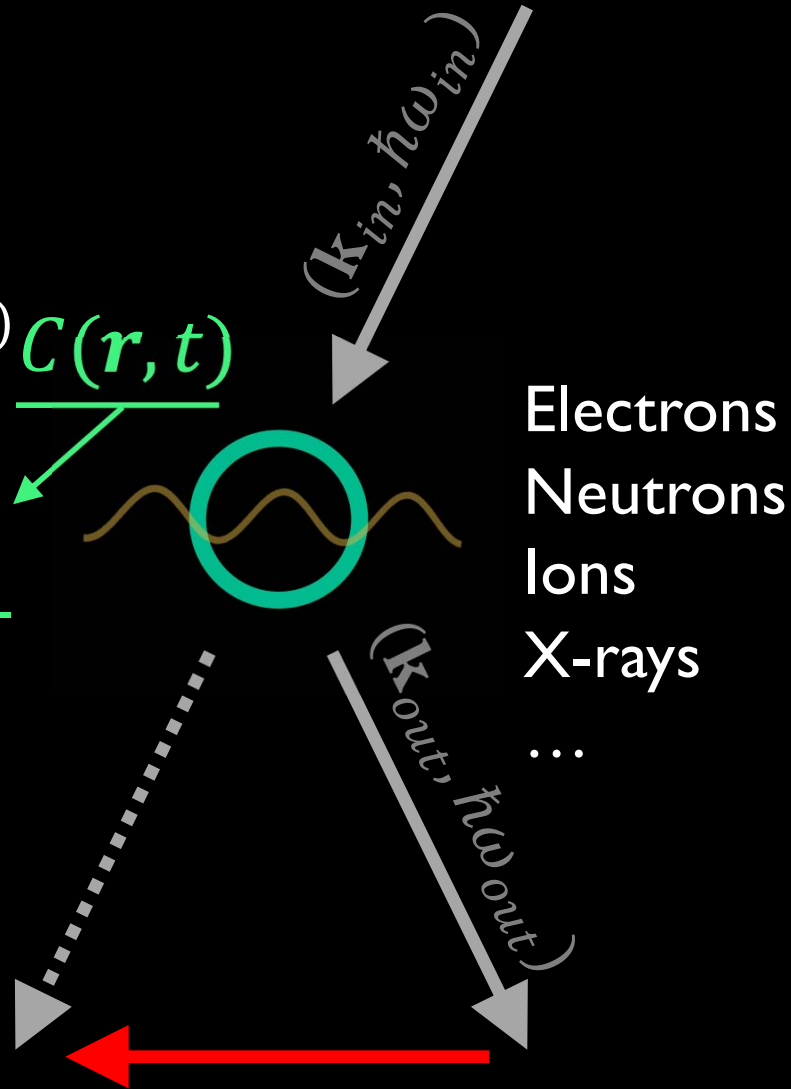
# Scattering probes

## Structure factor

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

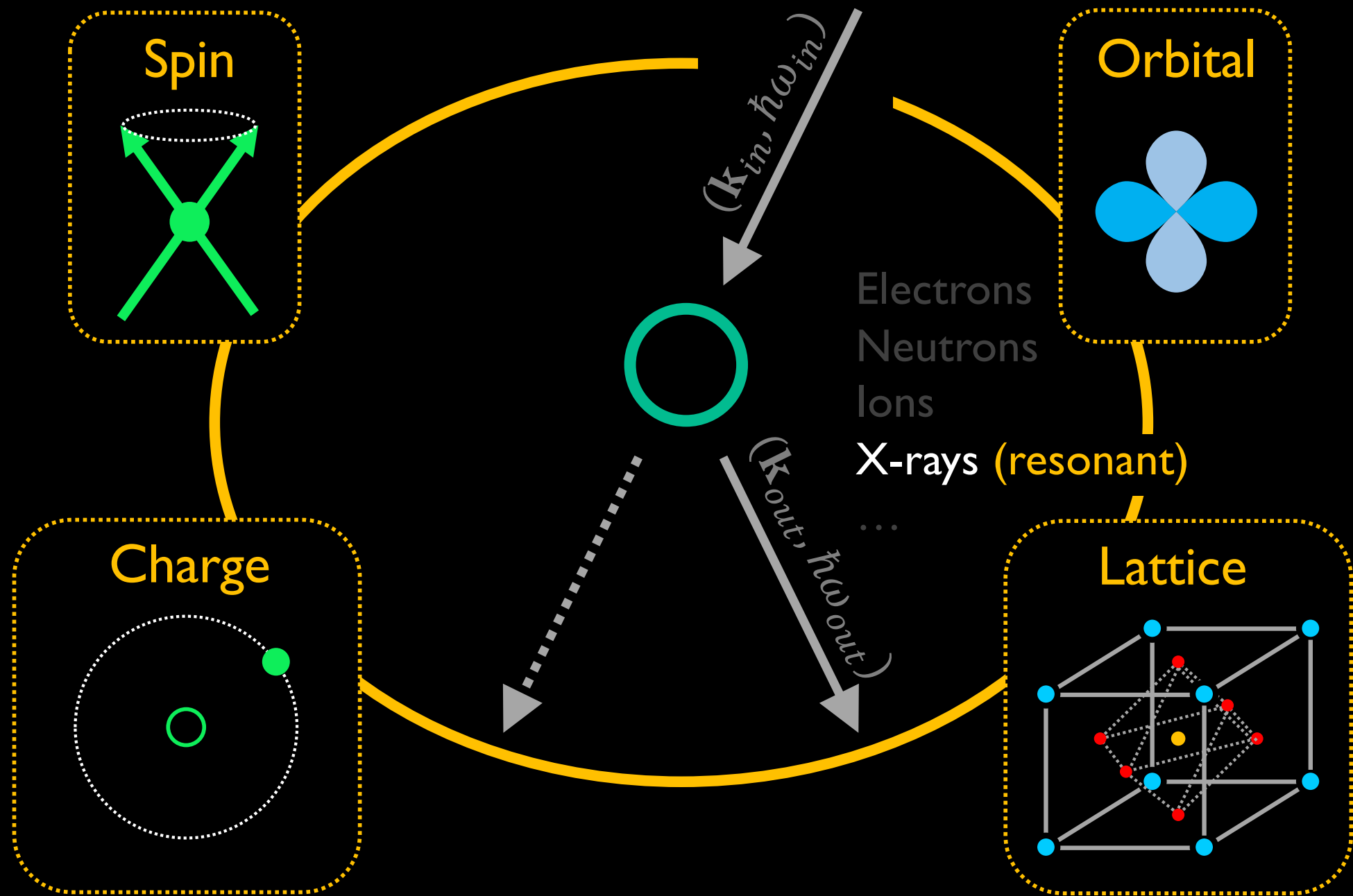
$$= \int dt d\mathbf{r} e^{i(\mathbf{Q} \cdot \mathbf{r} - \omega t)} \underline{C(\mathbf{r}, t)}$$

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



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

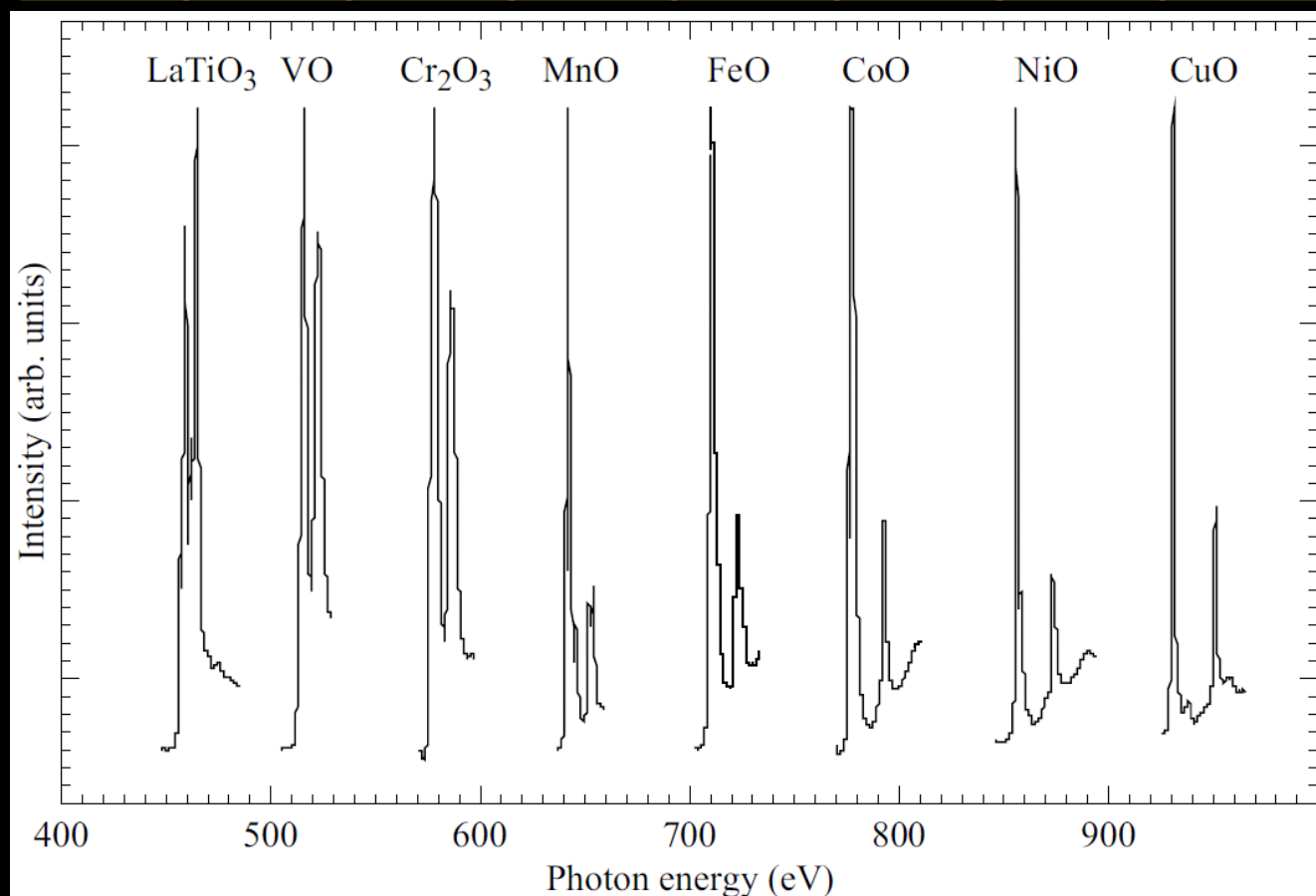
# Scattering probes





# Resonant scattering

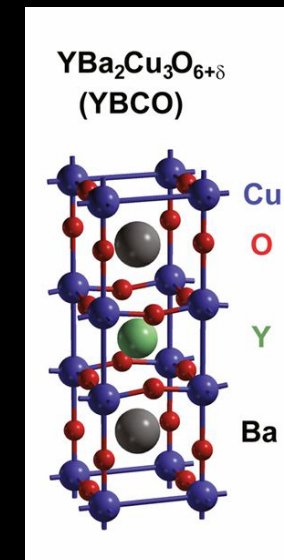
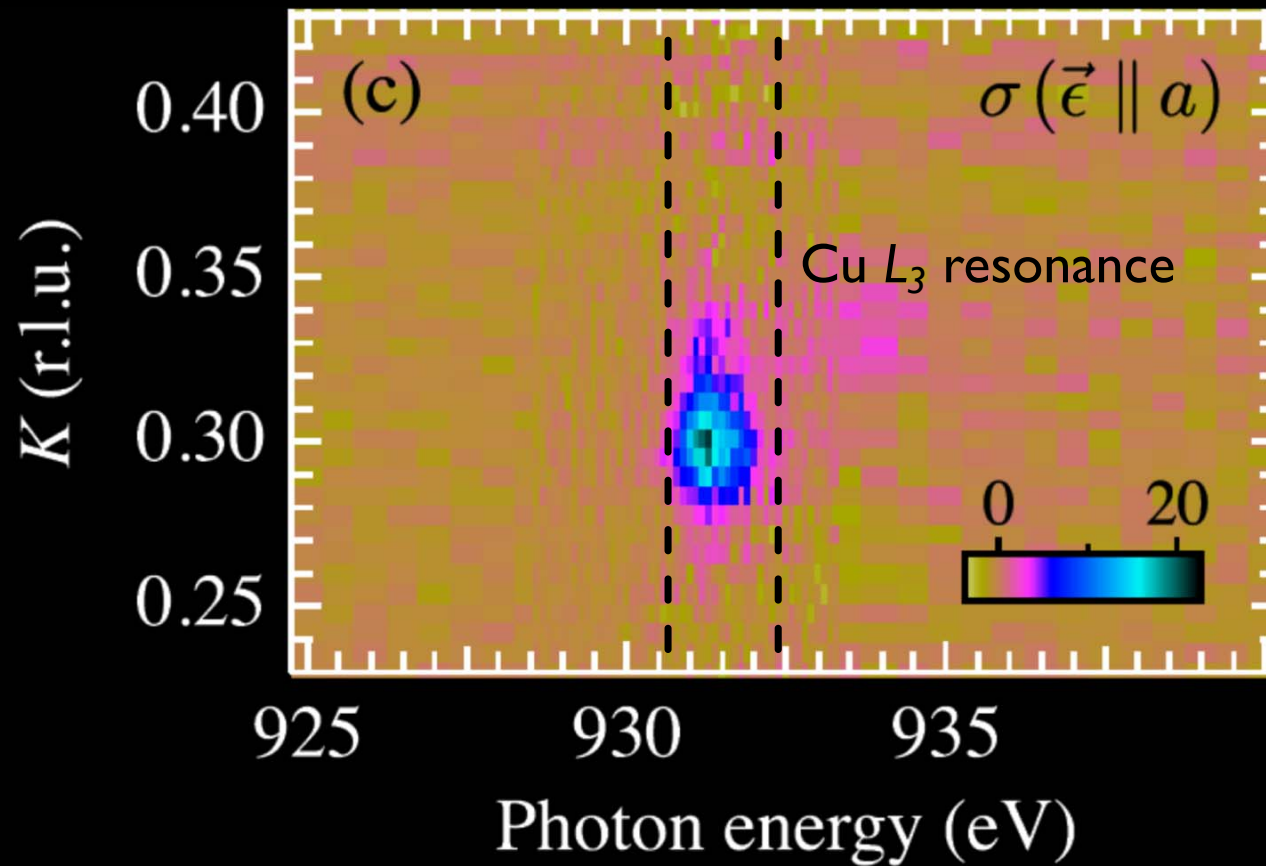
<sup>22</sup> Ti	<sup>23</sup> V	<sup>24</sup> Cr	<sup>25</sup> Mn	<sup>26</sup> Fe	<sup>27</sup> Co	<sup>28</sup> Ni	<sup>29</sup> Cu
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Strongly energy-dependent X-ray scattering amplitude

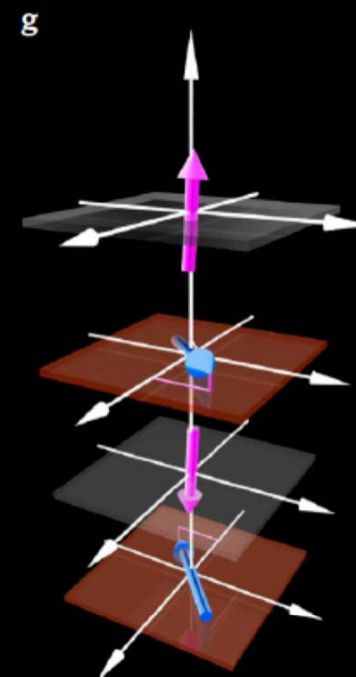
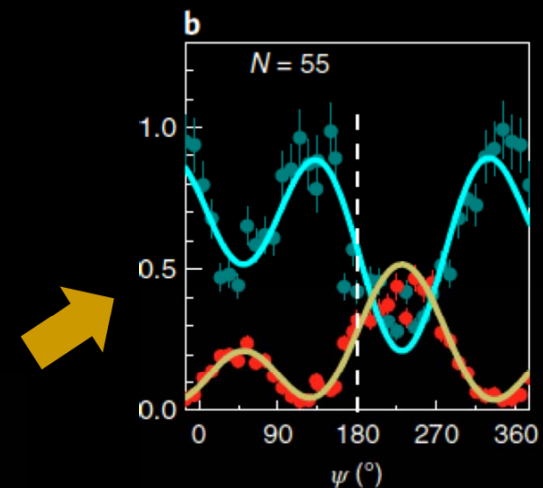
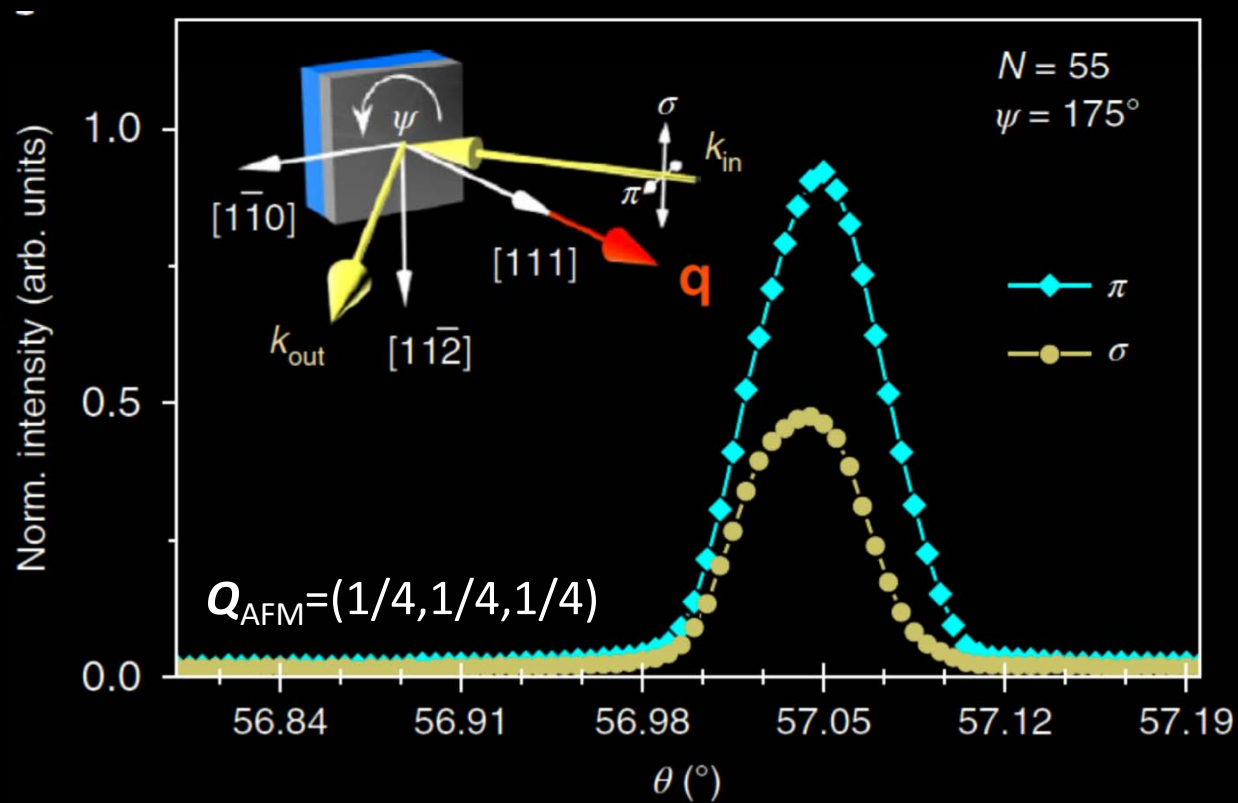
# Resonant scattering

## Charge order in copper oxide high-temperature superconductors



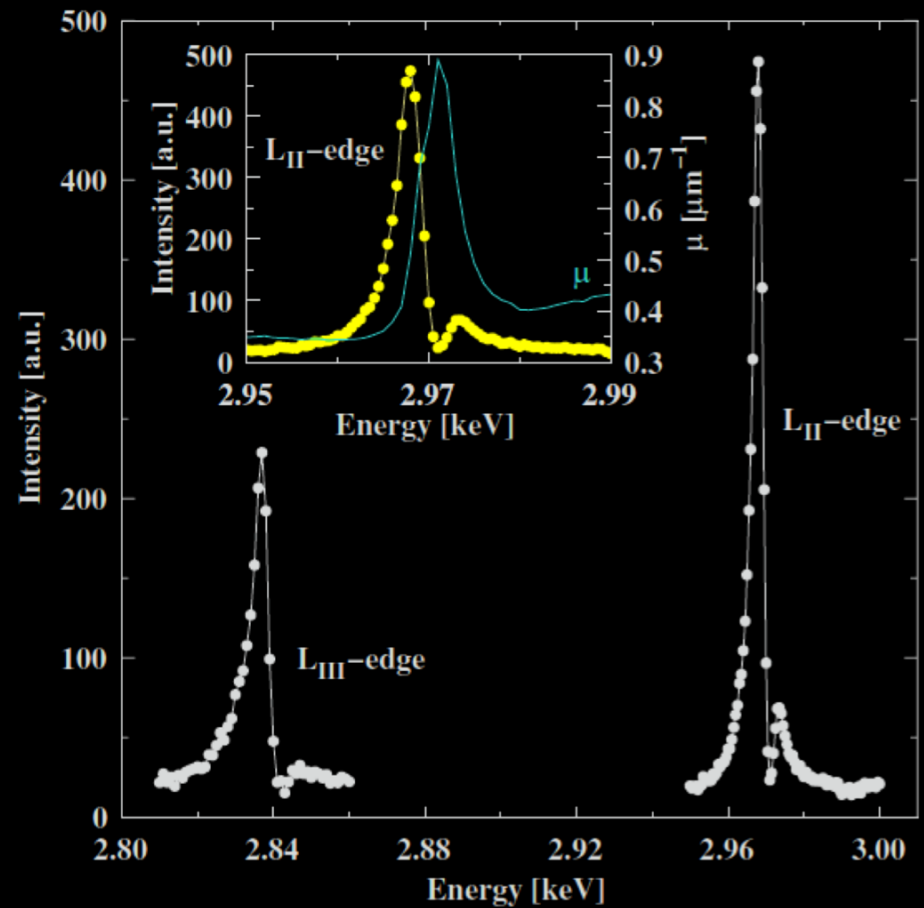
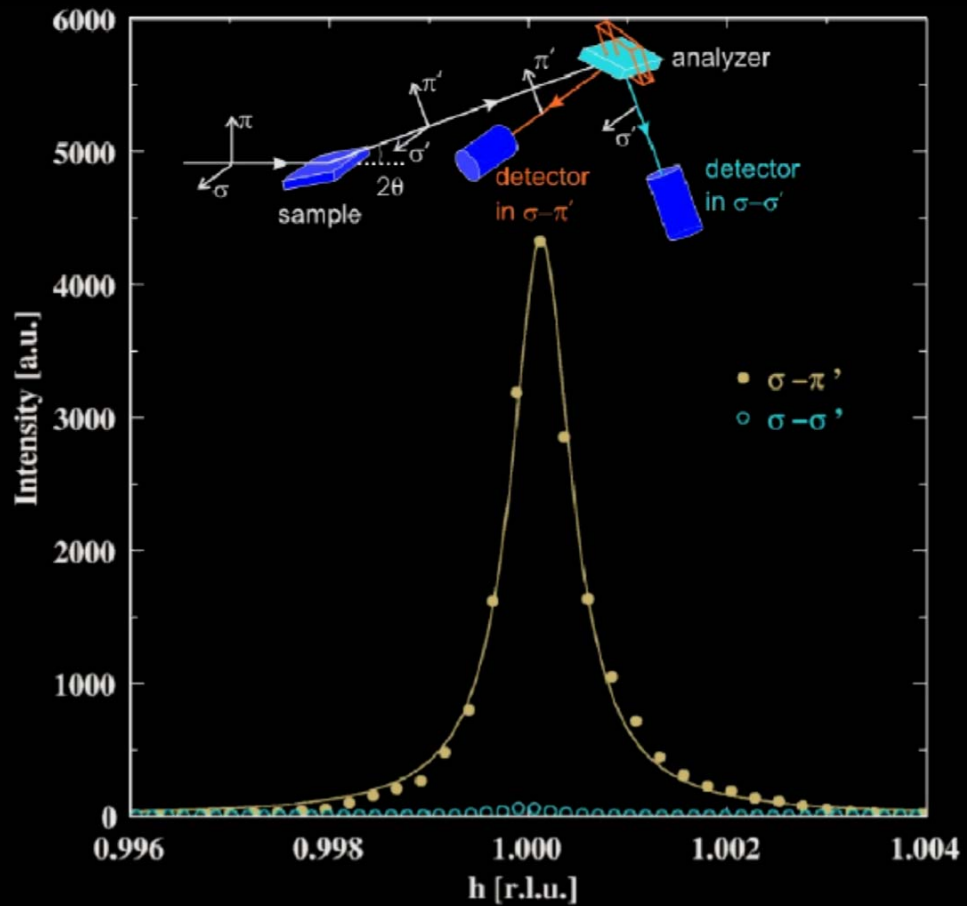
# Resonant scattering

## Antiferromagnetic order in rare earth nickelates



# Resonant scattering

## Orbital (+ magnetic) ordering in layered ruthenate $\text{Ca}_2\text{RuO}_4$

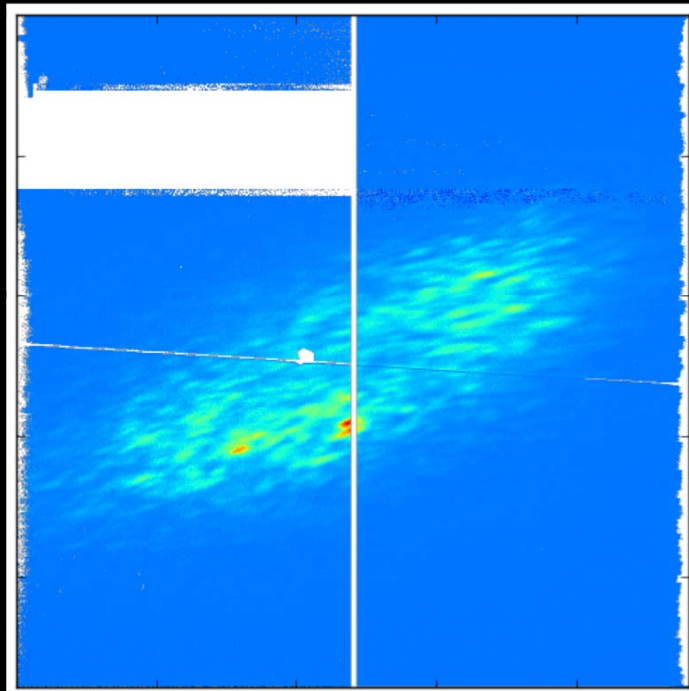


# Nanoscale electronic textures and coherent X-ray imaging

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# Electronic orders at the nanoscale

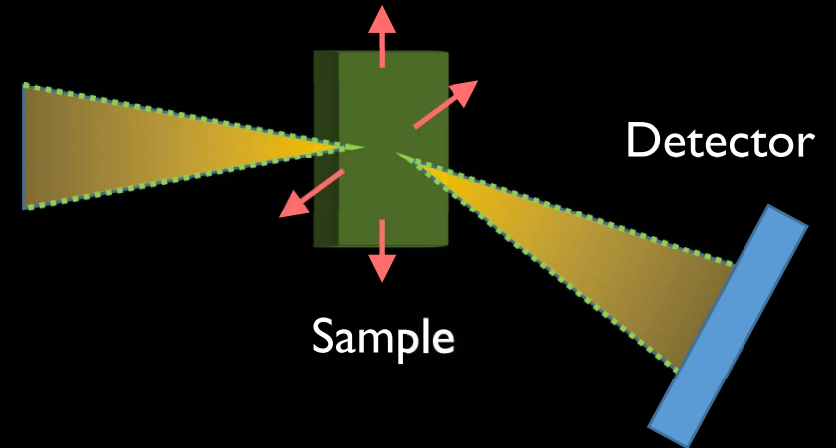
Reciprocal space  
(scattering)



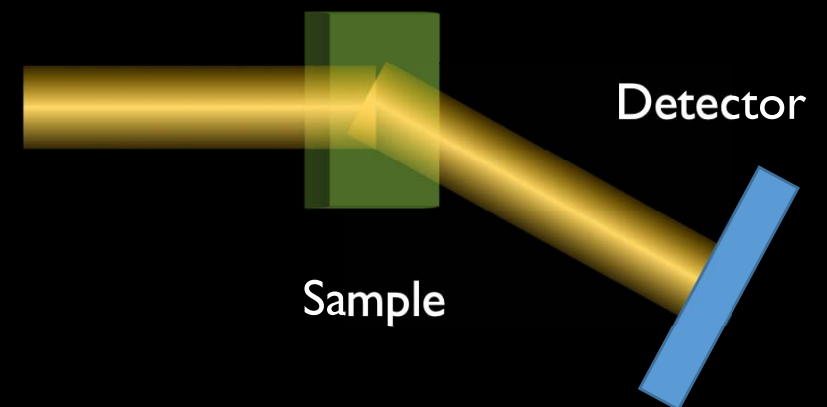
Recover real-space information

HOW

Scanning nanodiffraction (50 nm)



Full-field lensless imaging (10 nm)



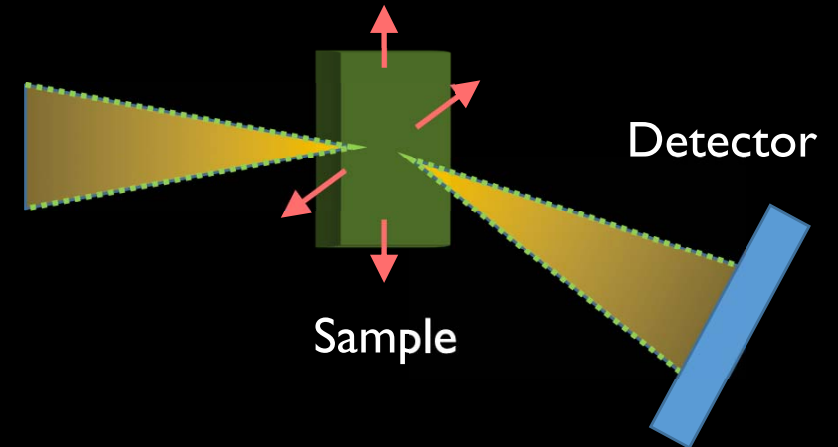
# Electronic orders at the nanoscale

## WHY

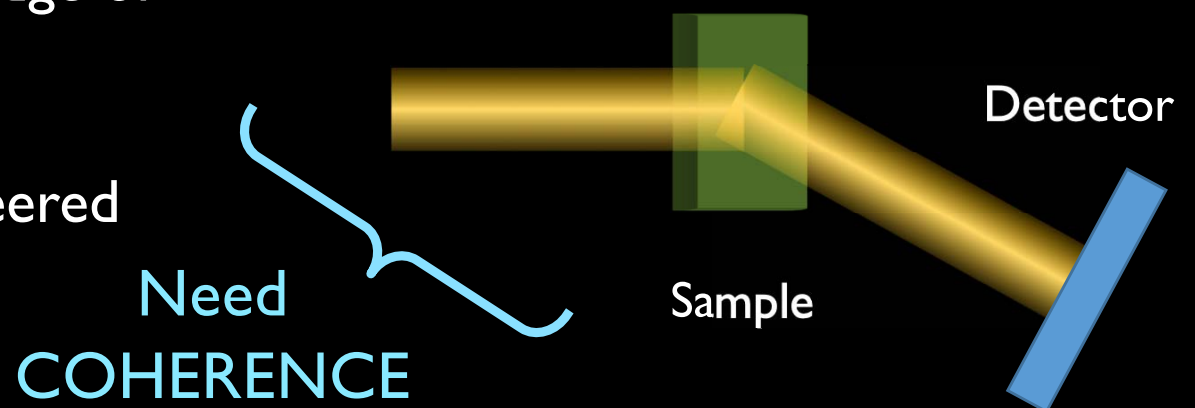
- Nanoscale granularity:
  - Intrinsic (phase competition & segregation)
  - Extrinsic (disorder, defects, doping, ...)
- Scale-invariant phenomena:
  - Extended range of length scales (10 nm to 10  $\mu\text{m}$ )
- Emergent physics at the edge or boundary:
  - Domain walls; lateral interfaces; nanoengineered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

## HOW

### Scanning nanodiffraction (50 nm)



### Full-field lensless imaging (10 nm)

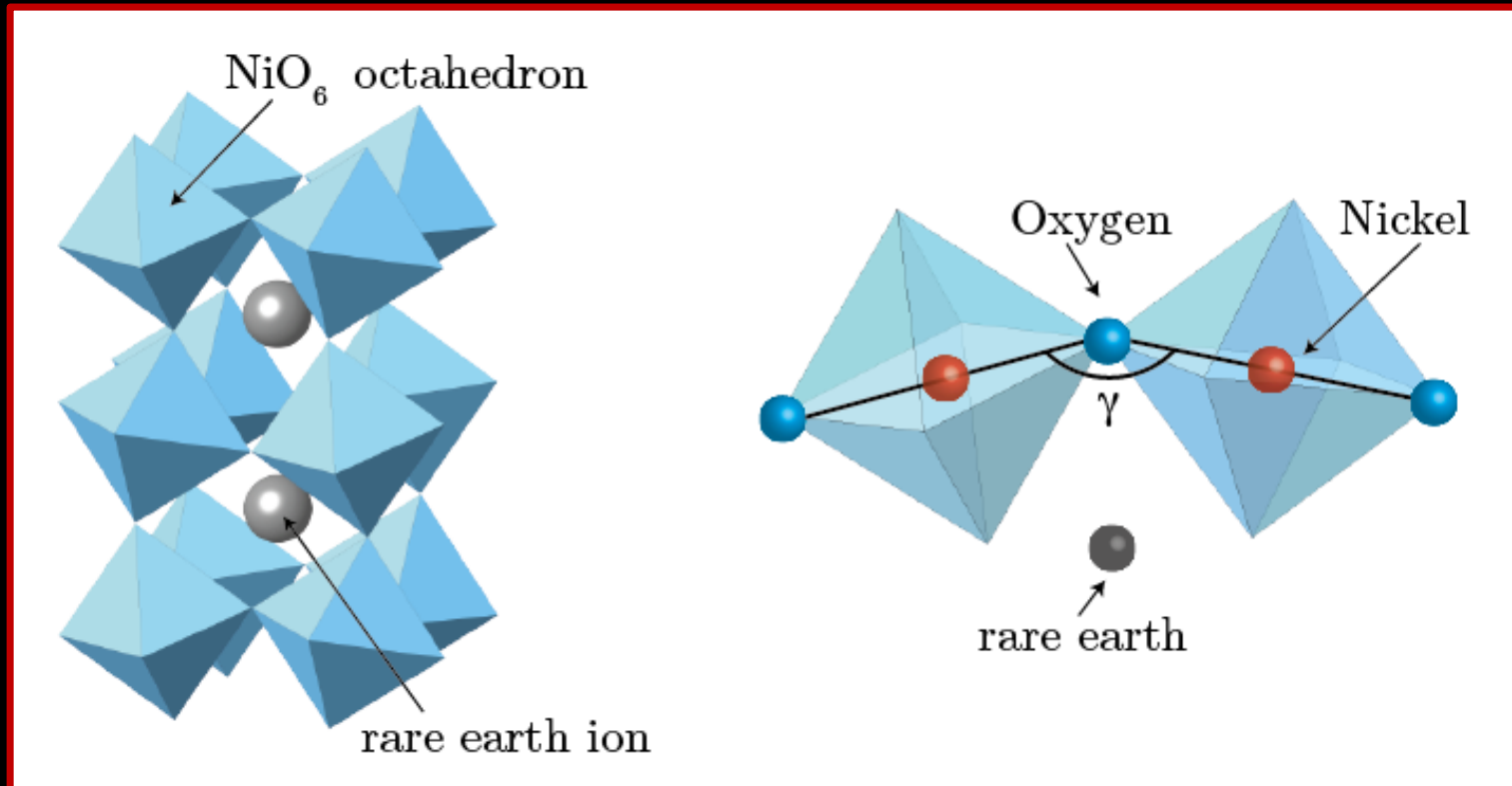


# Spin-density-waves and scale-invariant spin textures in nickel oxides

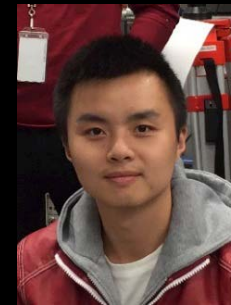
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# Rare earth nickelates



Jiarui Li

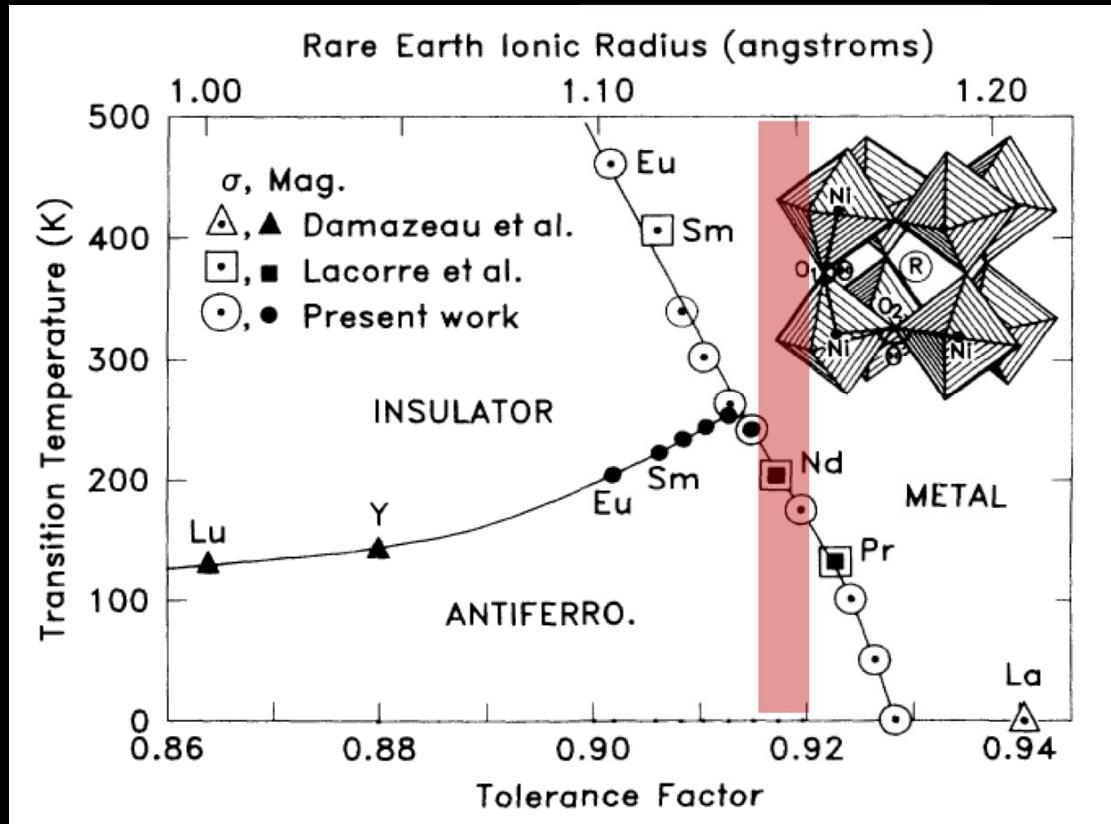


Johnny Pellicciari



# Rare earth nickelates

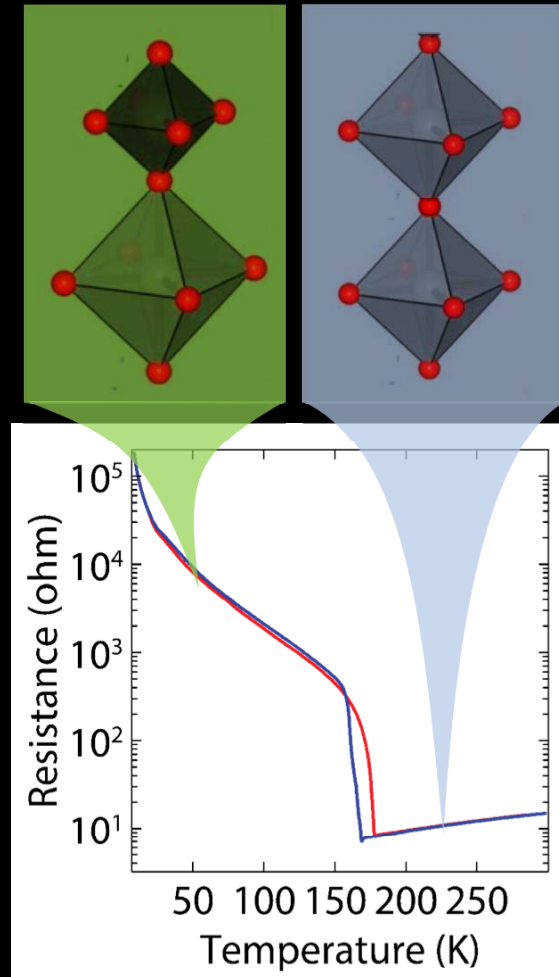
Torrance et al. PRB 1992



← Quantum tuning parameter →

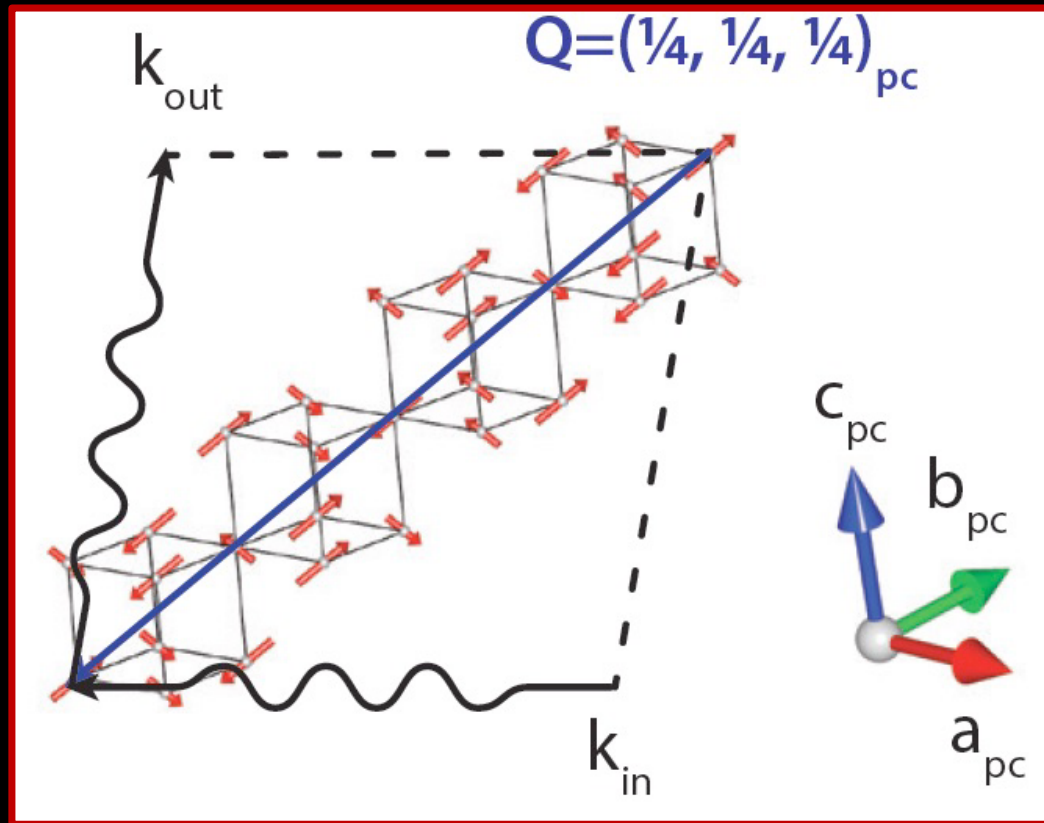
$P2_1/n$   
(insulator)

$Pbnm$   
(metal)



Metal insulator transition

# Rare earth nickelates

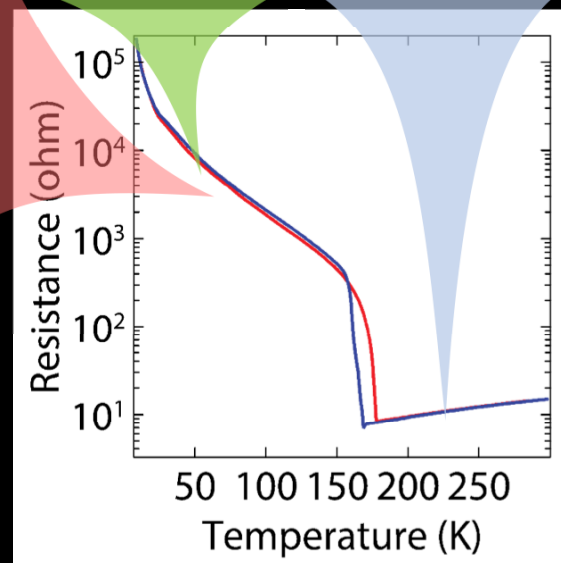
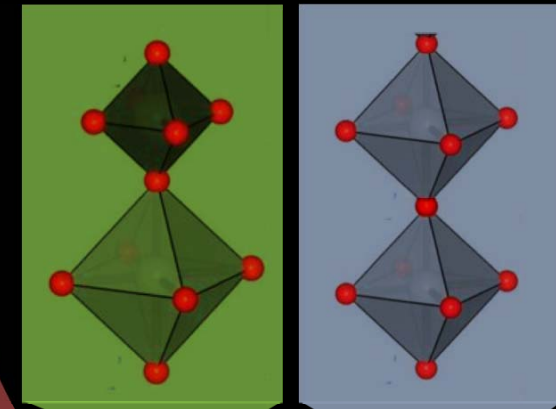


Magnetic order

**Goal:** map the charge and spin textures across the metal-insulator/Neel transition

$P2_1/n$   
(insulator)

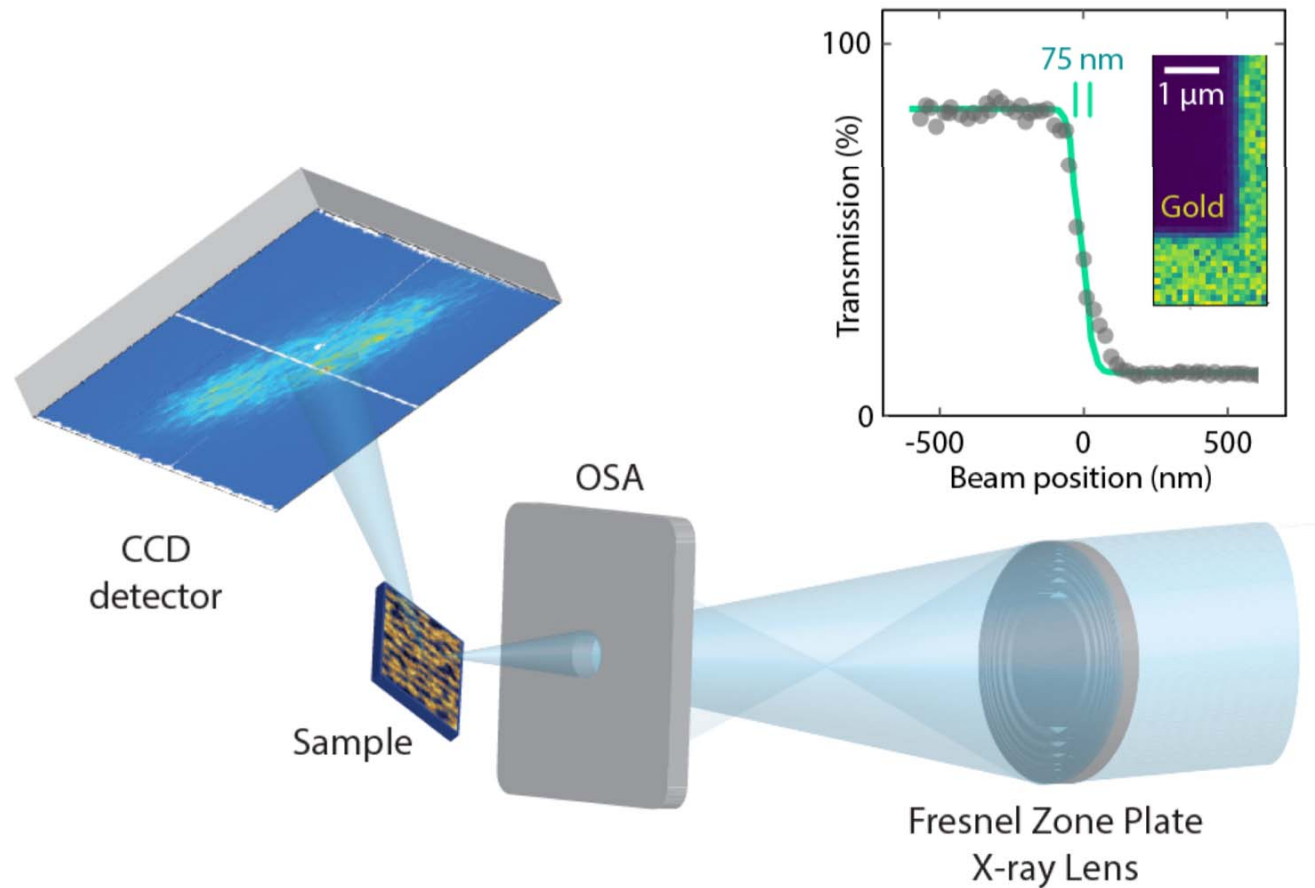
$Pbnm$   
(metal)



Metal insulator transition

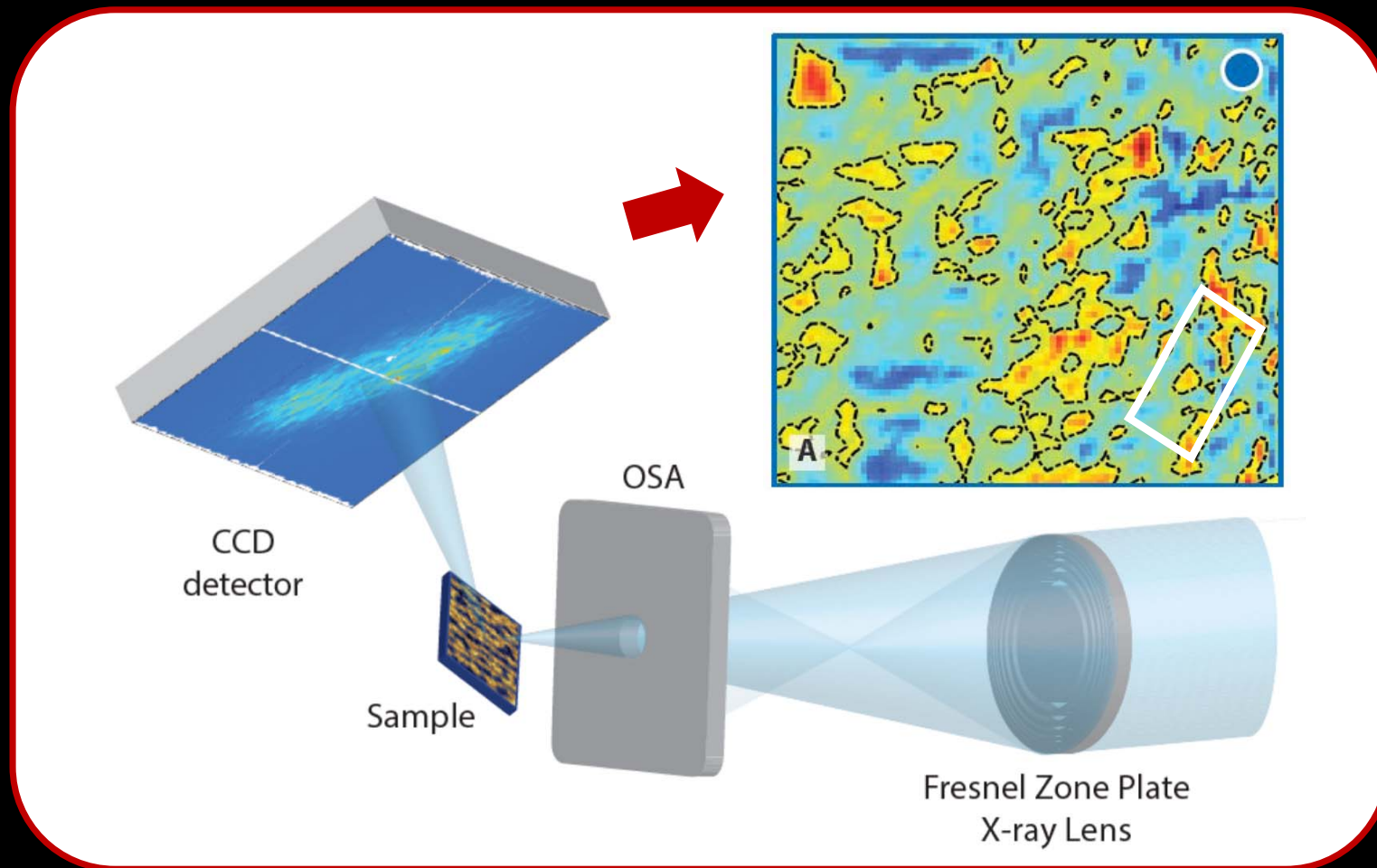
# Spin textures in nickelates

## Resonant scattering at the nanoscale



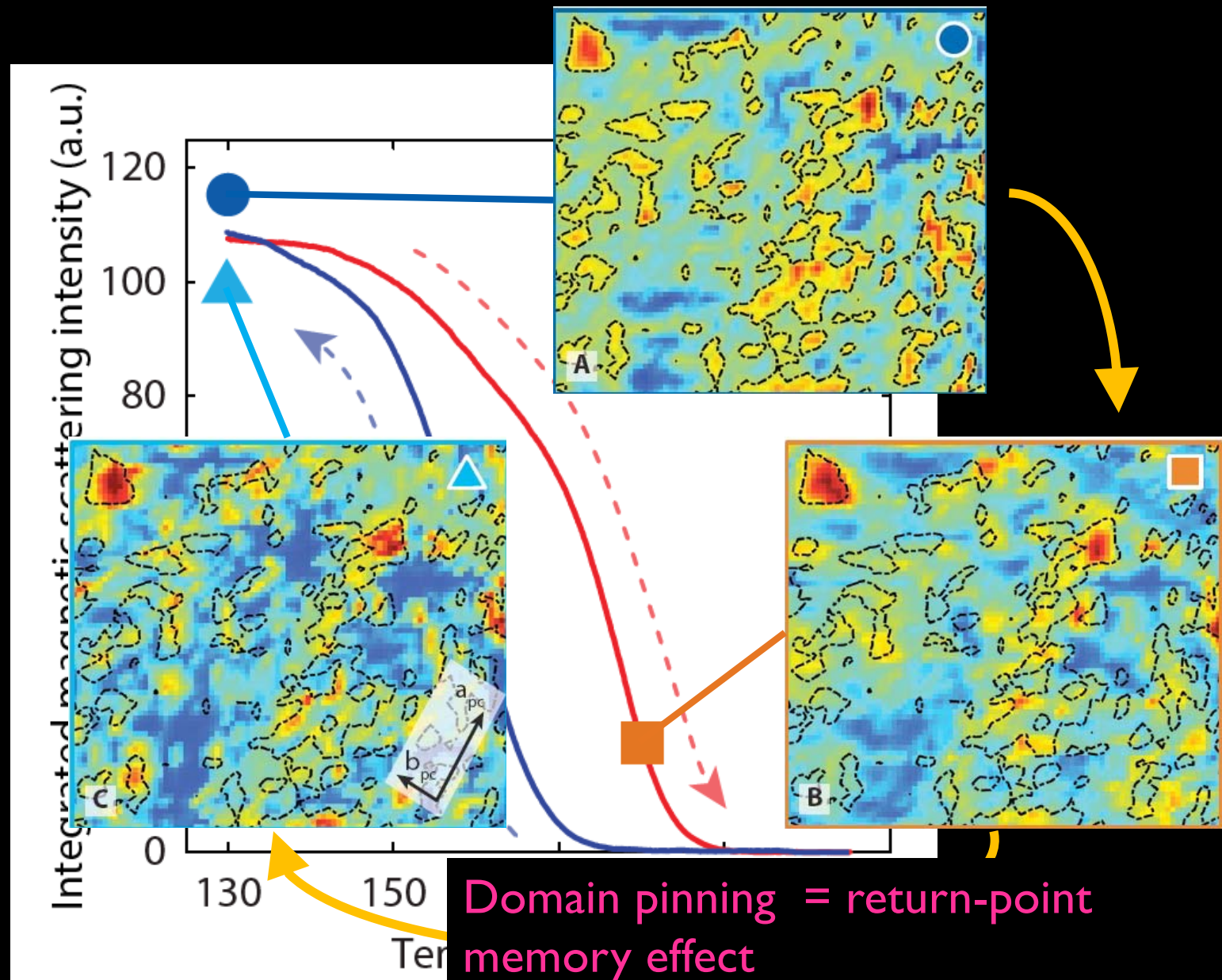
# Spin textures in nickelates

## Resonant scattering at the nanoscale



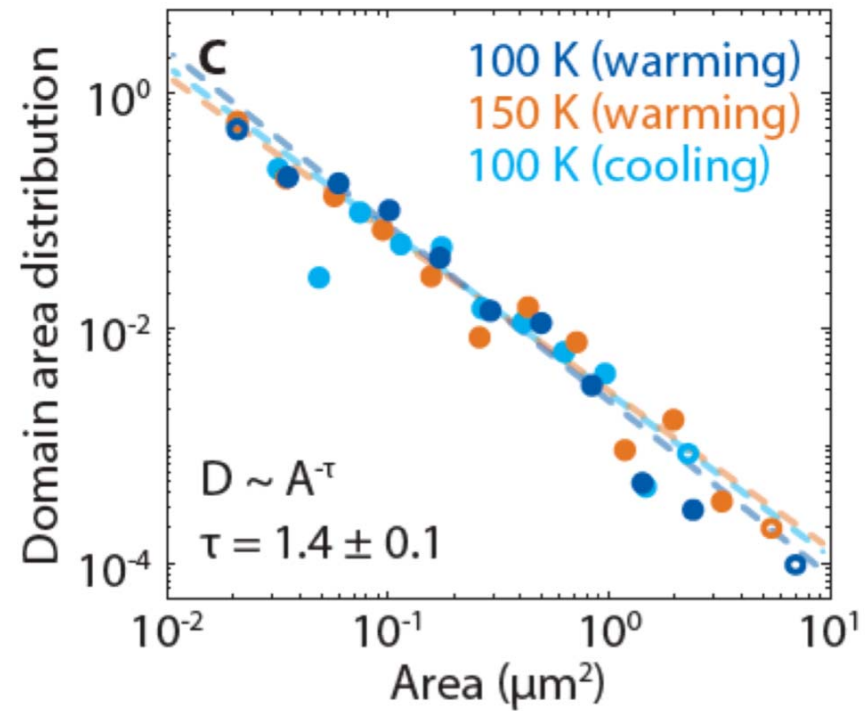
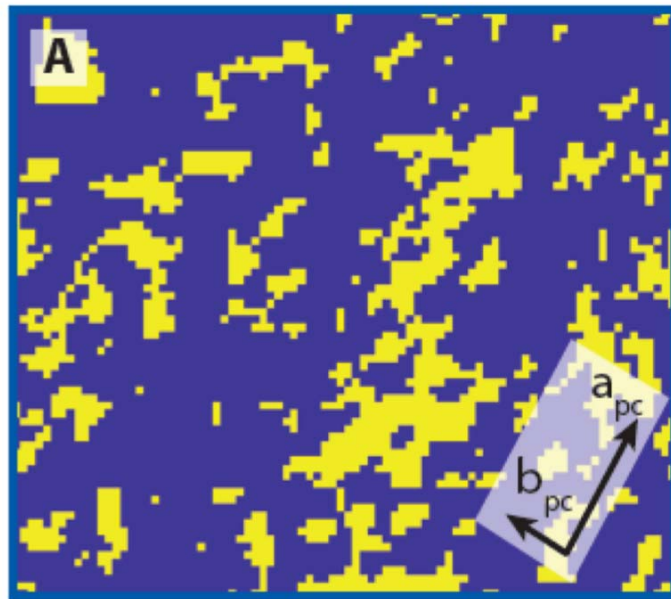
# Spin textures in nickelates

1  $\mu\text{m}$  x 1  $\mu\text{m}$



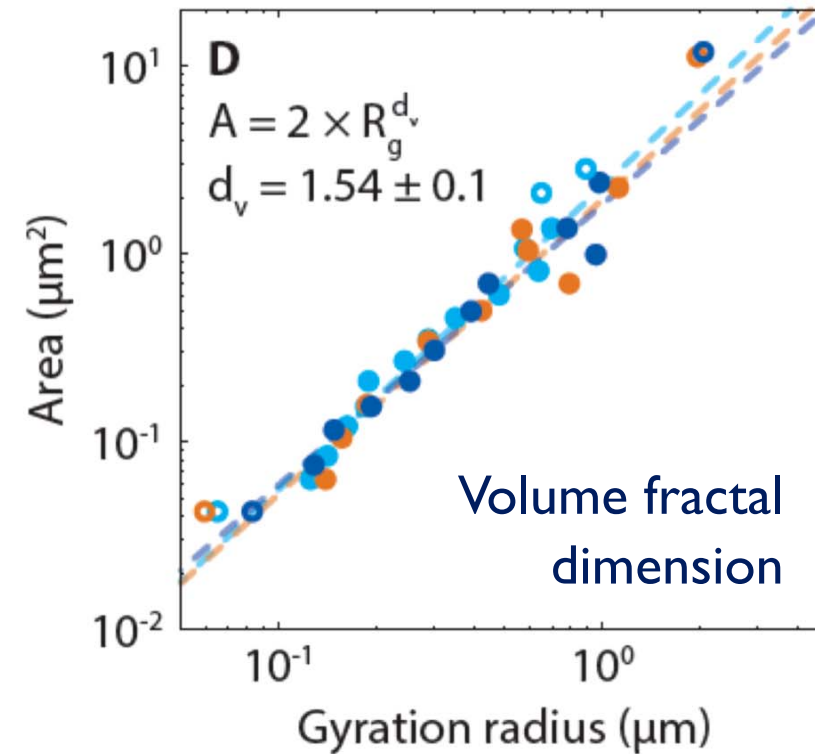
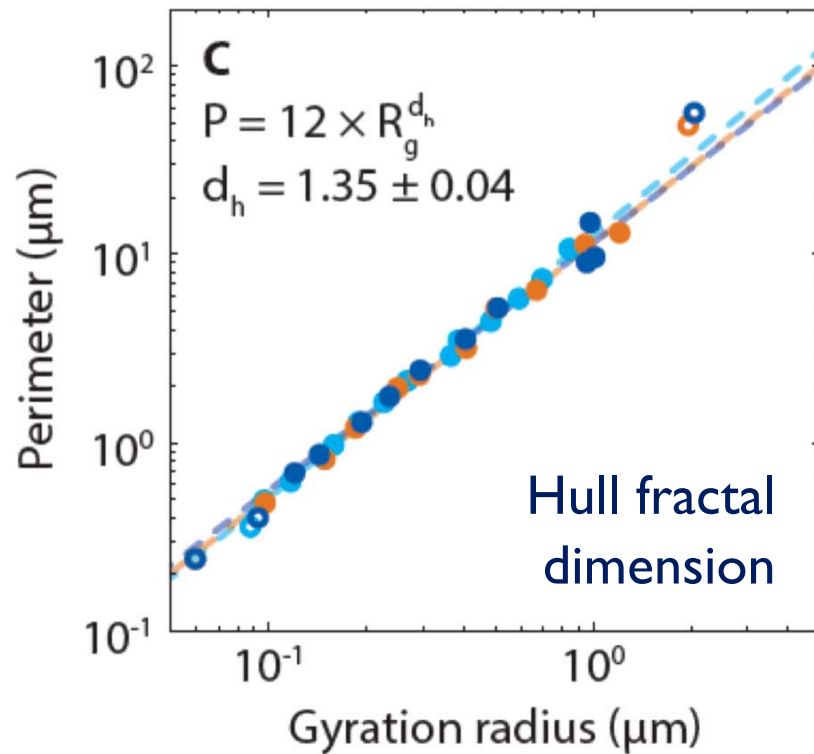
# Spin textures in nickelates

Domain map



Scale-invariant (power-law) domain distributions

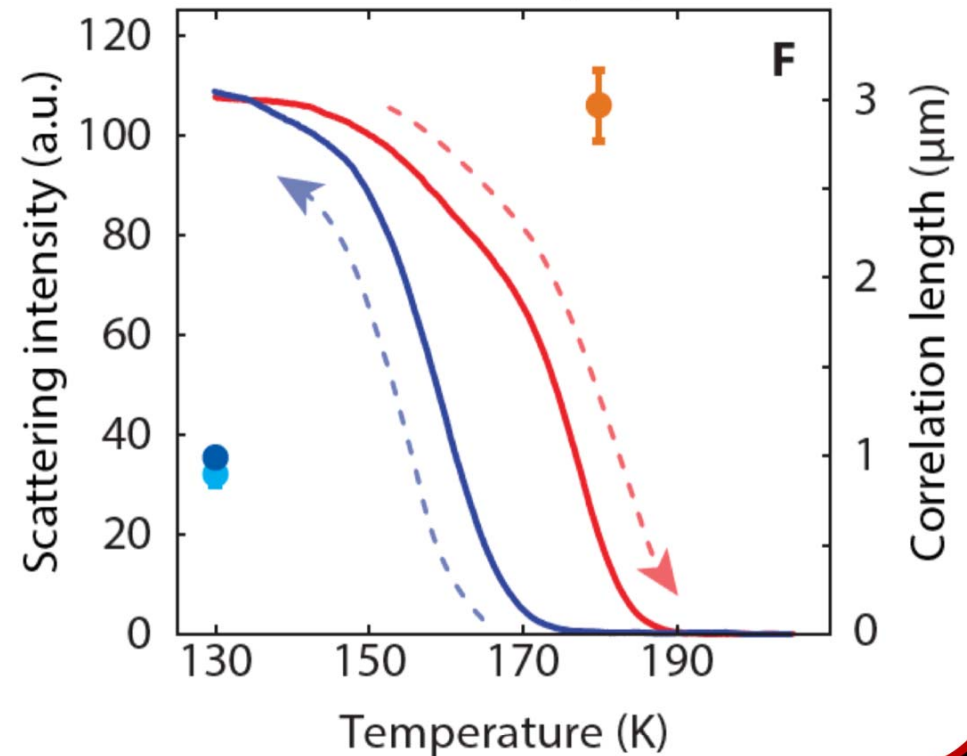
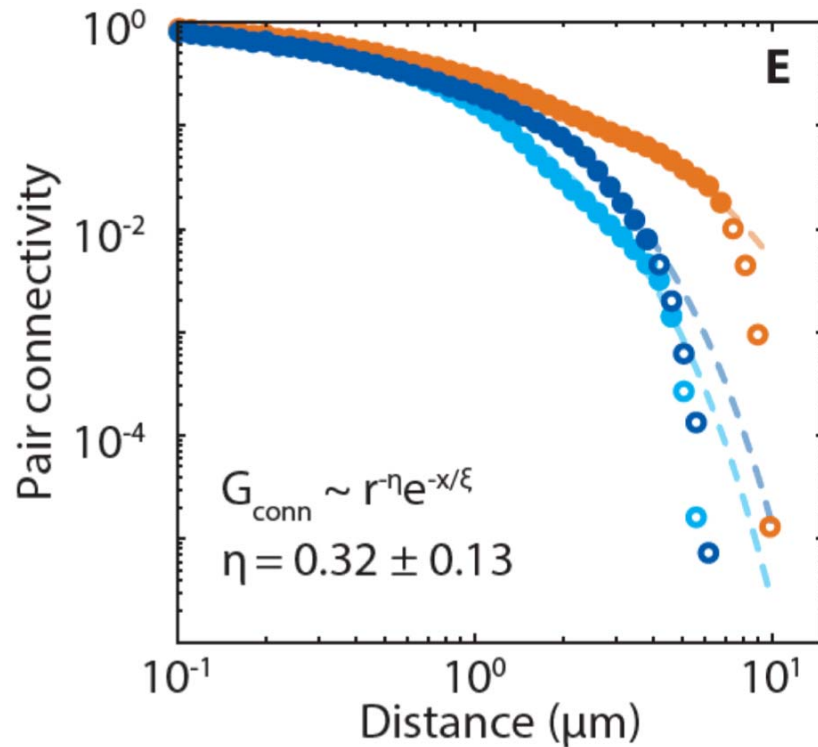
# Spin textures in nickelates



Non-Euclidean scaling between geometrical descriptors  
**Fractal** magnetic texture



# Spin textures in nickelates



Increasing pair connectivity correlation length  
near the Neel transition

# Resonant coherent diffractive imaging at 4<sup>th</sup> generation X-ray facilities

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# Coherent Lensless Imaging

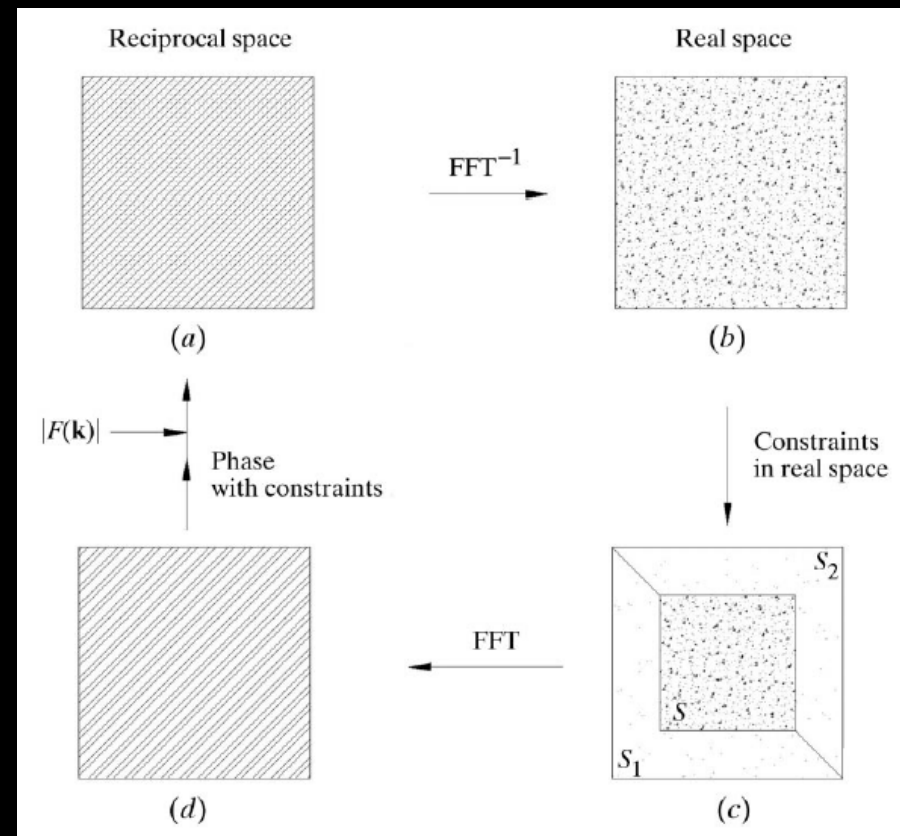
## Extending the methodology of X-ray crystallography to allow imaging of micrometre-sized non-crystalline specimens

Jianwei Miao\*, Pambos Charalambous†, Janos Kirz\* & David Sayre\*‡



## On possible extensions of X-ray crystallography through diffraction-pattern oversampling

J. Miao\*† and D. Sayre‡



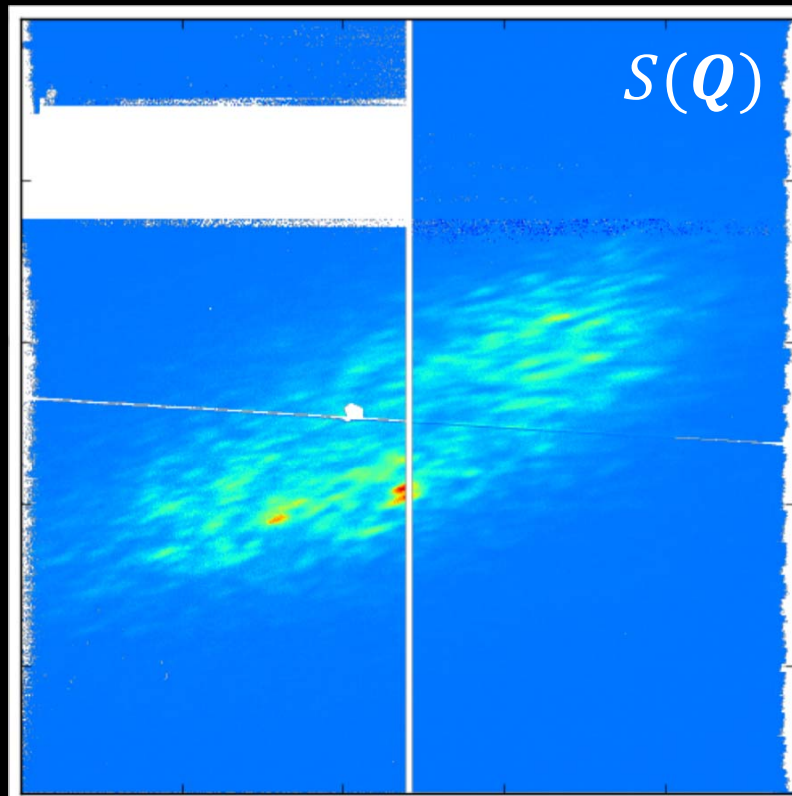
J. Miao, *et al.*, *Nature* **400**, 342 (1999)

J. Miao & D. Sayre, *Acta Cryst.* **A56**, 596 (2000)

D. Sayre, *Acta Cryst.* **5**, 843 (1952)

# Coherent Lensless Imaging

## RECIPROCAL SPACE



Coherent magnetic Bragg diffraction

$$S(\mathbf{Q}) = |\tilde{f}(\mathbf{Q})|^2$$



$$\tilde{f}(\mathbf{Q}) = \sqrt{S(\mathbf{Q})} \cdot \exp[i\phi(\mathbf{Q})]$$



$$\tilde{f}(\mathbf{r})$$

REAL SPACE

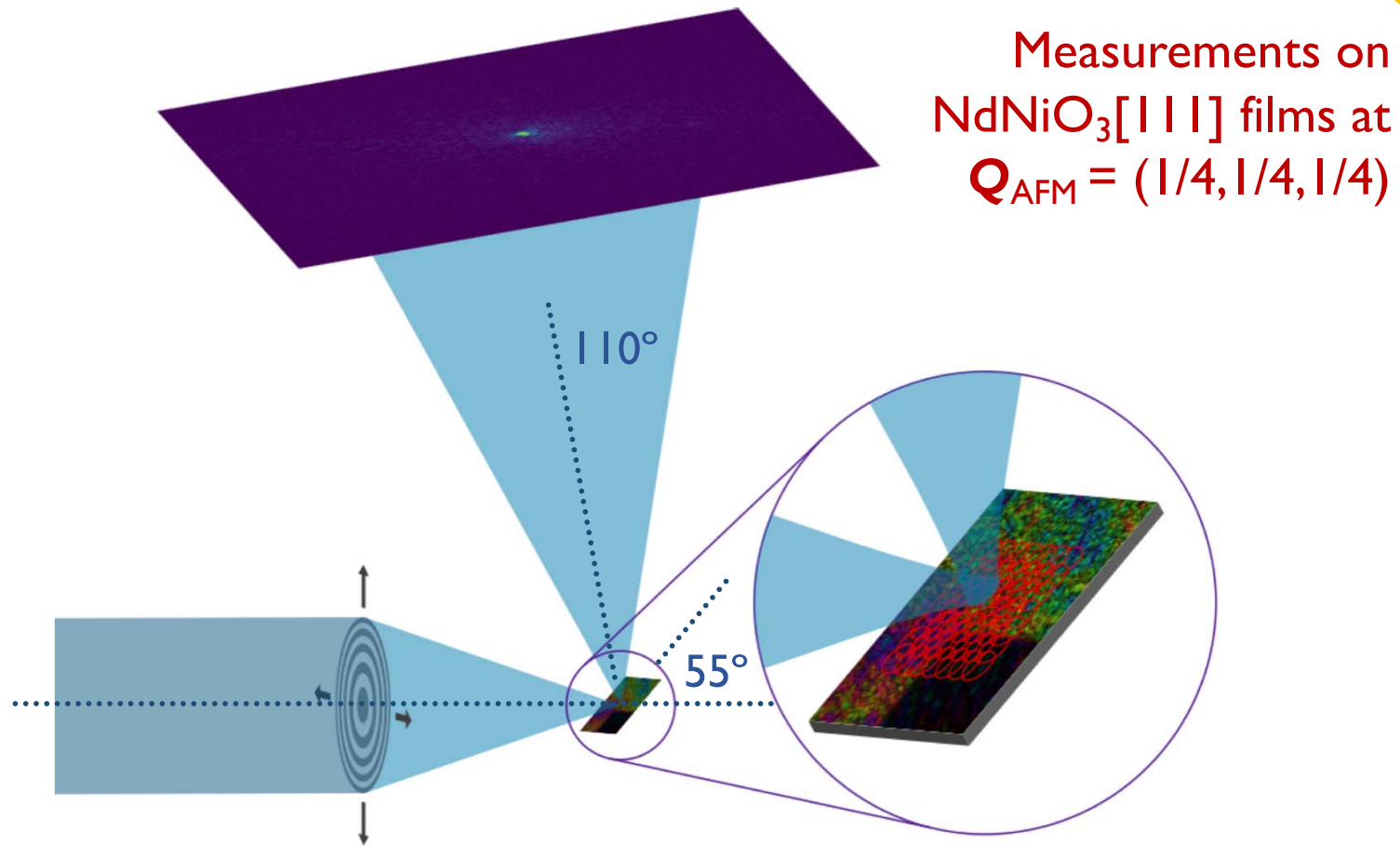
From measurement

We need to recover  
the phases of the  
exit waves

COMPUTATIONAL IMAGING

# Coherent Lensless Imaging

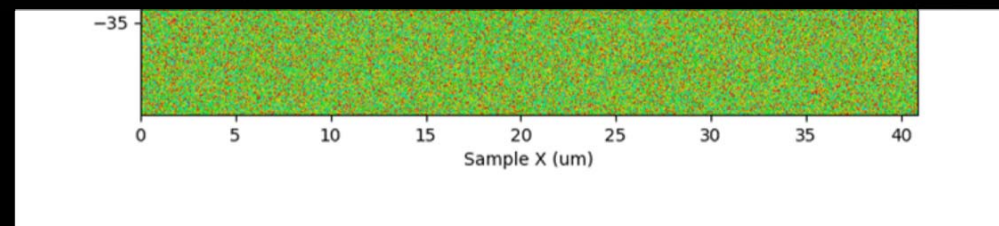
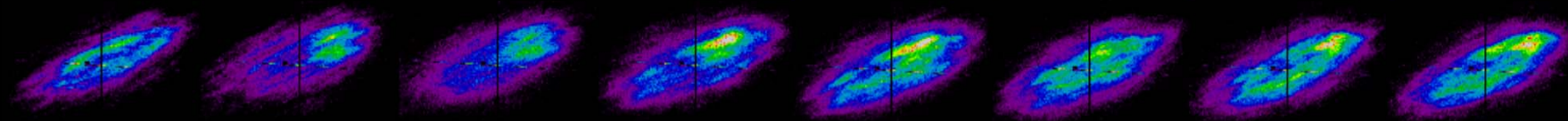
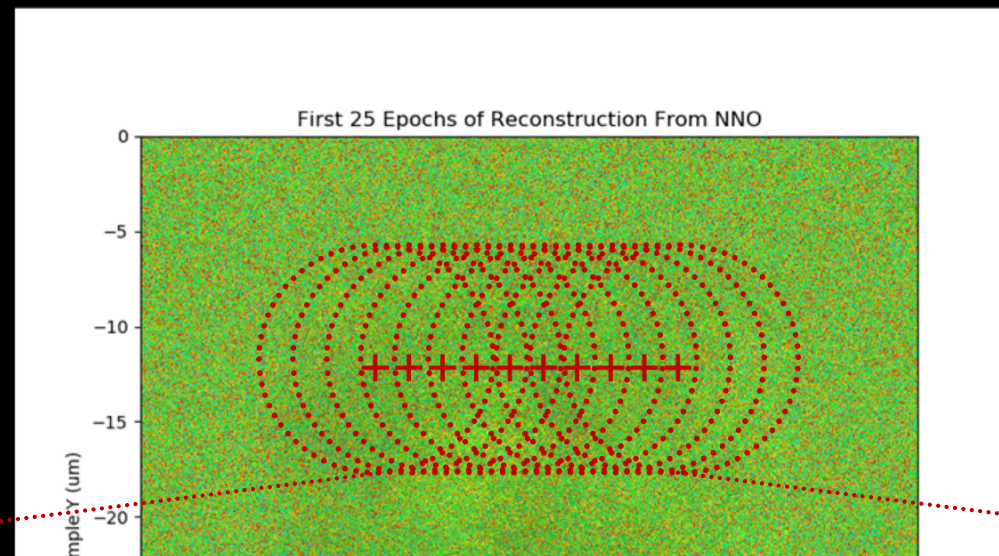
## Bragg ptychography



# Coherent Lensless Imaging

## Bragg ptychography

Measure overlapping regions and enforce a single-valued real field

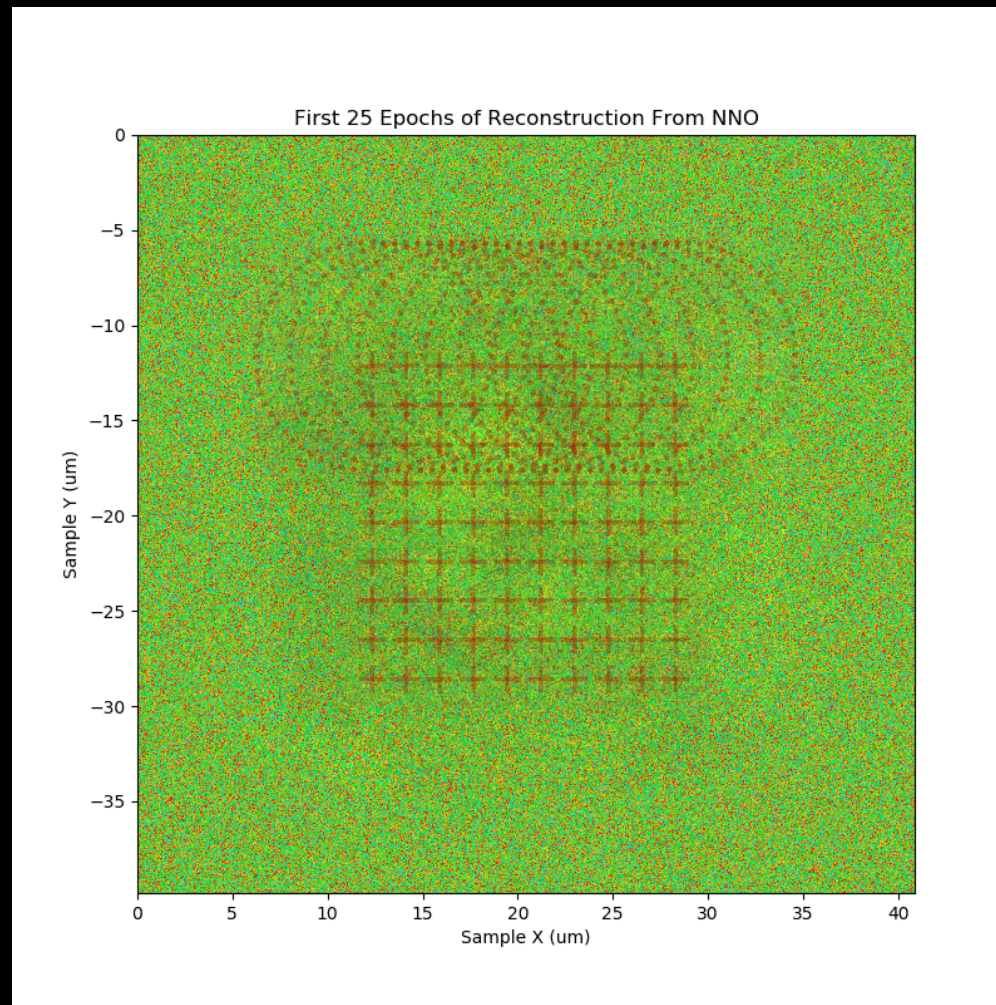


# Coherent Lensless Imaging

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## Bragg ptychography

Magnetic domain structure and nanoscale strain in rare earth nickelates



# Coherent Diffractive Imaging

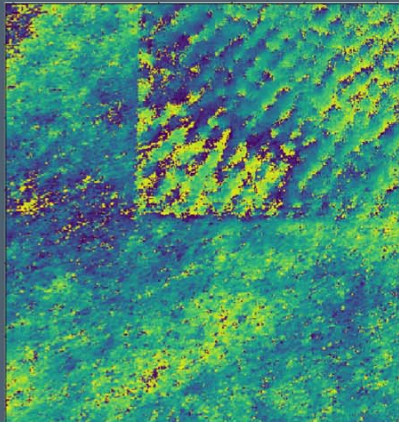
$$S(\mathbf{r}) = A(\mathbf{r}) \cos(\mathbf{Q}_{AFM} \cdot \mathbf{r} + \phi(\mathbf{r}))$$

Measurements on  $\text{NdNiO}_3[111]$   
thin films at  $\mathbf{Q}_{AFM} = (1/4, 1/4, 1/4)$

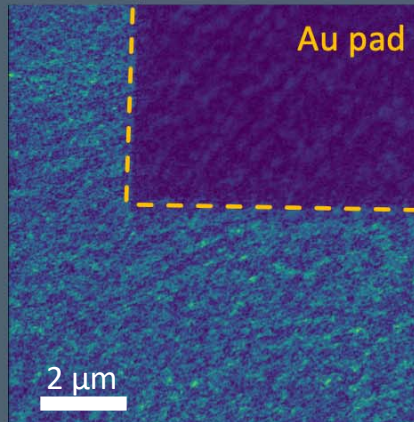


## Object reconstruction

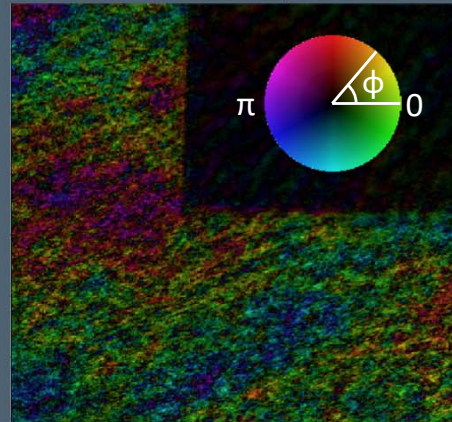
Phase  $\phi(\mathbf{r})$



Amplitude  $A(\mathbf{r})$

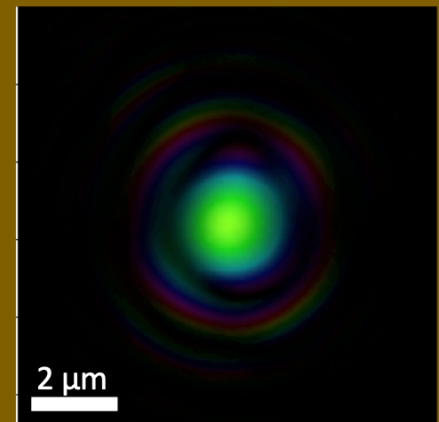


Phase + Amplitude



## Probe reconstruction

Phase + Amplitude

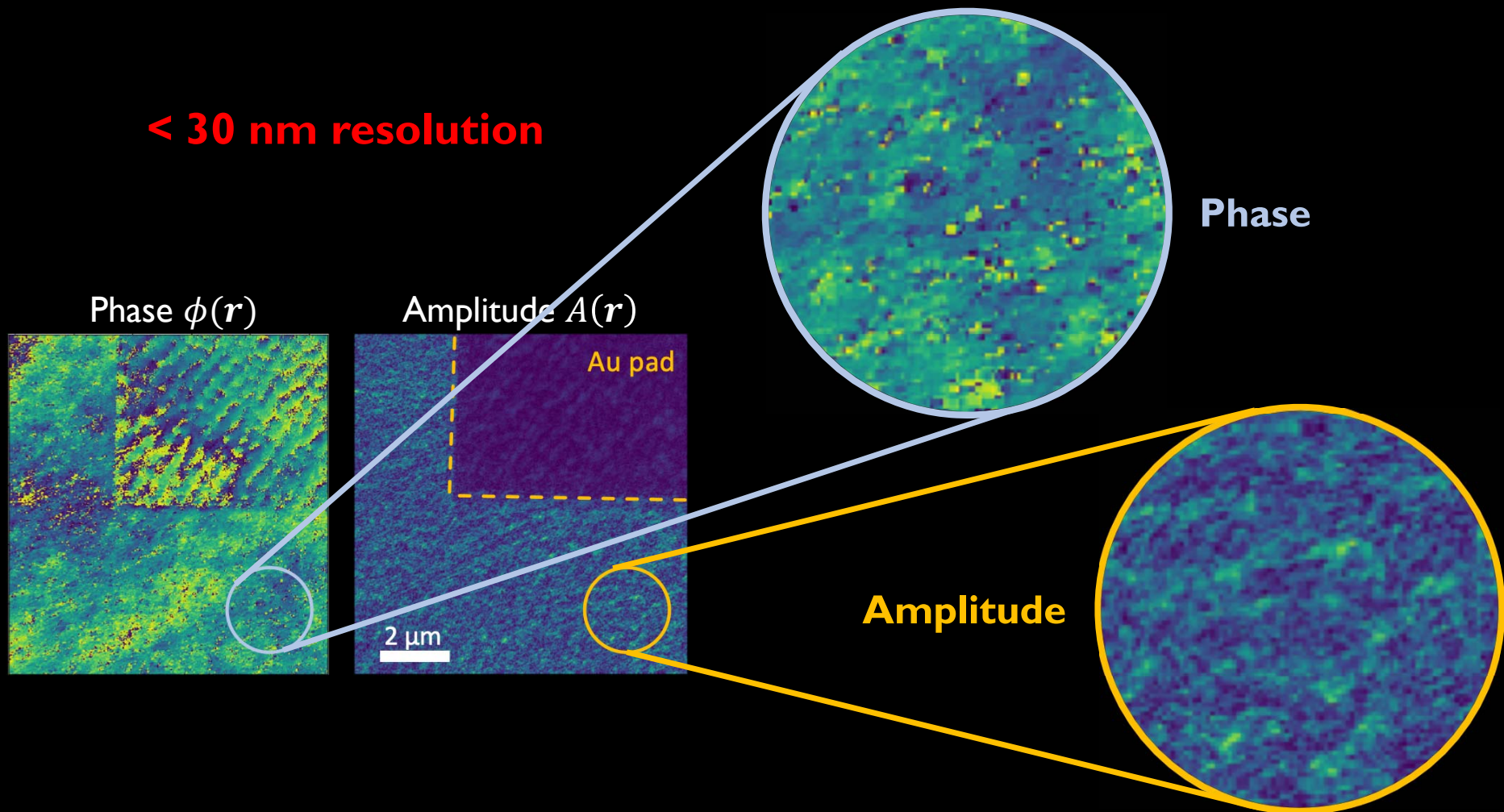




# Coherent Diffractive Imaging

$$S(\mathbf{r}) = A(\mathbf{r}) \cos(\mathbf{Q}_{AFM} \cdot \mathbf{r} + \phi(\mathbf{r}))$$

**< 30 nm resolution**



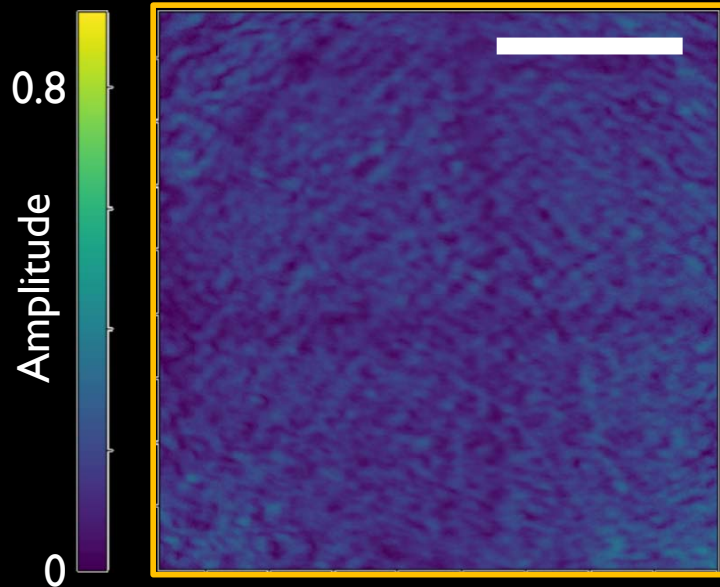
# Coherent Lensless Imaging

## Bragg ptychography

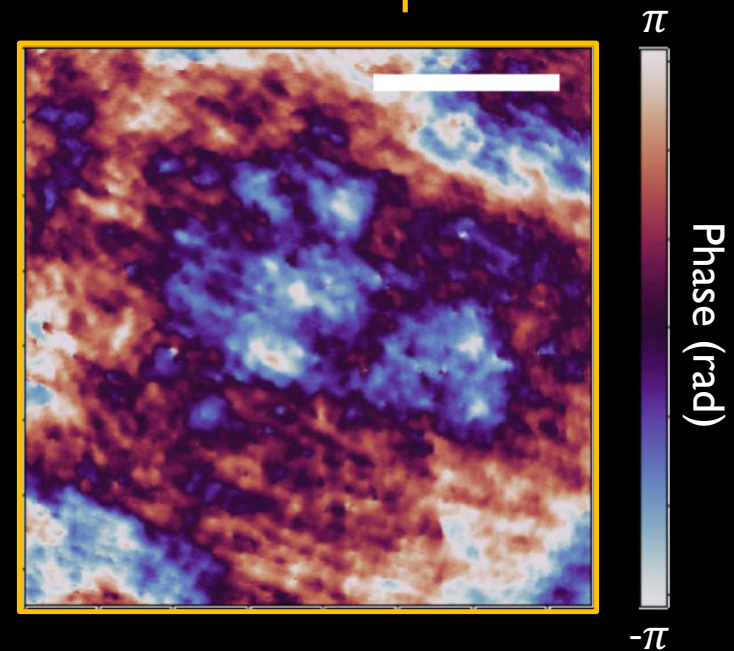
$$S(\mathbf{r}) = A(\mathbf{r}) \cos(\mathbf{Q}_{AFM} \cdot \mathbf{r} + \phi(\mathbf{r}))$$

ORDER  
PARAMETER

Amplitude map



Phase map

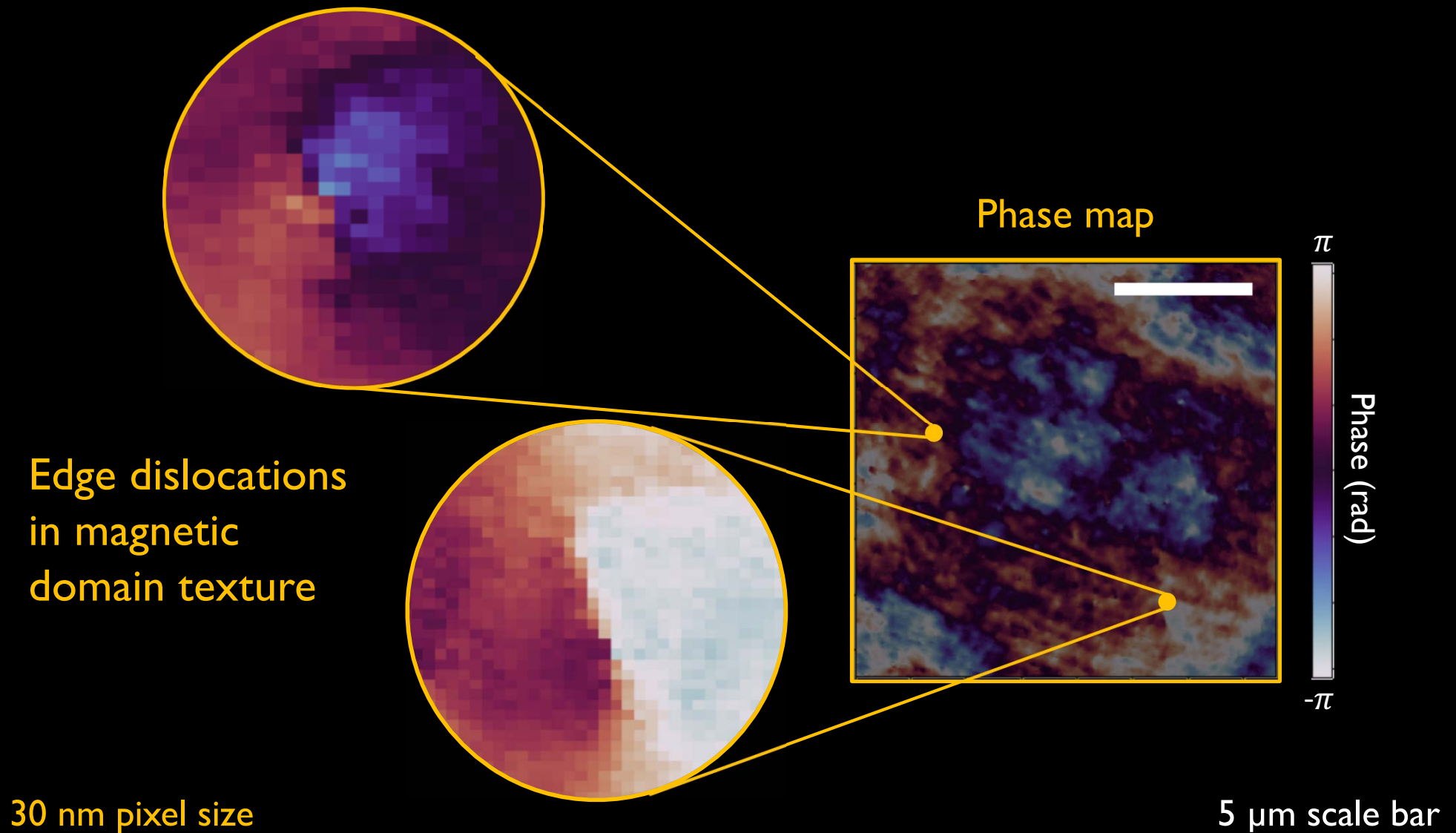


30 nm pixel size

5  $\mu\text{m}$  scale bar

# Coherent Lensless Imaging

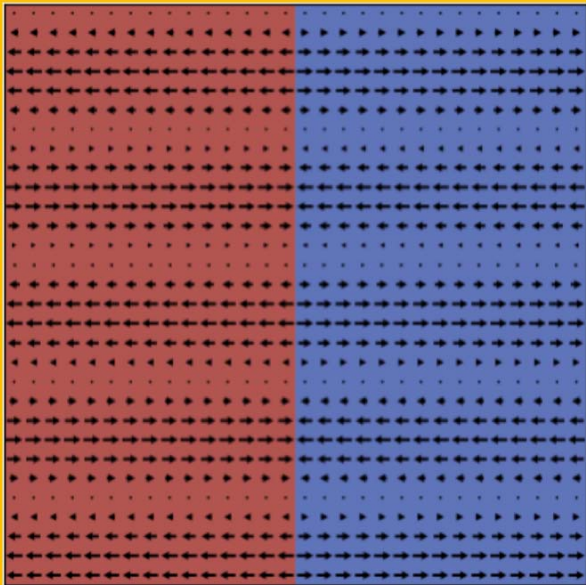
## Bragg ptychography



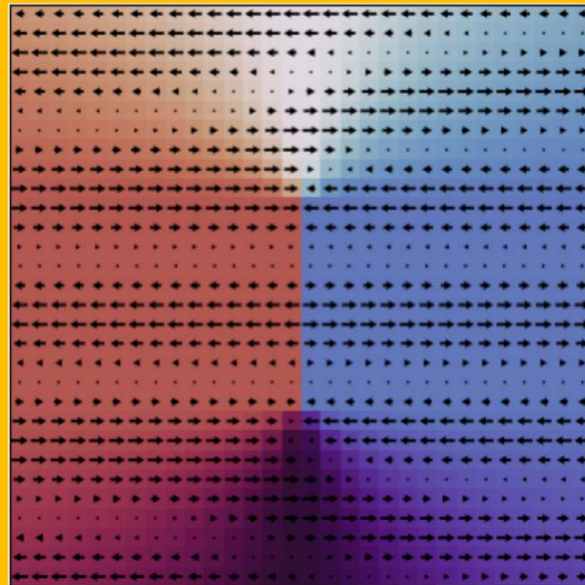
# Coherent Lensless Imaging

## Defects in magnetic order parameter

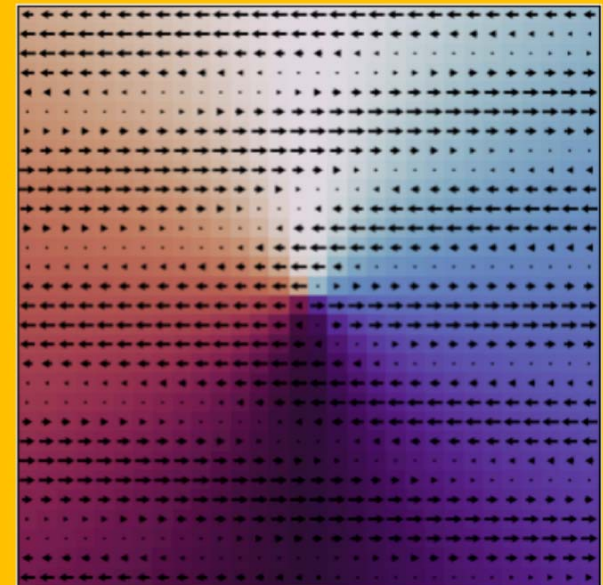
Line domain wall



Edge dislocation



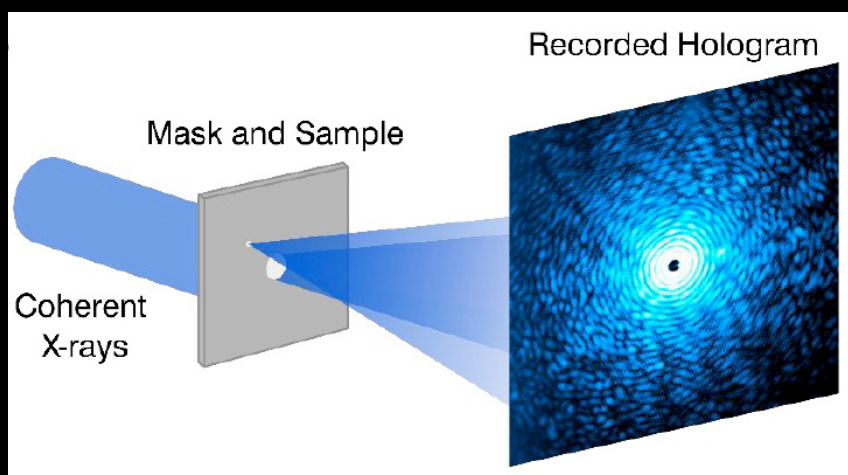
Point defect



# Coherent Diffractive Imaging

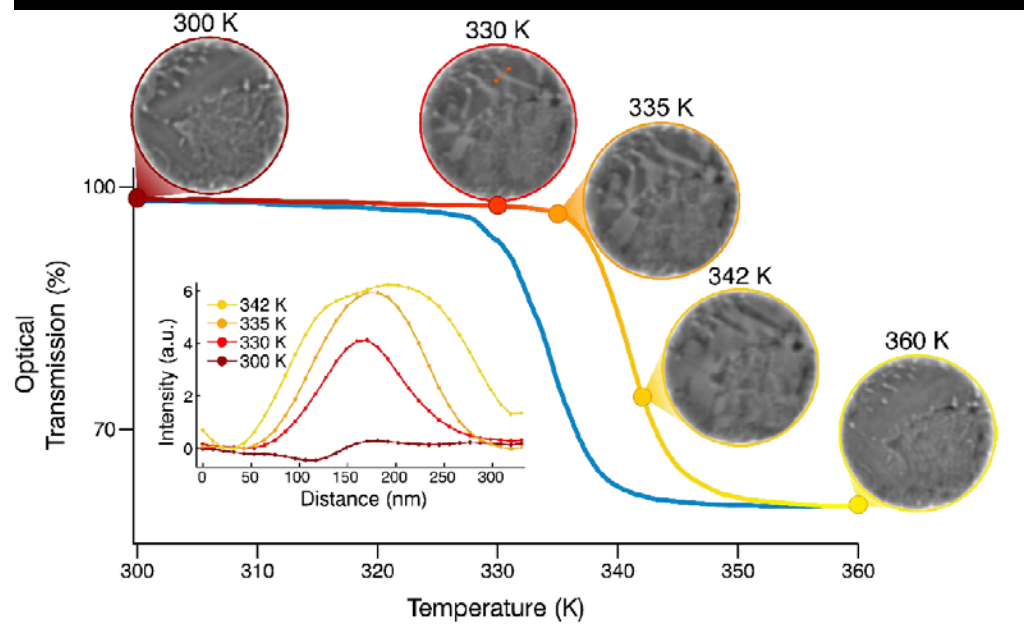
## Moving forward to single-shot imaging

### Resonant holography



### Metal-insulator transition in $\text{VO}_2$ (~40 nm resolution)

Imaging charge/spin textures  
around  $Q \sim 0$  (forward  
scattering)



# Coherent Diffractive Imaging @ CXFEL

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## Typical probing conditions for resonant soft X-ray CDI:

- 500-1000 eV range, tunable
- Polarization control
- Single mode probe improves quality and robustness of reconstruction
- $10^{12}$  ph/s flux yields peak count rate:  $\sim 10$ -100 kHz/pix
- Transverse (longitud.) coherence length  $> 10 \mu\text{m}$  (500 nm)
- Need focusing down to  $\sim 1$ -5  $\mu\text{m}$

vs.

## SX seeded FEL @ FERMI possible targets?:

Hopefully!

YES

Seeded beam is more ideal than SASE

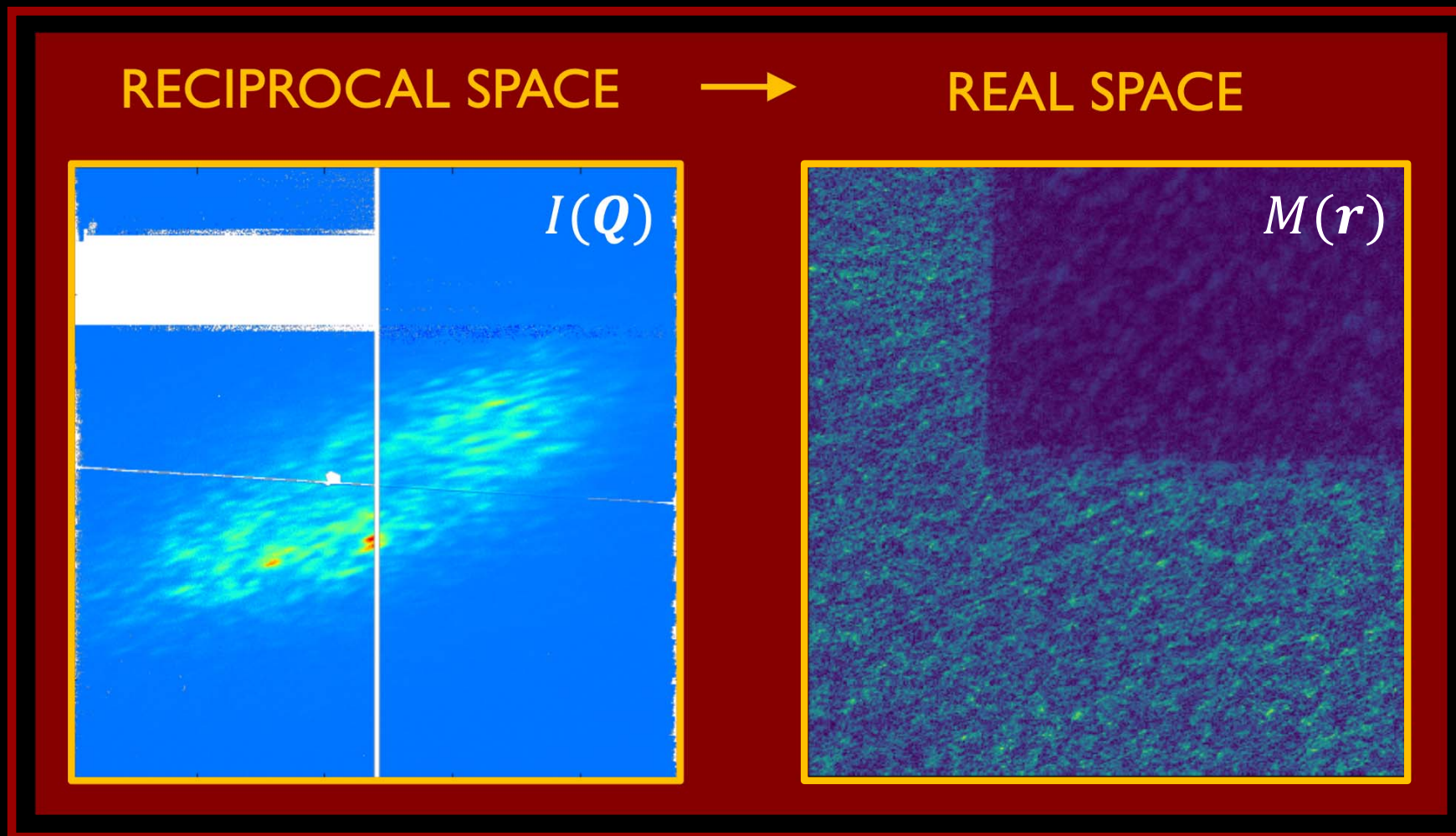
$10^{11-12}$  ph/pulse for single shot experiments

Long. OK –  $\lambda/\Delta\lambda > 1000$

Transv. presumably also OK

sub- $\mu\text{m}$  with appropriate optics

# Summary



Spatiotemporal imaging requires a full-field, single-shot probe of real-space textures with stable wavelength. New opportunities for soft X-ray diffractive imaging to reveal the nanoscale dynamics of collective states of matter

*Thank you for your attention!*



