Coherent Diffraction Investigation of Domain Excitations in Quantum Materials

Ian Robinson
David Yang
Ross Harder
Dina Sheyfer
Longlong Wu
Jack Griffiths
Emil Bozin
Mark Dean
Jialun Liu
Hengdi Zhao
Gang Cao

Angel Rodriguez-Fernandez
Jan-Etienne Pudell
Roman Shayduk
James Wrigley
Alexey Zozulya
Ulrike Boesenberg
Joerg Hallmann
Anders Madsen

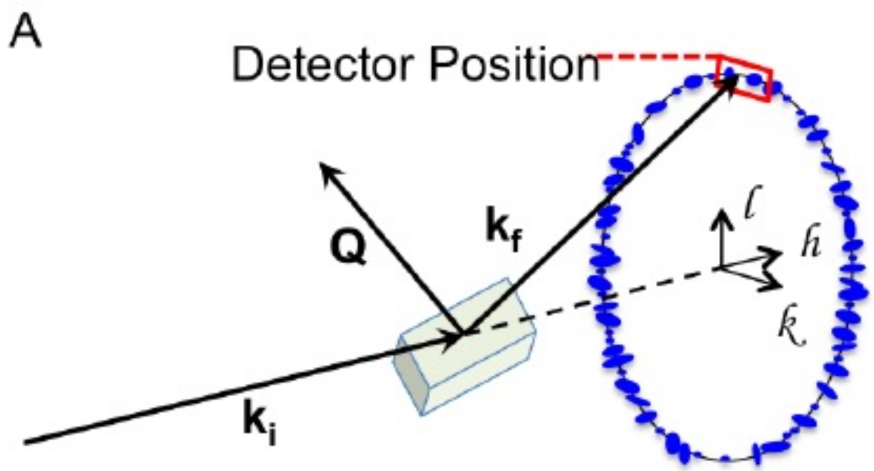
University College London
Brookhaven National Laboratory
Argonne National Laboratory
University of Colorado at Boulder
European XFEL

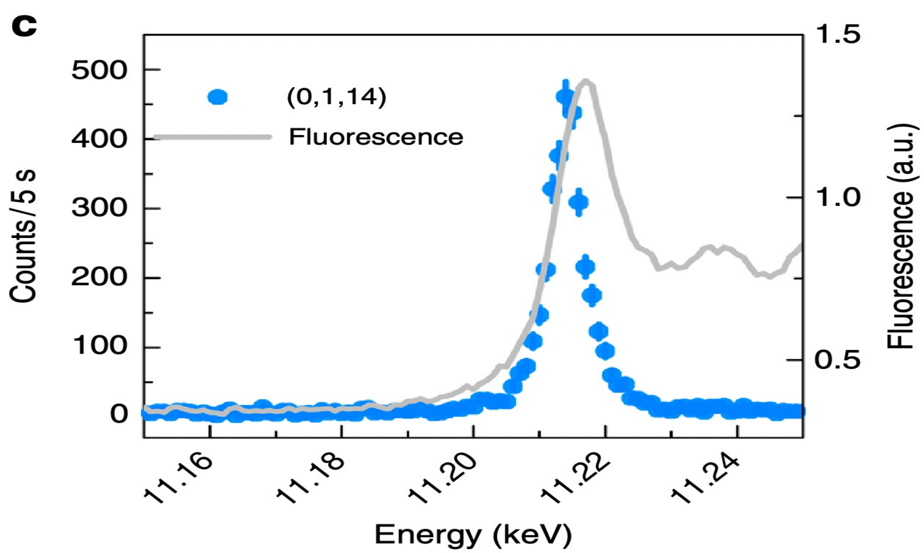
Emerging ultrafast diffraction facilities in the UK: LUXD, EPAC and RUEDI
Rutherford Appleton Laboratory, Harwell, UK, November 2025

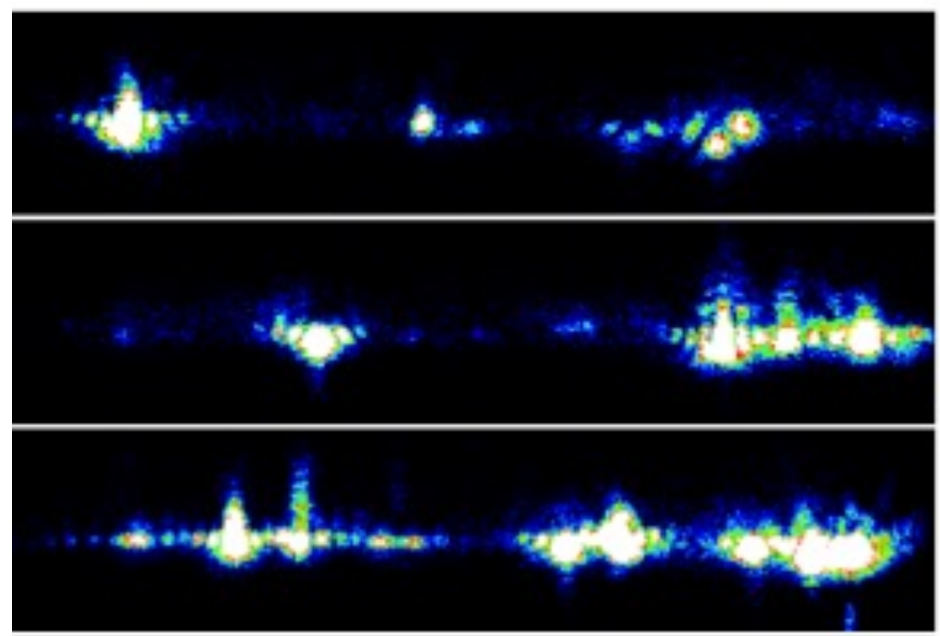
Outline

- Bragg Coherent Diffraction Imaging
- Sensitivity to strain
- Phase domain structures
- Antiferromagnetism of Sr_2IrO_4
- Pump-probe XFEL experiment
- Migration of magnetic domains
- Future RUEDI experiment

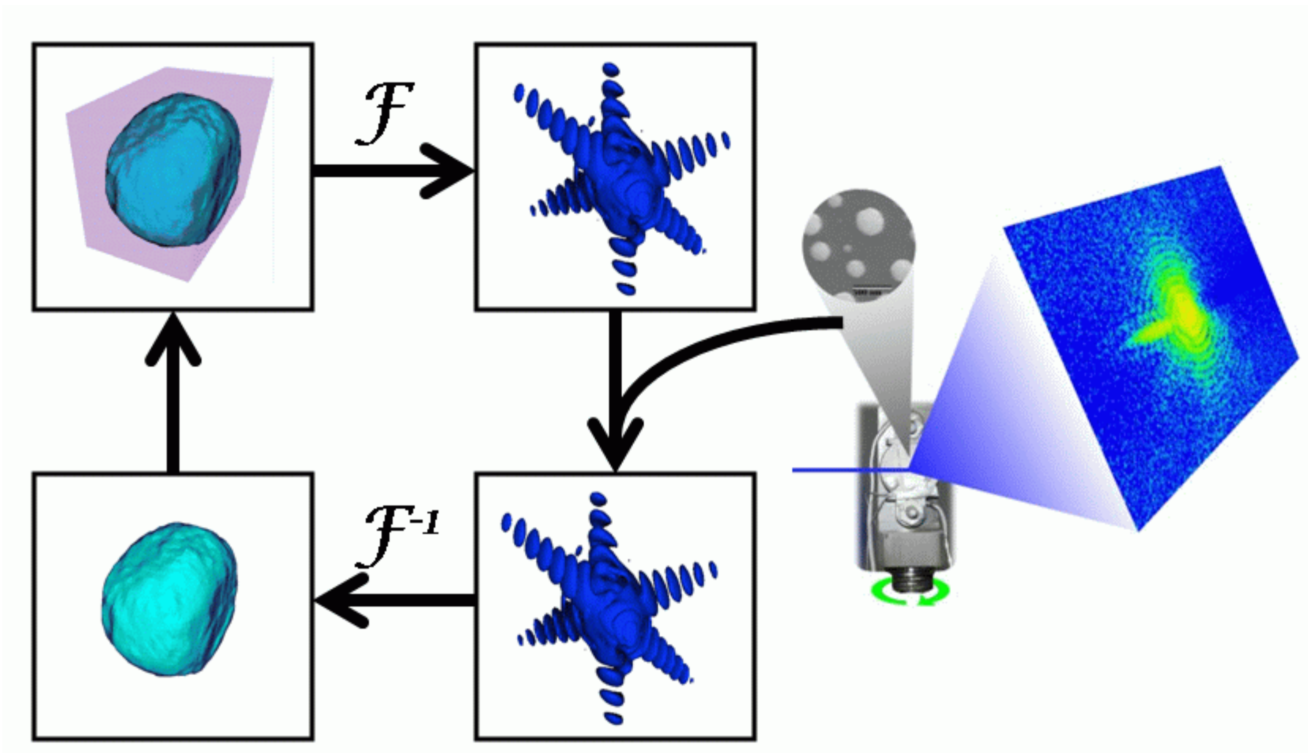
Magnetic Coherent X-ray Diffraction







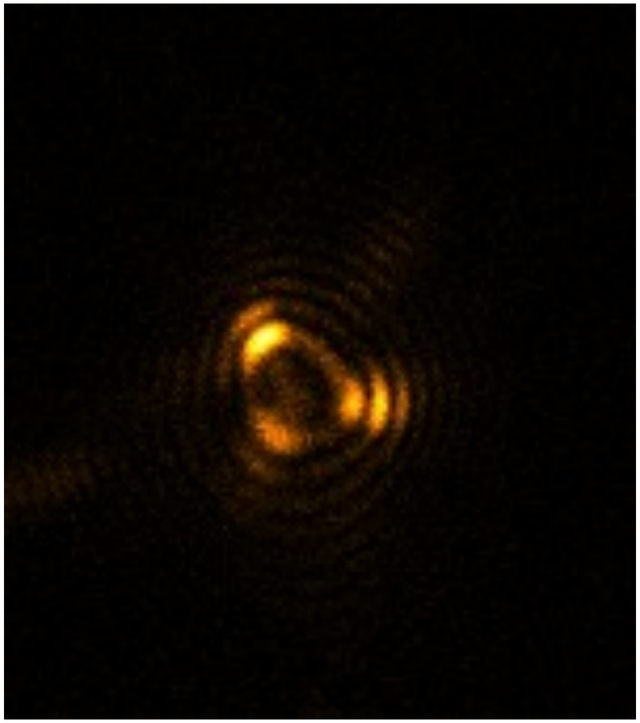
Generic "Error Reduction" method

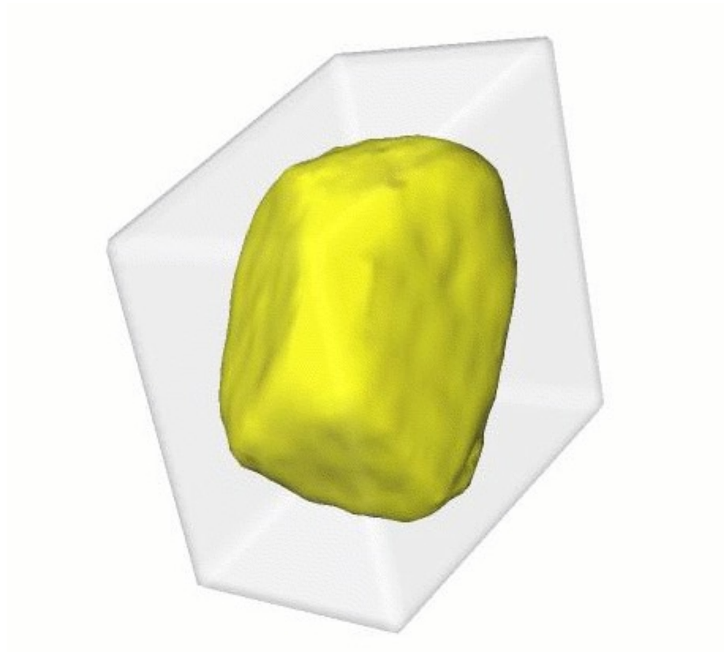


J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

Gold nanocrystal reconstruction

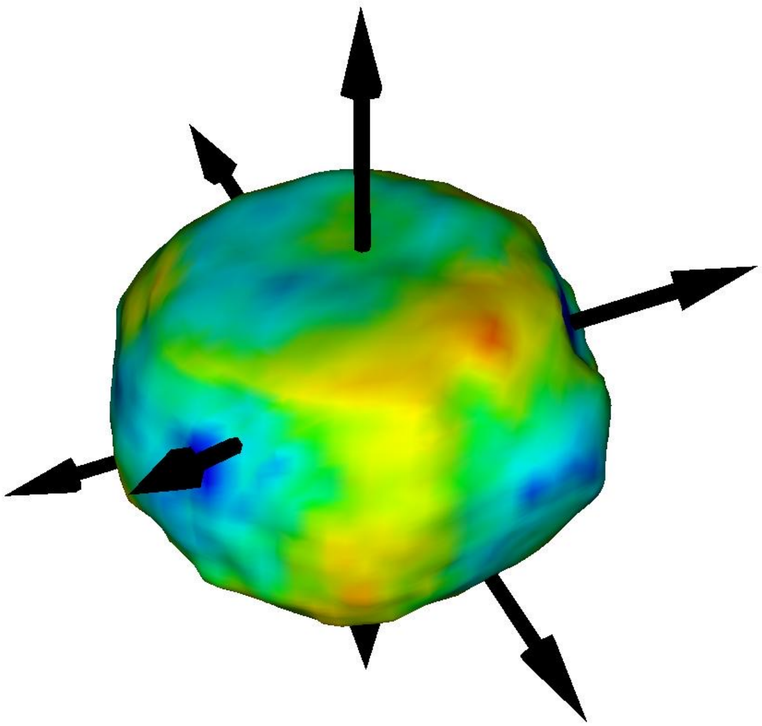
showing support used for 20 HIO followed by 10 ER

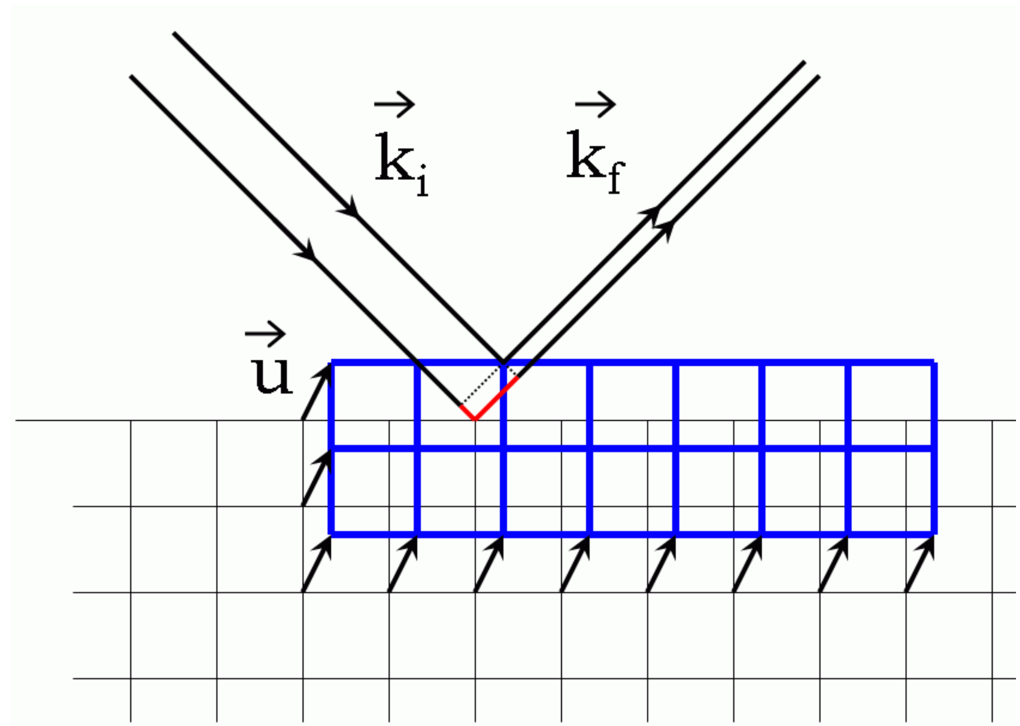




Phase isosurface showing residual strain

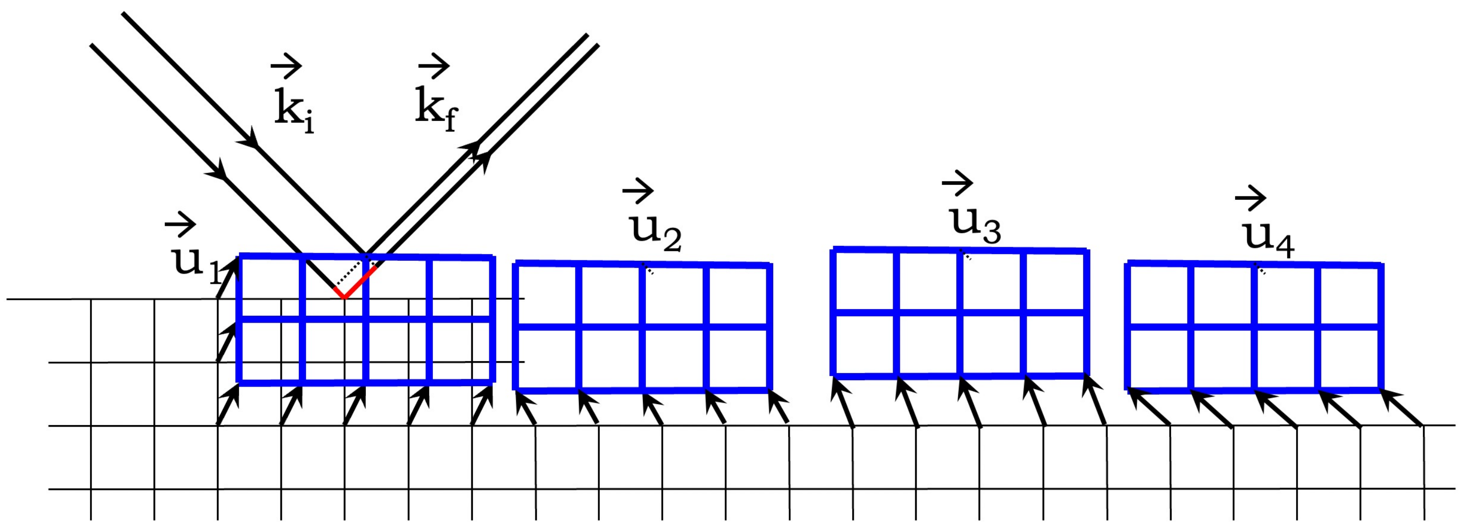
$$\Delta\phi = \mathbf{k_f} \cdot \mathbf{u} - \mathbf{k_i} \cdot \mathbf{u} = \mathbf{Q} \cdot \mathbf{u}$$





Sensitivity to domain offsets

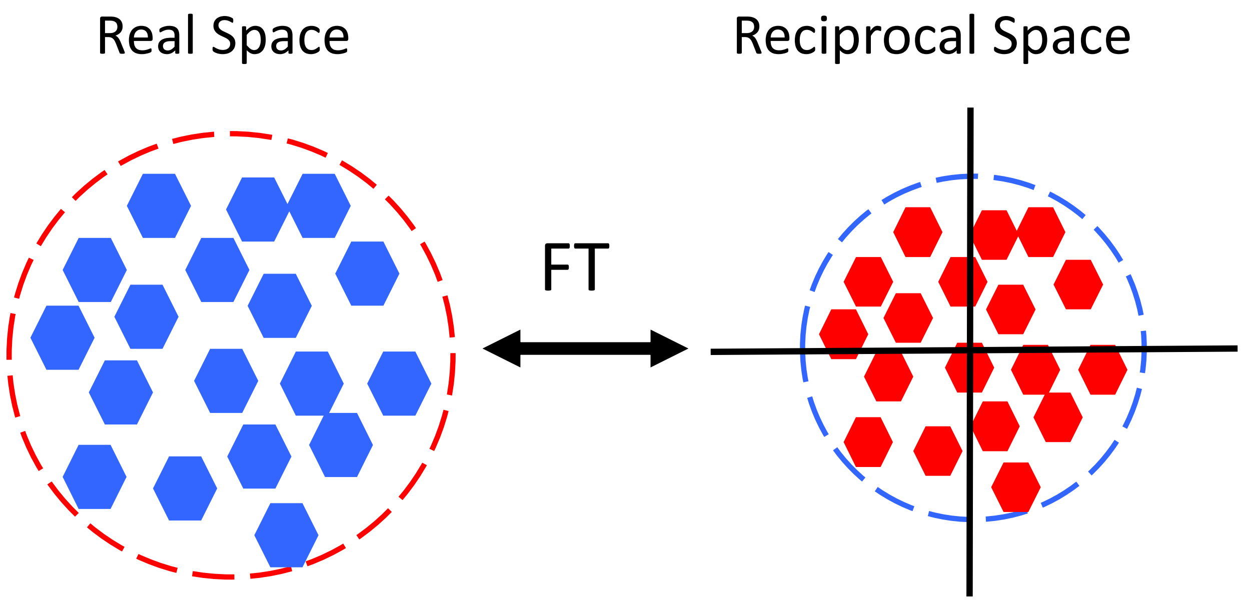
$\Delta\phi = \mathbf{k_f} \cdot \mathbf{u} - \mathbf{k_i} \cdot \mathbf{u} = \mathbf{Q} \cdot \mathbf{u}$



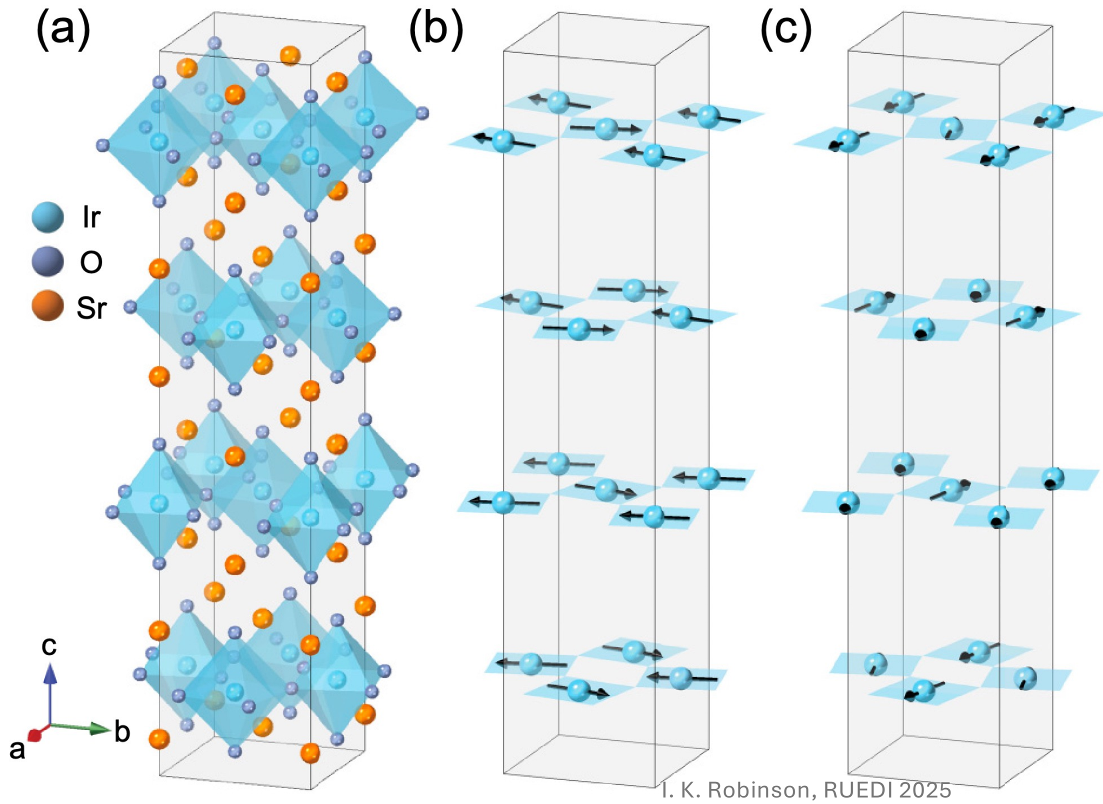
I. K. Robinson, Synchrotron Radiation News (2024)

Domain Counting by Coherent Diffraction

Ian Robinson et al, J. Superconductivity and Novel Magnetism (2019)



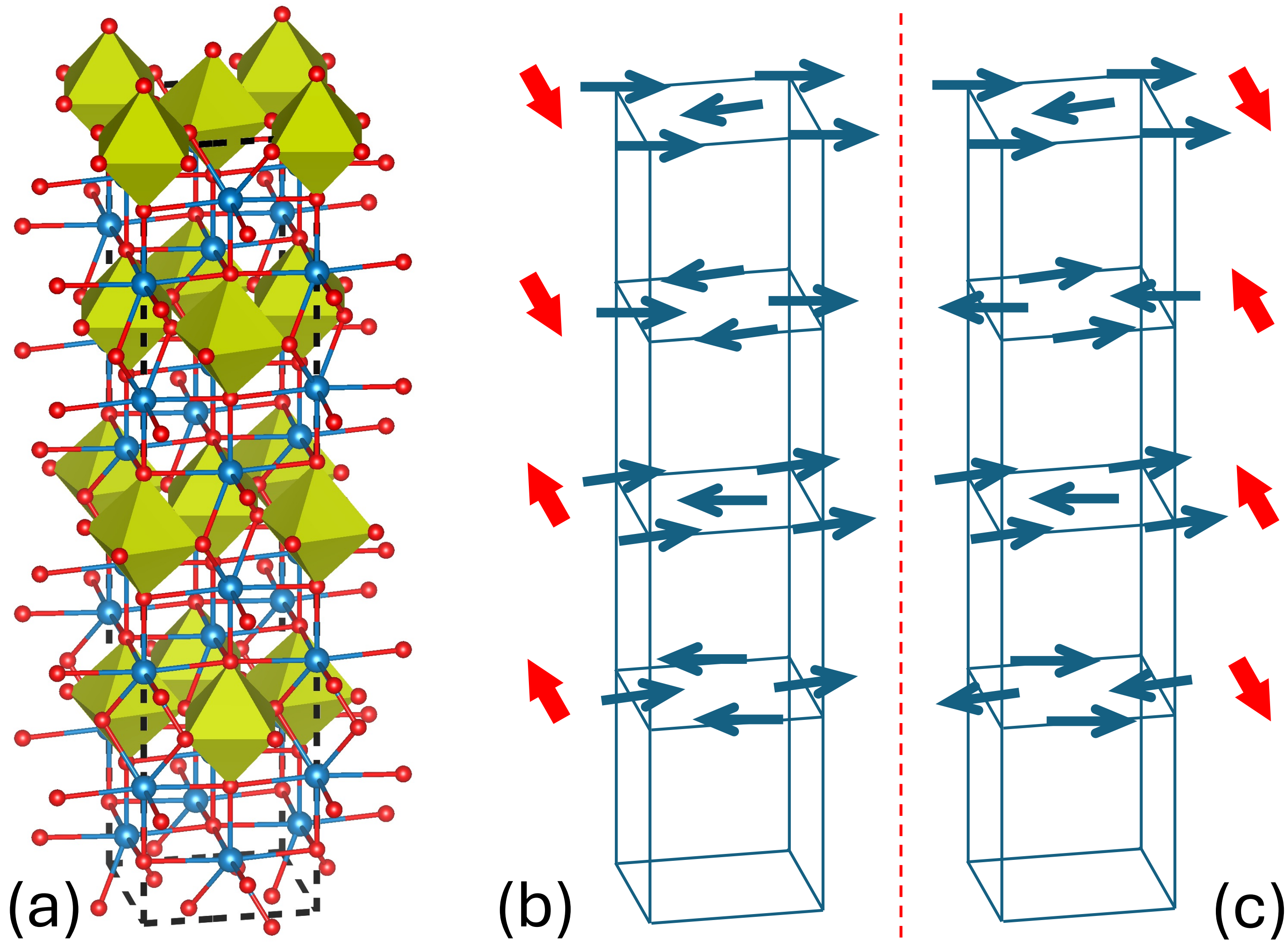
Sr_2IrO_4 has "214" Layered square planar structure like $La_{2-x}Sr_xCuO_4$ superconductors



Strong AFM peaks at
106 first domain
016 second domain
108 second domain

pseudo tetragonal
unit cell

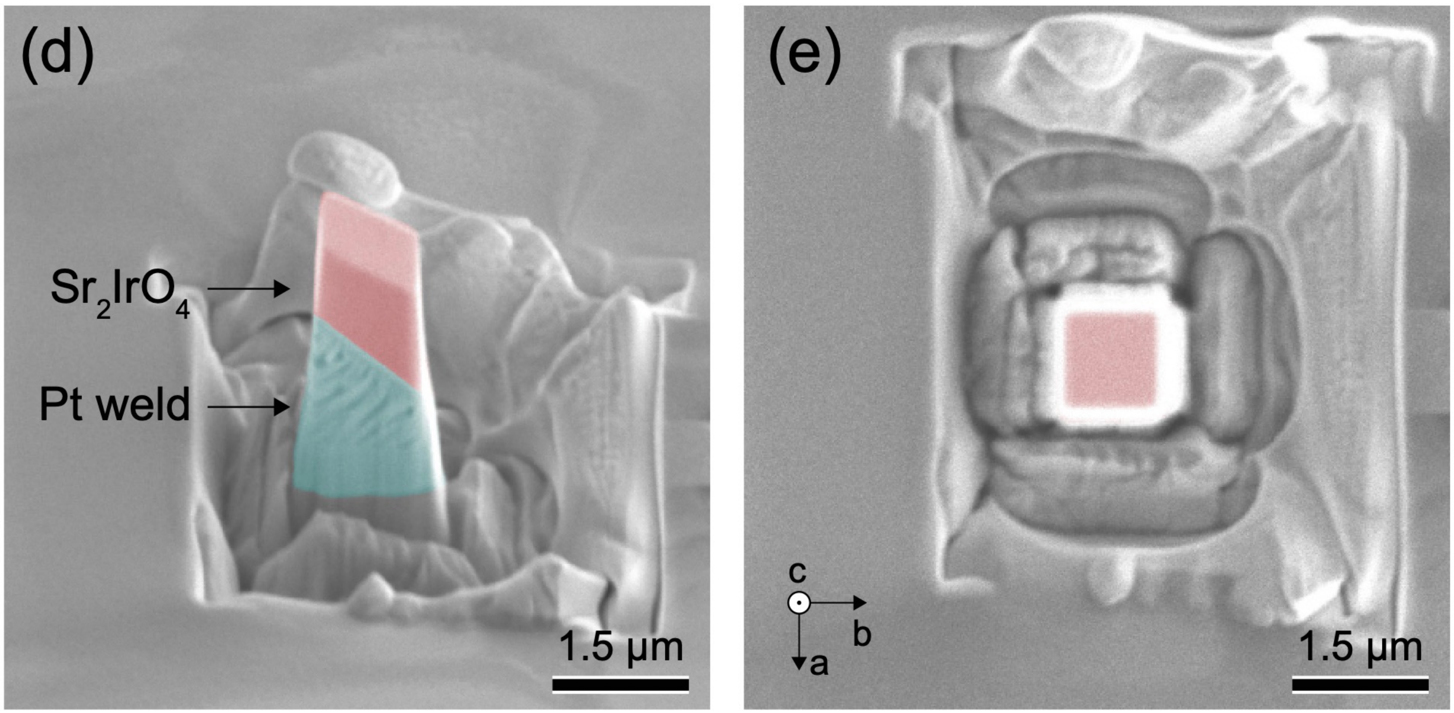
resonance at Ir L_3
11.215 keV


(a)
(b)
(c)

FIB sample preparation (Kim Kisslinger CFN BNL)

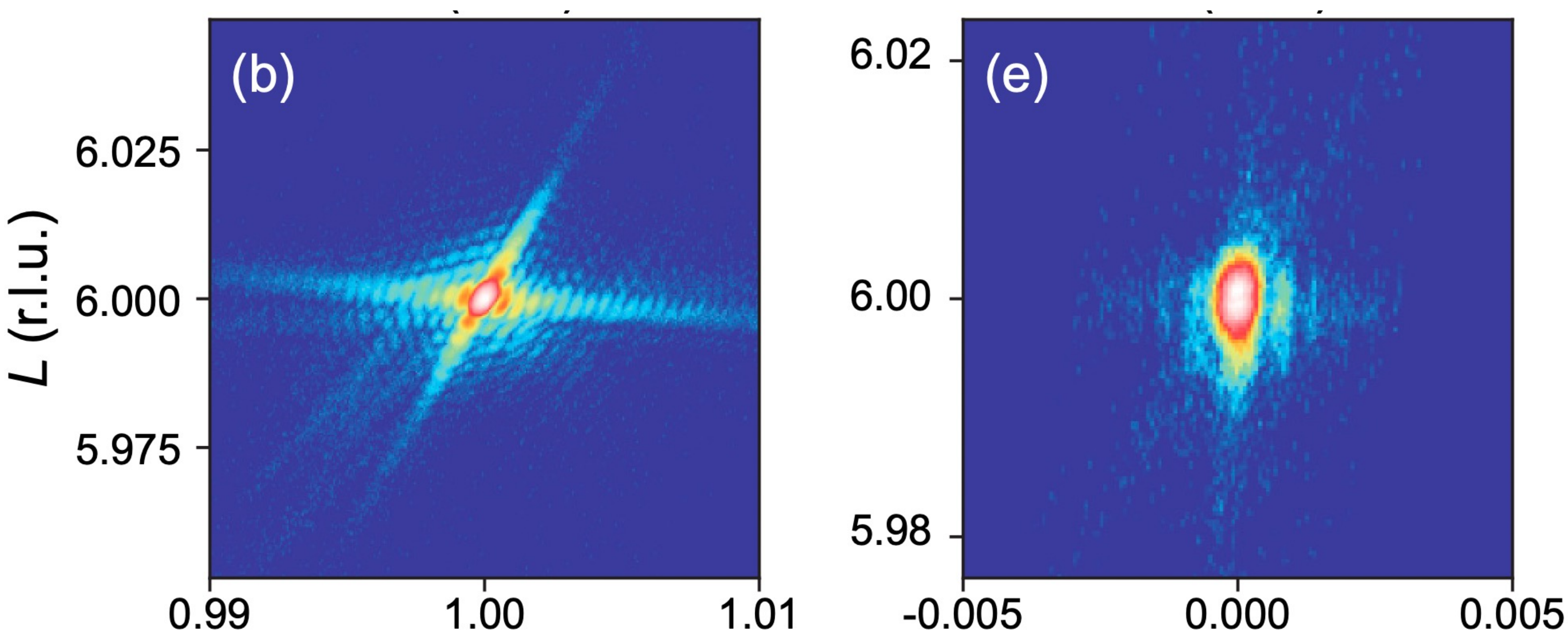
34-ID-C Medipix at 2.5m: 1.8x1.8x1.8 mm crystals

MID-XFEL Jungfrau at 8m: 5x5x5 μm and 4x4x4 μm crystals



116 charge and 106 magnetic diffraction peaks

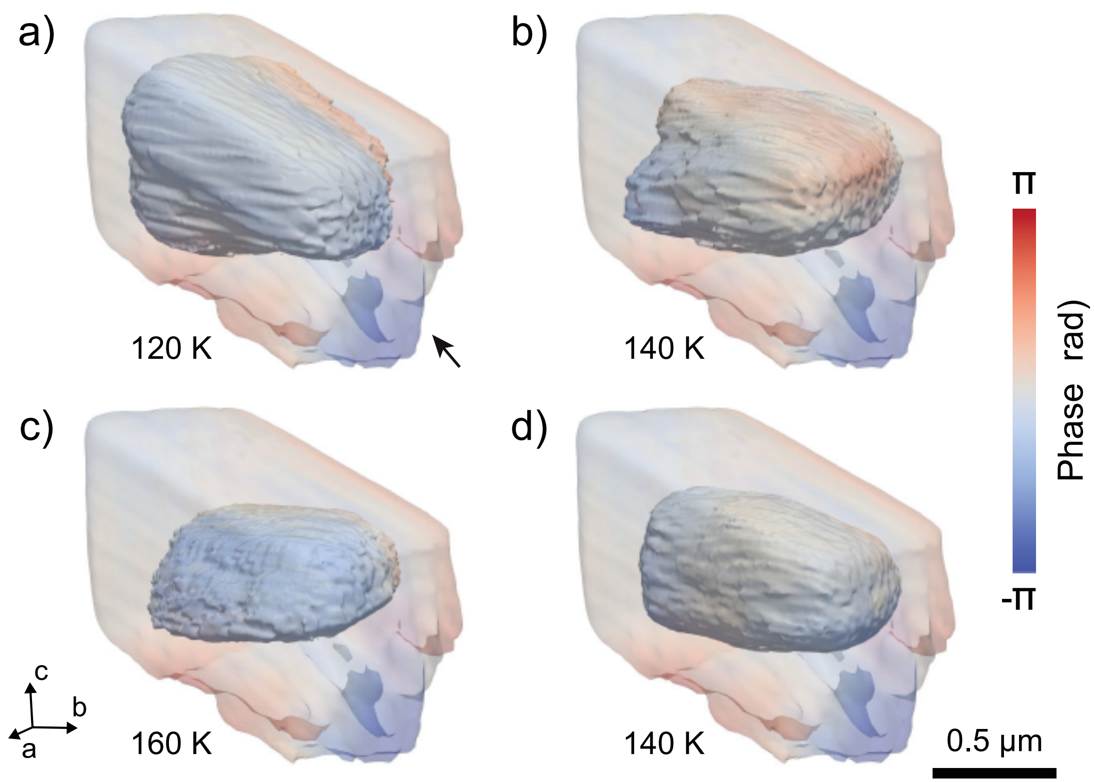
Resonant on Ir L_3 at 11.215 keV



length (nm)	(116) peak	120 K	140 K	160 K
ξ_H	1153 ± 6	1137 ± 4	1158 ± 4	1030 ± 13
ξ_K	970 ± 3	1019 ± 15	1083 ± 20	871 ± 54
ξ_L	902 ± 3	739 ± 2	610 ± 3	423 ± 9

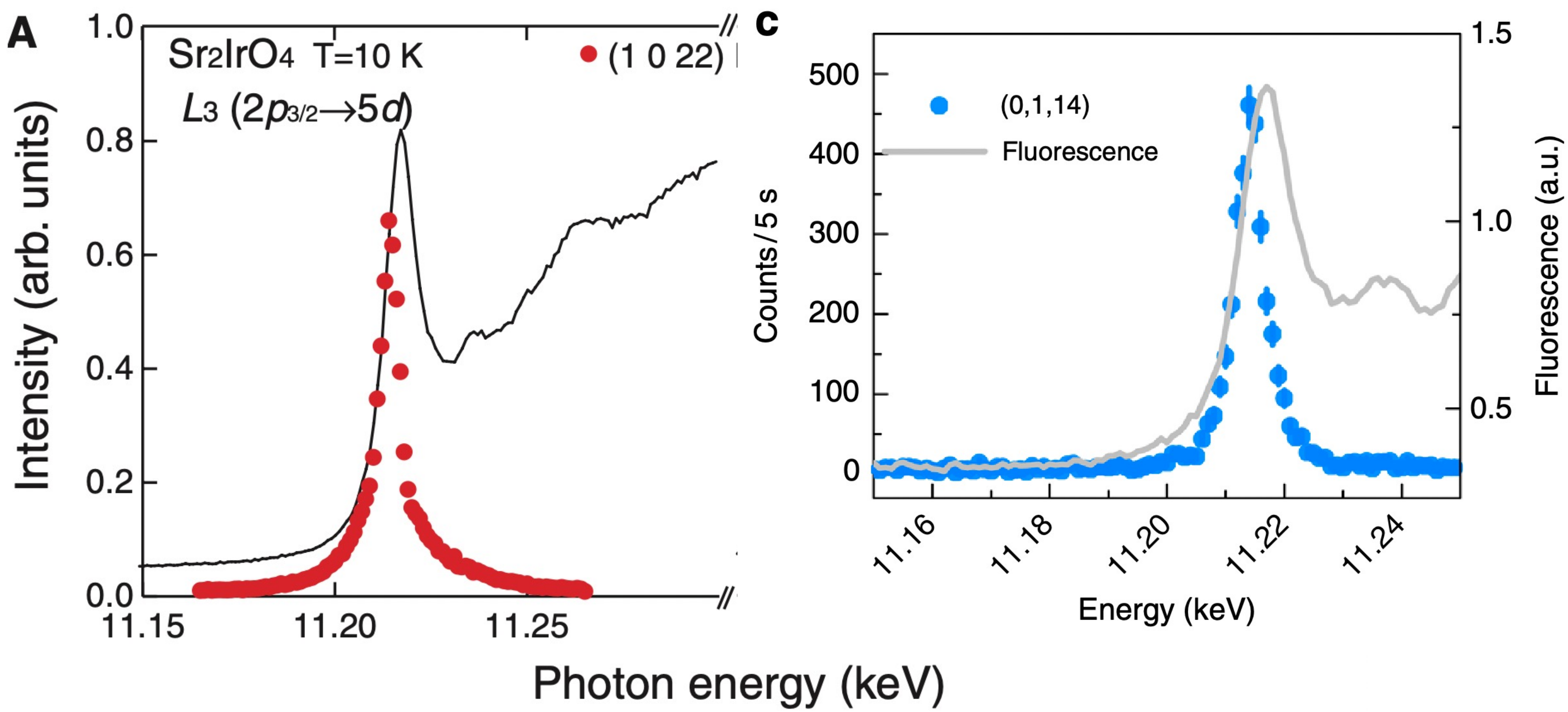
Magnetic BCDI images of single AFM domain

Anisotropy of Antiferromagnetic Domains in a Spin-orbit Mott Insulator
Longlong Wu et al Physical Review B 108 L020403 (2023)



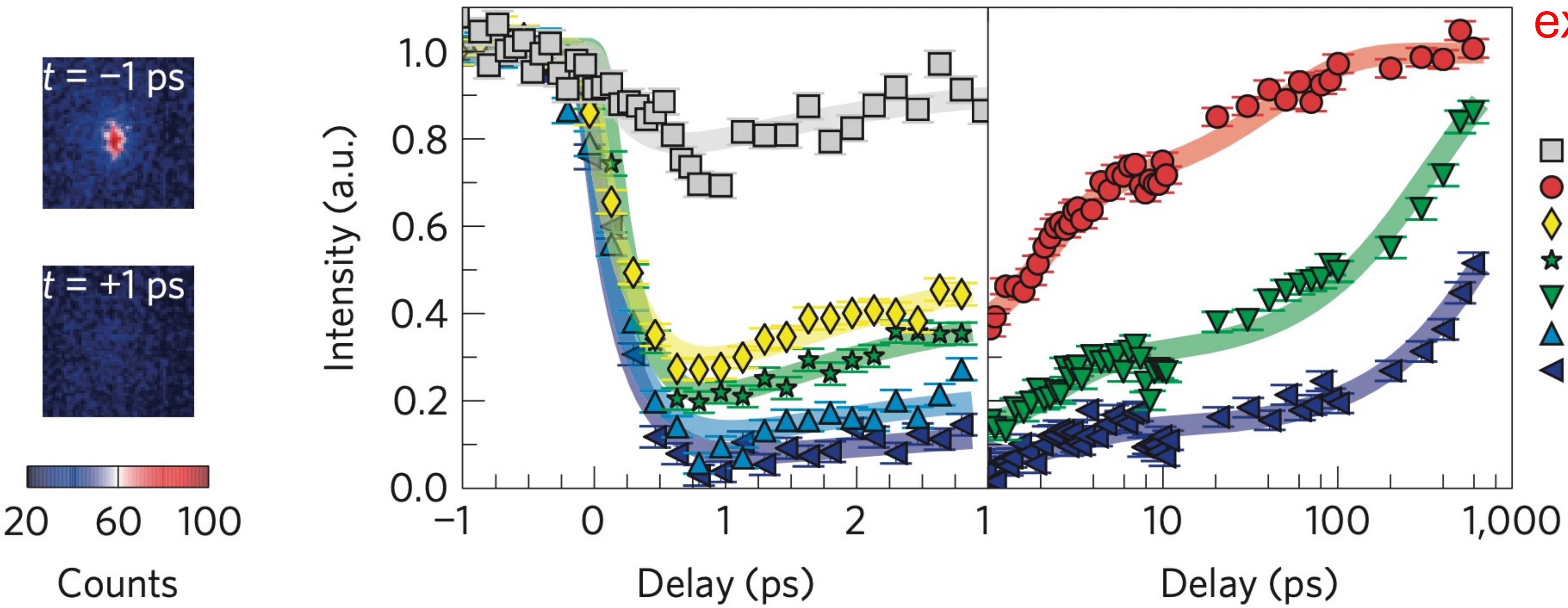
Magnetic Bragg peak resonance at 11.215 keV

B.J. Kim et al Science (2009); K. Finkelstein Nat Comm (2018)



Ultrafast energy- and momentum-resolved dynamics of magnetic correlations in the photo-doped Mott insulator Sr_2IrO_4 [XPP LCLS]
M. P. M. Dean et al Nature Materials 15 601 (2016)

800 nm laser excitation

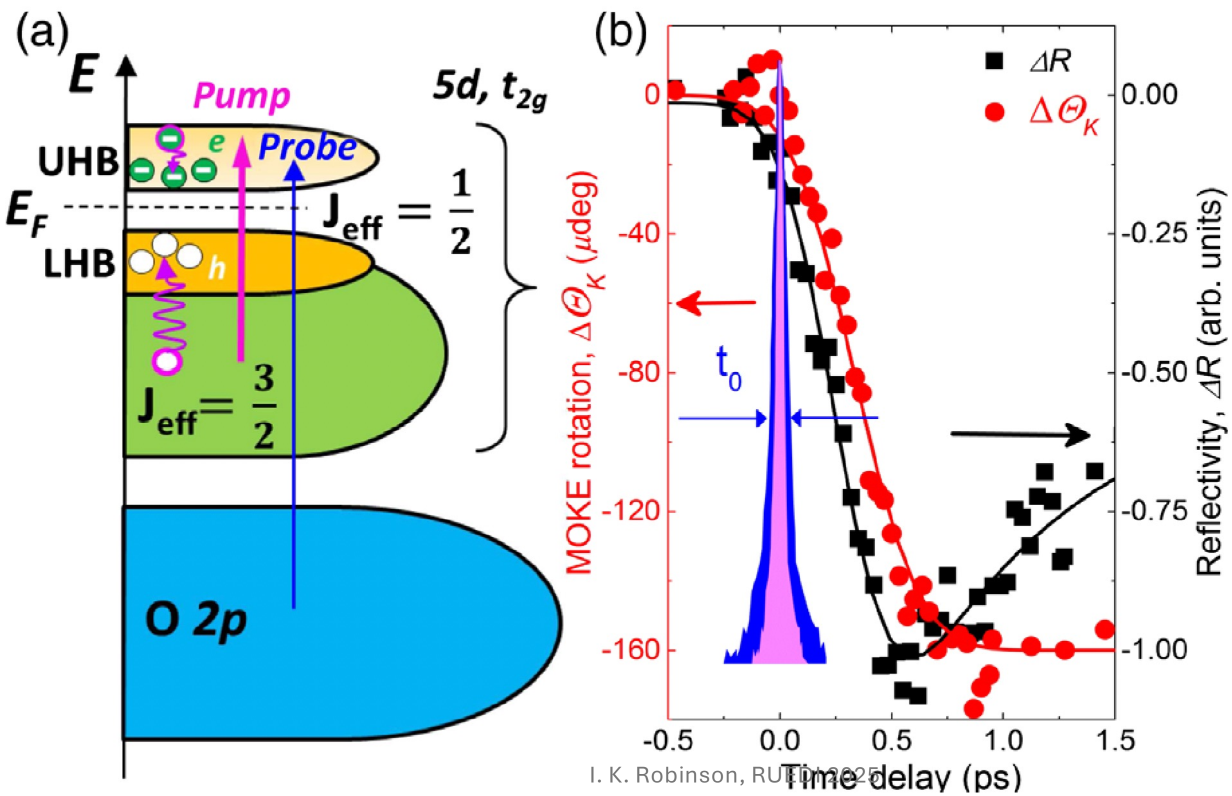


(−3 −2 28) magnetic Bragg peak (backscattering at 12.215 keV).

Laser reflectivity of AFM Demagnetisation by MOKE

Ultrafast Spin Dynamics in Photodoped Spin-Orbit Mott Insulator Sr_2IrO_4

D. Afanasiev et al Phys Rev X 9 021020 (2019)



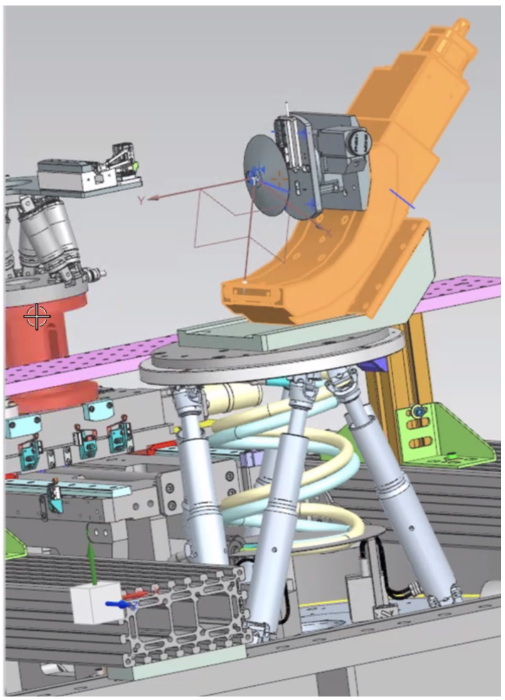
400 nm laser excitation

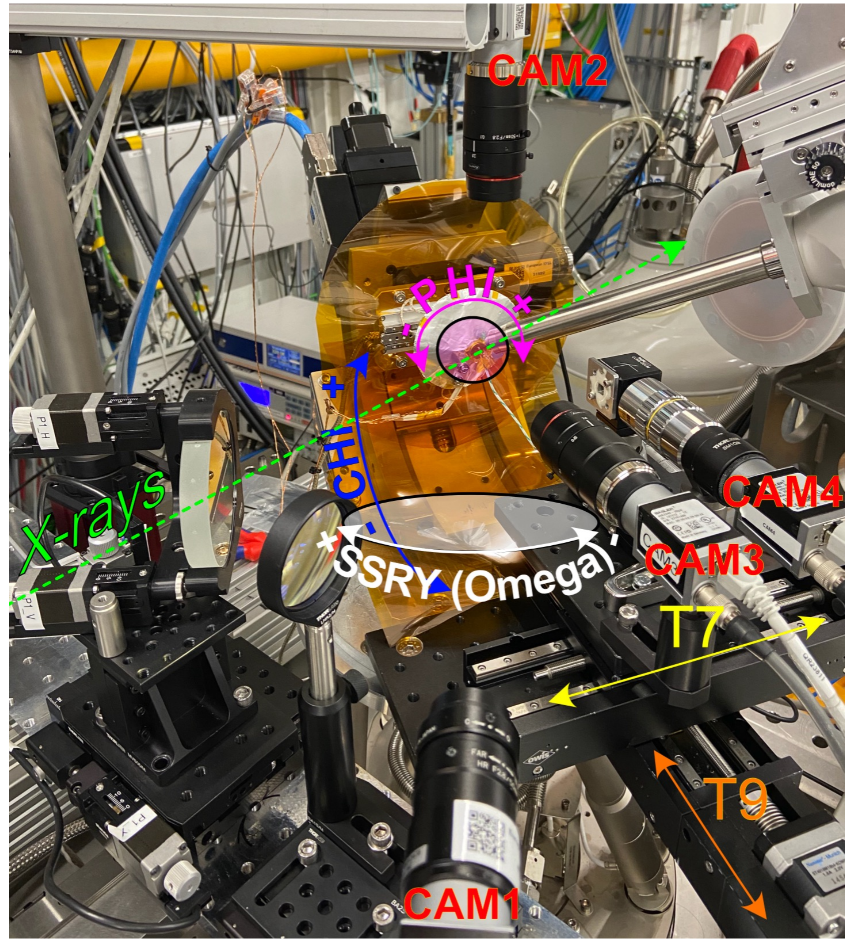
P3331 + P6156 experiments at MID of E-XFEL

- 2-day compensation beamtime (for cryo failure in April 2023)
- Challenging sample alignment
  - 5x5 μm crystal prealigned to 001 normal 100 parallel to edge
  - self seeding at exact resonant energy 11.215 keV
  - magnetic peaks only below 230K
  - (1 0 6) peak is tilted 51 degrees in 001/100 plane
  - spatial and temporal overlap of laser and 12 μm X-ray focus
- Pre-align on (2 0 12) charge peak at double the Bragg angle
- Planned to get 3D BCDI rocking curves vs PP delay and fluence

Set-up at MID E-XFEL April 2023 + April 2024

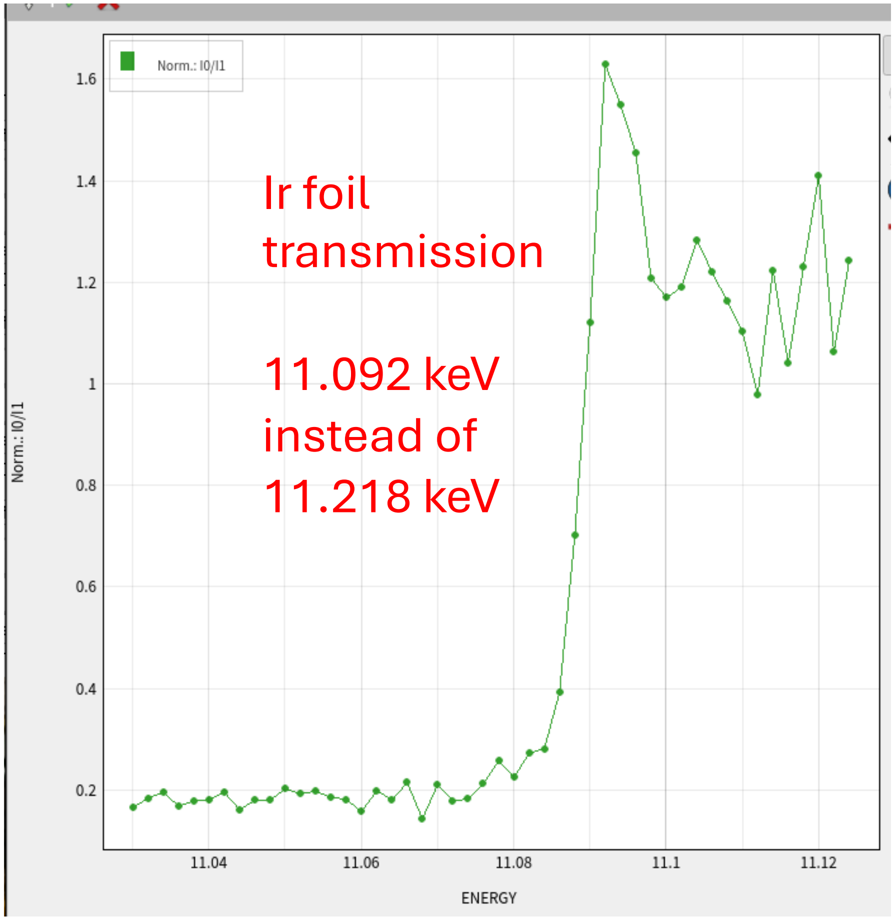
NAFO CRL focussing





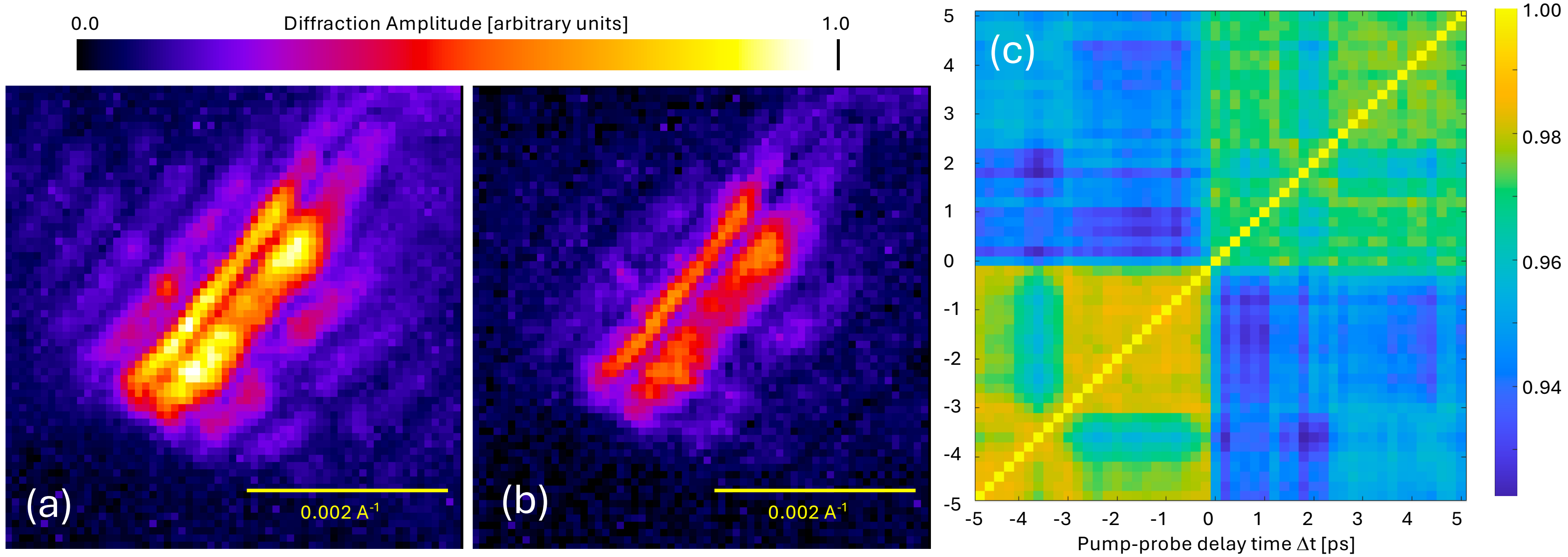
cryostream cooling

Single crystal sample of Sr_2IrO_4 at 100K



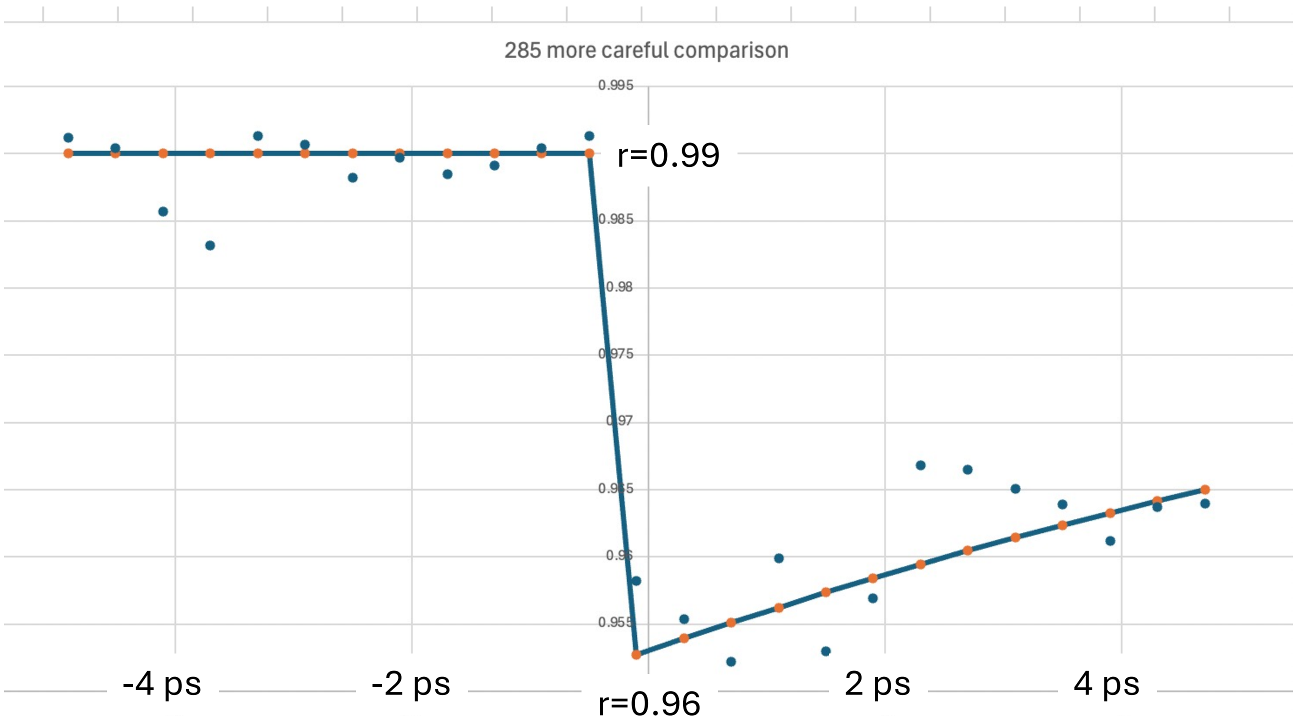
Coherent diffraction at 106 peak

T=100K E=11.215 keV



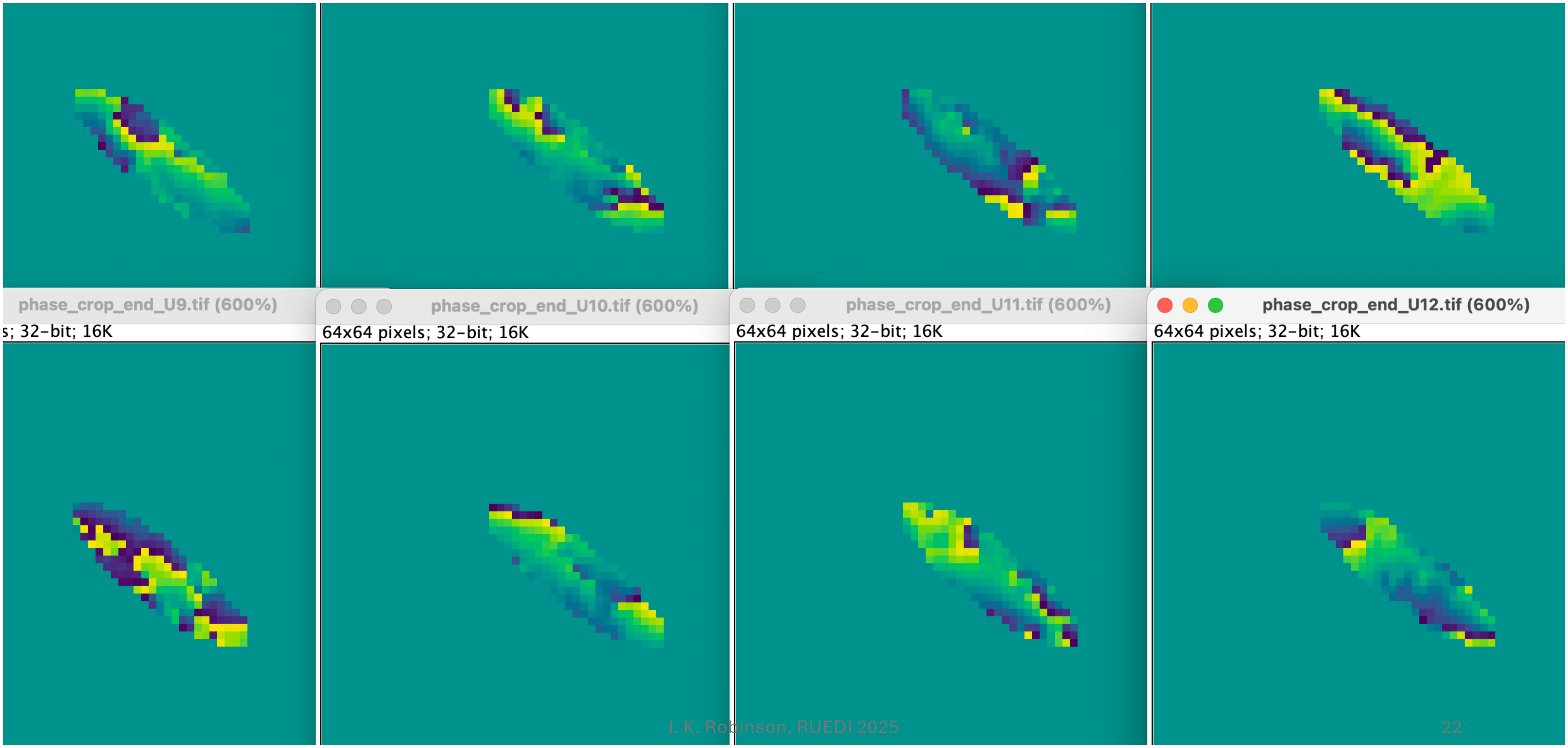
Run285 bin = 4 compare with P5 reconstruction

800 nm Laser at 45% = 15 mJ/cm^2

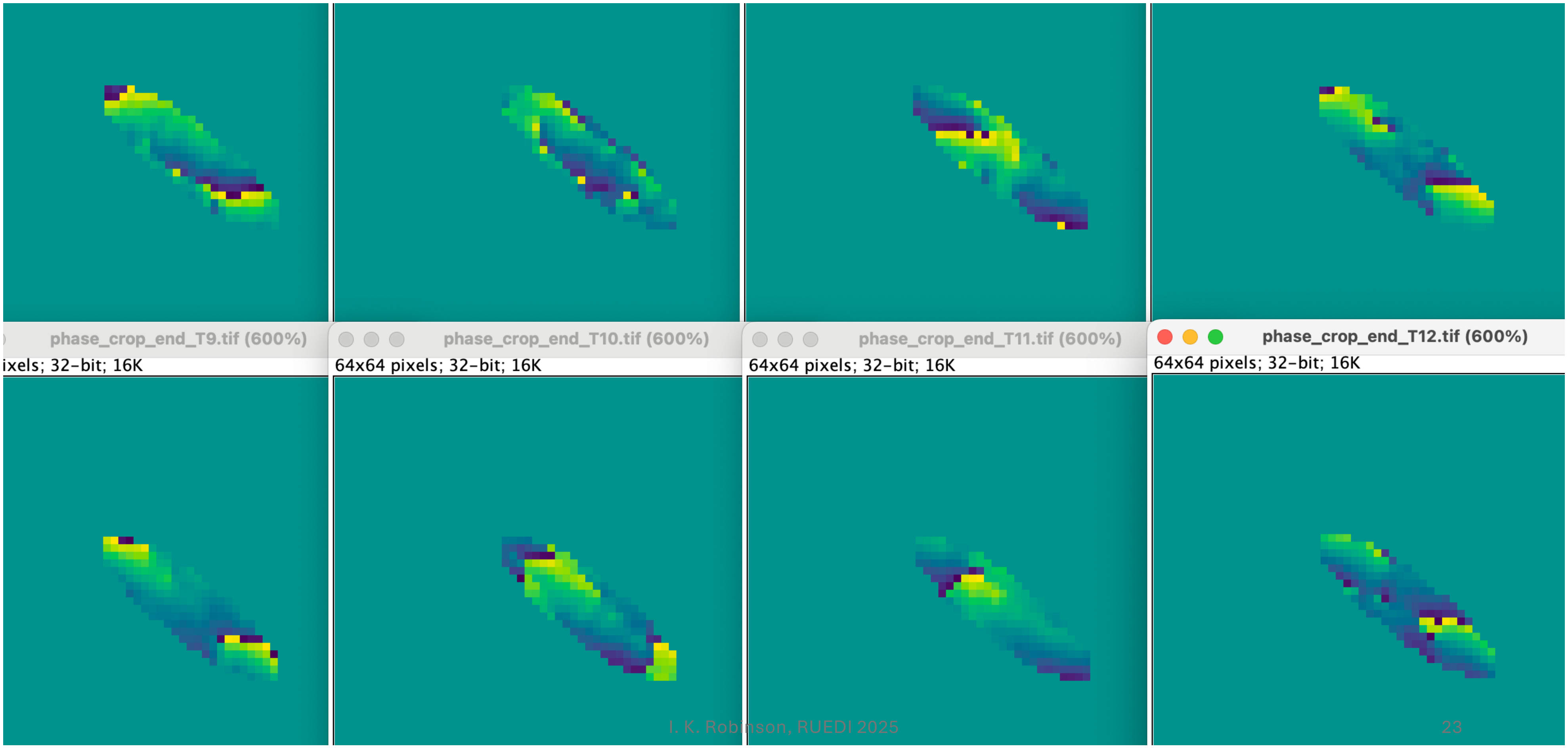


5.3	offset
0.037	amp
12 ps	decay
0.99	scale

500 cycles ER-HIO, fixed support, seeds=5-12

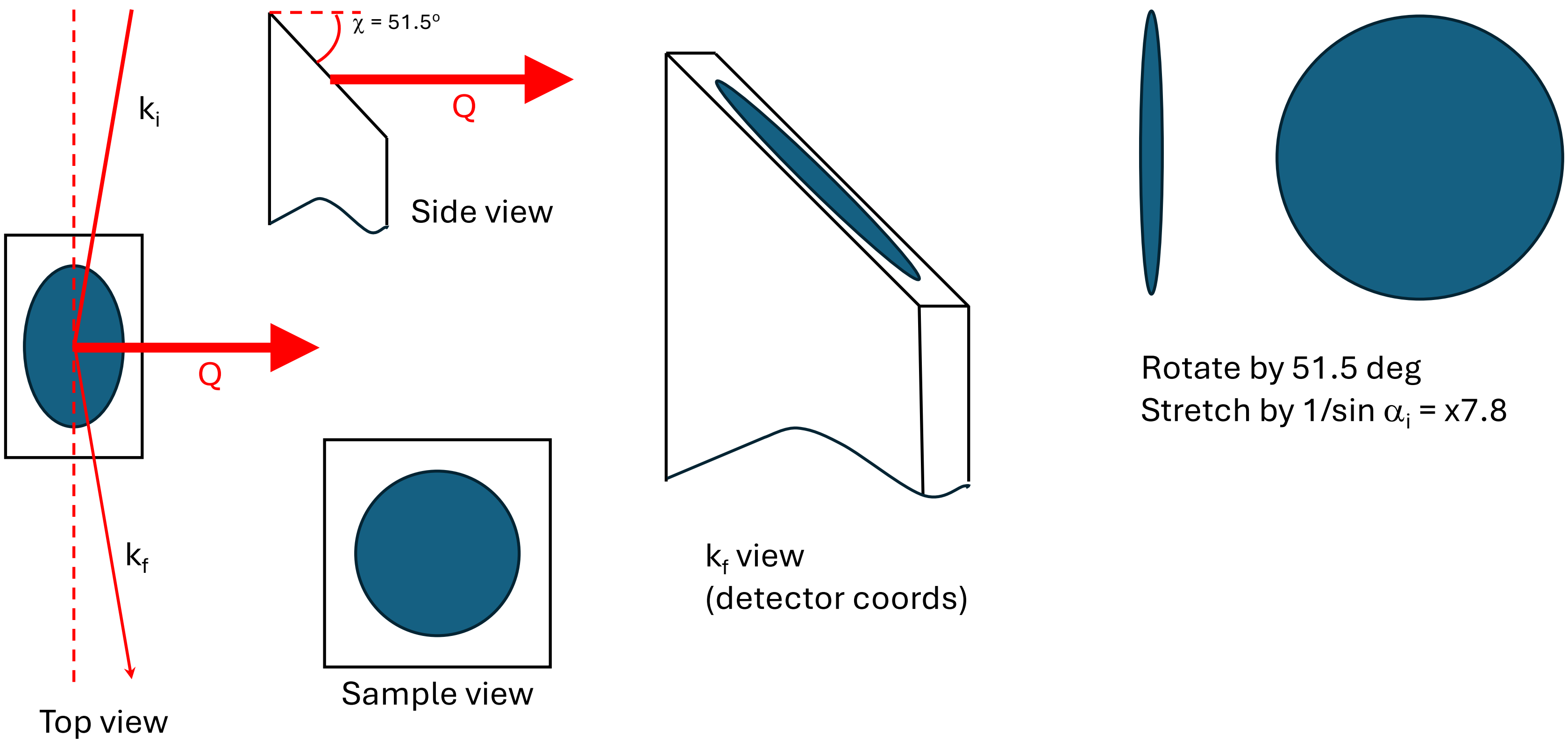


Guided HIO, 5 generations, seeds=5-12



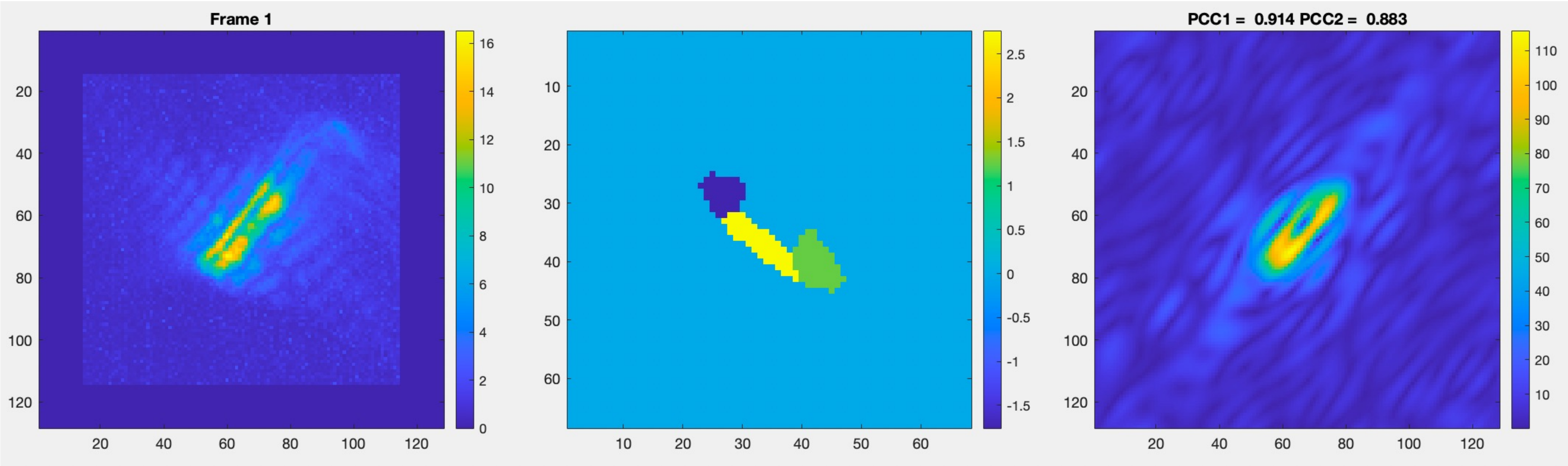
Domain sizes + Coordinate transformation

Ignoring penetration depth ~0.65 μm



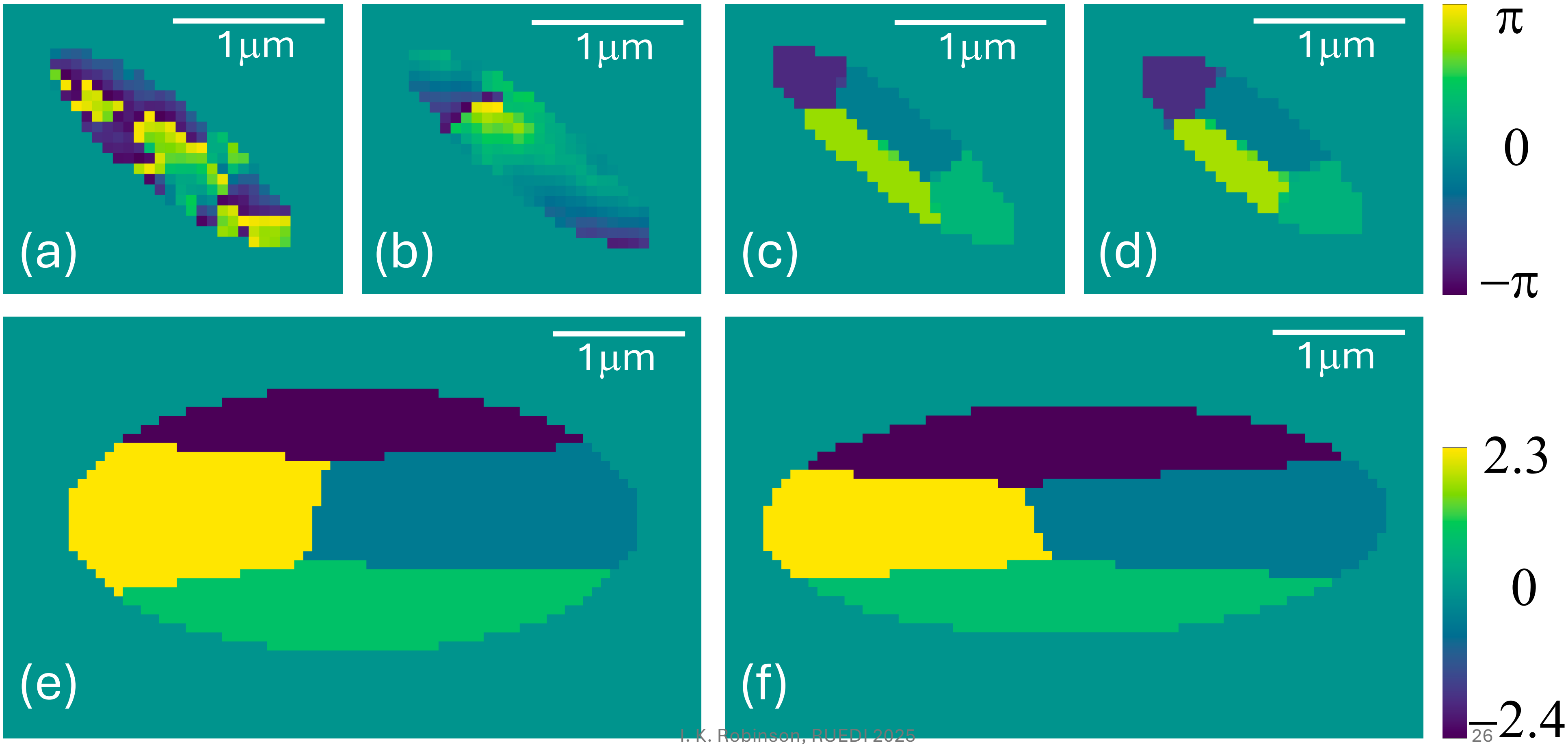
Rotate by 51.5 deg
Stretch by 1/sin α_i = x7.8

4-domain model motivated by reconstruction

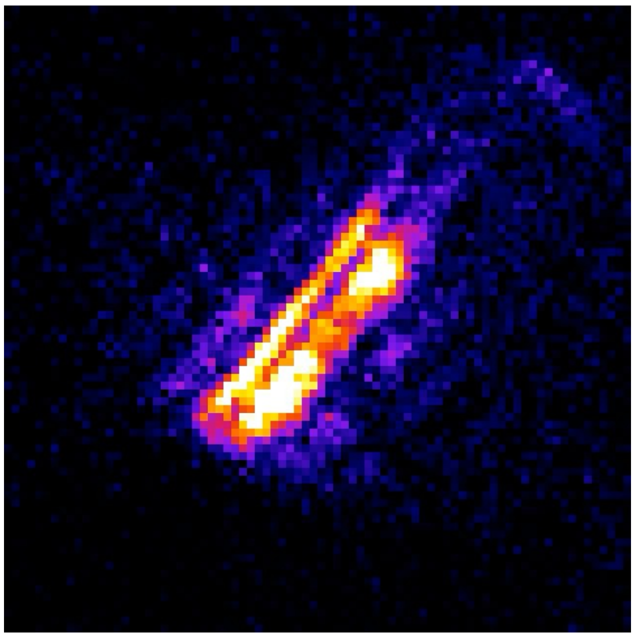


Evolution of fitted model with coord transformation

Time delay at -5 ps and +1 ps



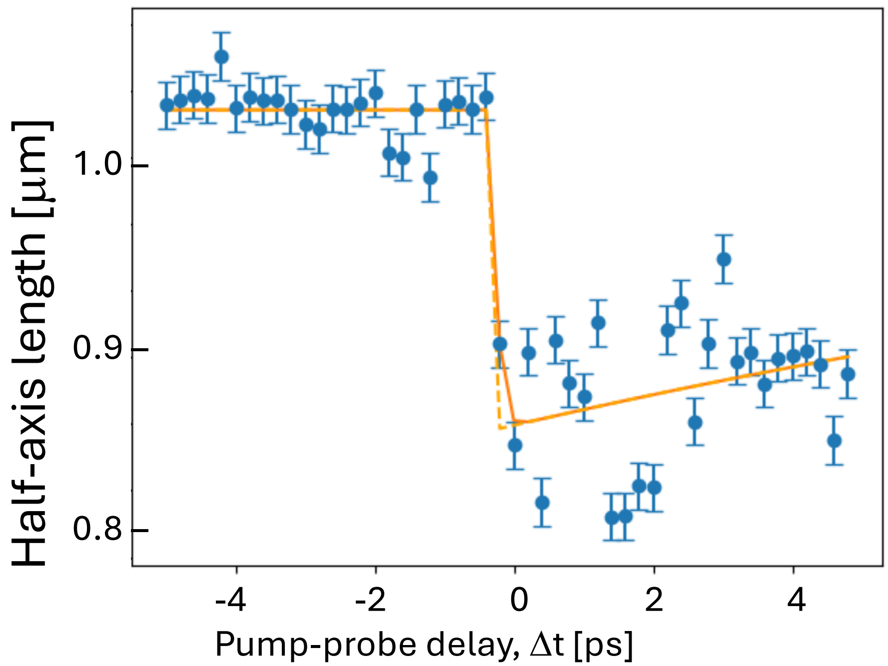
Movie of 4-domain model versus pump-probe delay -5 ps < t < 5 ps



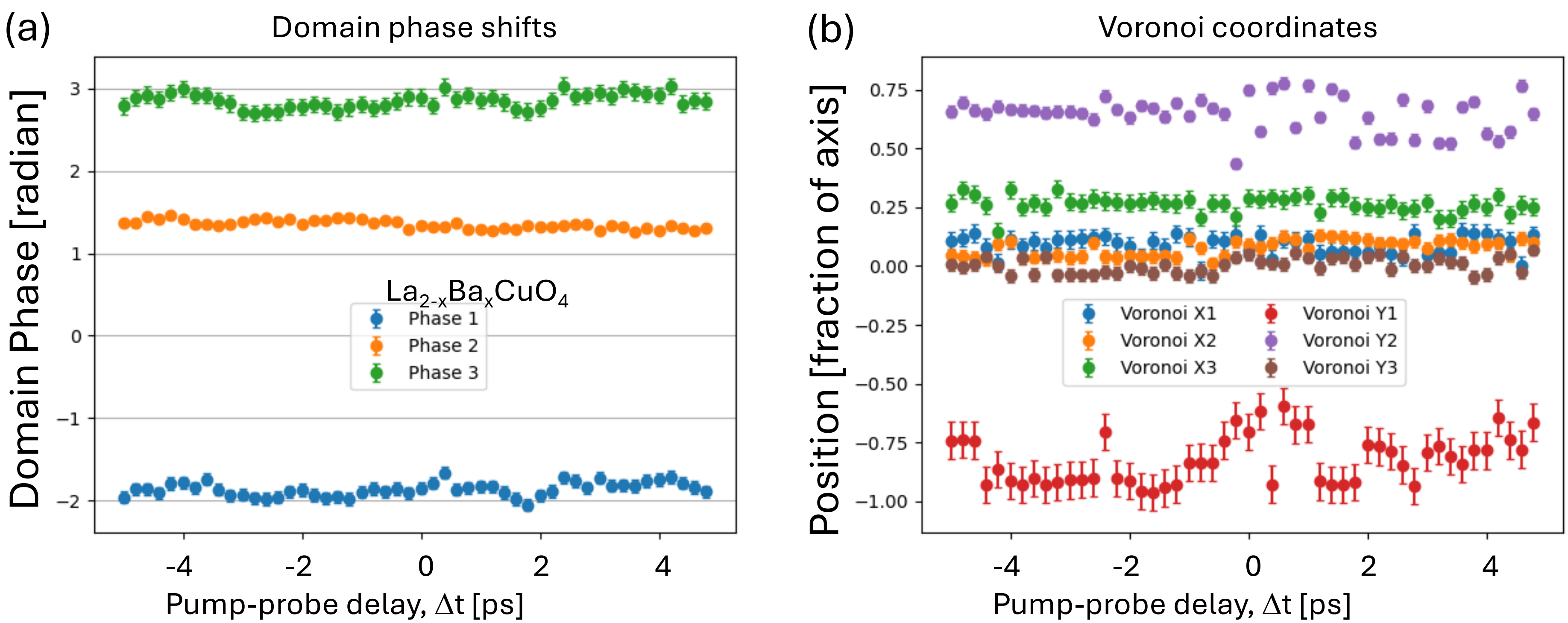
Fit of ellipse length with 200 fs time bins

40±30 fs drop followed by 20 ps rise

arXiv.2511.07359 (2025)



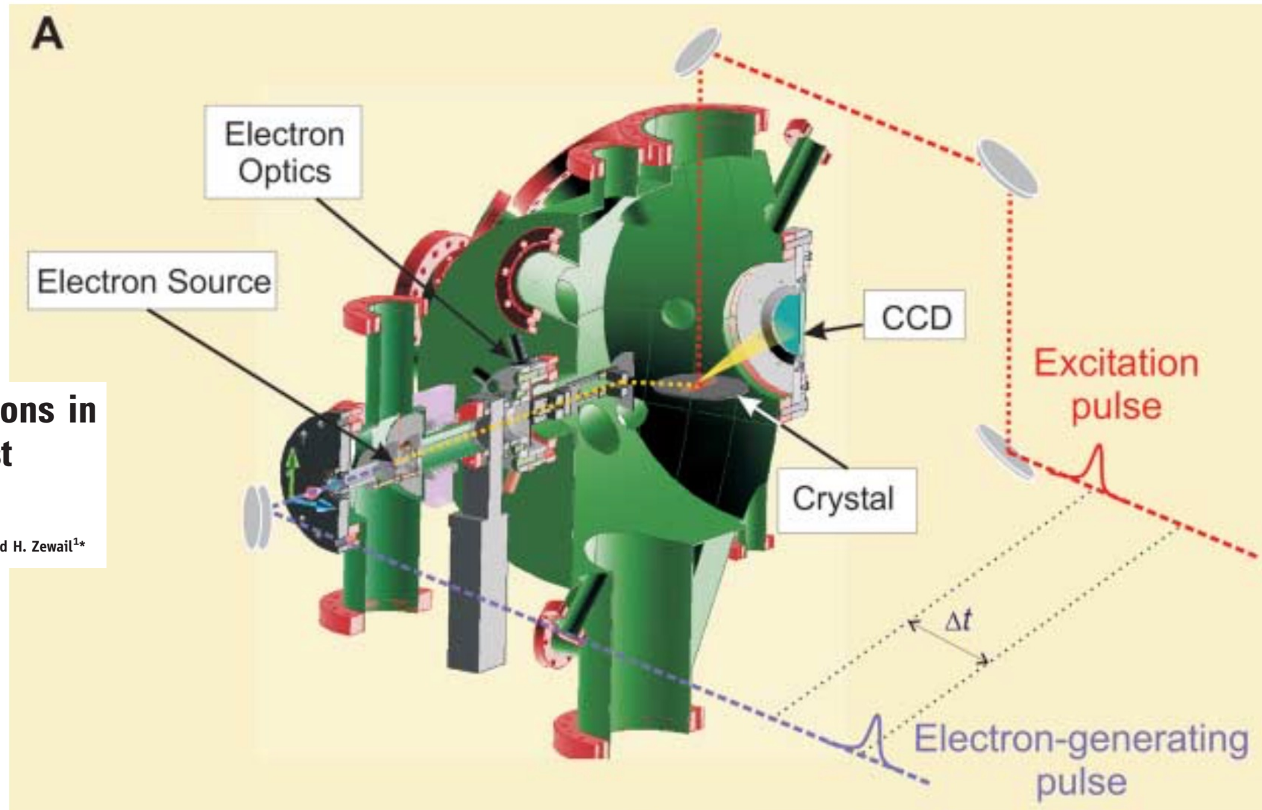
Fit of model params with 200 fs time bins

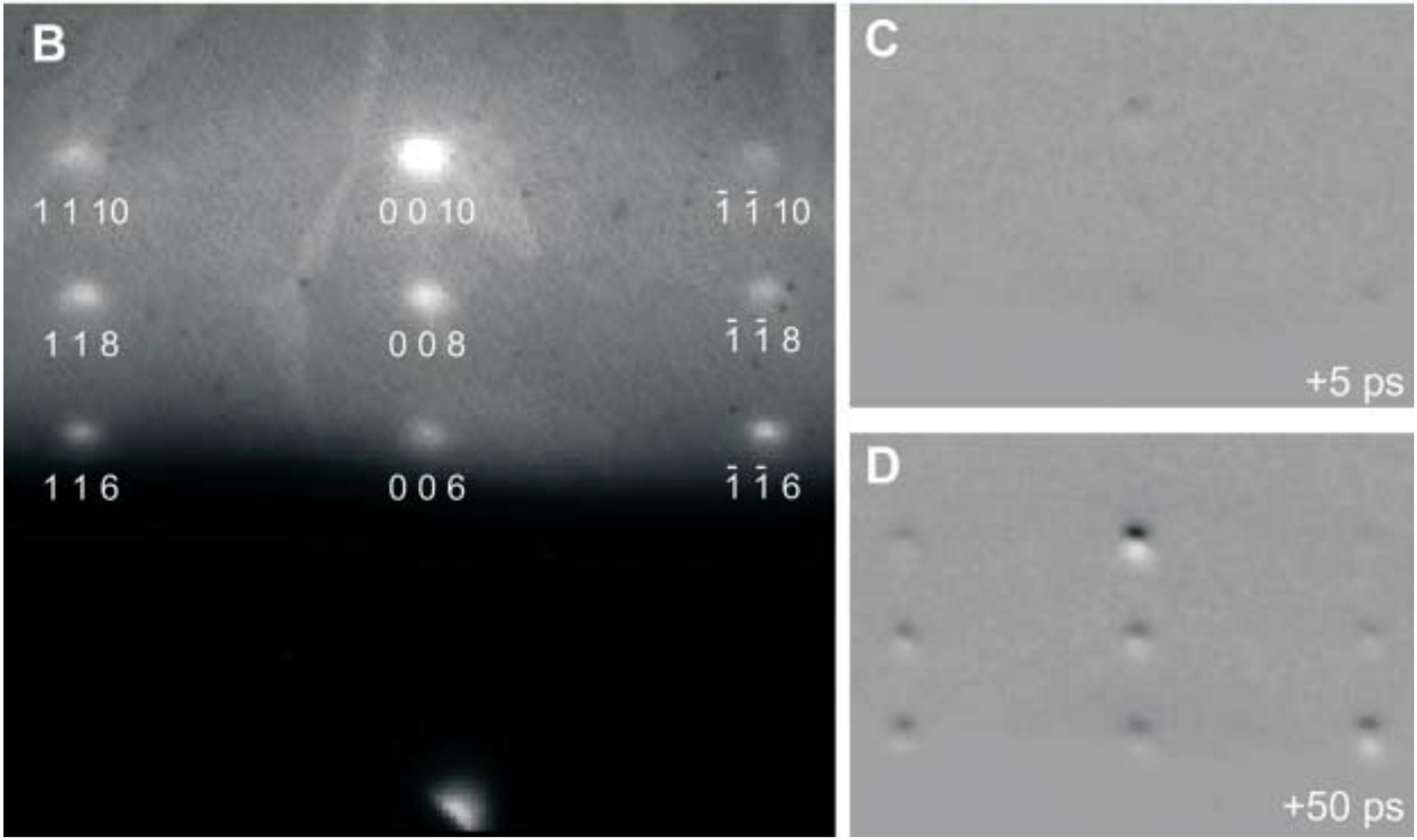


Outline

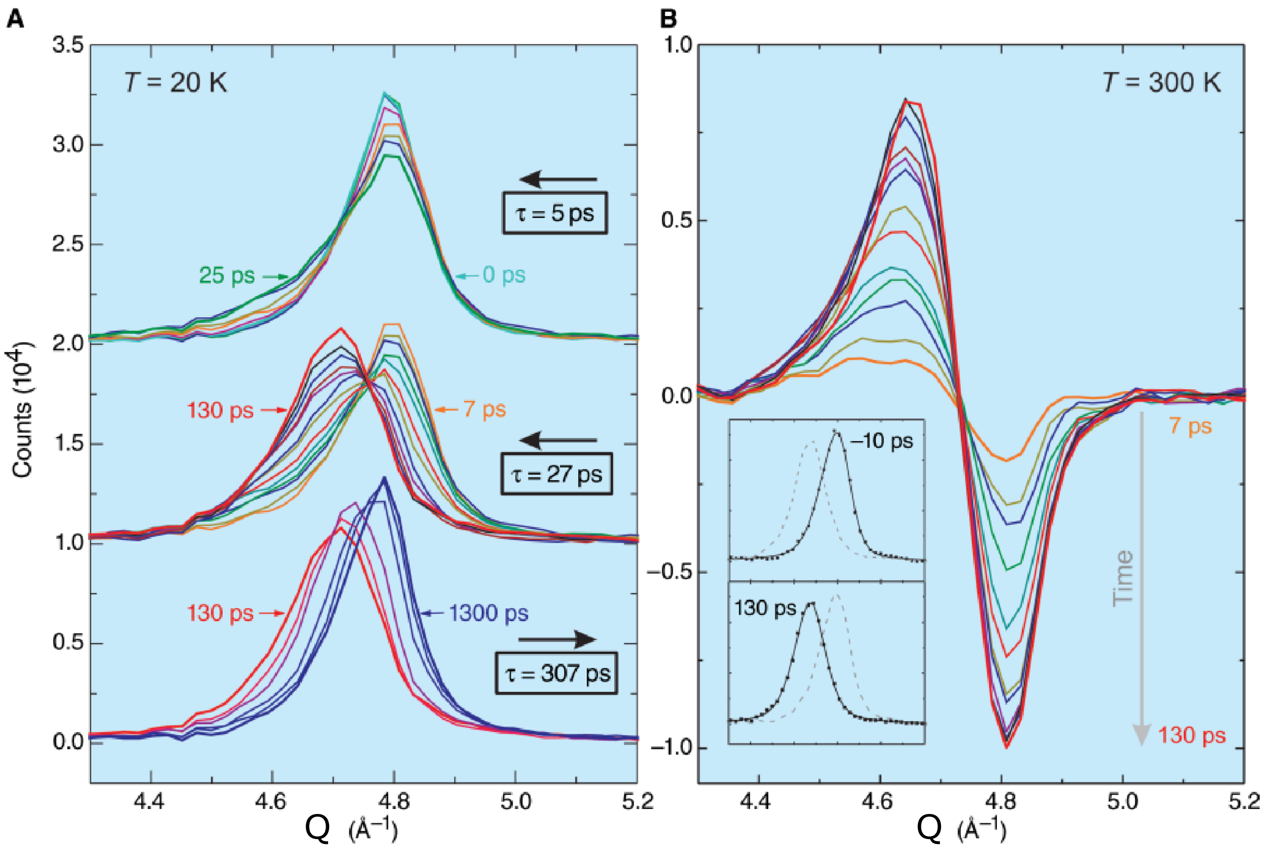
- Bragg Coherent Diffraction Imaging
- Sensitivity to strain
- Phase domain structures
- Antiferromagnetism of Sr_2IrO_4
- Pump-probe XFEL experiment
- Migration of magnetic domains
- Future RUEDI experiment

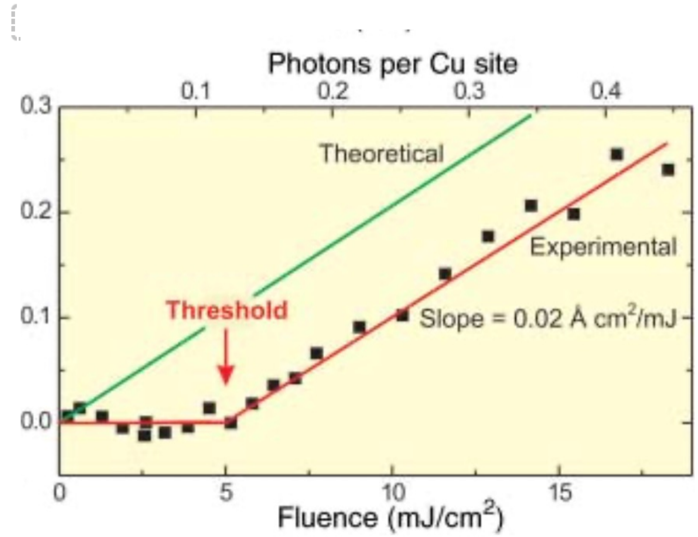
30 keV RHEED Science 316 425 (2007) $La_2CuO_{4-\delta}$




B
1 1 10
0 0 10
$\bar{1}$ $\bar{1}$ 10
1 1 8
0 0 8
$\bar{1}$ $\bar{1}$ 8
1 1 6
0 0 6
$\bar{1}$ $\bar{1}$ 6
C
+5 ps
D
+50 ps

Gedik-Bozovic-Zewail: UED from $La_2CuO_{4-\delta}$
N. Gedik et al Science 316 425 (2007)





2.5% lattice expansion
Fluence threshold 5mJ/cm^2
Electronic structure transition

Proposed UED from structured LBCO thin film

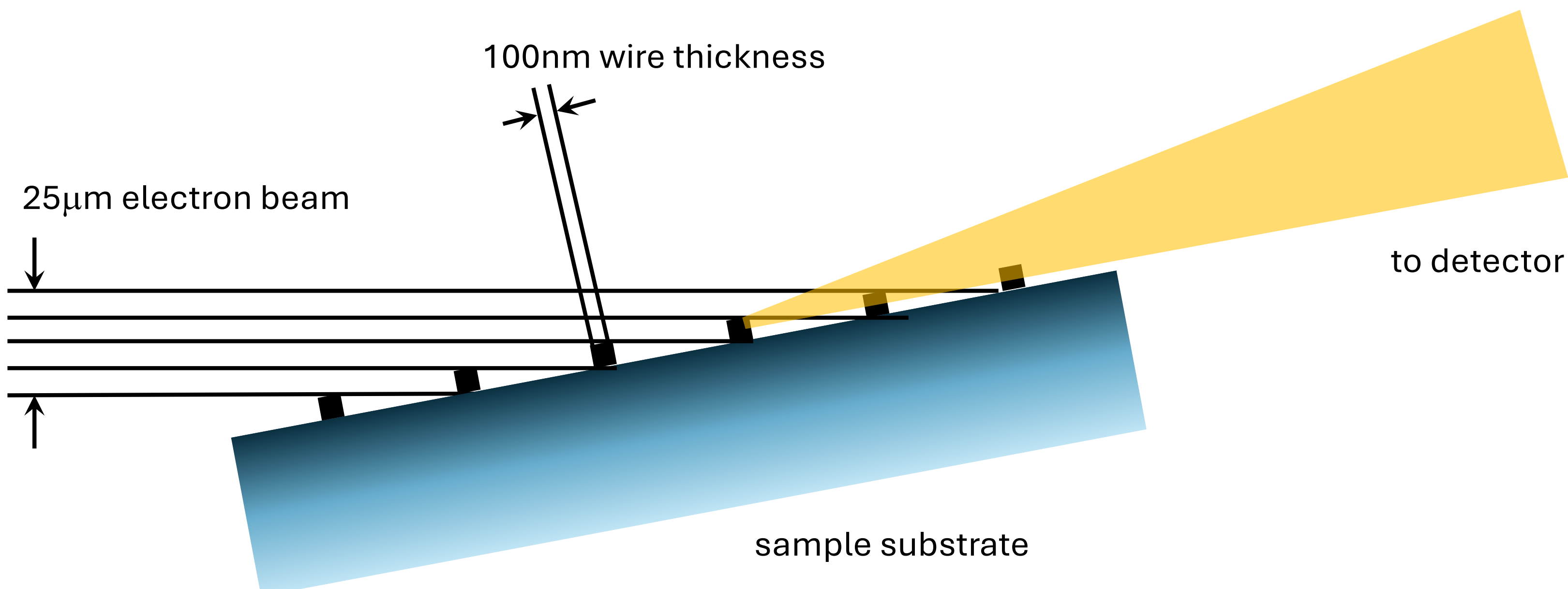
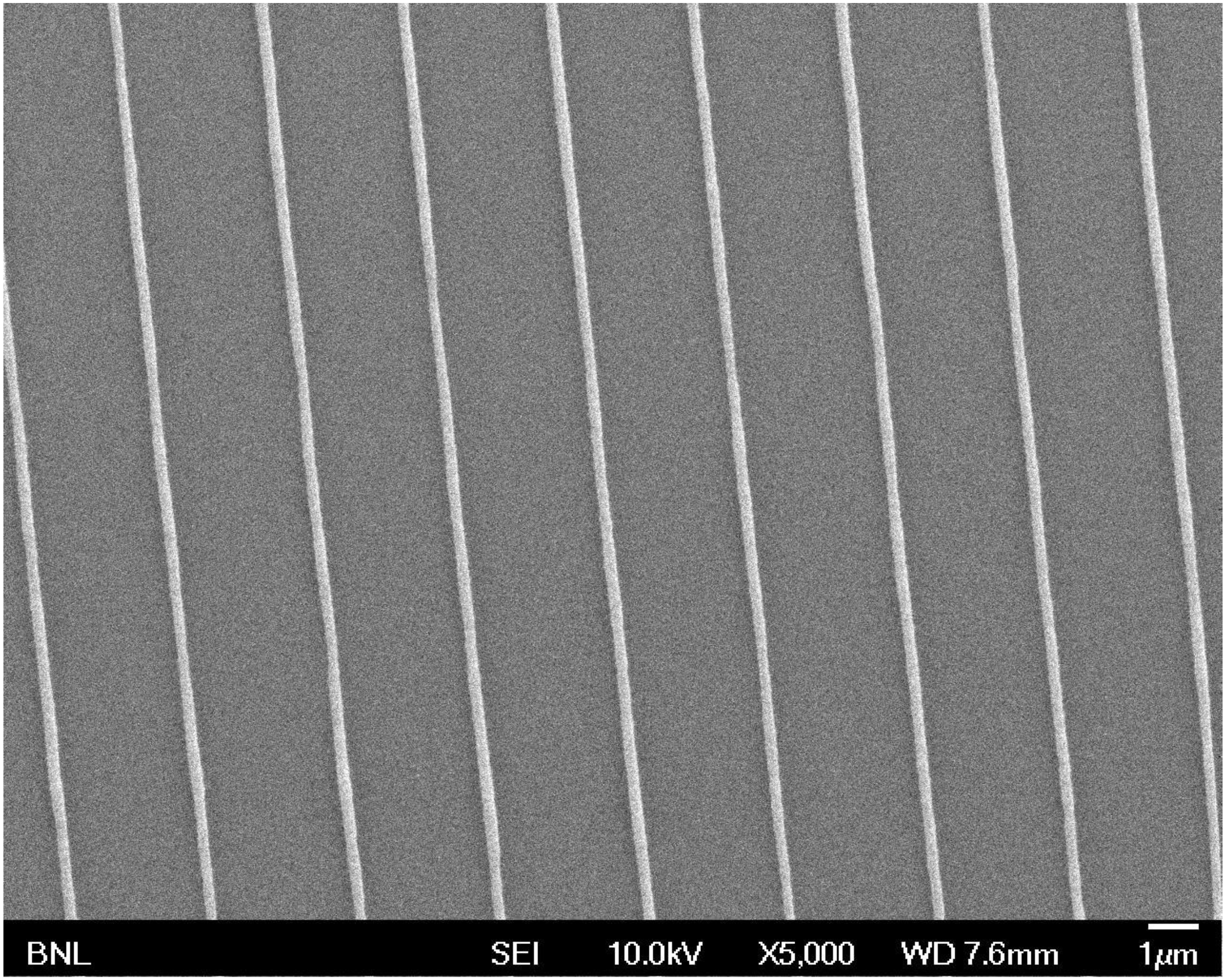


Figure 6. Sample geometry. The MeV-UED beam comes in at grazing angle, so that it penetrates just one of the 100nm thick lithographed wires before diffracting away from the sample to reach the detector.


BNL
SEI
10.0kV
X5,000
WD 7.6mm
1μm

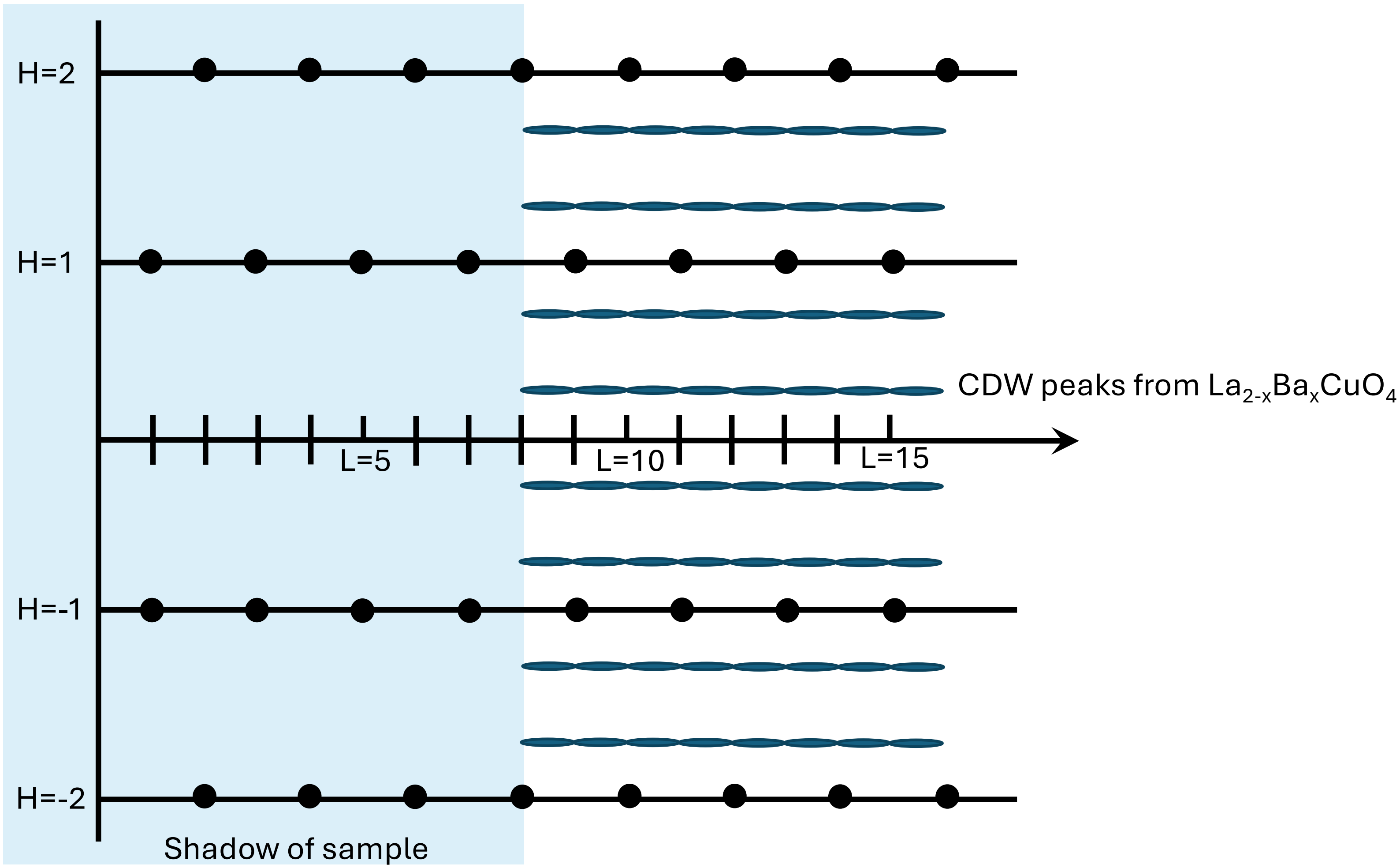


Figure 7. Expected UED diffraction pattern from LSCO. The low-L peaks are shadowed by the sample edge in the grazing geometry, but the high-L peaks of interest are accessible.

Magnetic Coherent X-ray Diffraction Imaging

- Bragg Coherent Diffraction Imaging at E-XFEL
- Domain images of and Sr_2IrO_4
- Laser driven migration of AFM domains in Sr_2IrO_4
- $4x10^6$ m/s velocity cannot involve phonons
- Spins migrate at $\sim V_{Fermi}$
- Proposed RUEDI experiment on $La_{2-x}Ba_xCuO_4$

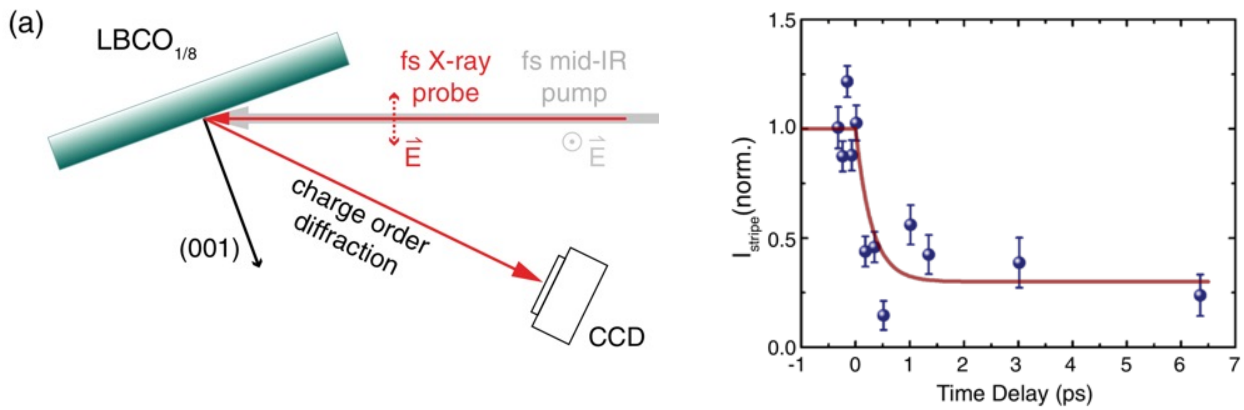
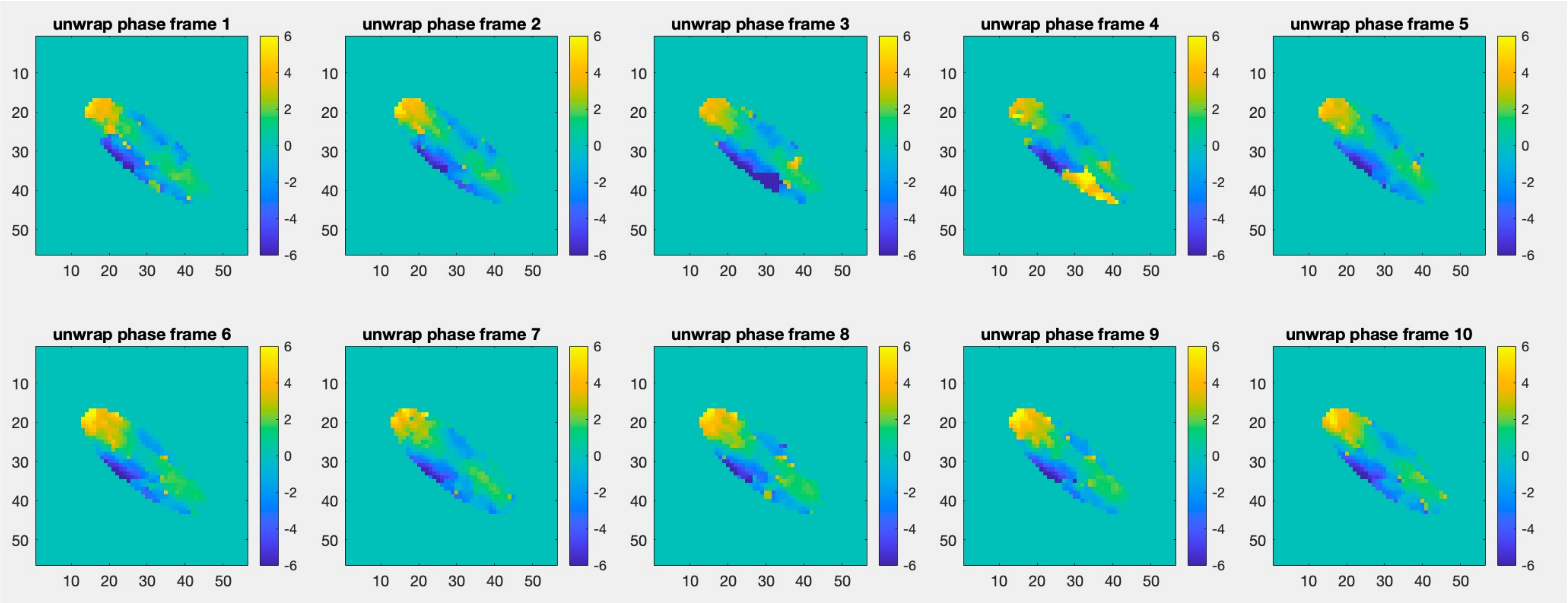


Fig 2. (a) Diffraction condition of the charge stripe order peak at (0.24 0 0.5) for LBCO (001). (b) Pump-probe response measured at SXR, LCLS. The red solid line is an exponential with 300 fs time constant, which was the resolution of the experiment [5].

M. Först, R. I. Tobey, H. Bromberger, S. B. Wilkins, V. Khanna, A. D. Caviglia, Y.-D. Chuang, W. S. Lee, W. F. Schlotter, J. J. Turner, M. P. Minitti, O. Krupin, Z. J. Xu, J. S. Wen, G. D. Gu, S. S. Dhesi, A. Cavalleri and J. P. Hill, "Melting of Charge Stripes in Vibrationally Driven $La_{1.875}Ba_{0.125}CuO_4$: Assessing the Respective Roles of Electronic and Lattice Order in Frustrated Superconductors" Phys. Rev. Lett. **112** 157002 (2014)

Phase transfer from 2-dom model + BCDI phasing

bin data into 10 bins of 1 ps   Unwrapped phases



Fit of ellispe length (px) to pump-probe delay time

12 fs drop followed by 11 ps rise or 1.1 ps oscillation period?

drop is 0.39 μm in 12 fs, moving at 3.2×10^7 m/s