

Probing the finite-momentum spectrum of light-induced superconductors

Matteo Mitrano

University of Illinois at Urbana-Champaign

Harvard University

Nonequilibrium Superconductivity

CCQ, Flatiron Institute, NYC Jan 16th 2020

People

Experiments

I ILLINOIS

A. Husain, S. Lee, G. de la Pena, X. Sun, P. Abbamonte

SLAC NATIONAL
ACCELERATOR
LABORATORY

A. H. Reid, S. F. Wandel, W. Schlotter, G. Coslovich, T. van Driel

NIST
National Institute of
Standards and Technology
U.S. Department of Commerce

Y. I. Joe

Theory

I ILLINOIS

M. Zhu, N. Goldenfeld



L.V. Delacretaz, S. Hartnoll

Samples

BROOKHAVEN
NATIONAL LABORATORY

G. D. Gu, J. Schneeloch

GORDON AND BETTY
MOORE
FOUNDATION



U.S. DEPARTMENT OF
ENERGY

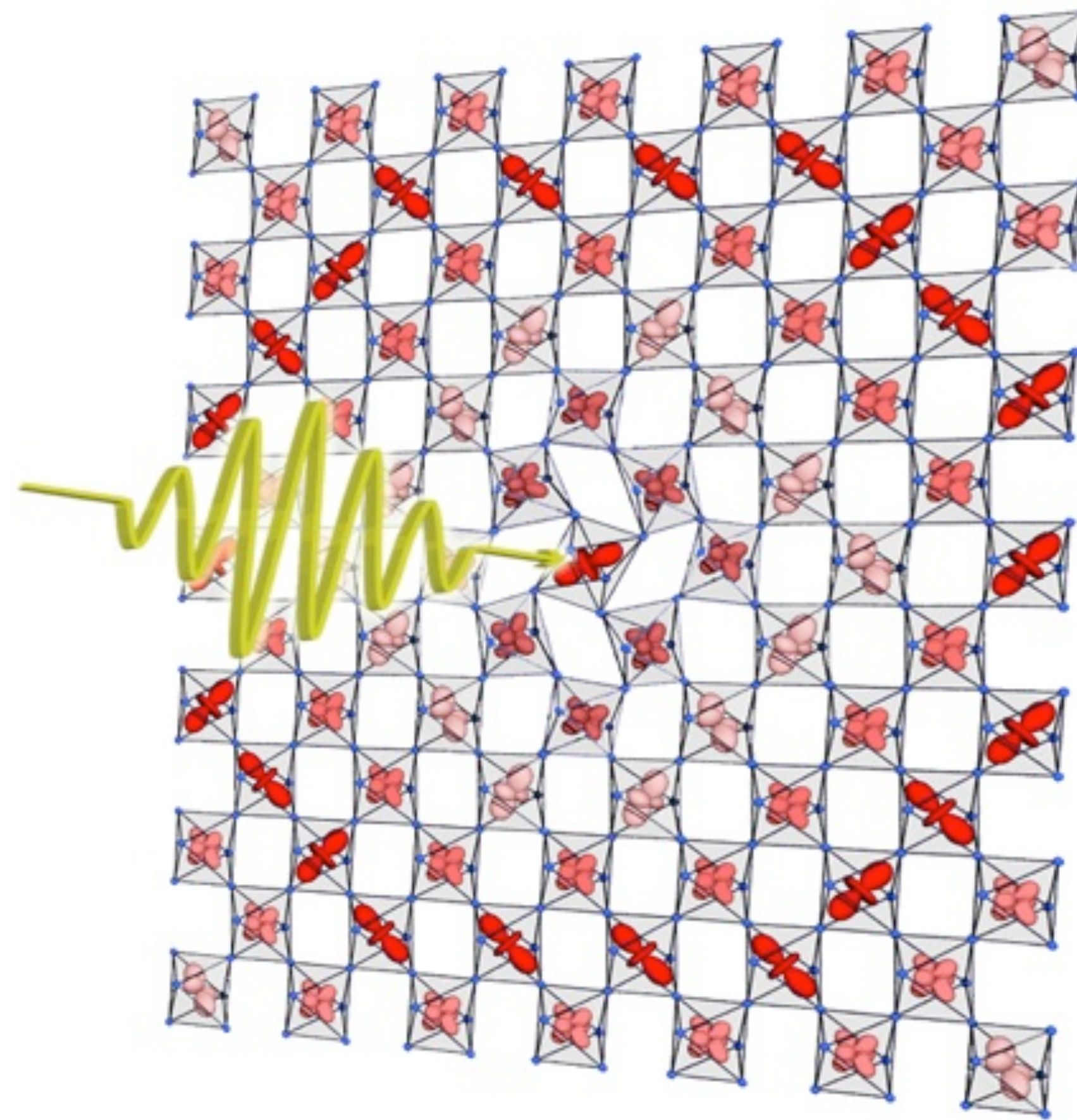
Office of
Science

Unterstützt von / Supported by



Alexander von Humboldt
Stiftung/Foundation

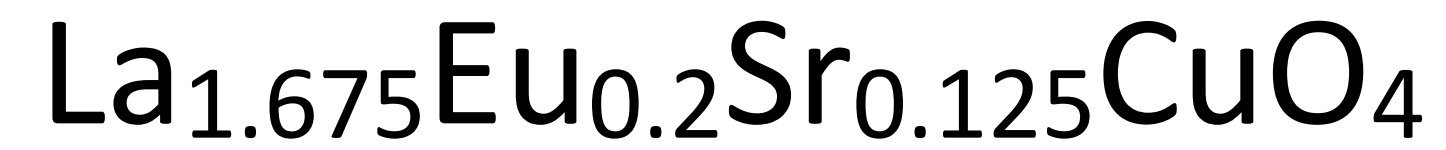
Light control of the lattice



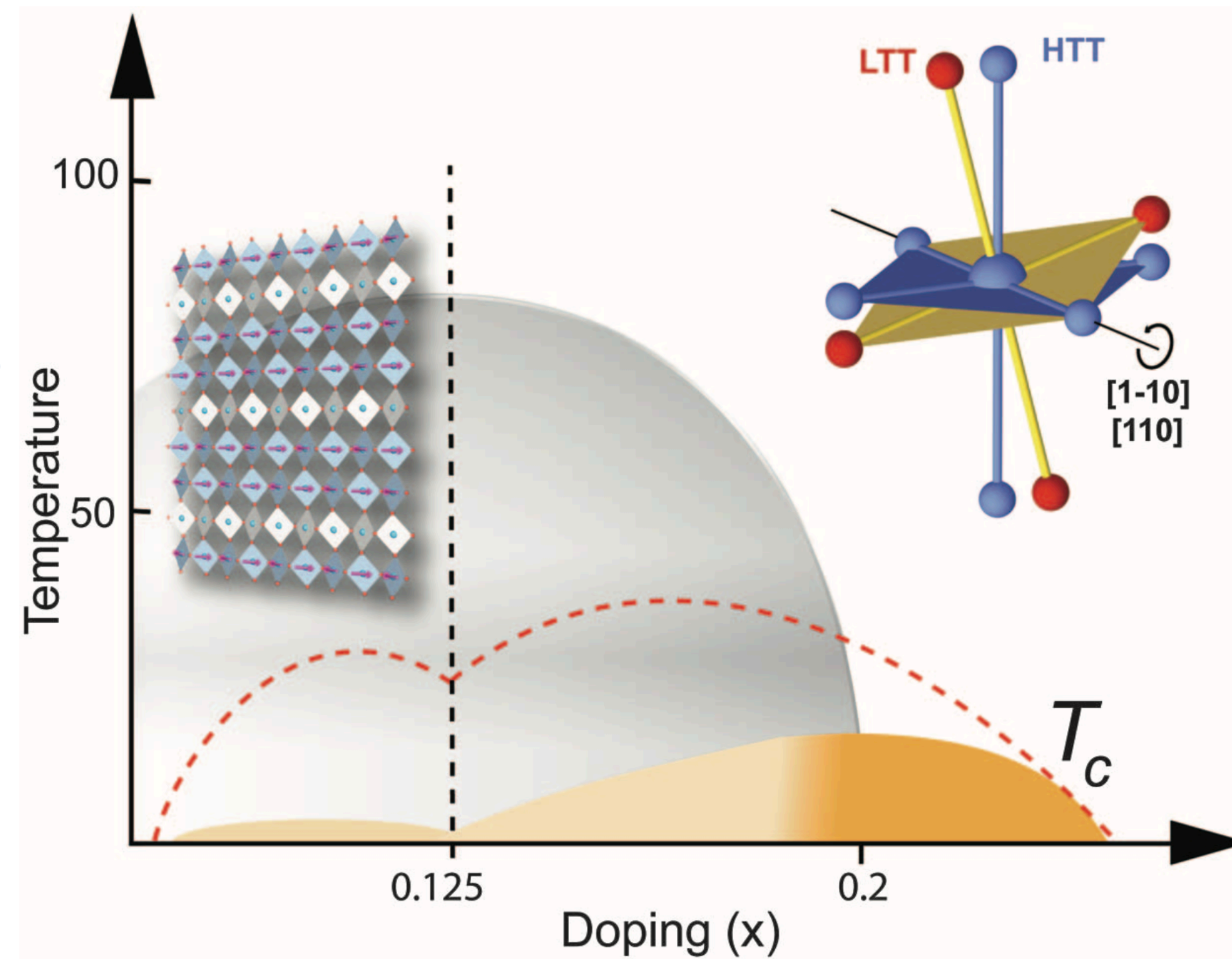
Andrea's group

Driving large amplitude displacements with laser pulses

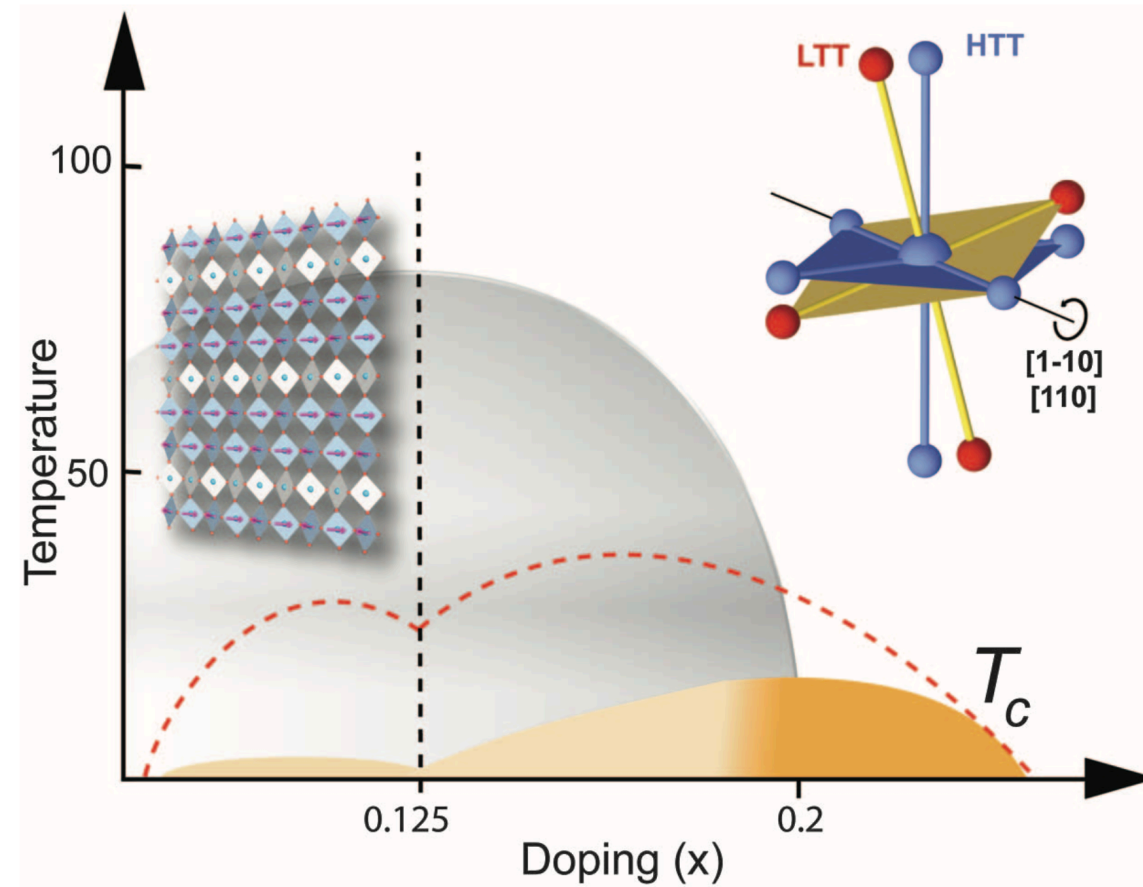
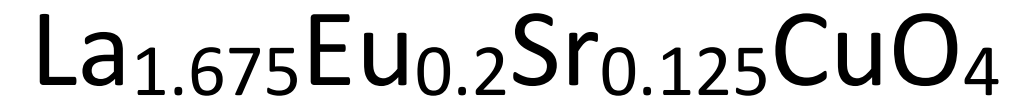
Light-induced superconductivity



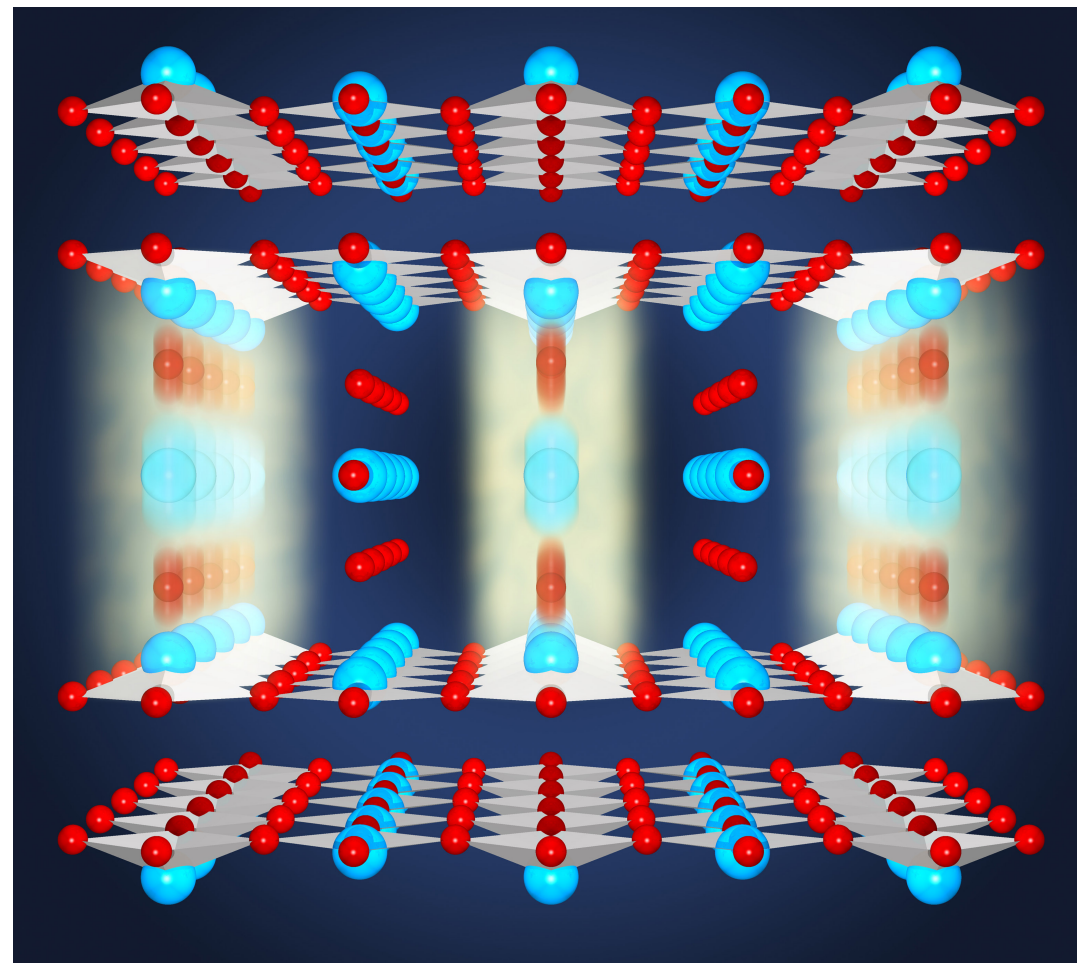
D. Fausti et al. Science **331**, 189 (2011)



Light-induced superconductivity

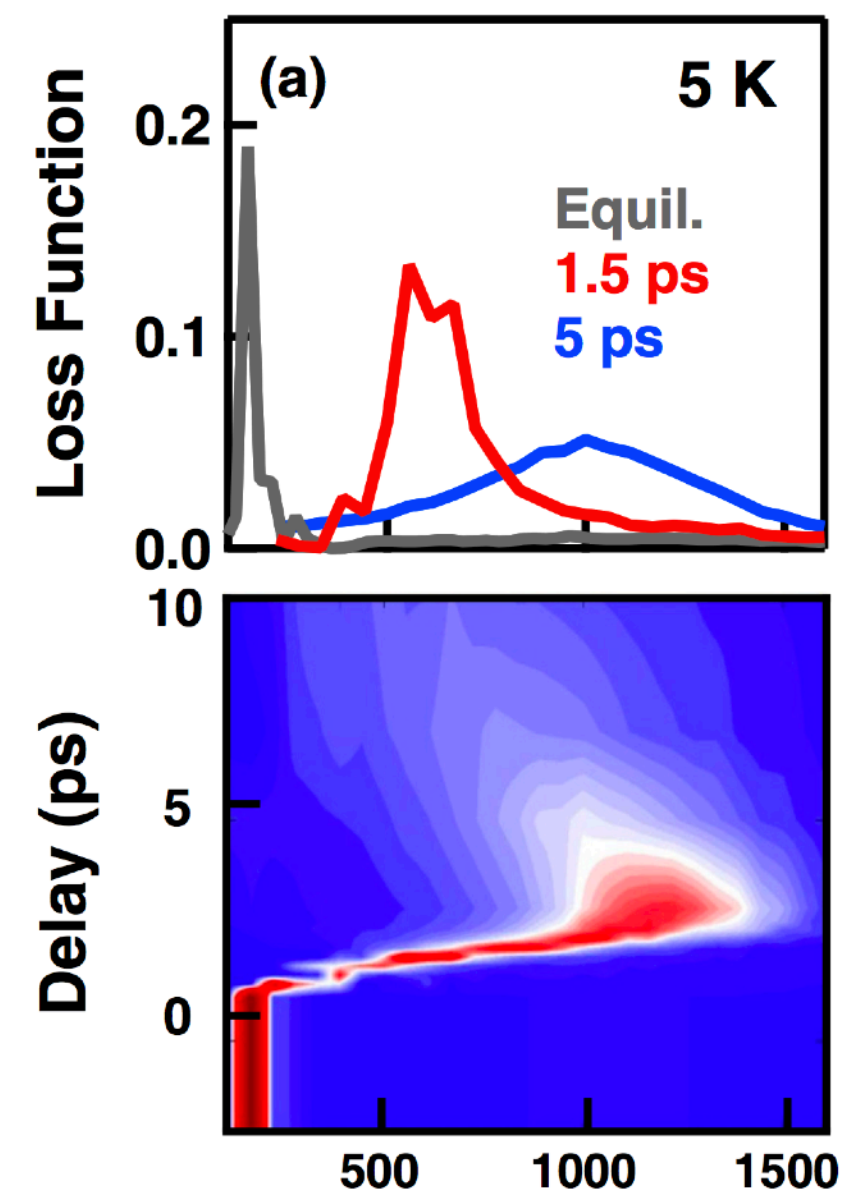


D. Fausti et al. *Science* **331**, 189 (2011)



W. Hu et al. *Nature Materials*, **13**, 705–711 (2014)

S. Kaiser et al. *Phys. Rev. B* **89**, 184516 (2014)

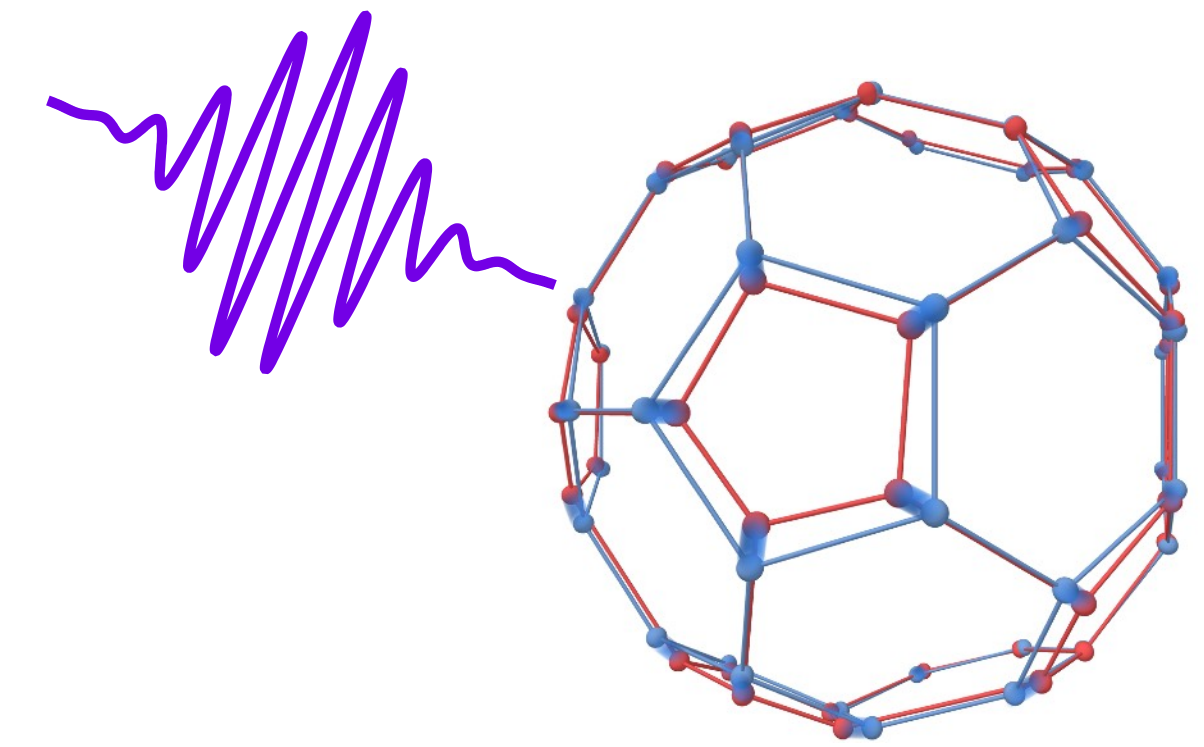
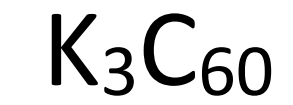


D. Nicoletti et al. *Phys. Rev. B* **90**, 100503(R) (2014)

D. Nicoletti et al. *Phys. Rev. Lett.* **121**, 267003 (2018)

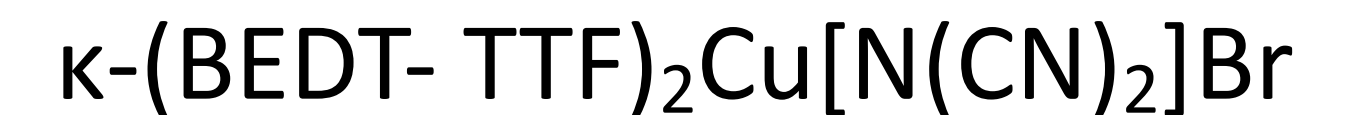
M. Först et al. *Phys. Rev. Lett.* **112**, 157002 (2014)

K. A. Cremin et al. *PNAS* **116**, 19875 (2019)

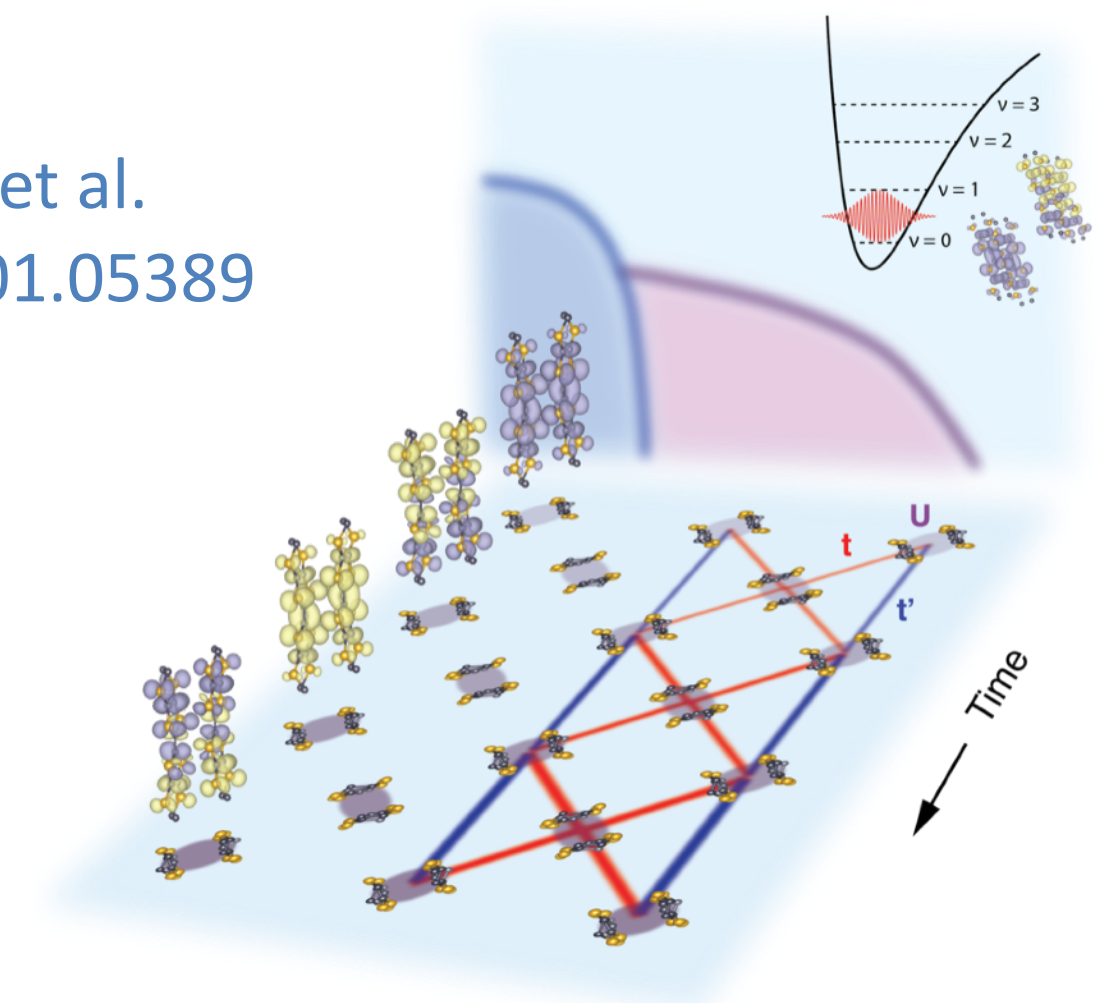


M. Mitrano et al. *Nature*, **530**, 461–464 (2016)

A. Cantaluppi et al. *Nature Physics*, **14**, 837–841 (2018)

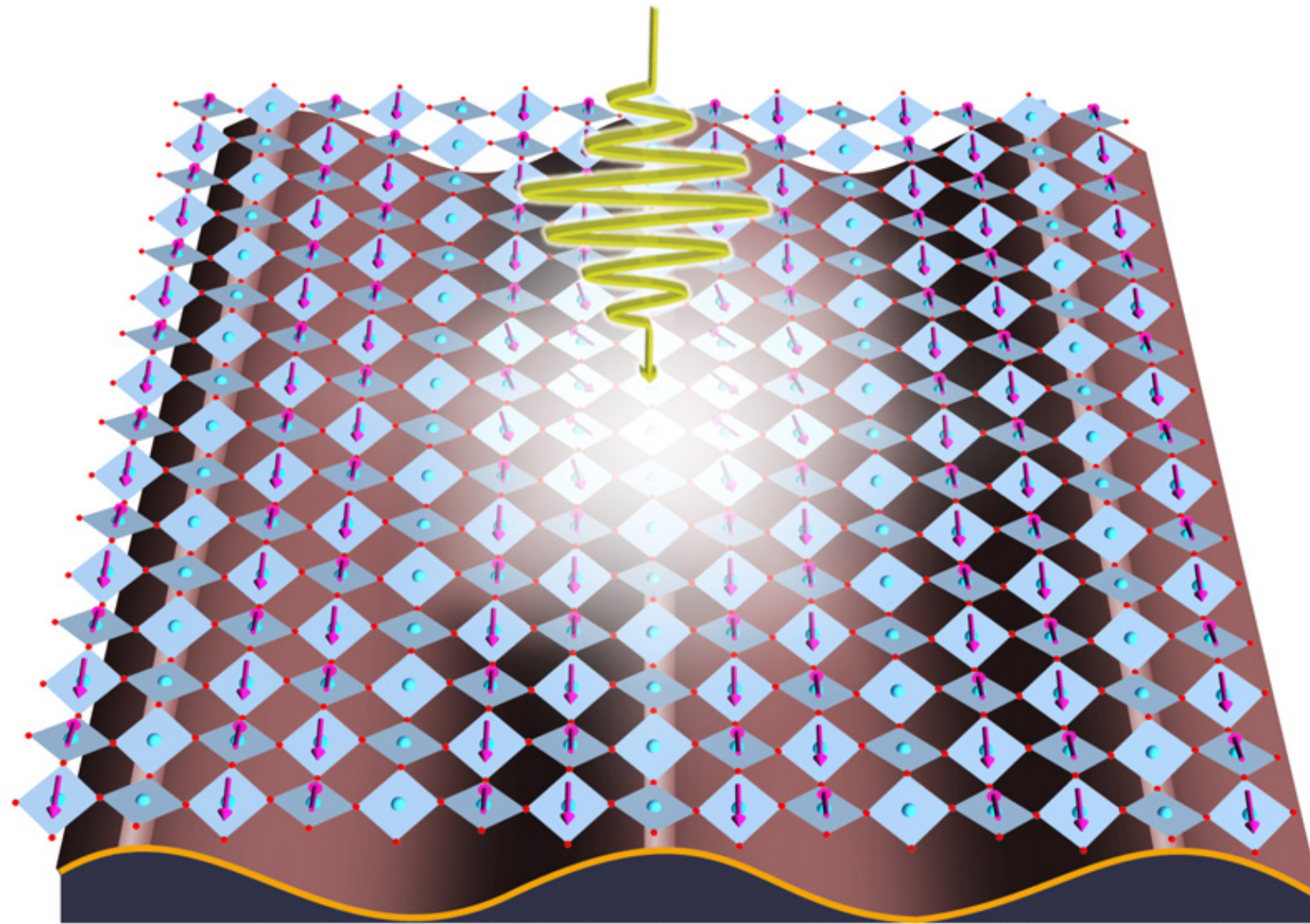


M. Buzzi et al.
arXiv:2001.05389



Is there a unique underlying mechanism?

Possible mechanisms



Z. M. Raines et al. *Phys. Rev. B* **91**, 184506 (2015)

R. Höppner et al. *Phys. Rev. B* **91**, 104507 (2015)

M. Sentef et al. *Phys. Rev. B* **93**, 144506 (2016)

M. Kim et al. *Phys. Rev. B* **94**, 155152 (2016)

M. Knap et al. *Phys. Rev. B* **94**, 214504 (2016)

A. Komnik et al. *Euro. Phys. Journal B* **89**, 244 (2016)

J. Okamoto et al. *Phys. Rev. Lett.* **117**, 227001 (2016)

A. Patel & A. Eberlein *Phys. Rev. B* **93**, 195139 (2016)

Y. Murakami et al. *Phys. Rev. B* **96**, 045125 (2017)

M. Babadi et al. *Phys. Rev. B* **96**, 014512 (2017)

M. Sentef et al. *Phys. Rev. Lett.* **118**, 087002 (2017)

G. Mazza & A. Georges *Phys. Rev. B* **96**, 064515 (2017)

D. M. Kennes et al. *Nature Physics* **13**, 479–483 (2017)

J. R. Coulthard et al. *Phys. Rev. B* **96**, 085104 (2017)

P. Werner et al. *Phys. Rev. B* **95**, 195405 (2018)

A. Nava et al. *Nature Physics* **14**, 154–159 (2018)

G. Chiriac et al. *Phys. Rev. B* **98**, 220510 (2018)

P. Werner et al. *Nature Comms.* **10**, 5556 (2019)

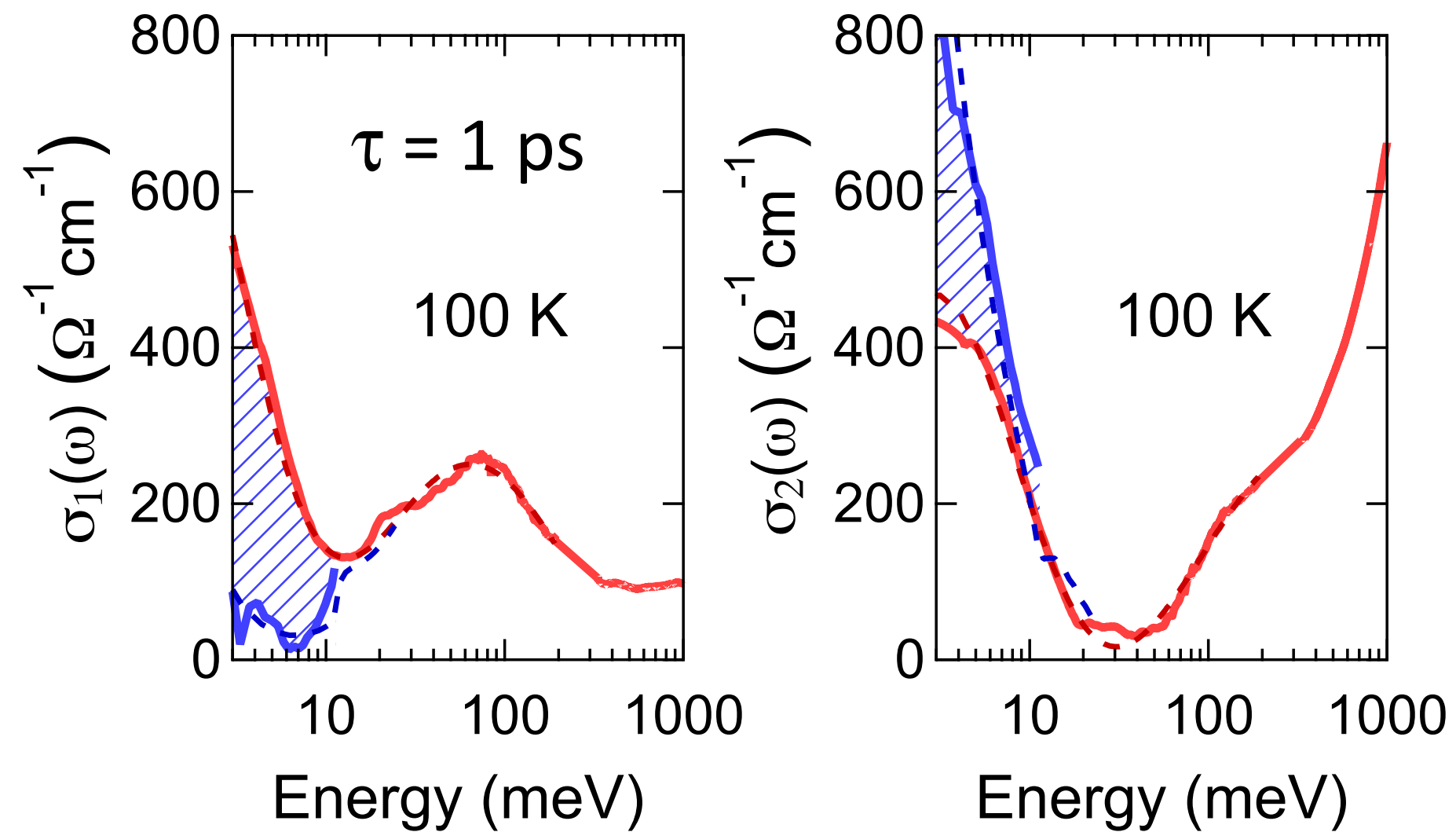
N. Bittner et al. *J. Phys. Soc. Jpn.* **88**, 044704 (2019)

and more...

Need to measure new observables

Experimental probes

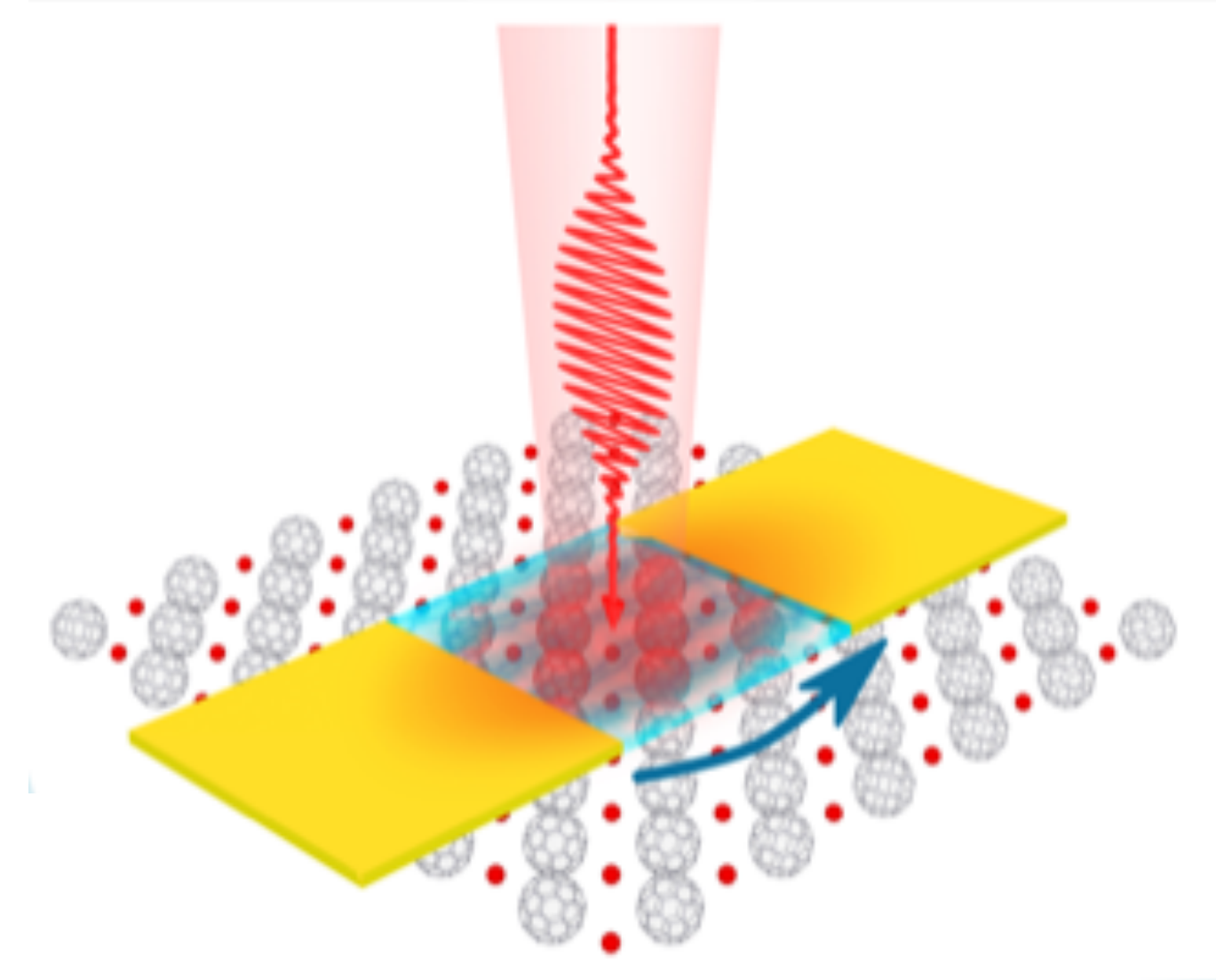
Time-resolved optical spectroscopy



D. Fausti et al. *Science* **331**, 189 (2011)
W. Hu et al. *Nature Materials*, **13**, 705 (2014)
S. Kaiser et al. *Phys. Rev. B*, **89**, 184516 (2014)
M. Mitrano et al. *Nature*, **530**, 461 (2016)
A. Cantaluppi et al. *Nature Physics*, **14**, 837 (2018)

D. Nicoletti et al. *PRB* **90**, 100503(R) (2014)
D. Nicoletti et al. *PRL* **121**, 267003 (2018)
M. Först et al. *PRL* **112**, 157002 (2014)
K. A. Cremin et al. *PNAS* **116**, 19875 (2019)
R. Mankowsky et al. *PRB* **91**, 094308 (2015)

Time-resolved transport

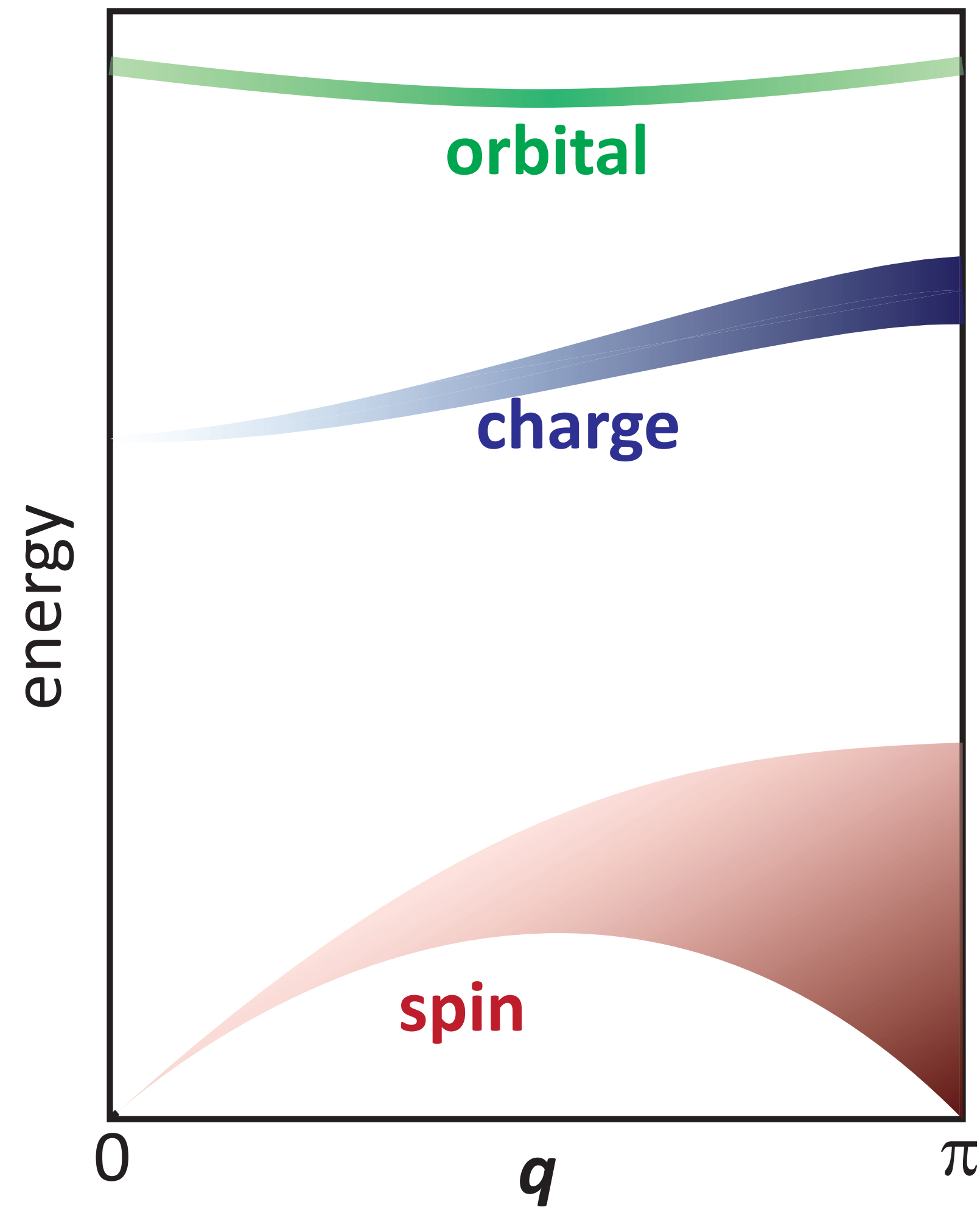


M. Budden et al. forthcoming

Problem: time-resolved observables are either q-integrated or at q=0

Solutions: trARPES, tr-nanoIR, tr-scattering, ...

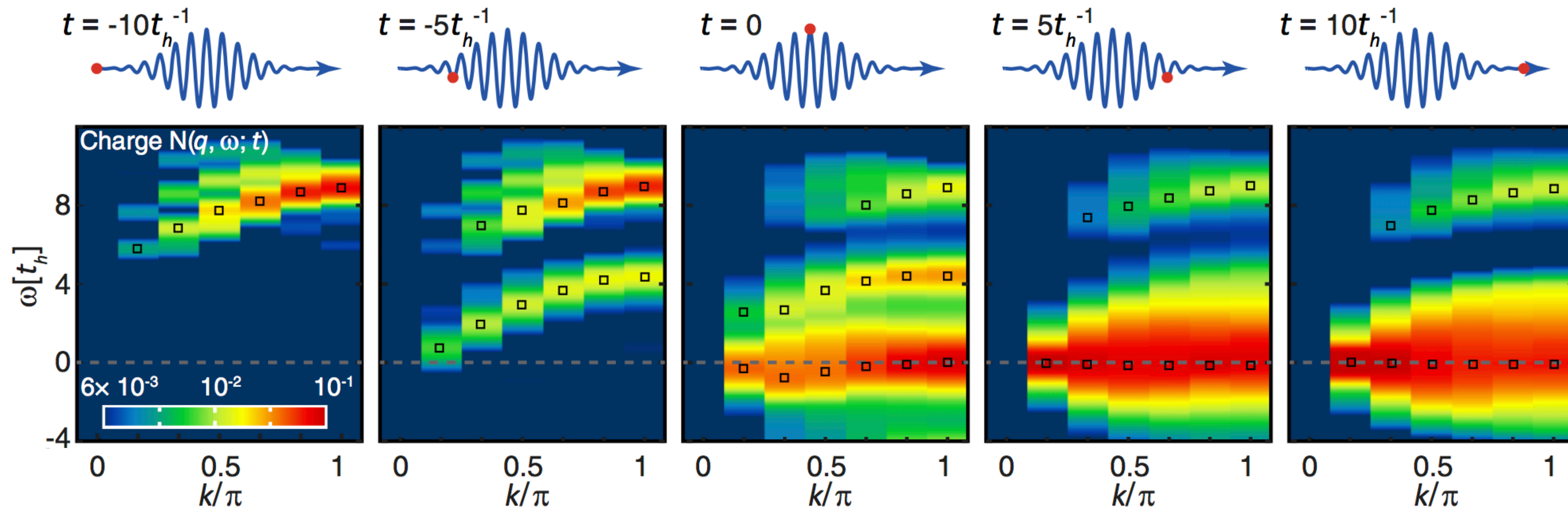
What can we learn from the inelastic spectrum?



Courtesy of Y. Wang (Harvard)

- Propagating collective excitations
- Coupled degrees of freedom
- Microscopic order parameters

What can we learn from the inelastic spectrum?



e.g. Devereaux group

Y. Wang et al. *Phys. Rev. B* **96**, 235142 (2017)

Y. Chen et al. *Phys. Rev. B* **99**, 104306 (2019)

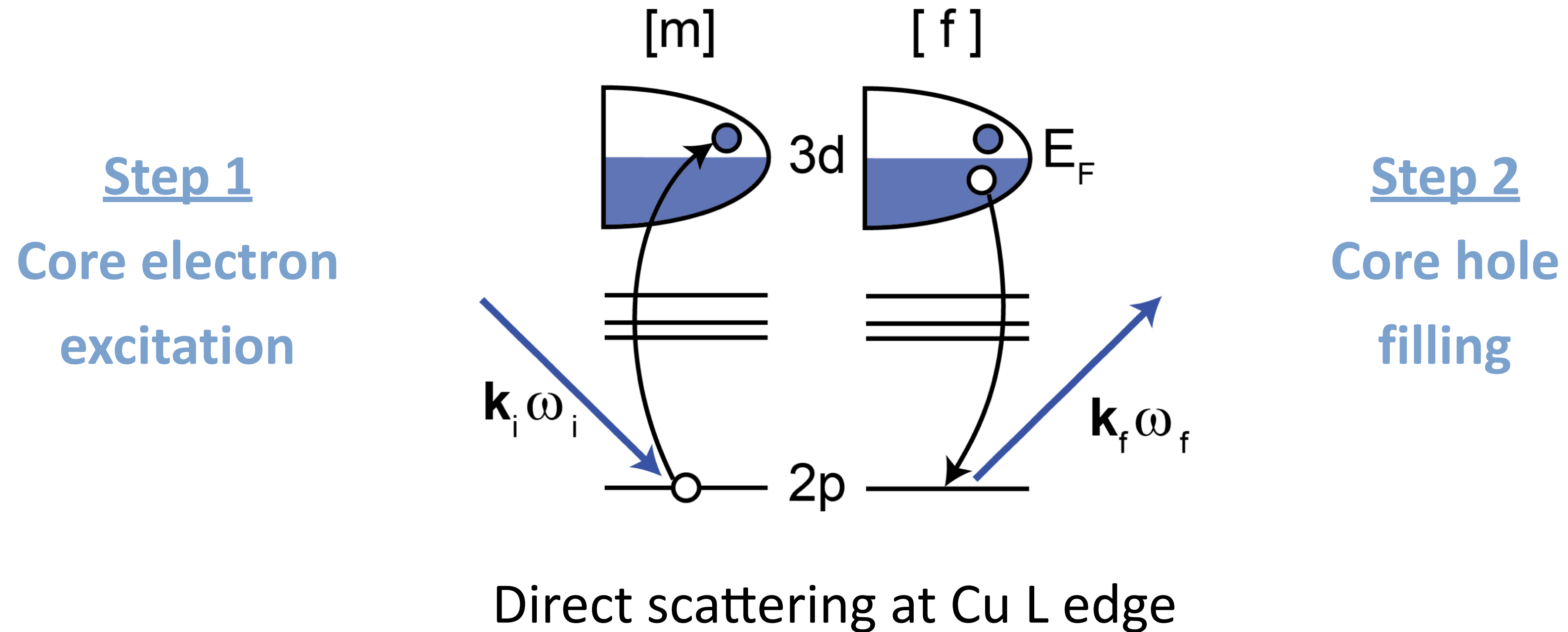
Y. Wang et al. *Nat. Rev. Mat.* **3**, 312–323 (2018)

Open questions for light-induced superconductivity

- Symmetry of photo-induced gaps
- Dynamics of spin excitations
- Renormalization of phonon dispersion and damping
- Floquet driving of plasmonic spectrum

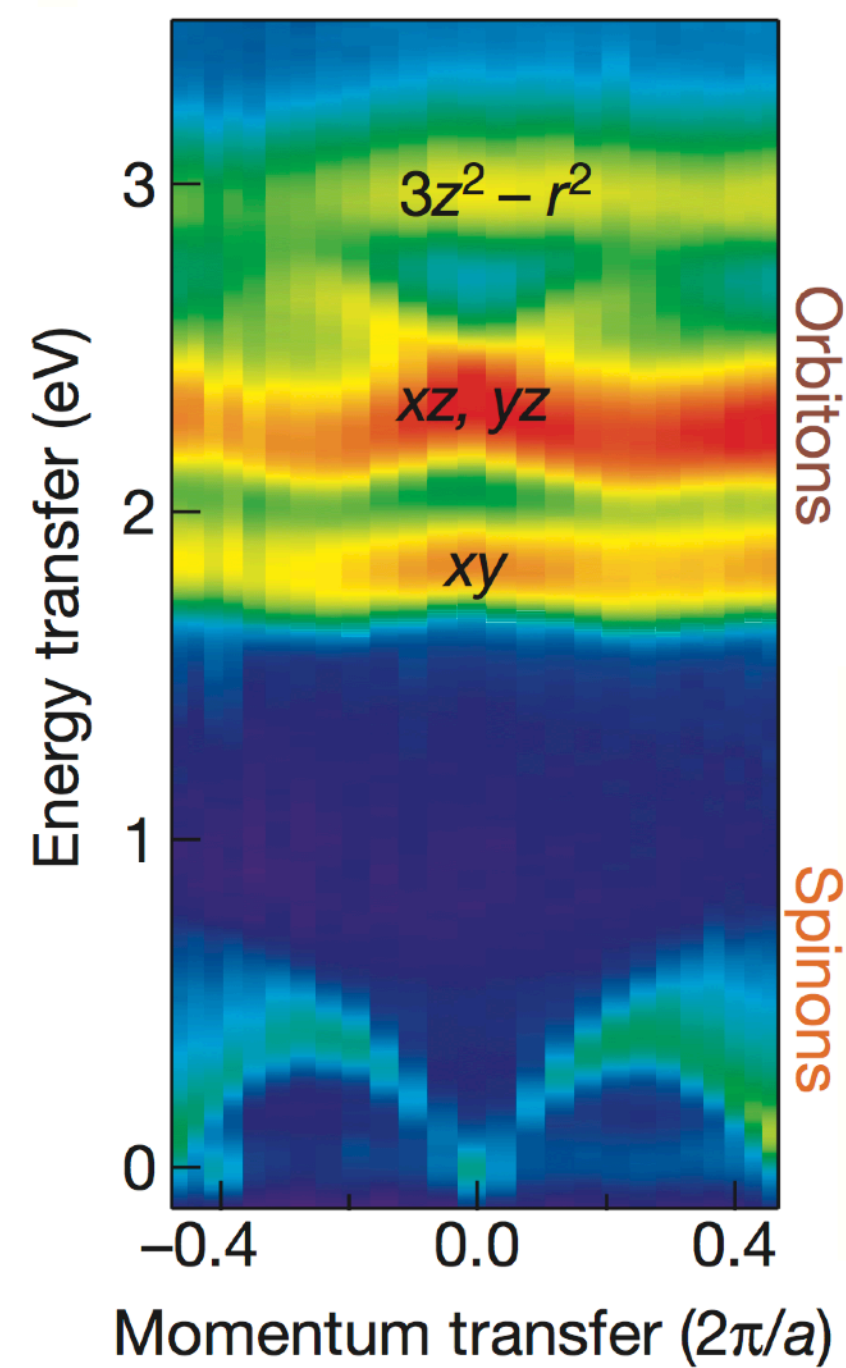
How to probe time-dependent, finite-momentum
inelastic dynamics?

Resonant inelastic X-ray scattering (RIXS)

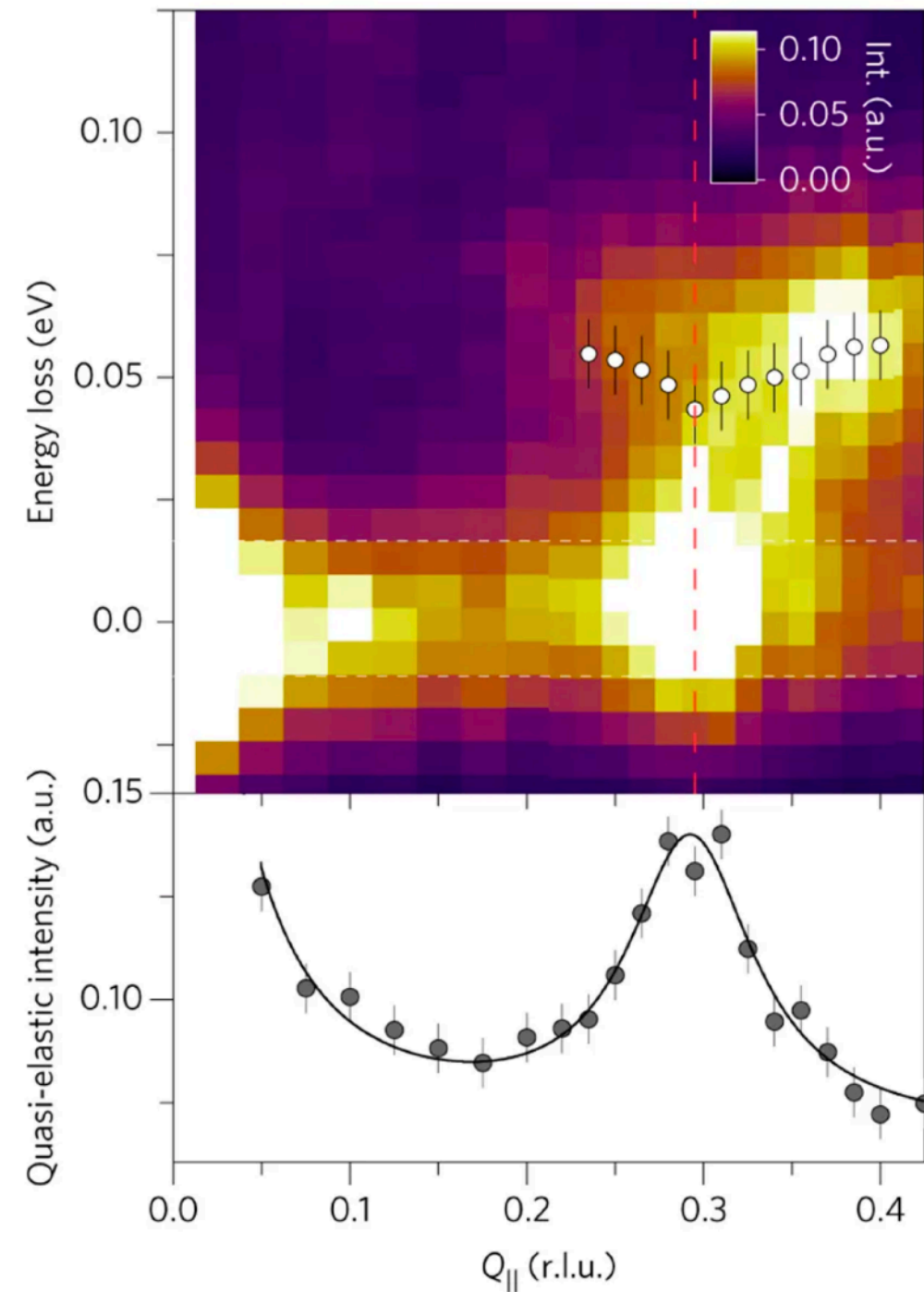


- Resonance increases **intensity** and **valence-band sensitivity**
- X-rays are a **finite-momentum** probe

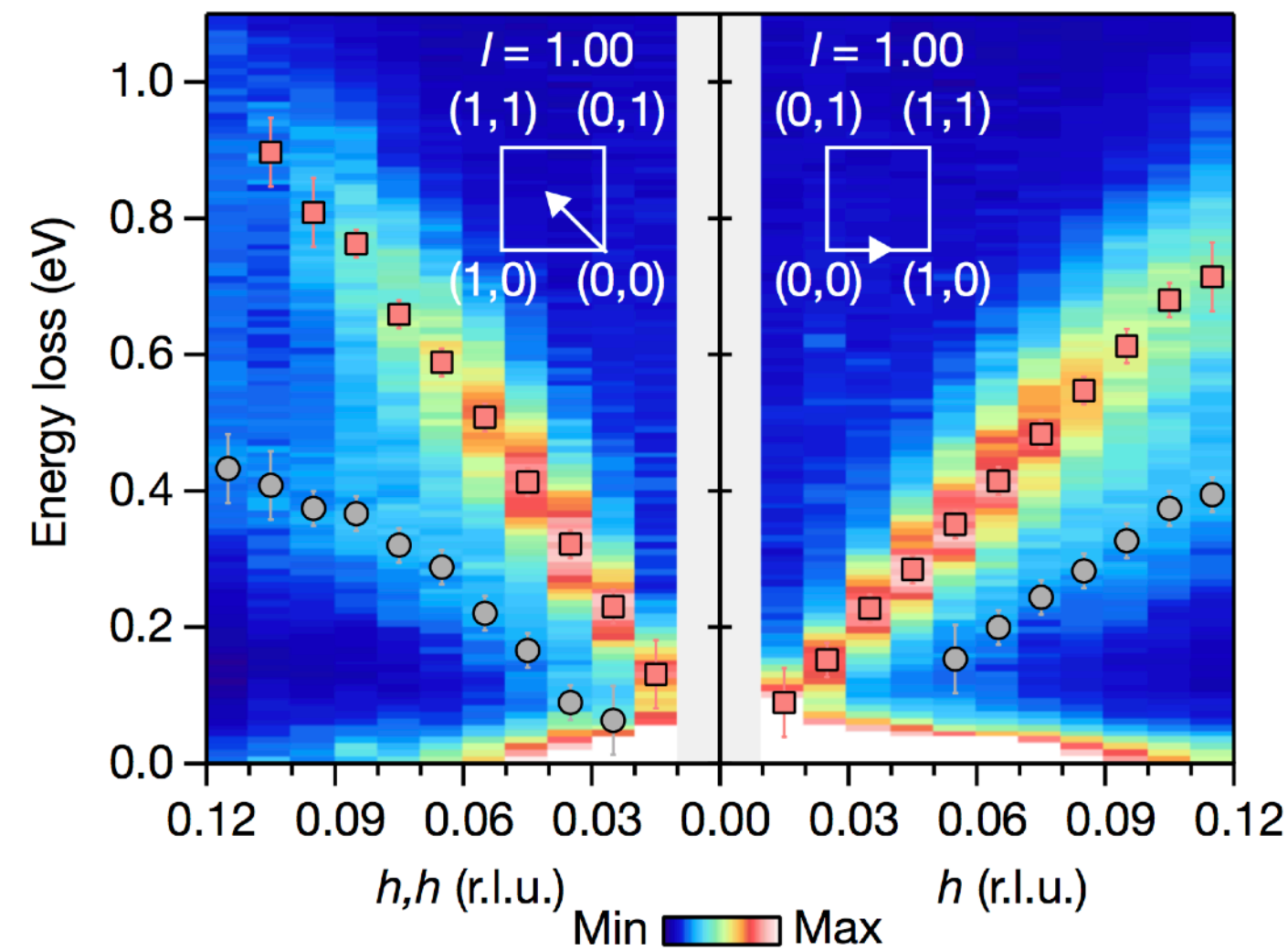
Resonant inelastic X-ray scattering (RIXS)



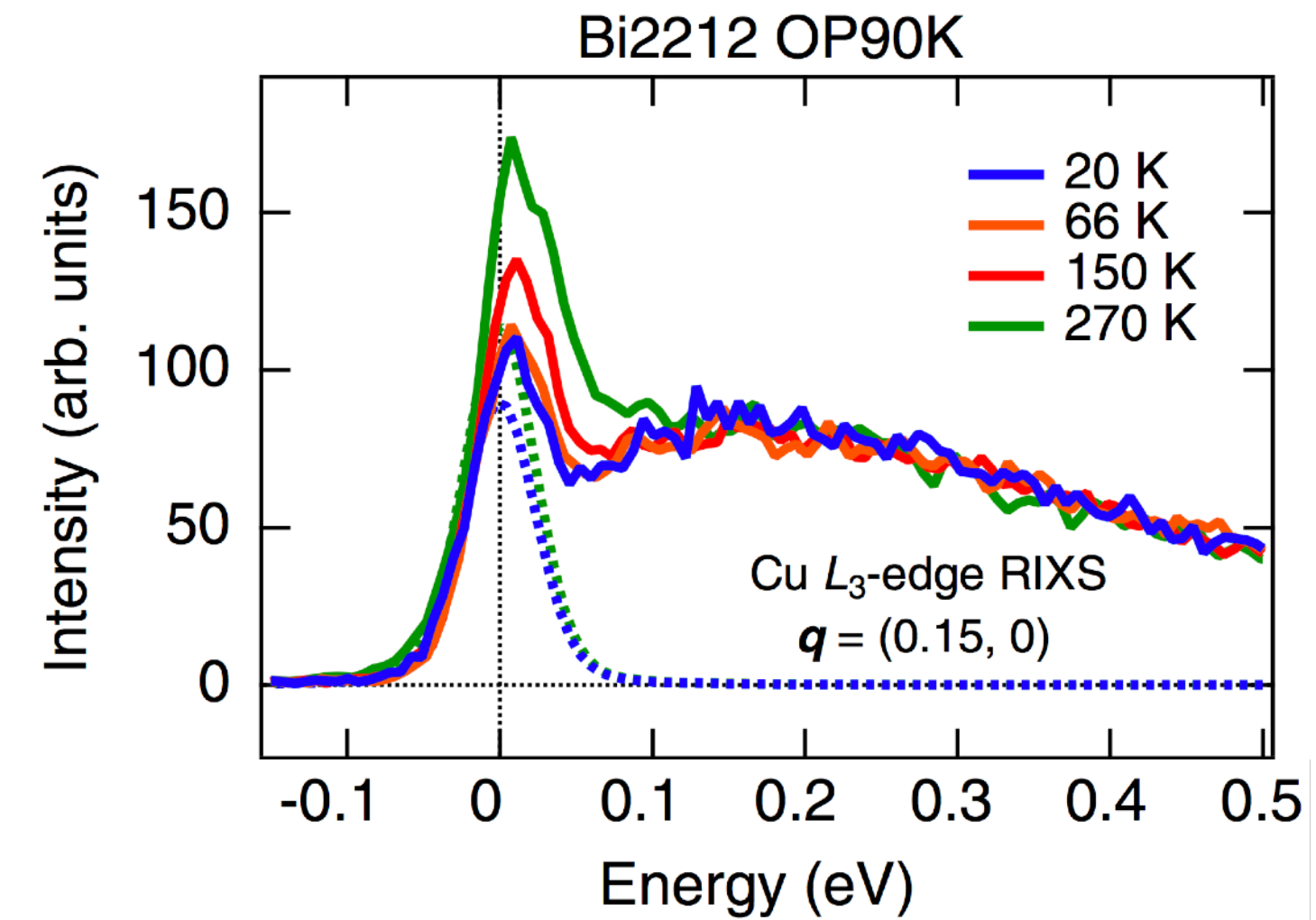
J. Schlappa et al. Nature
485, 82–85(2012)



L. Chaix et al. Nature Physics
485, 82–85(2012)



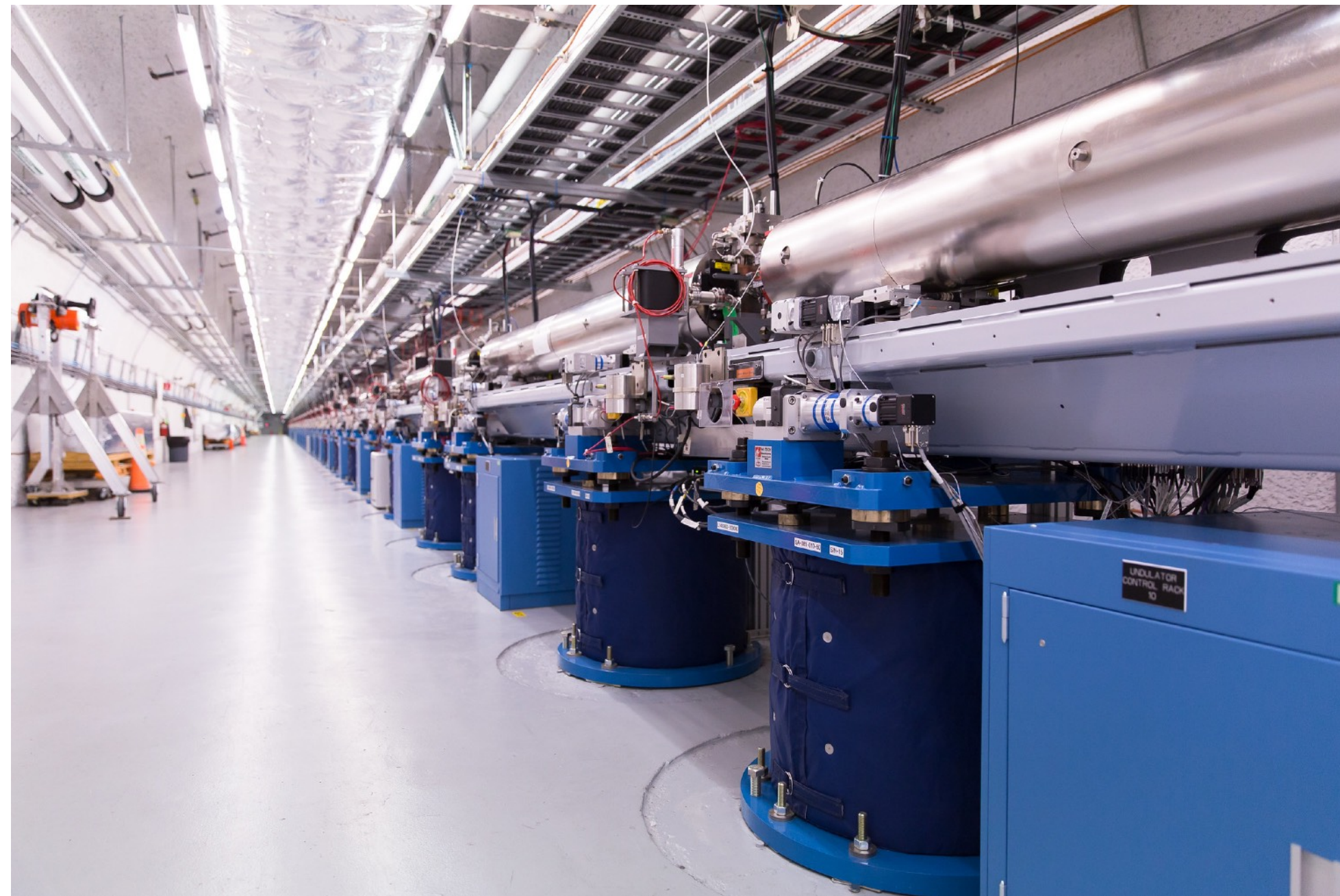
M. Hepting et al. Nature 563,
374–378 (2018)



H. Suzuki et al. npj Quantum
Materials 65 (2018)

- Resonance increases **intensity** and **valence-band sensitivity**
- X-rays are a **finite-momentum** probe
- Sensitive to collective excitations (phonons, magnons, plasmons)

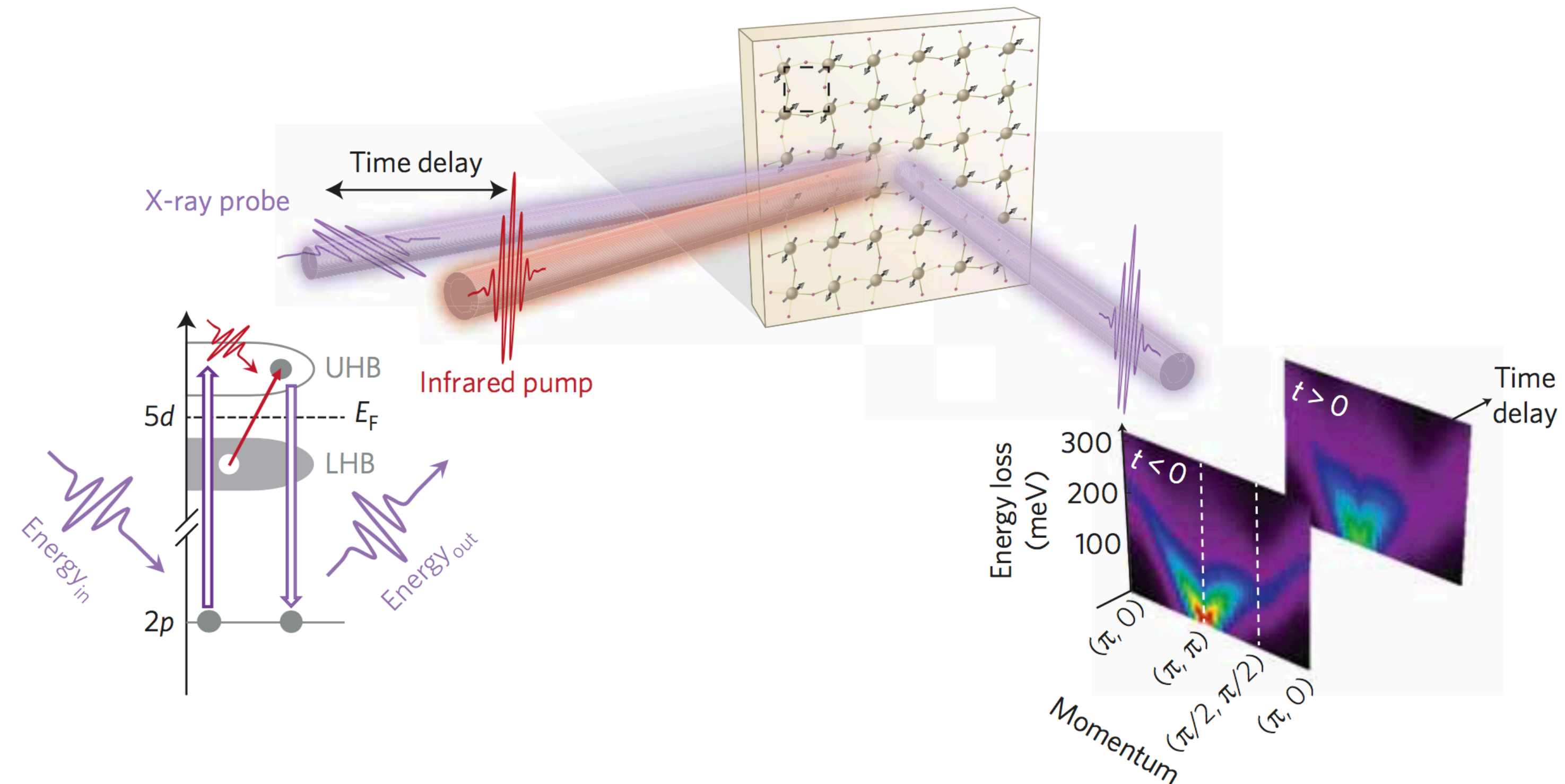
Time-resolved RIXS at free electron lasers



LCLS @ SLAC

- LCLS pioneered trRIXS both in **hard and soft X-rays**
- First trRIXS experiment: **spin dynamics** in Sr_2IrO_4 (hard X-rays, Ir L-edge)
- Last two years: extension to soft X-ray (Cu, V, Ni L edge, O K edge)

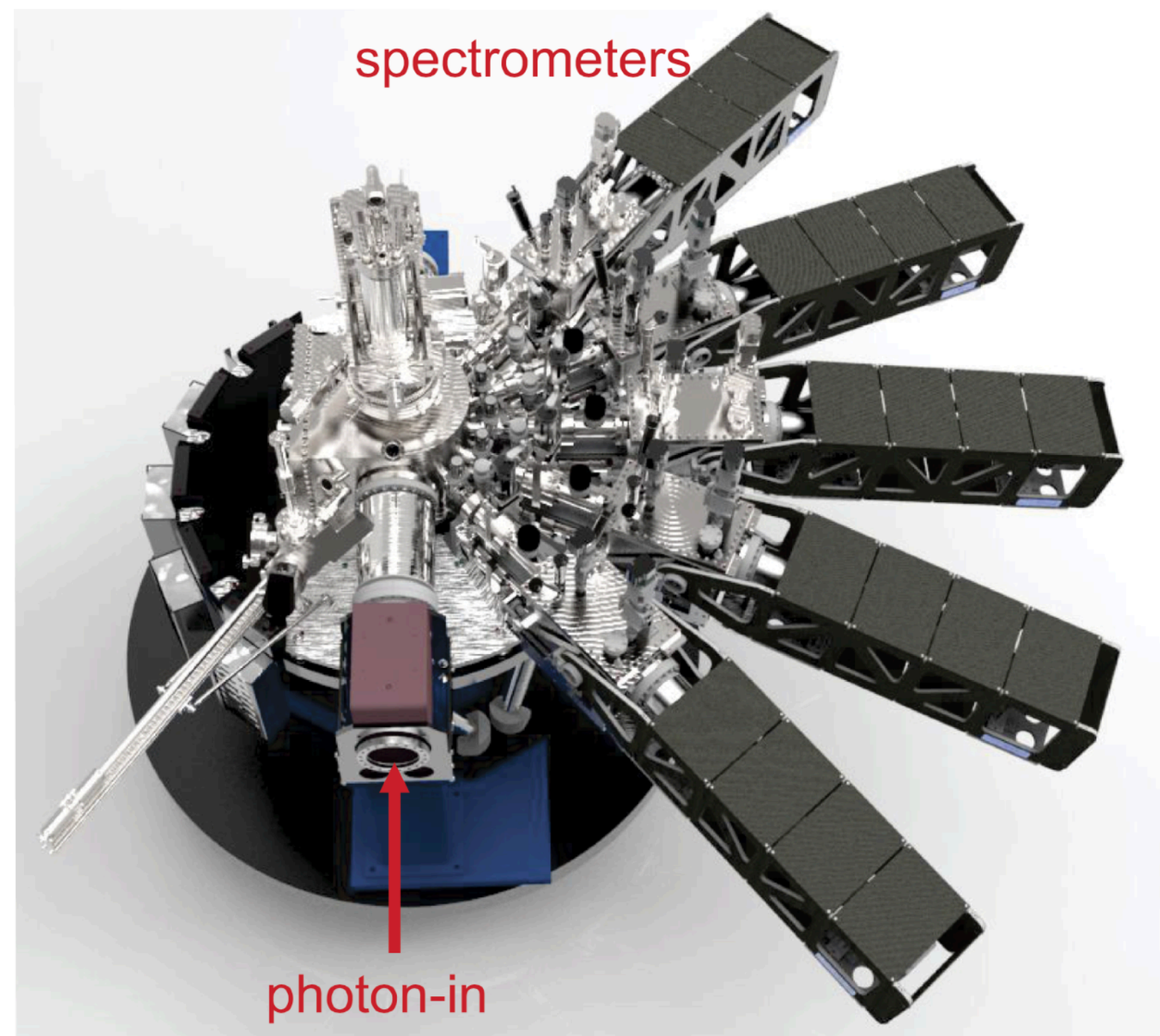
M. P. M. Dean et al. Nature Materials **15**, 601 (2016)



This talk and S. Parchenko et al. arXiv: 1908.02603 (2019)

Soft X-ray Time-resolved RIXS

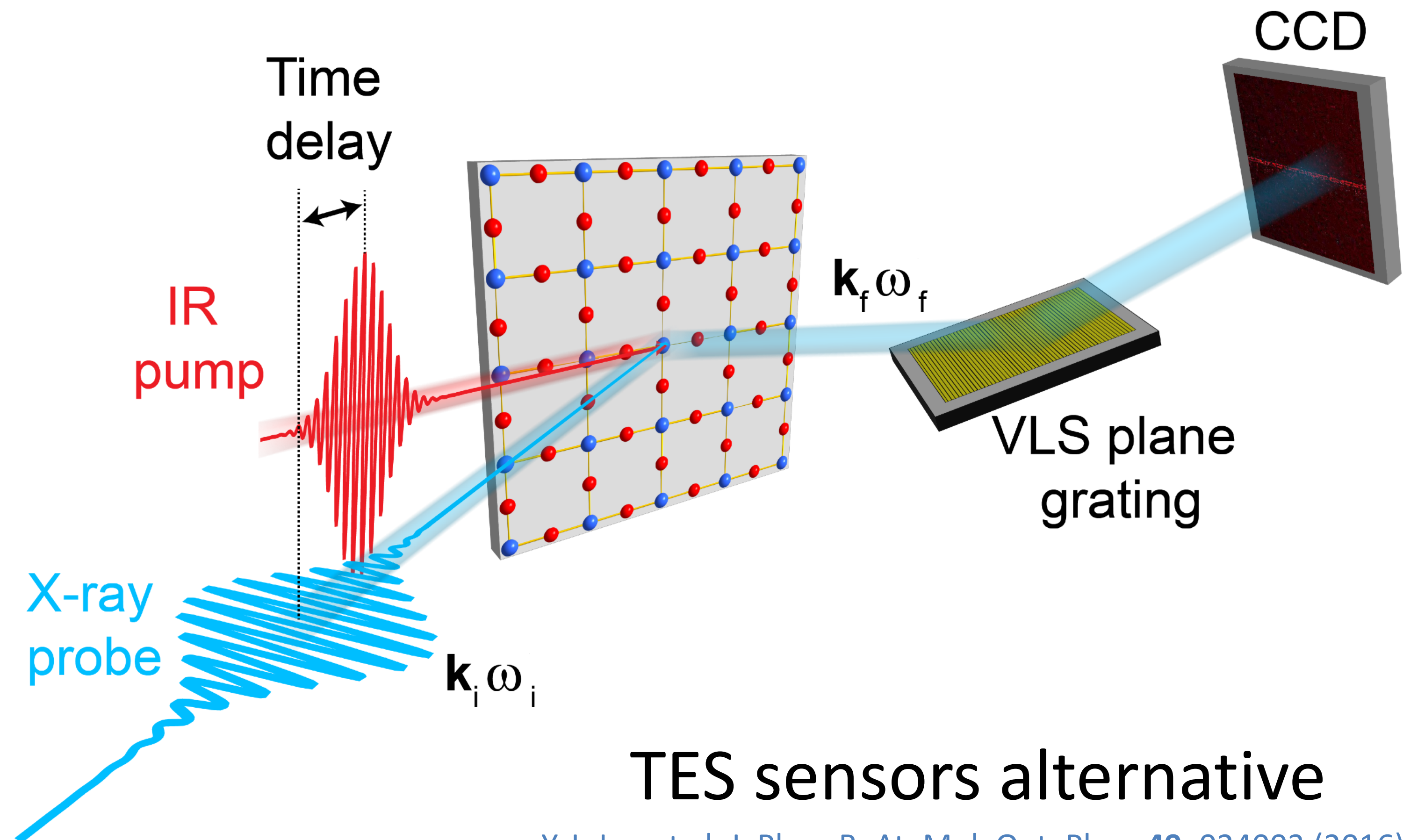
RIXS is photon-hungry (more later), and LCLS is a 120 Hz linac



Y. D. Chuang et al. Rev. Sci. Instr. **88**, 013110 (2017)

Y. D. Chuang et al. J. El. Spectr., in press 146897 (2019)

qRIXS spectrometer (1.2m)
LCLS, PAL-XFEL, ALS, ...



TES sensors alternative

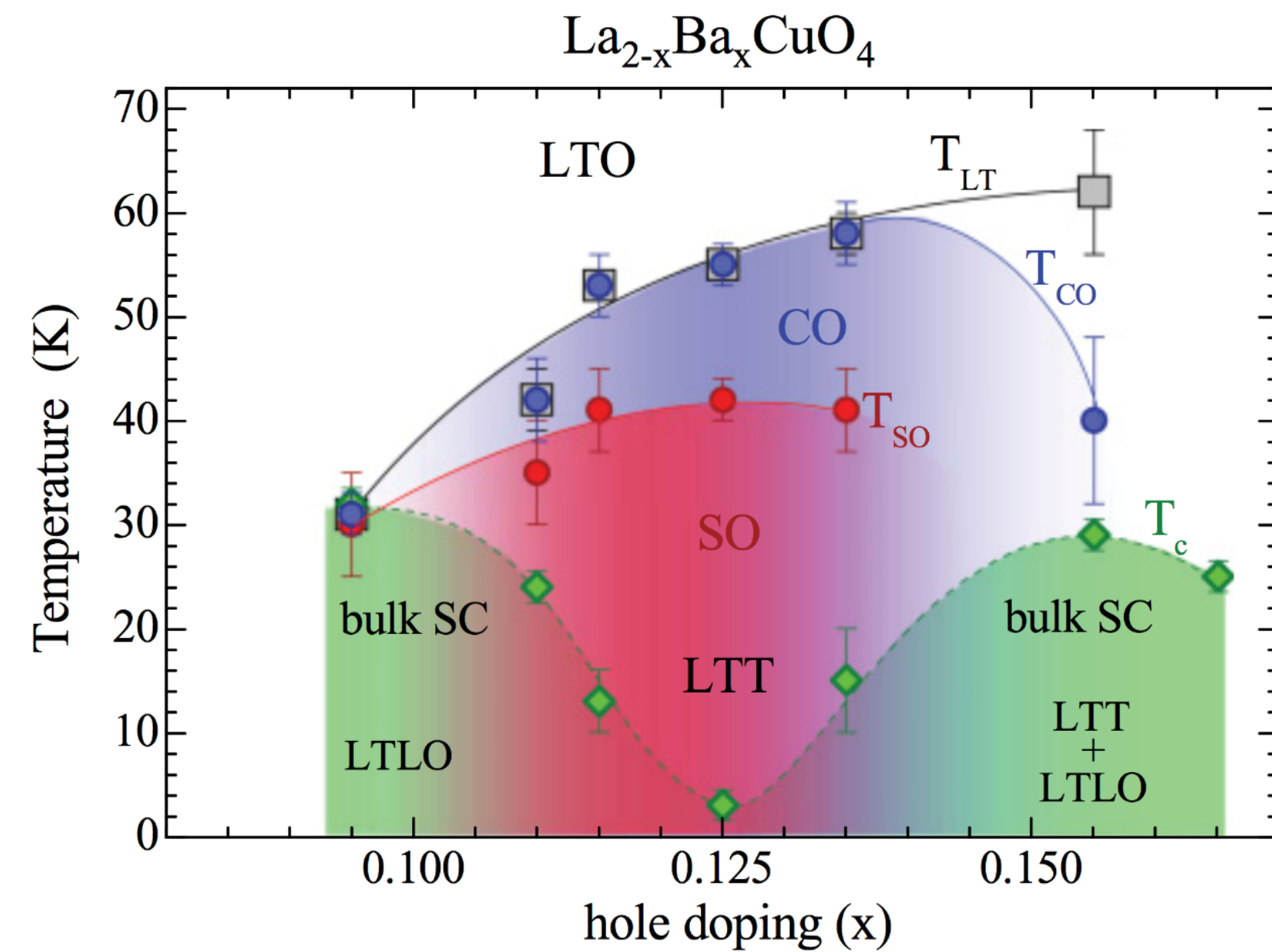
Y. I. Joe et al. J. Phys. B: At. Mol. Opt. Phys. **49**, 024003 (2016)

J.N. Ullom et al. Synchr. Rad. News **27**, 24-27 (2014)

J. Uhlig et al. Phys. Rev. Lett. **110**, 138302 (2013)

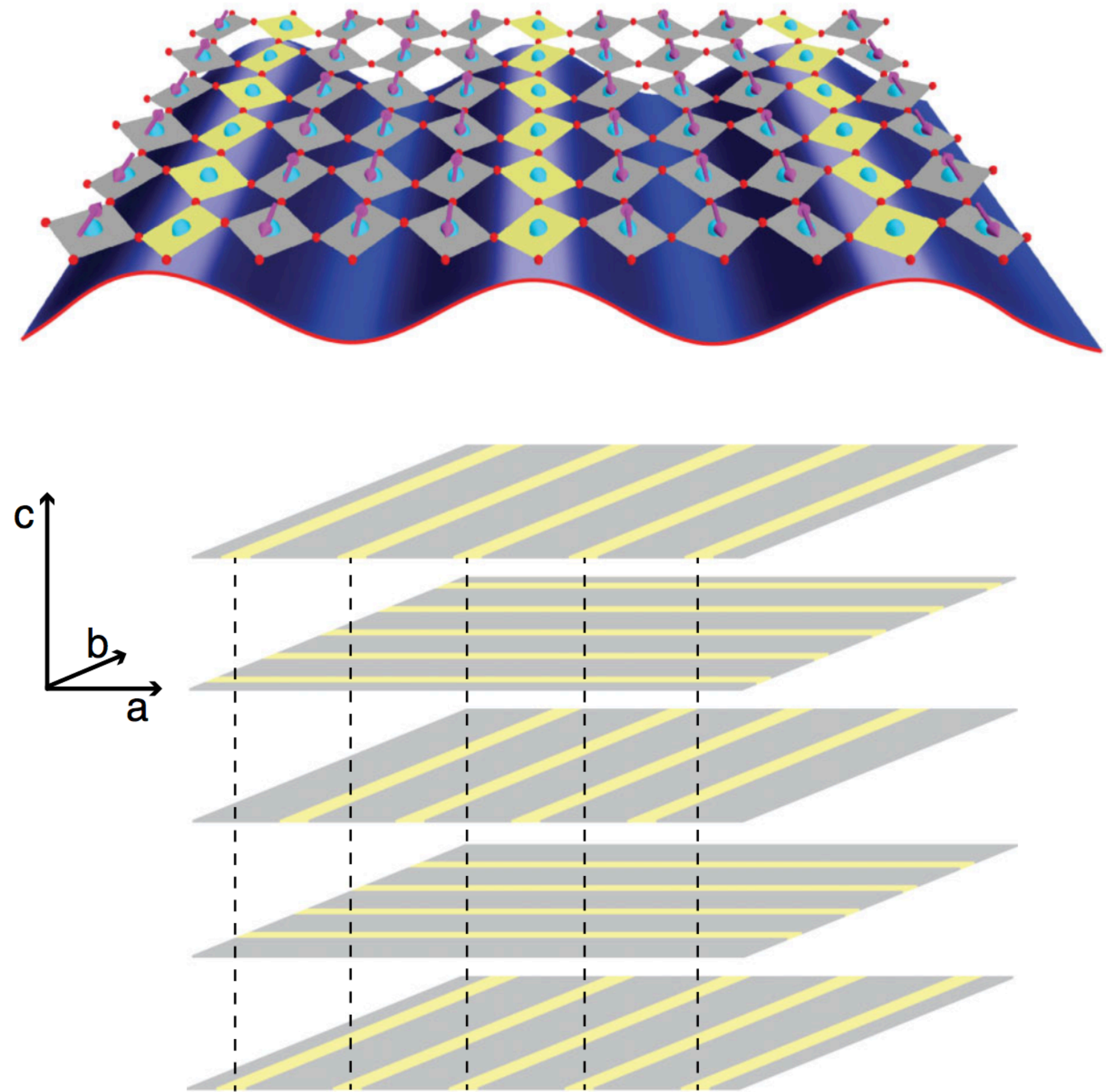
Photo-excited superconductivity in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$

Charge order and superconductivity in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$



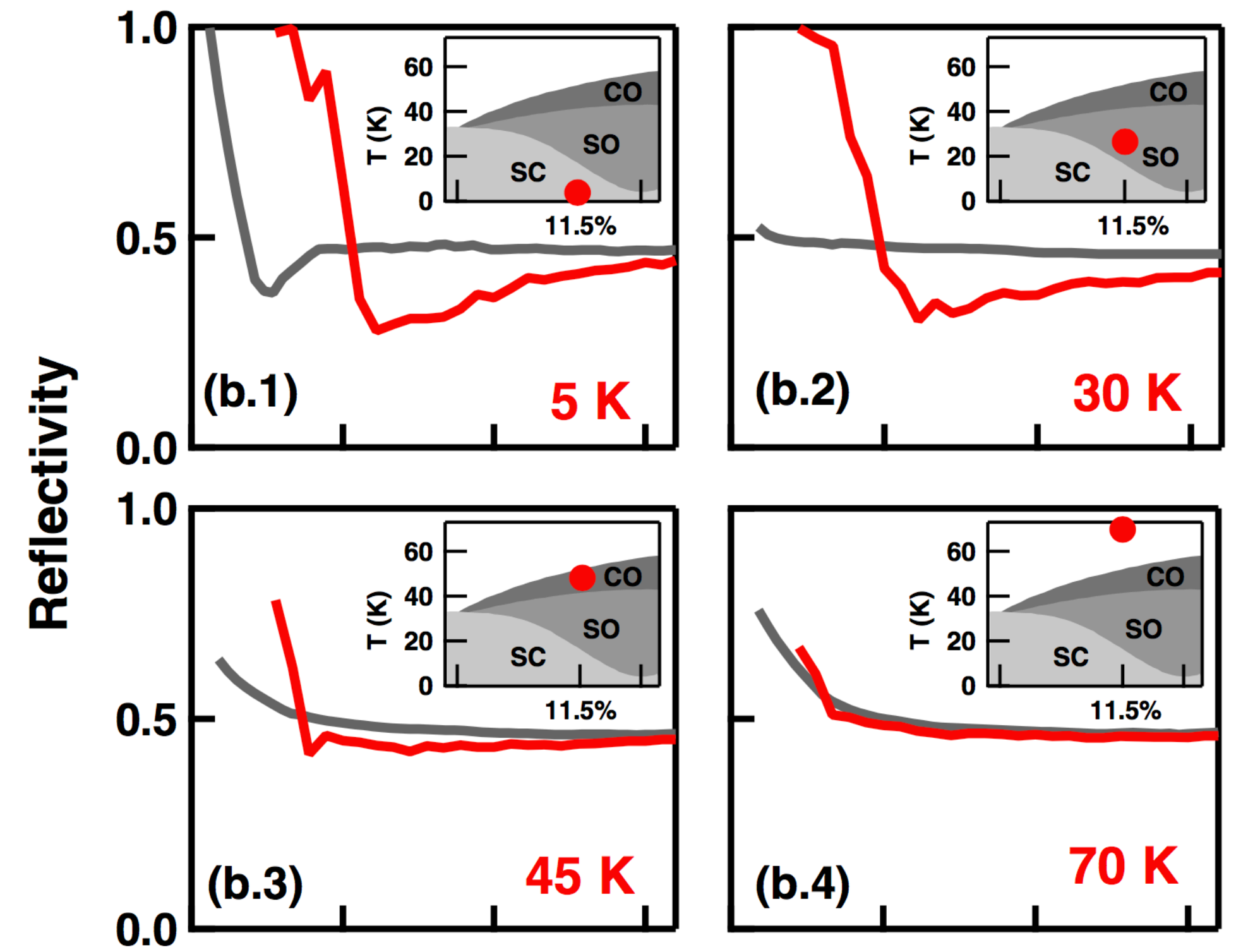
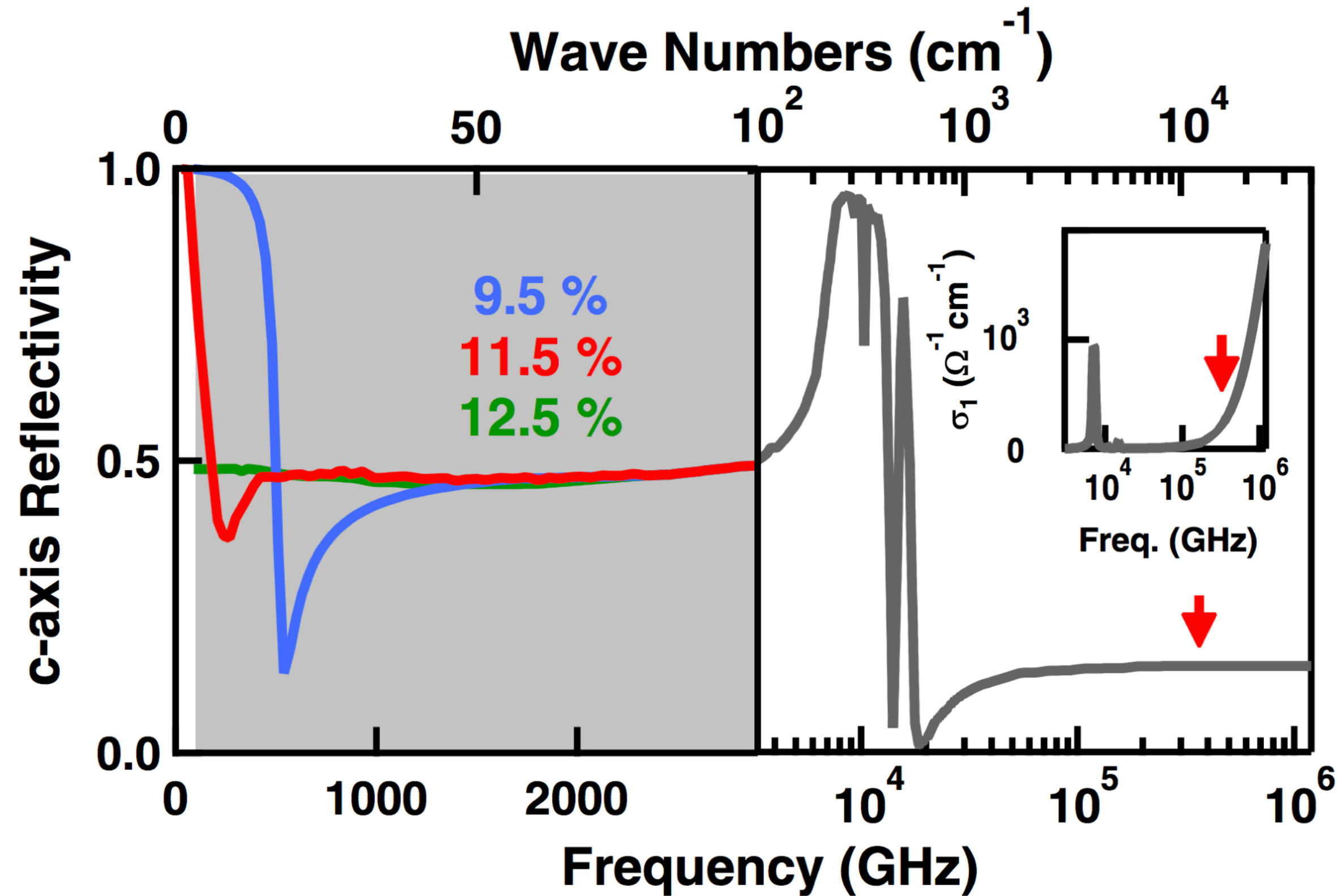
Hücker et al. Phys. Rev. B **83**, 104506 (2011)

- Striped single layer cuprate
- Strong 1/8 anomaly
- 2D high-T SC fluctuations



Först et al. Phys. Rev. Lett. **112**, 157002 (2014)

Light-induced superconductivity in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$

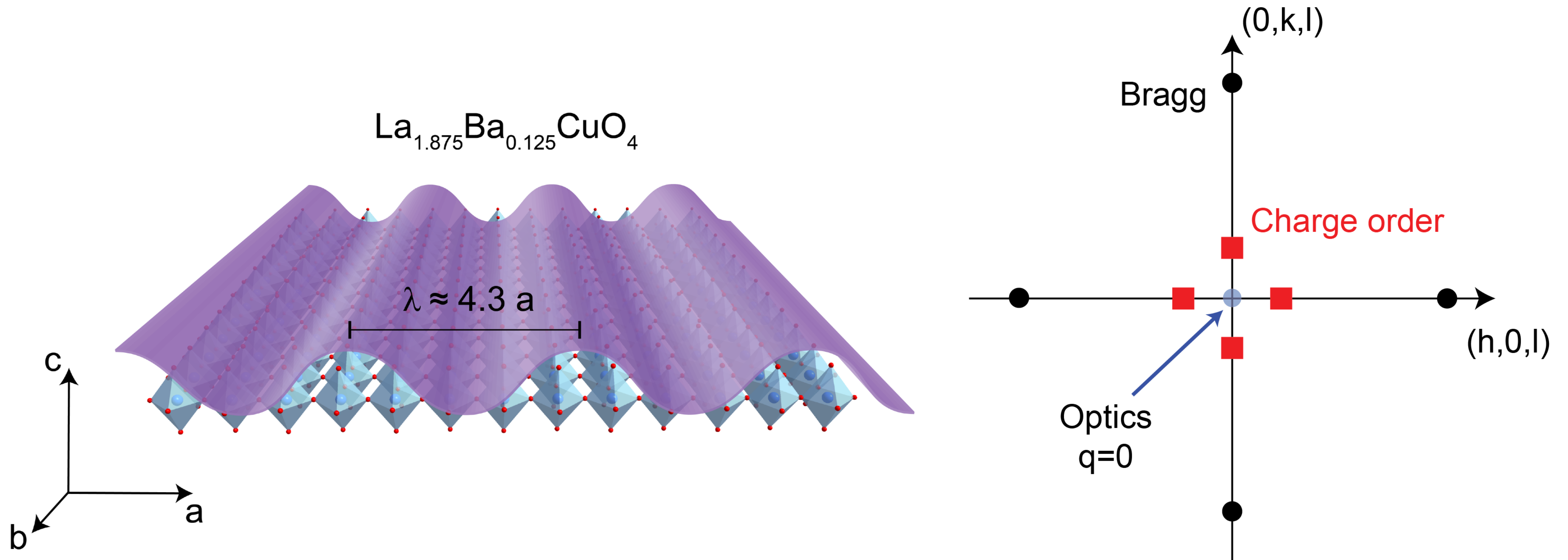


Transient c-axis reflectivity

- JPR blueshift with 1.55 eV pump
- Enhancement below and above equilibrium T_c

D. Nicoletti et al. Phys. Rev. B **90**, 100503(R) (2014)
V. Khanna et al. Phys. Rev. B **93**, 224522 (2016)
E. Casandruc et al. Phys. Rev. B **91**, 174502 (2015)
K. A. Cremin et al. PNAS **116**, 19875 (2019)

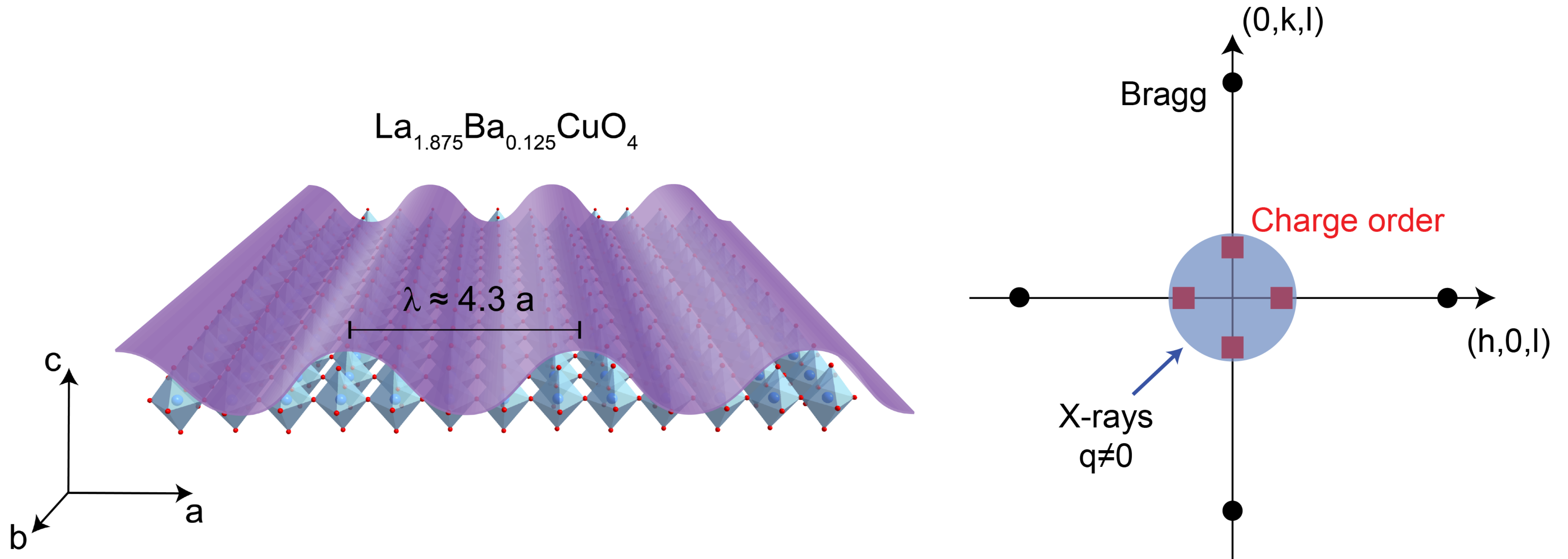
Charge order parameter structure



Charge order modulation gives rise to finite-q order parameter

J. M. Tranquada et al. Nature 429, 534-538 (2004)
P. Abbamonte et al. Nature Physics 1, 155-158 (2005)

Charge order parameter structure



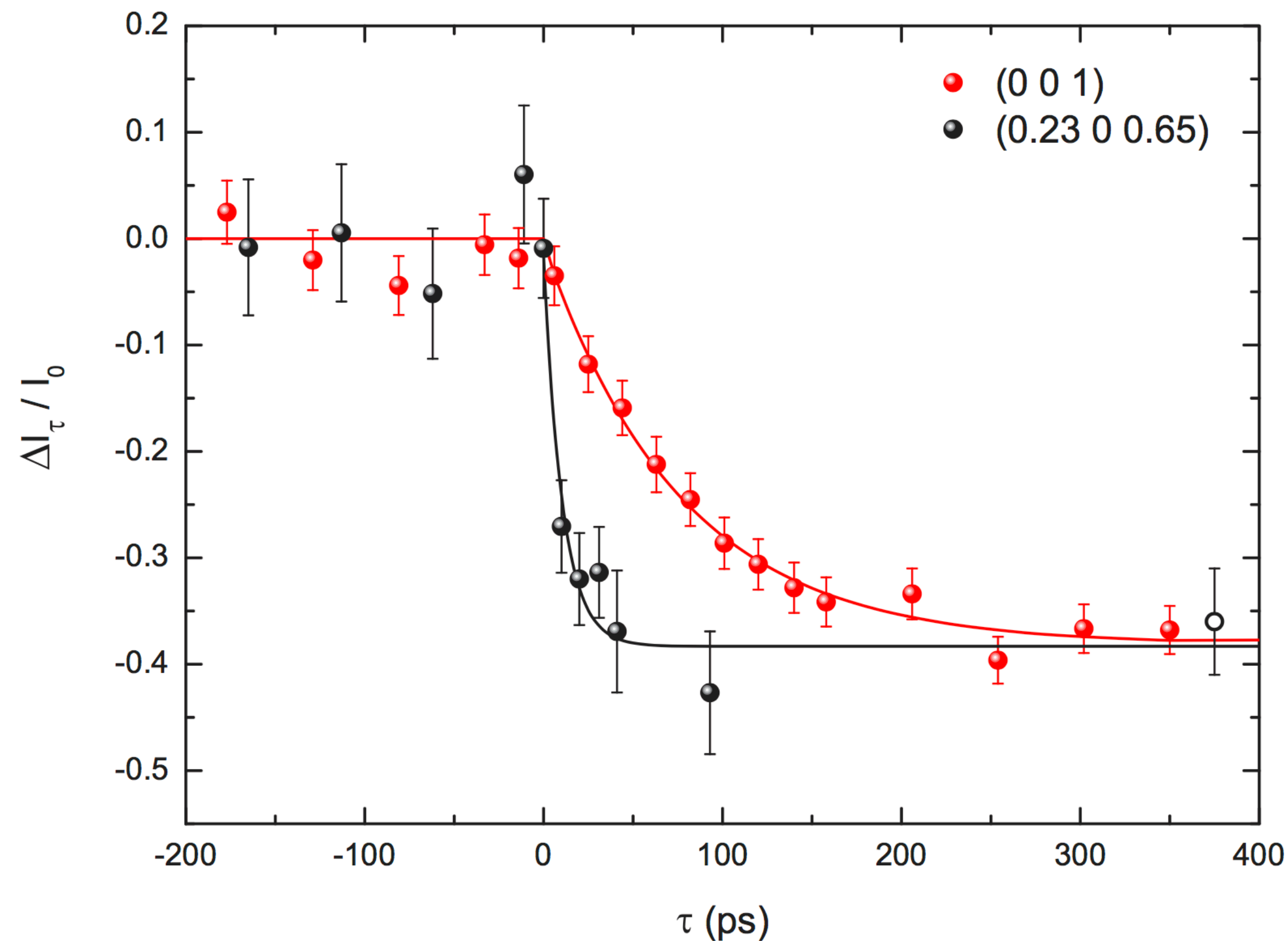
Charge order modulation gives rise to finite- q order parameter

J. M. Tranquada et al. Nature 429, 534-538 (2004)
P. Abbamonte et al. Nature Physics 1, 155-158 (2005)

Transient suppression of charge order

Pump: 1.55 eV, ac pol.

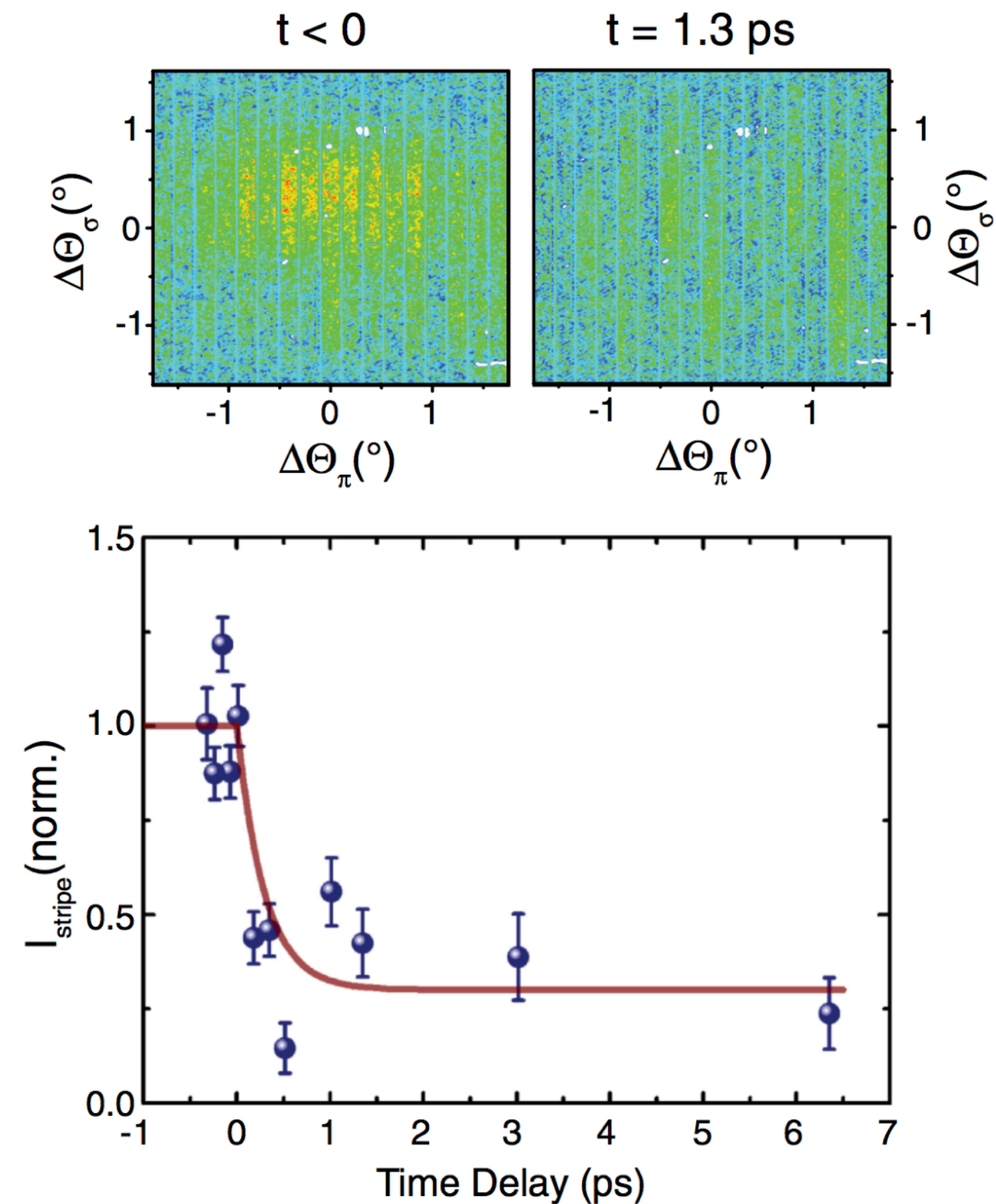
Probe: O K edge



Khanna et al. Phys.Rev. B. **93**, 224522 (2016)

Pump: 85 meV, ab pol. (Cu-O stretching)

Probe: O K edge

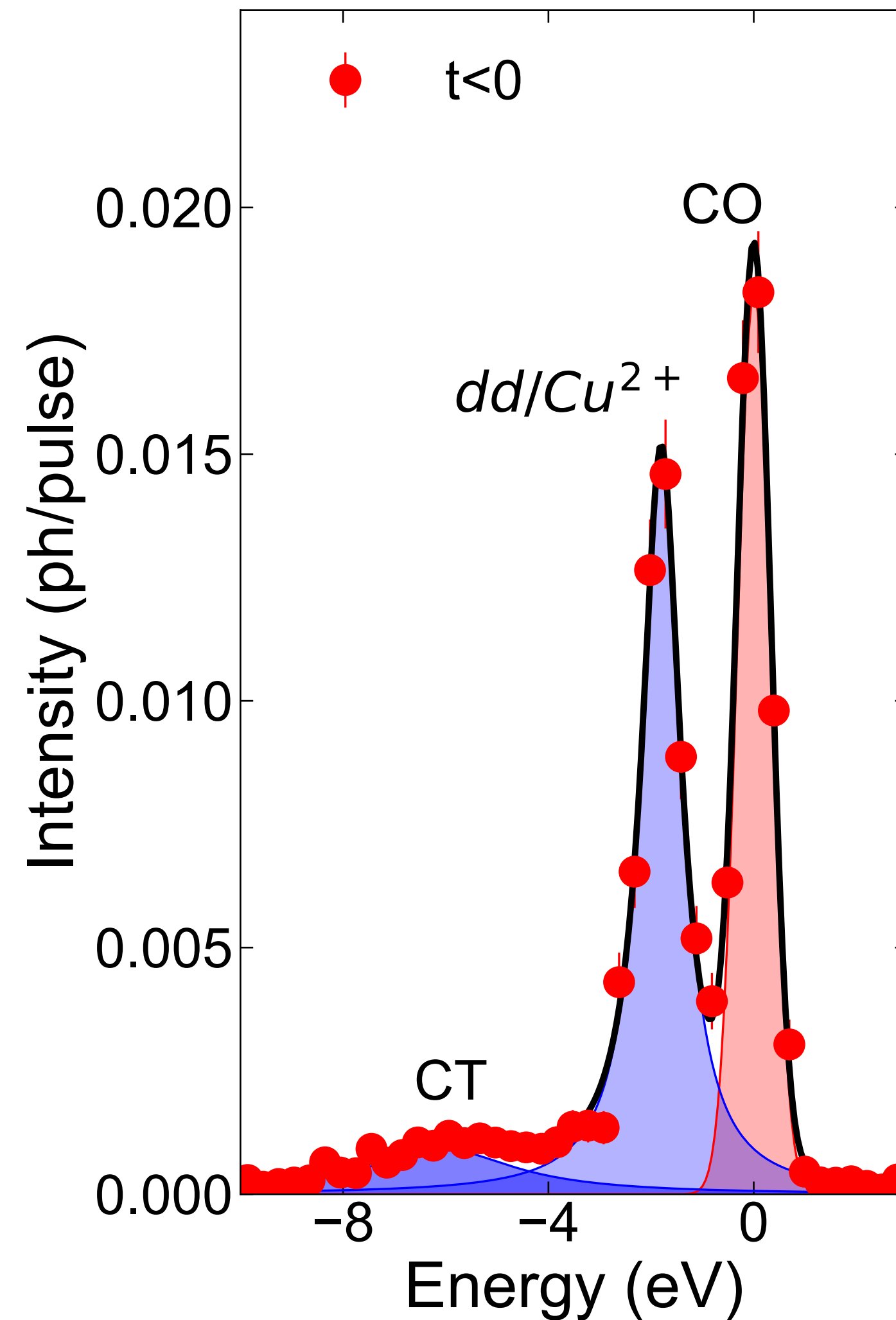


Först et al. Phys.Rev. Lett. **112**, 157002 (2014)

Early experiments detected transient charge order suppression

TrRIXS spectrum of photo-excited $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$

Equilibrium RIXS spectrum of $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$



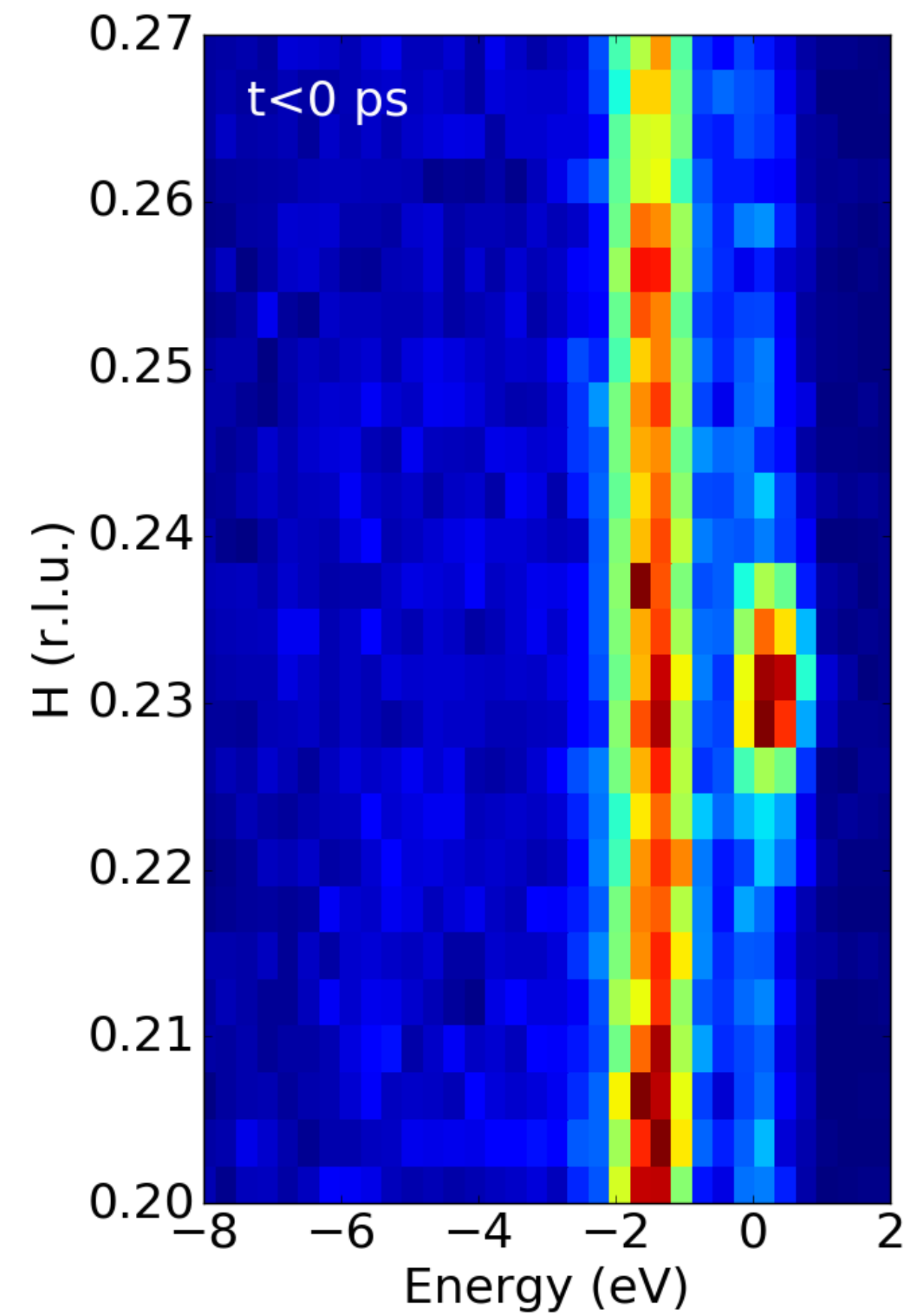
Cu L-edge

Spectral components:

- Elastic CO scattering
- dd excitations $d_{x^2-y^2} \rightarrow d_{z^2/zx/xy/yz}$
- Charge transfer $\text{Cu } 3d \rightarrow \text{O } 2p$

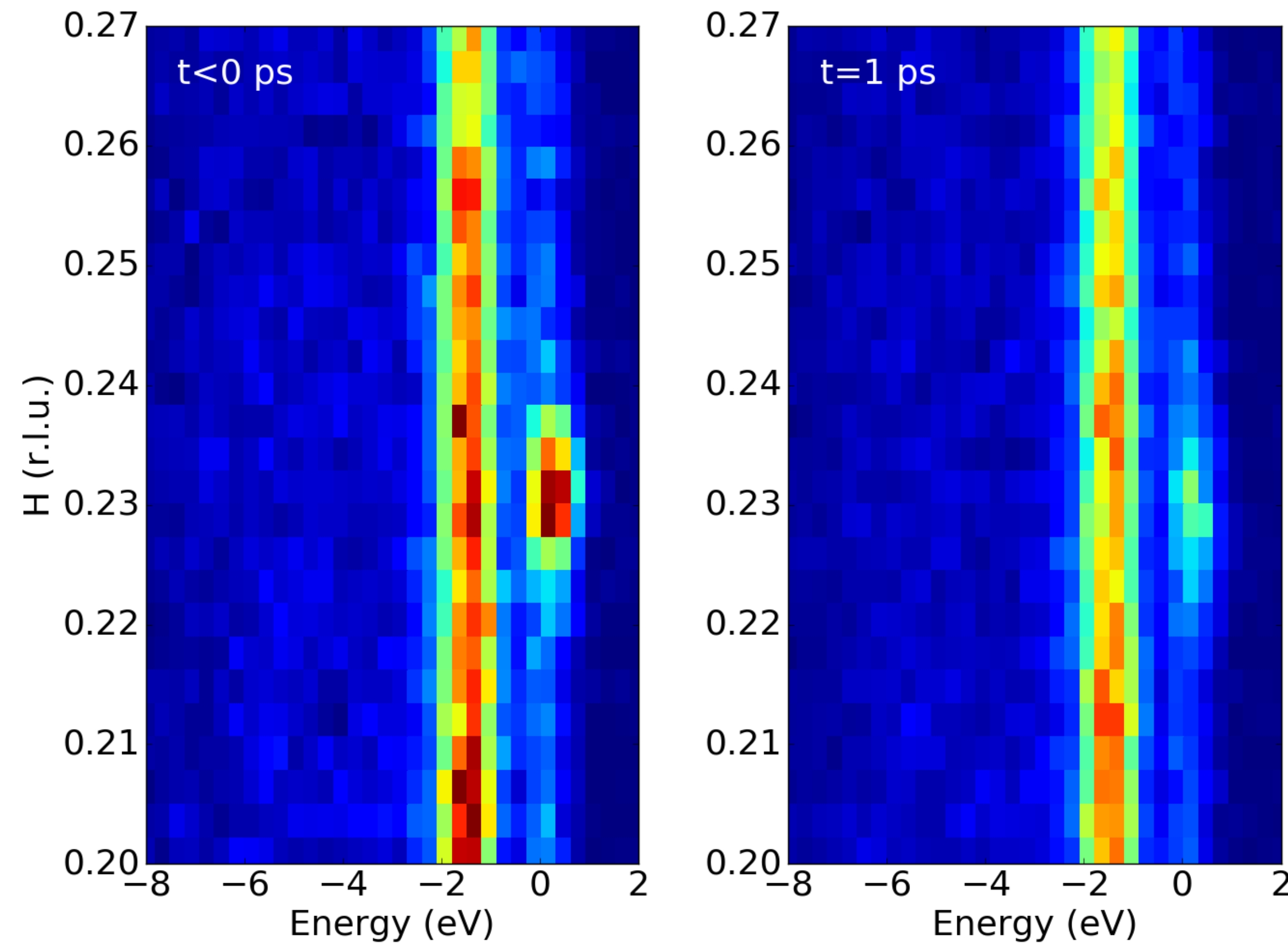
qRIXS: RP~1300 (2nd order), $\Delta E_{\text{res}}=0.7$ eV

Momentum-resolved order parameter quench



Momentum-resolved order parameter quench

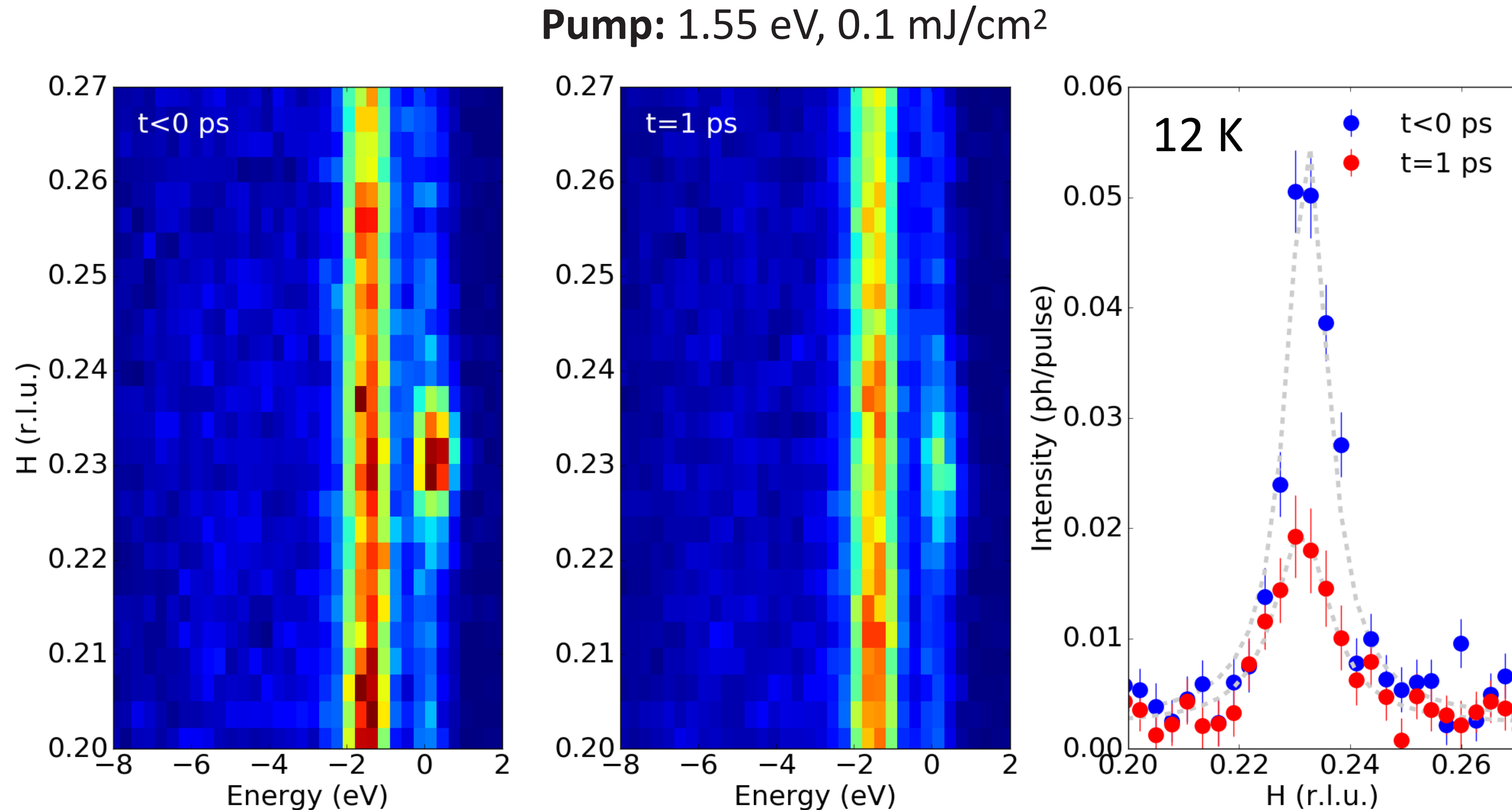
Pump: 1.55 eV, 0.1 mJ/cm²



MM et al. Science Advances **5**, eaax3346 (2019)

MM et al. PRB **100**, 205125 (2019)

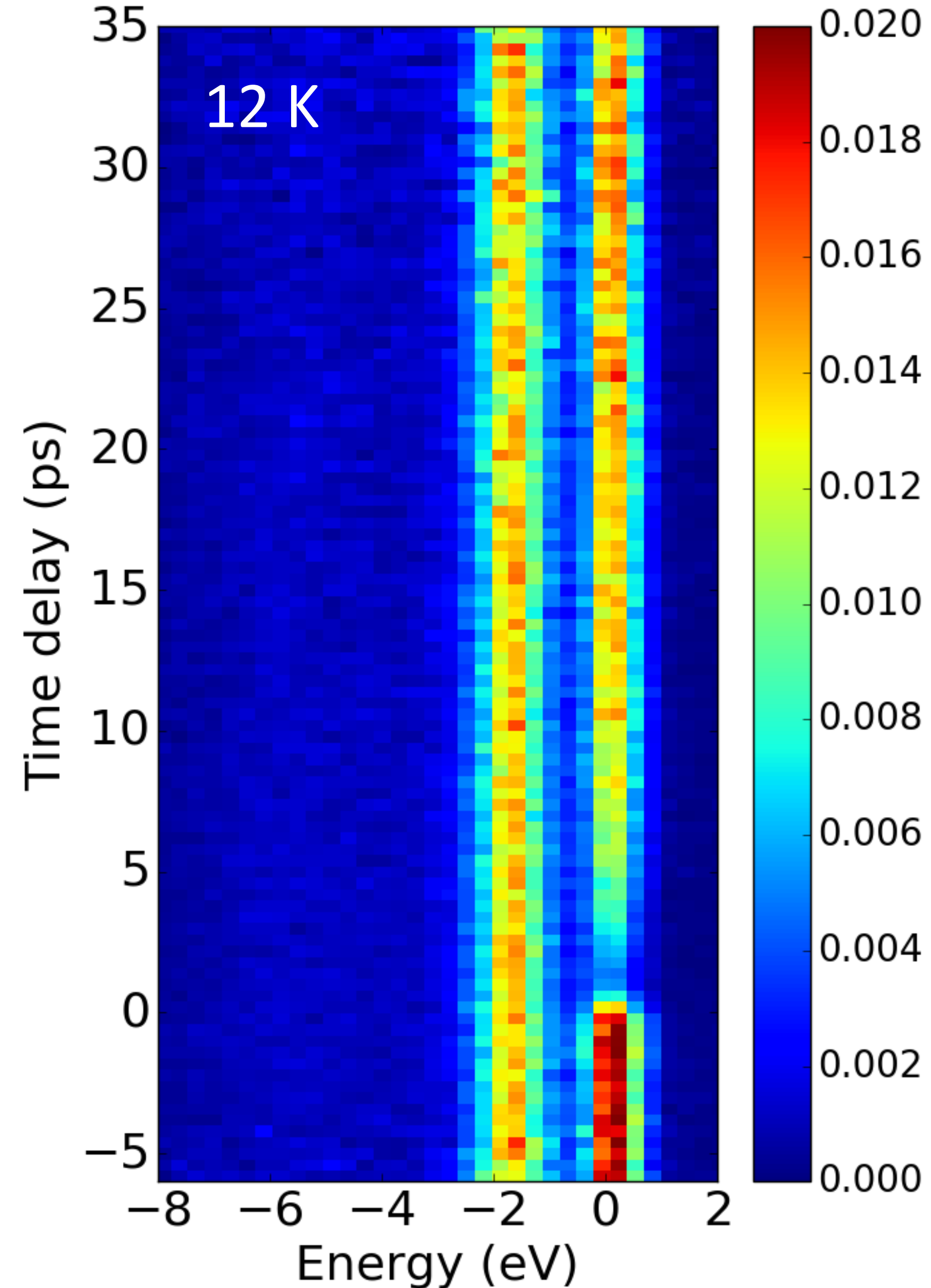
Momentum-resolved order parameter quench



- Order parameter suppressed and broadened in q space
- Pump only affects quasielastic scattering

Coherent charge order dynamics at early delays

Time-dependent RIXS spectrum

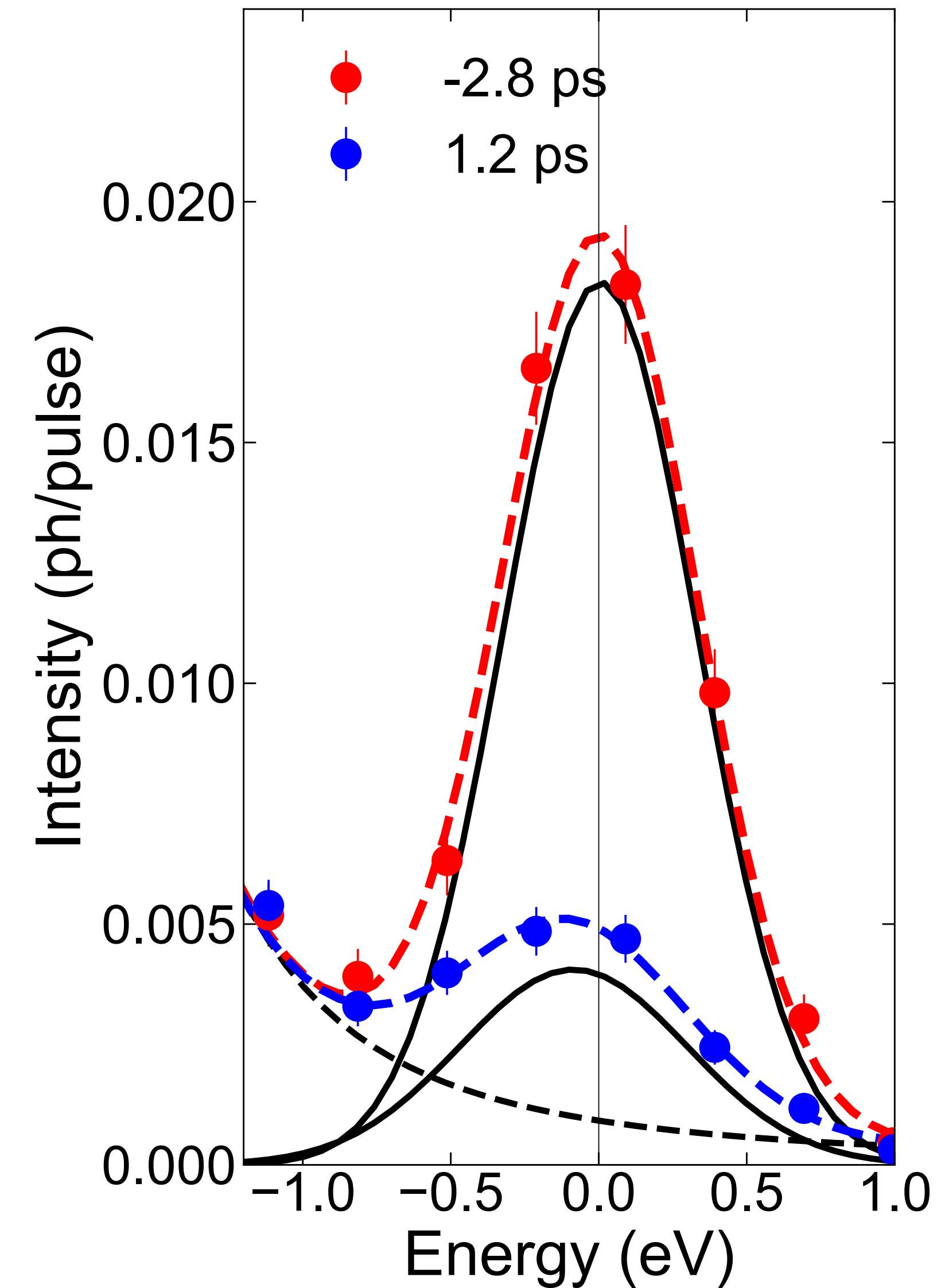
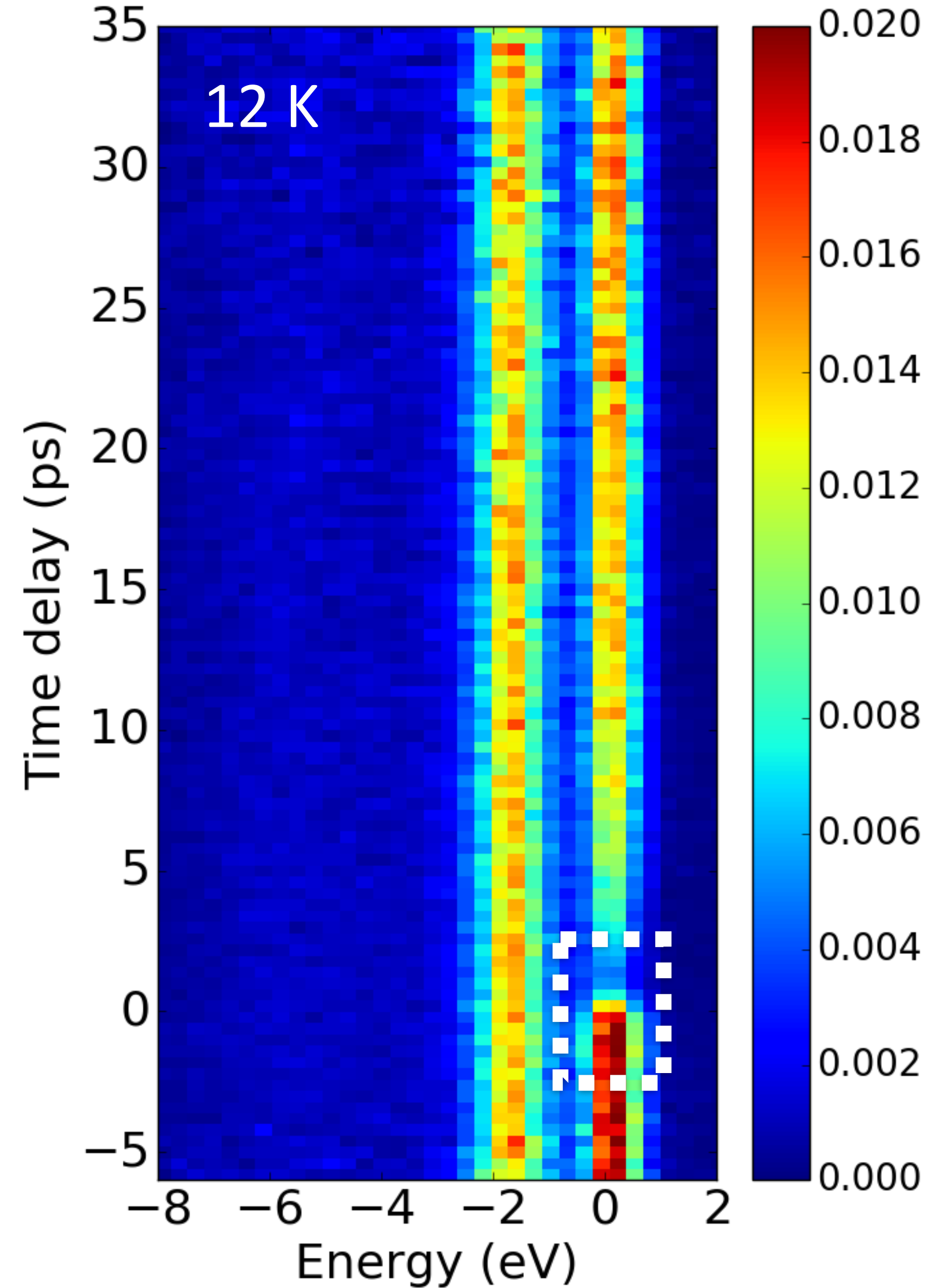


- No inelastic time evolution
- Charge order recovers in 3.8 ps
- Large ΔE_{res} enables dense delays

MM et al. Science Advances **5**, eaax3346 (2019)

MM et al. PRB **100**, 205125 (2019)

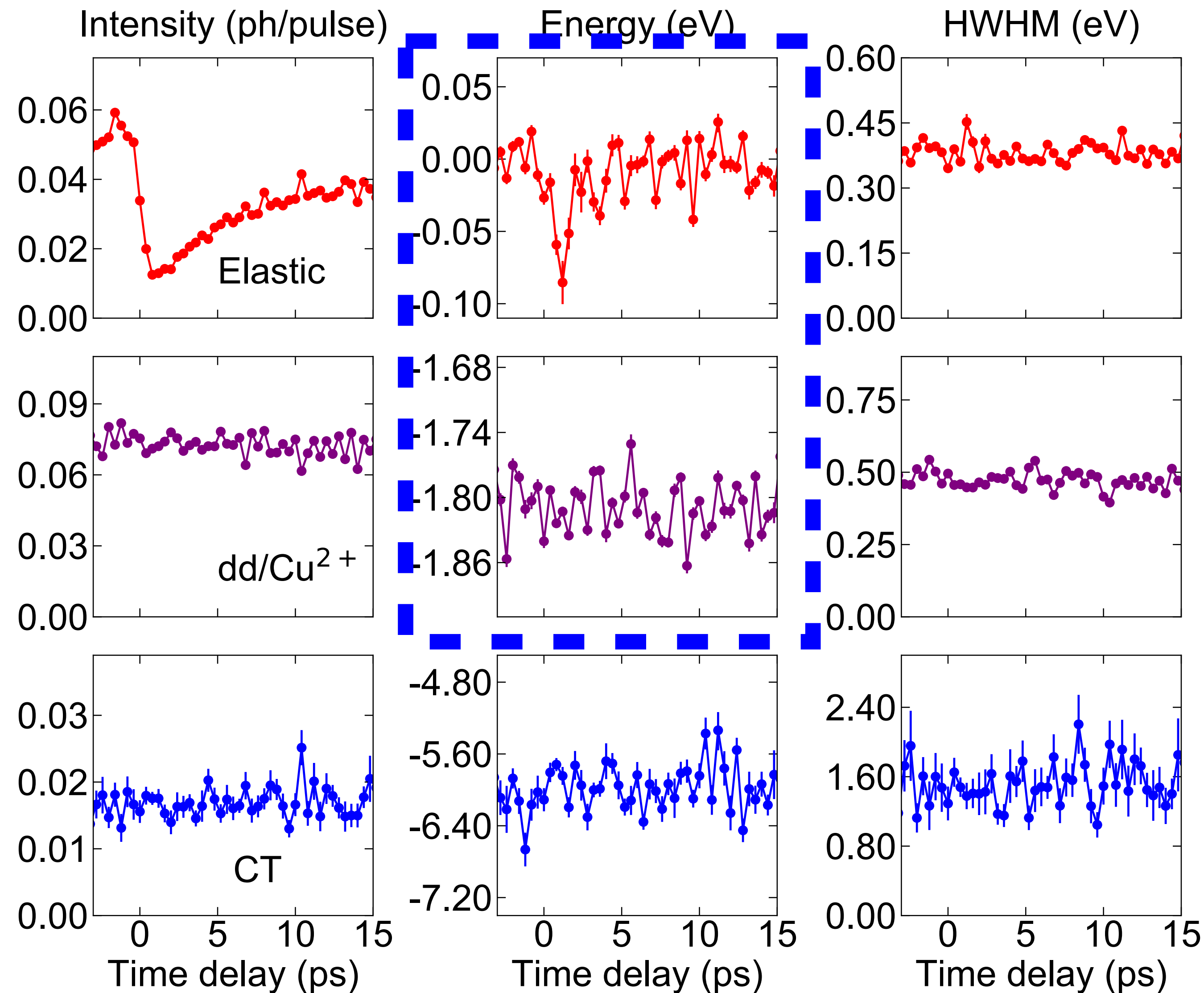
Time-dependent RIXS spectrum



MM et al. Science Advances 5, eaax3346 (2019)

MM et al. PRB 100, 205125 (2019)

The order parameter line redshifts

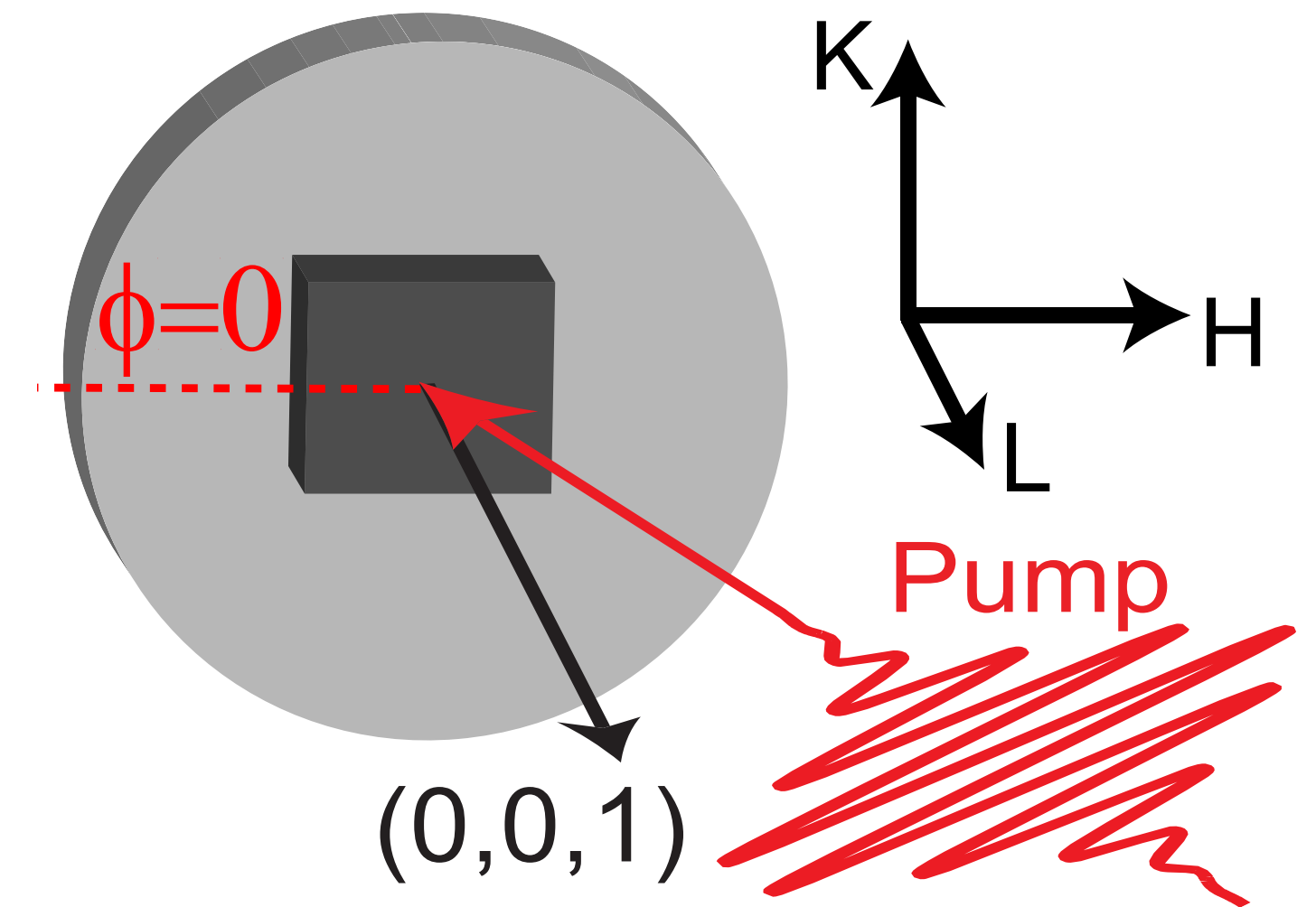
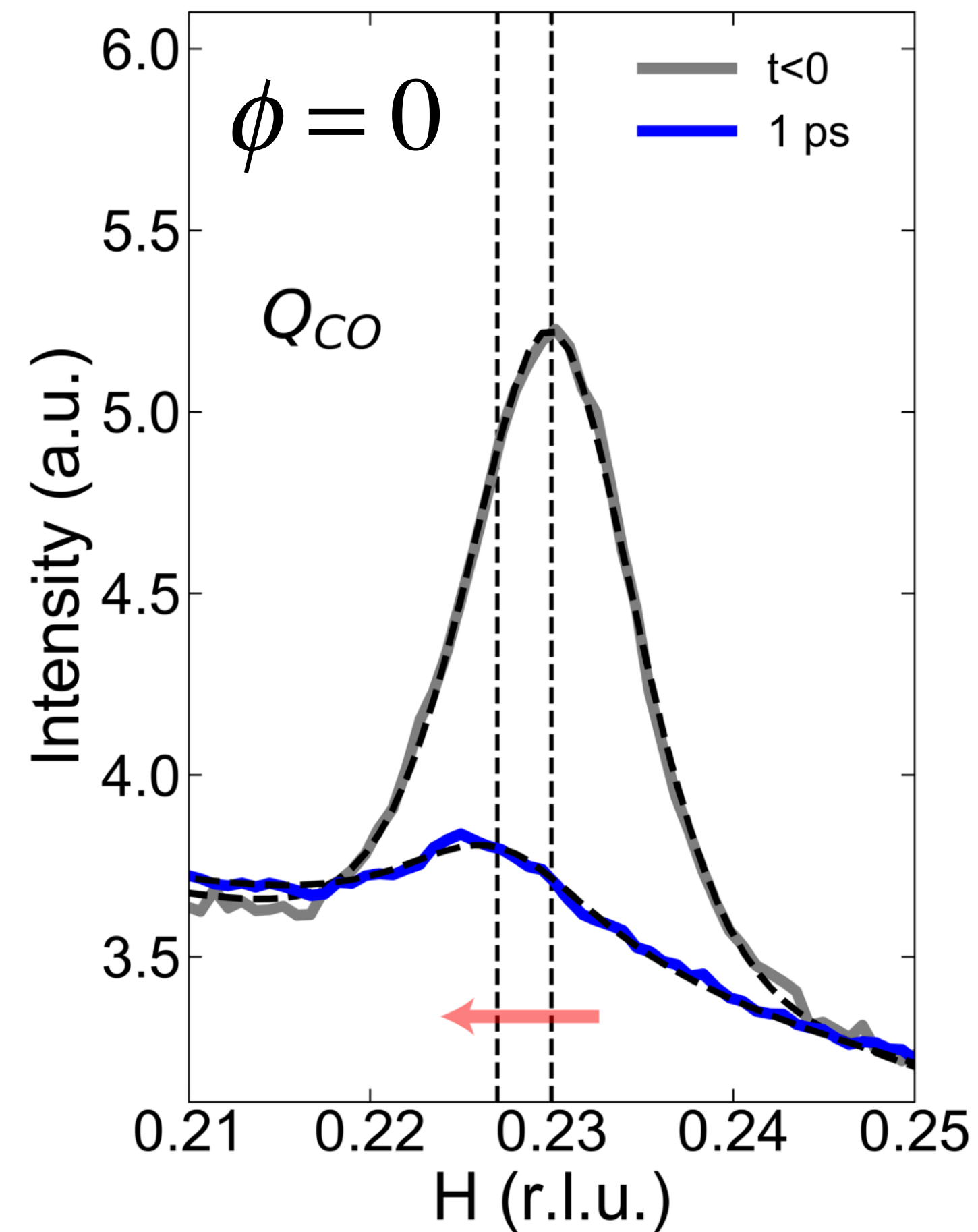


Two possibilities:

- Elastic line becomes inelastic
- Excitations below 0.3 eV

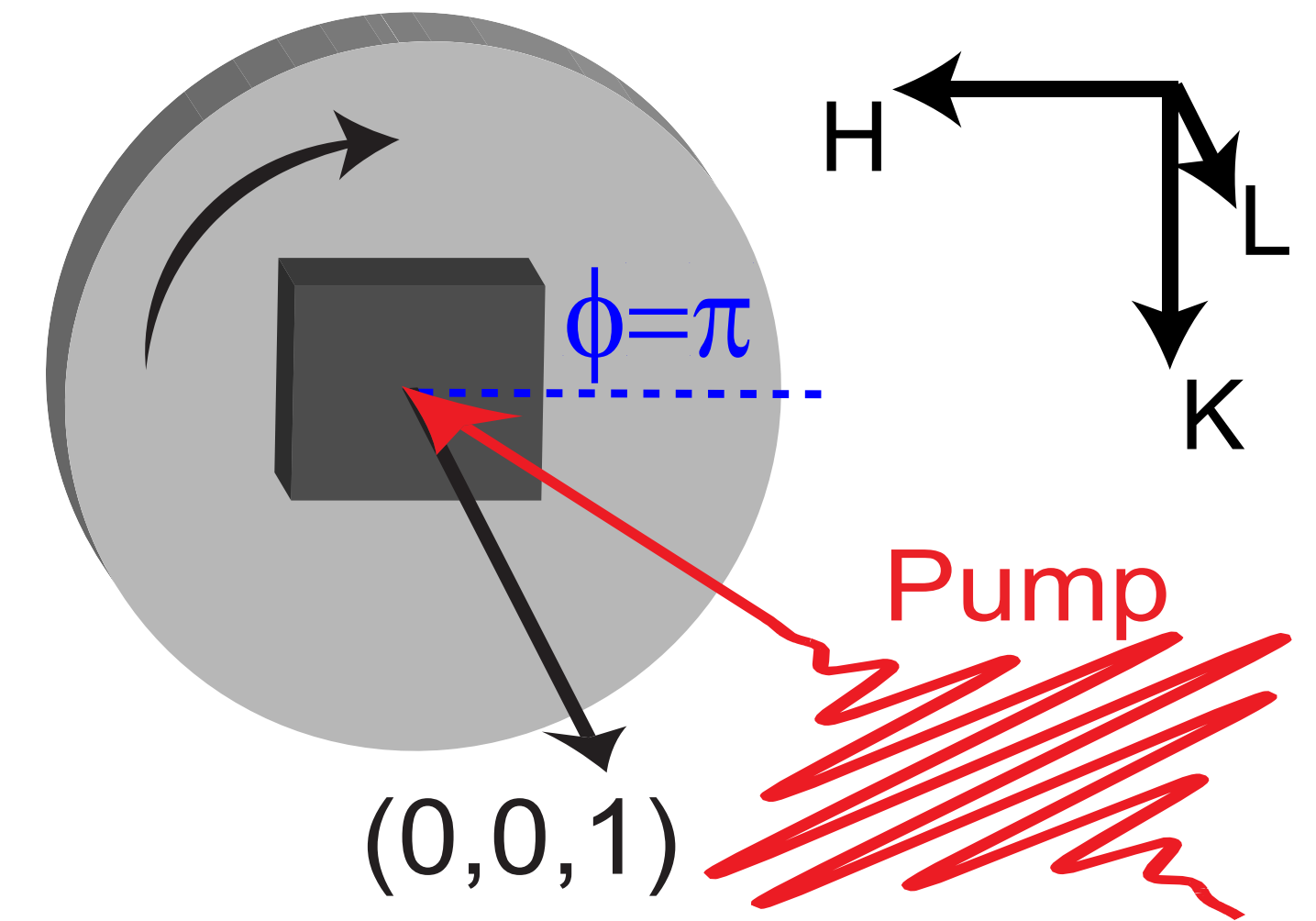
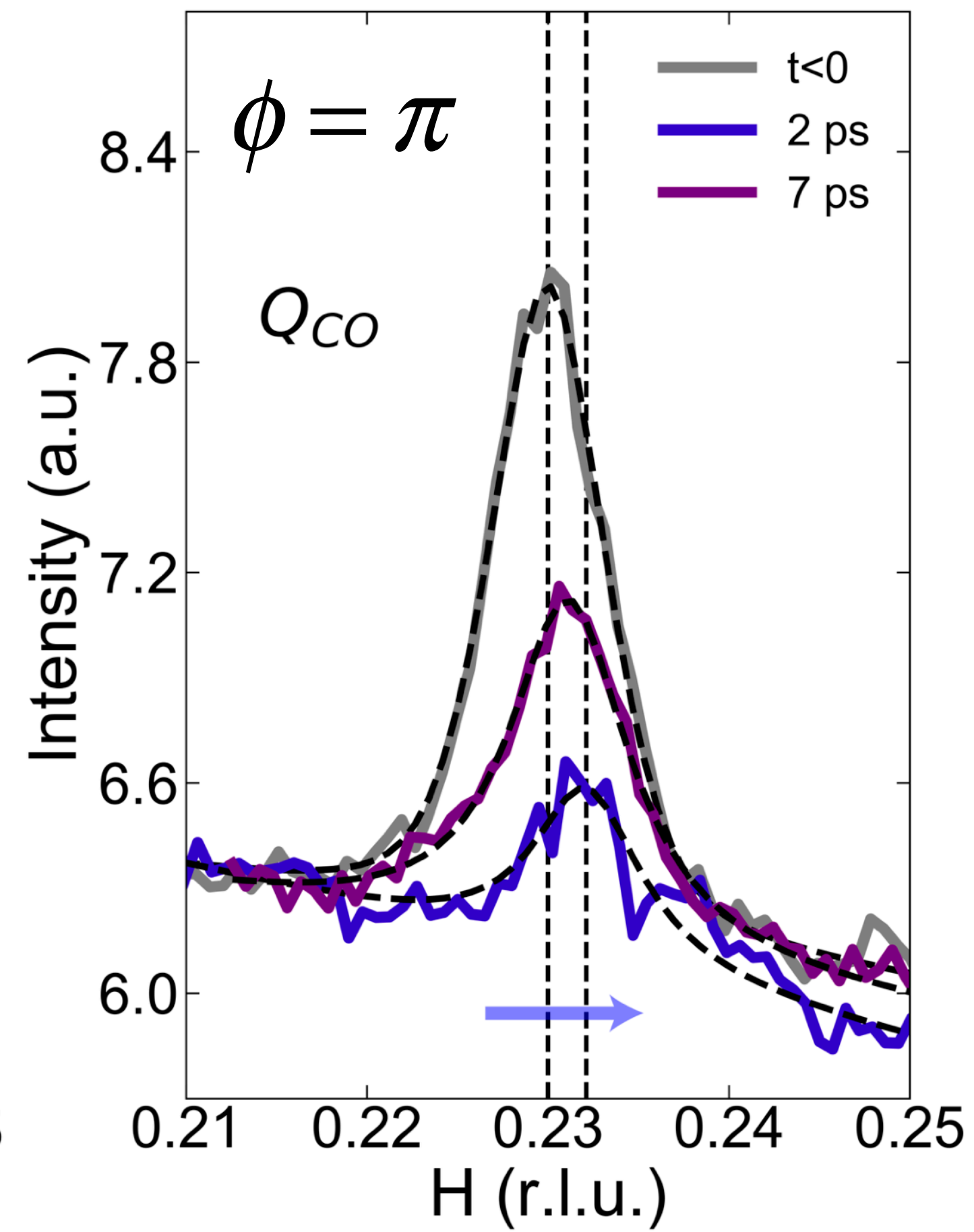
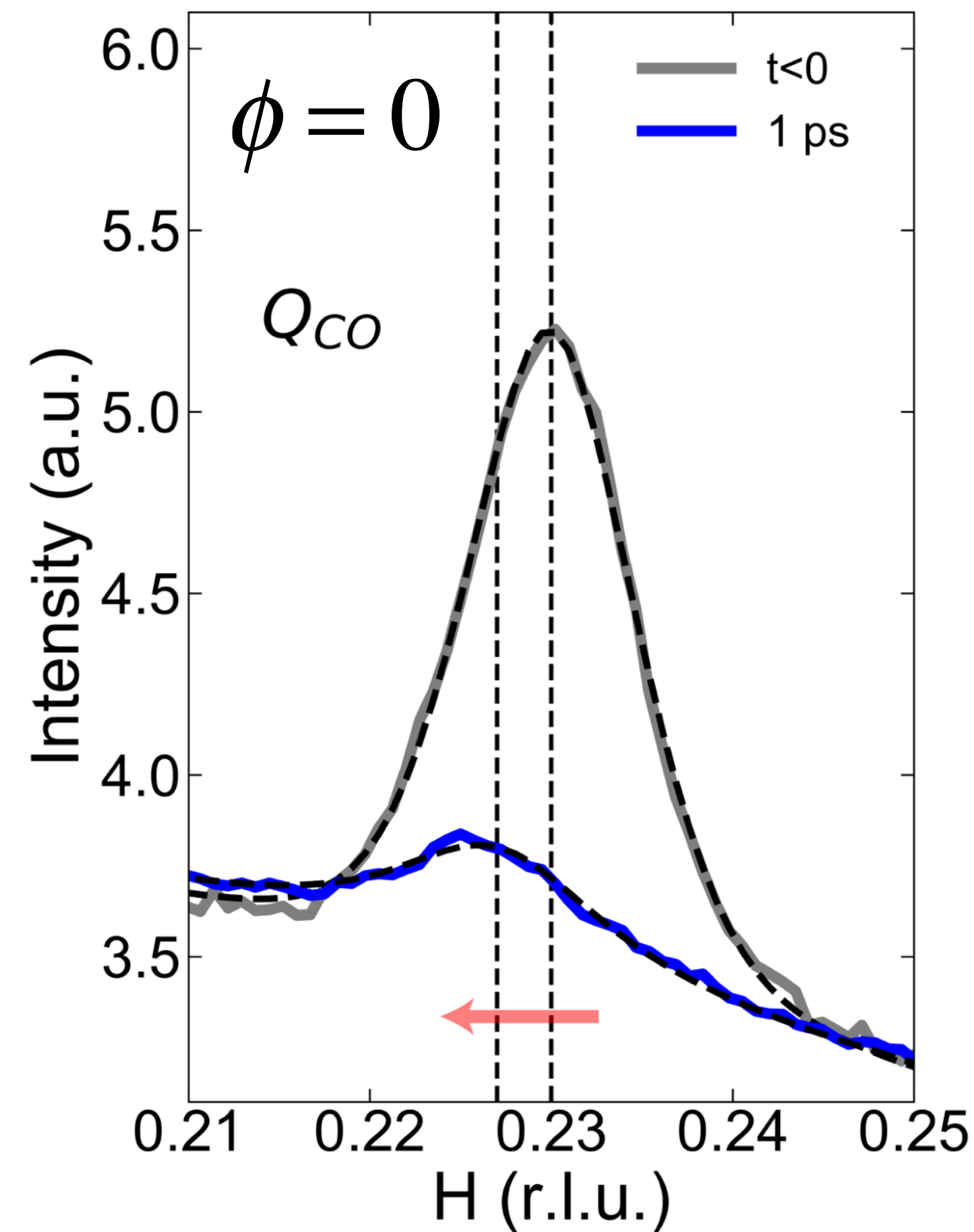
What about momentum?

Pump-induced momentum shift of the order parameter



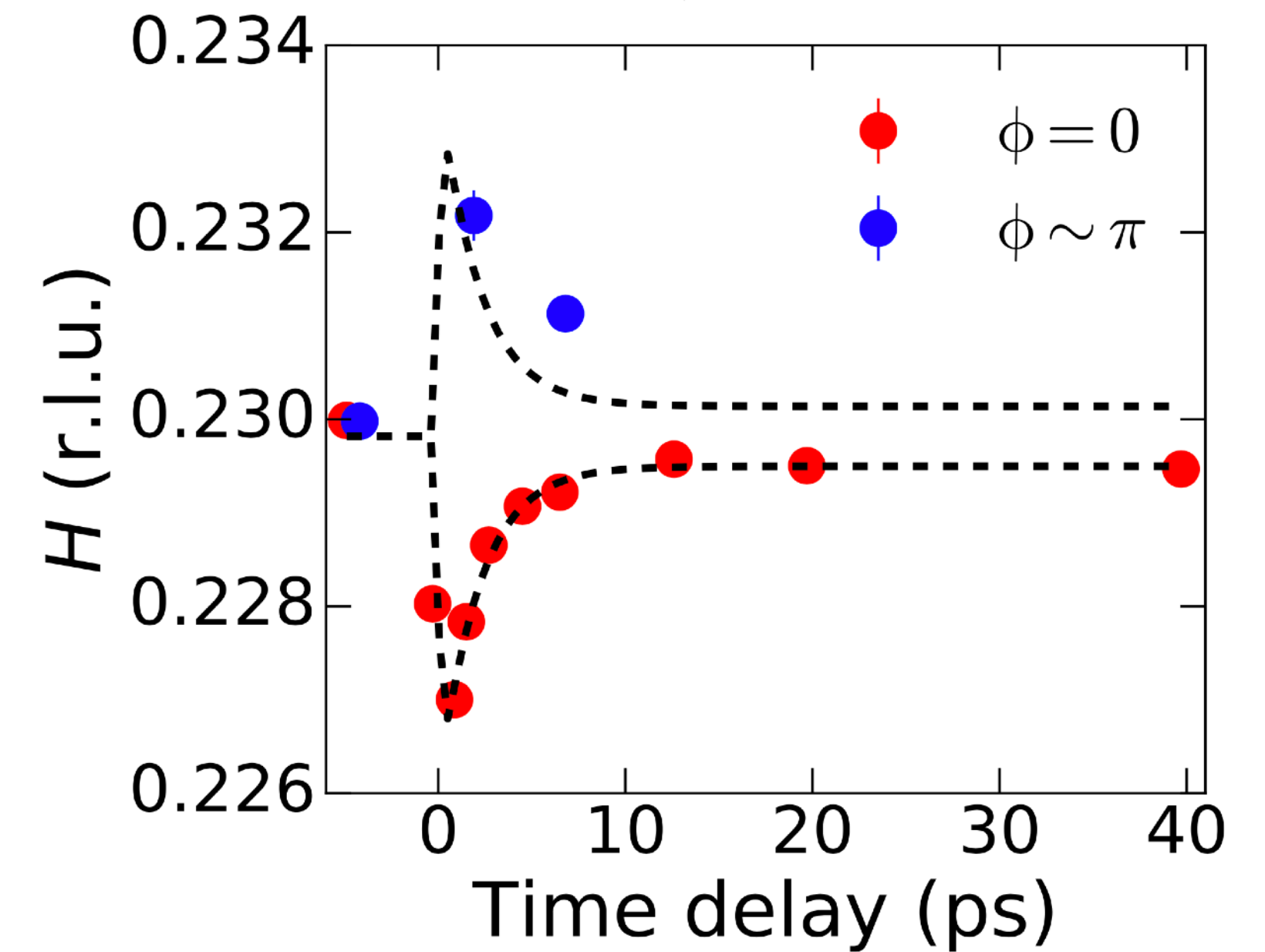
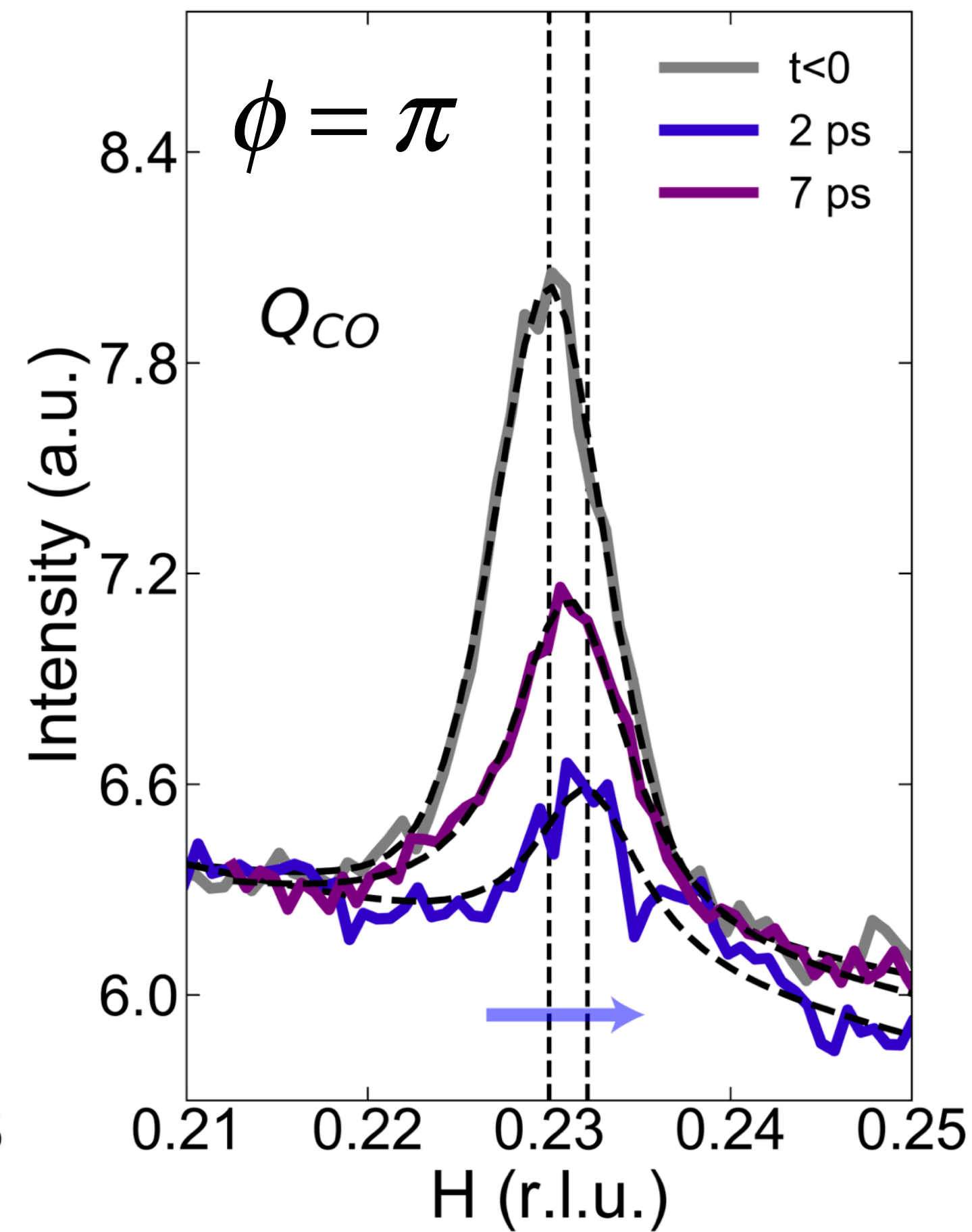
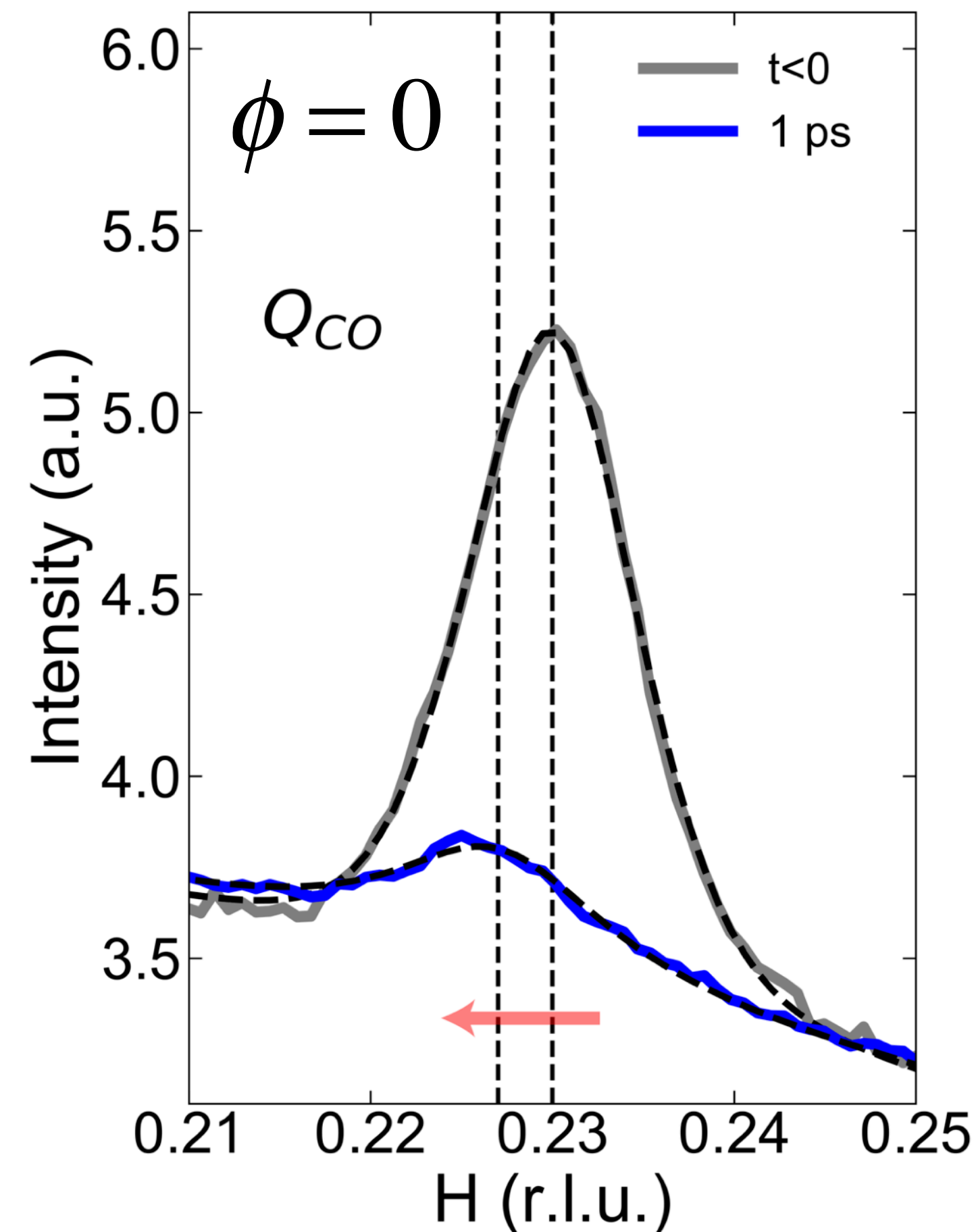
- Max shift 0.003 r.l.u.

Pump-induced momentum shift of the order parameter



- Max shift 0.003 r.l.u.
- Direction follows pump

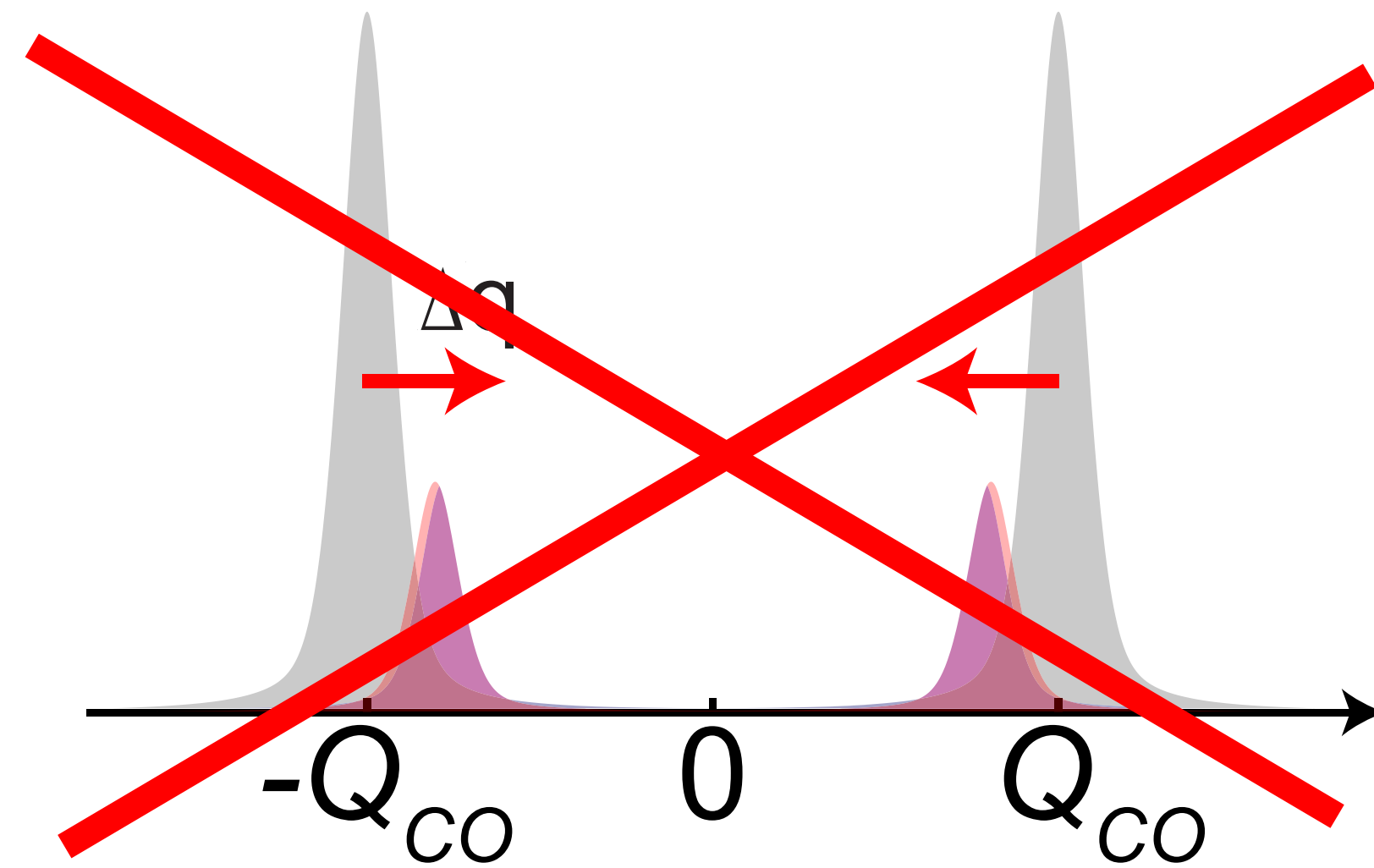
Pump-induced momentum shift of the order parameter



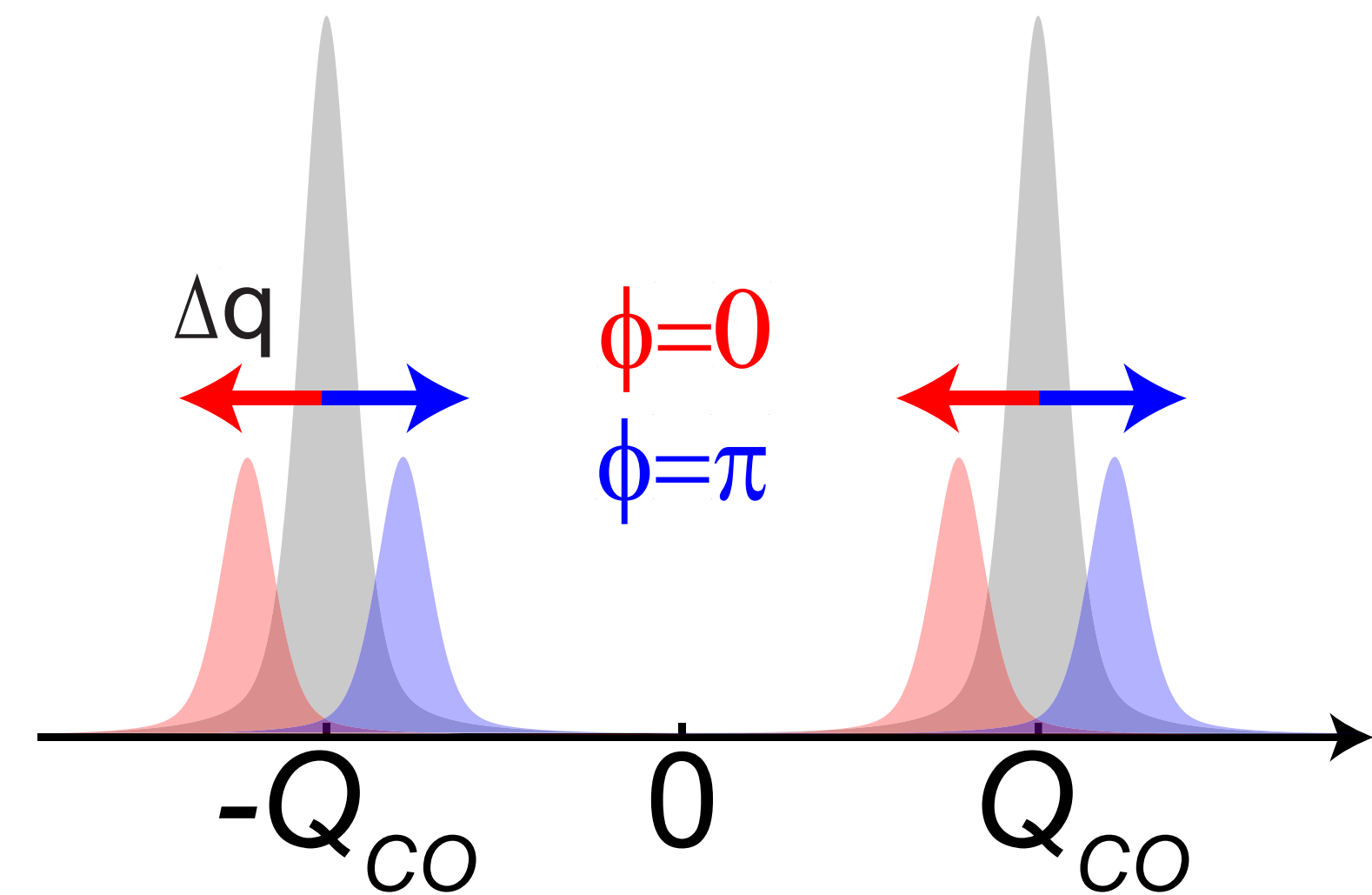
- Max shift 0.003 r.l.u.
- Direction follows pump

Charge order undergoes collective dynamics
beyond melting

Pump-induced momentum shift of the order parameter



CO deformation



Light-induced CO sliding

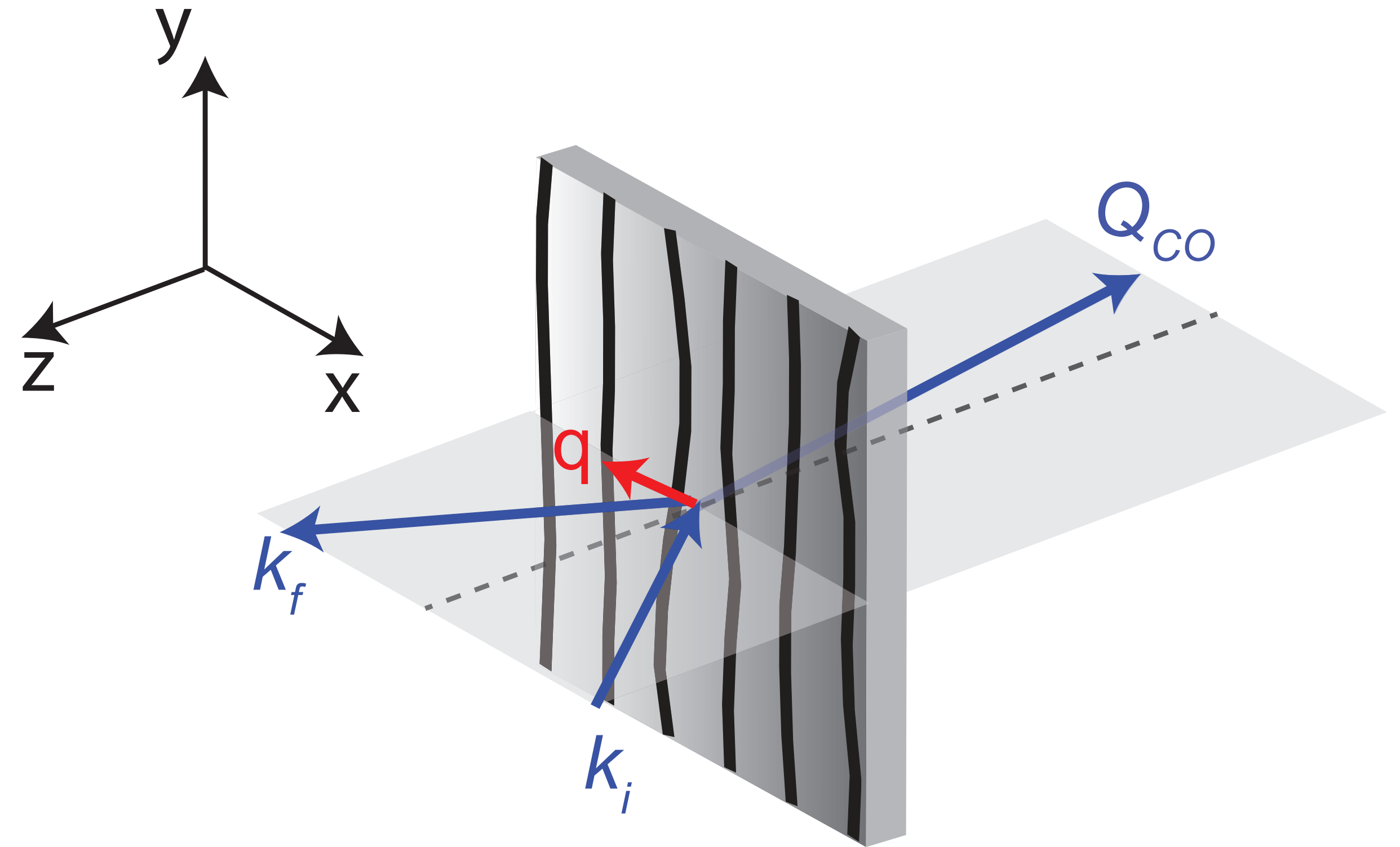
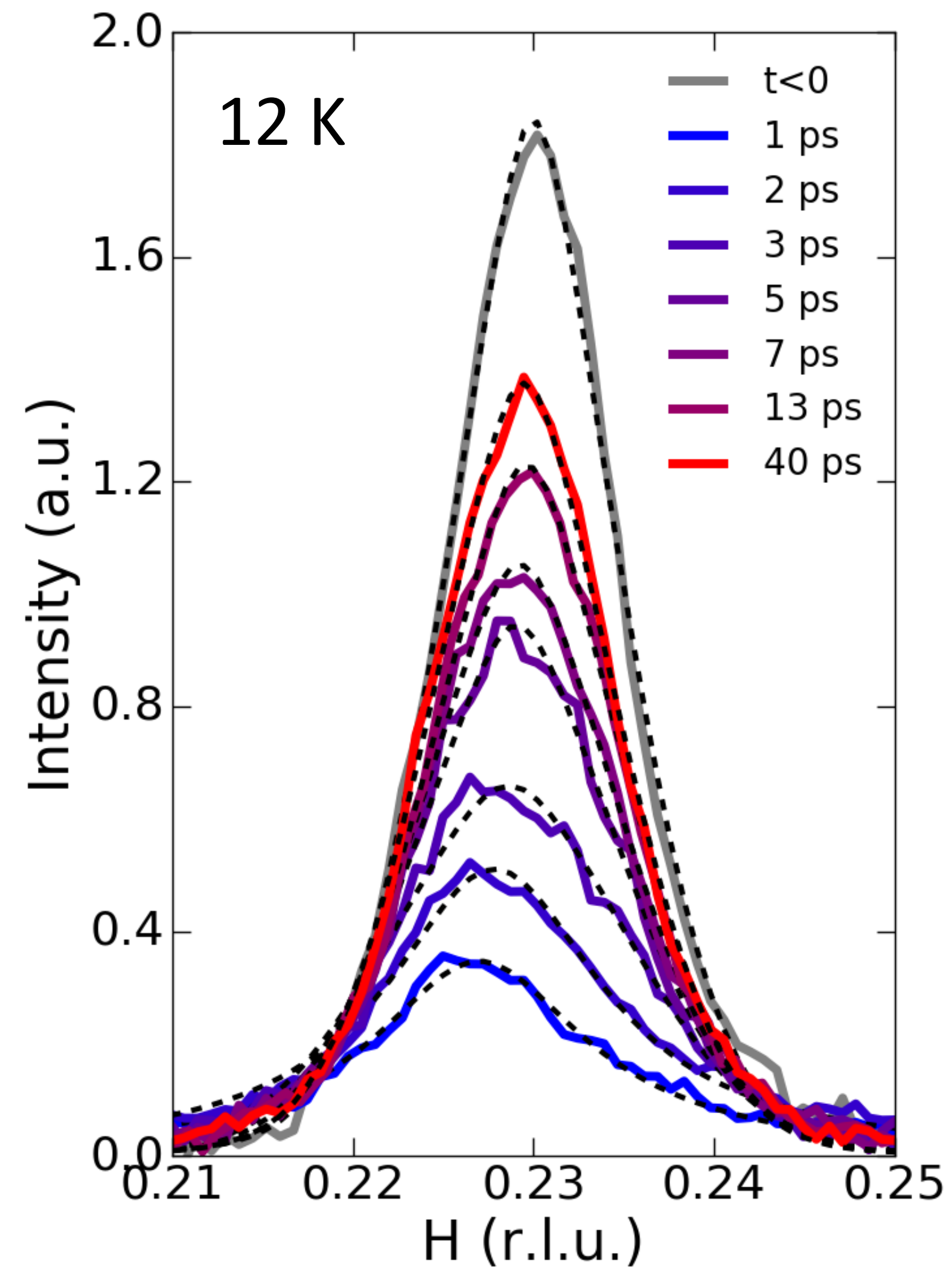
$$(Q_{CO}, \omega = 0) \rightarrow (Q_{CO} \pm \Delta q, \mp v \Delta q)$$

Charge order exhibits coherent dynamics in ω and q , possibly sliding

Caveat: if sliding, ω shift is not $v\Delta q$.

Ultralow energy spectroscopy of the charge order recovery

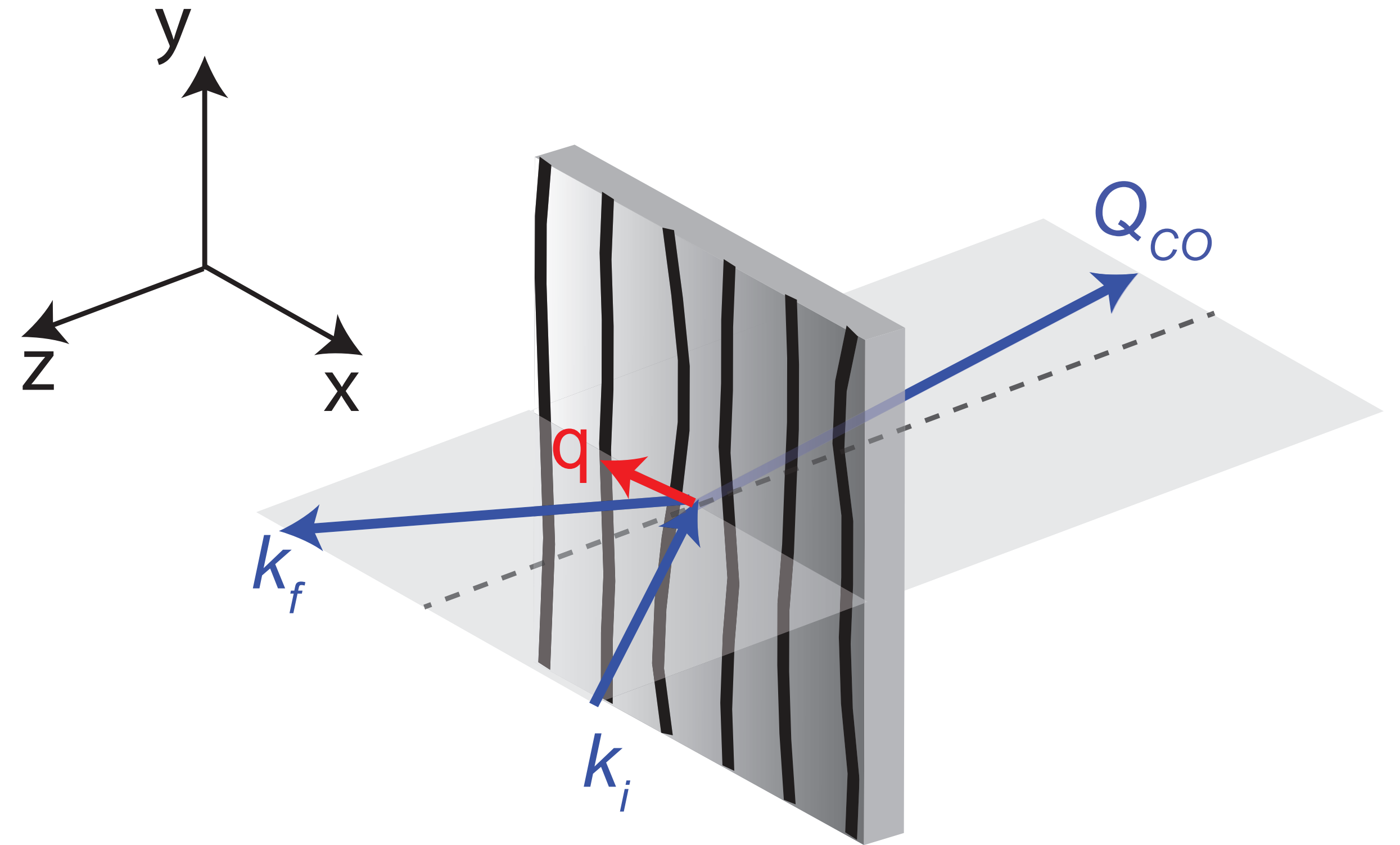
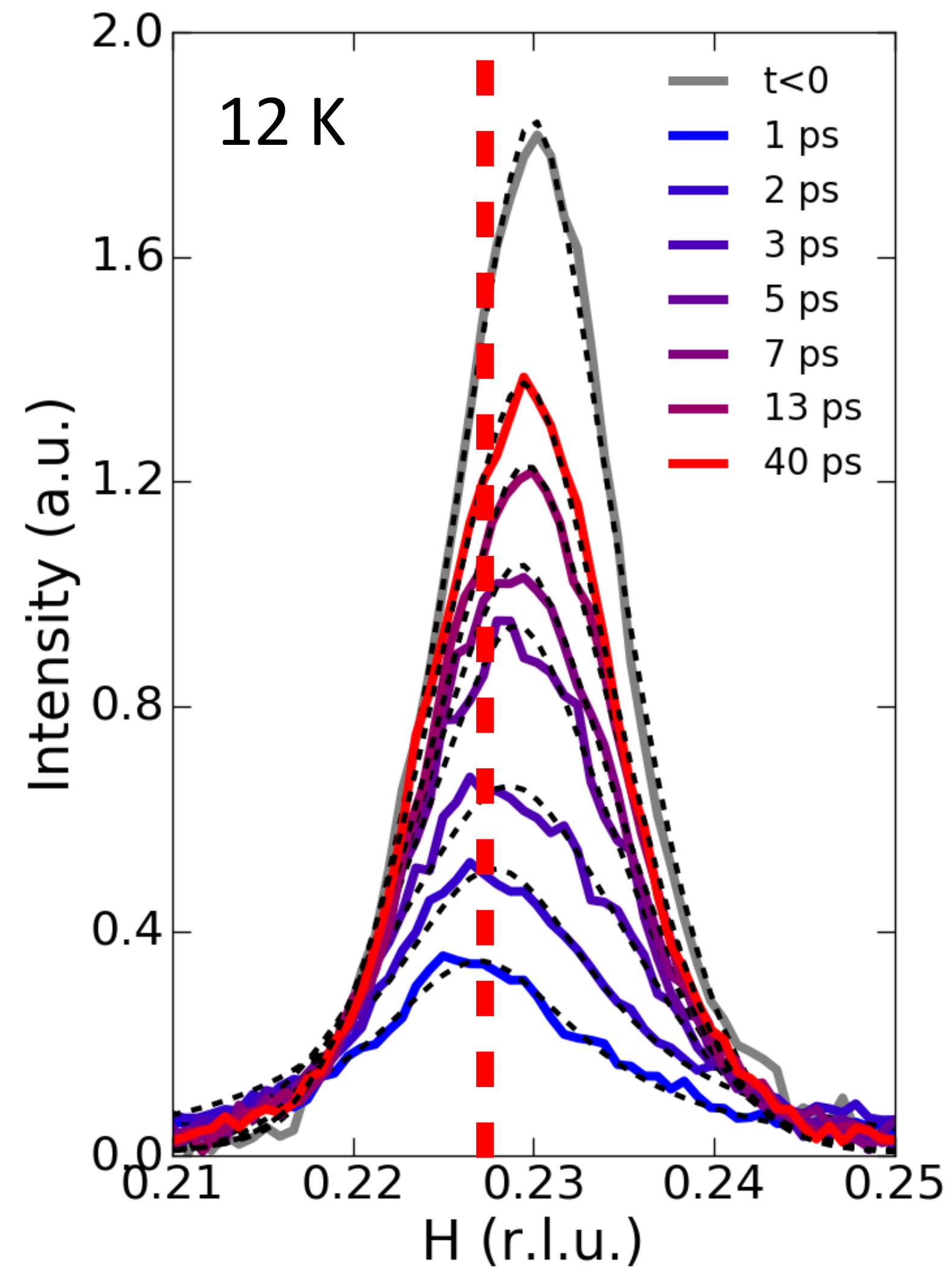
Dynamics of the charge order recovery



MM et al. Science Advances **5**, eaax3346 (2019)

MM et al. PRB **100**, 205125 (2019)

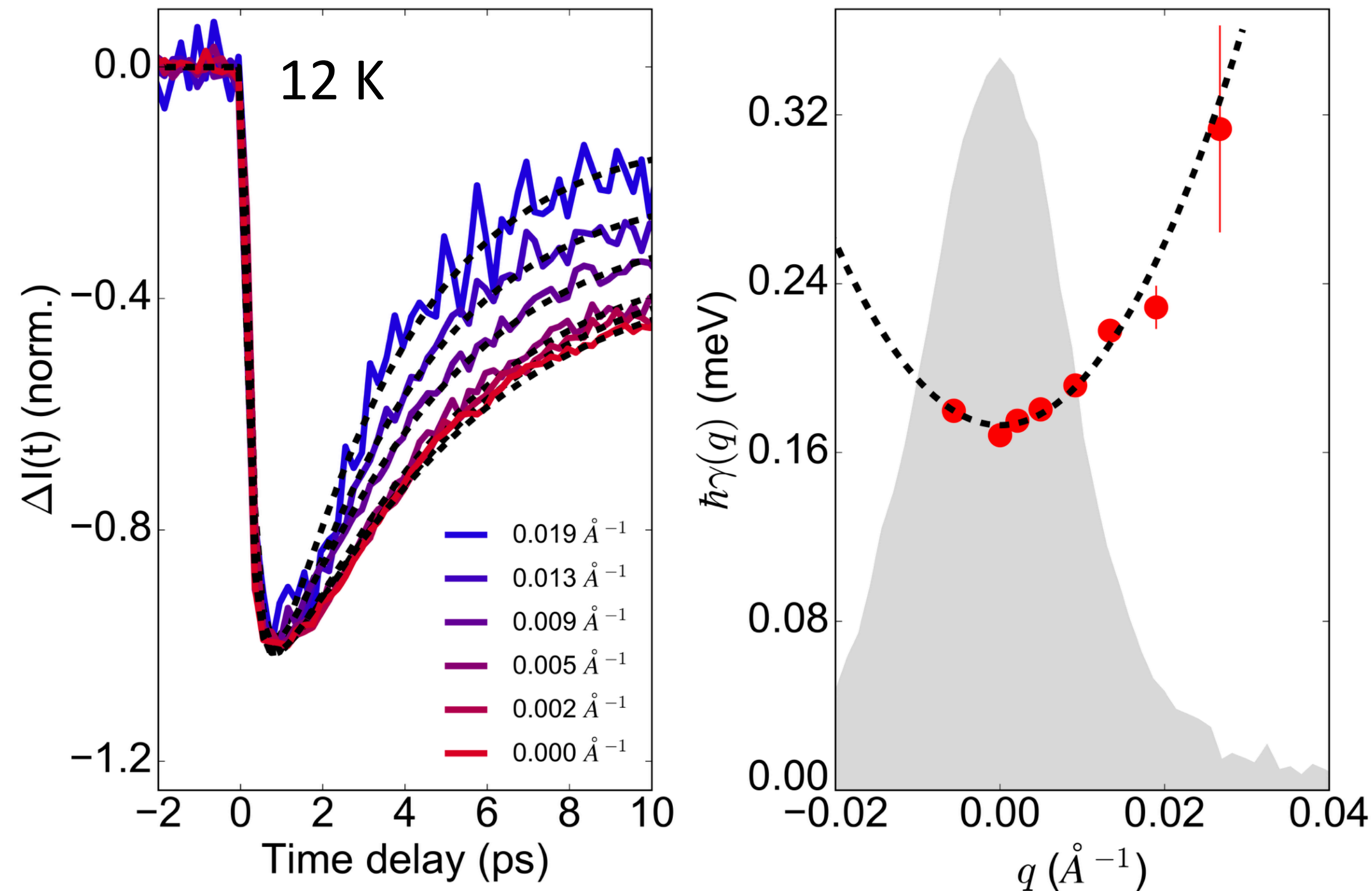
Dynamics of the charge order recovery



MM et al. Science Advances **5**, eaax3346 (2019)

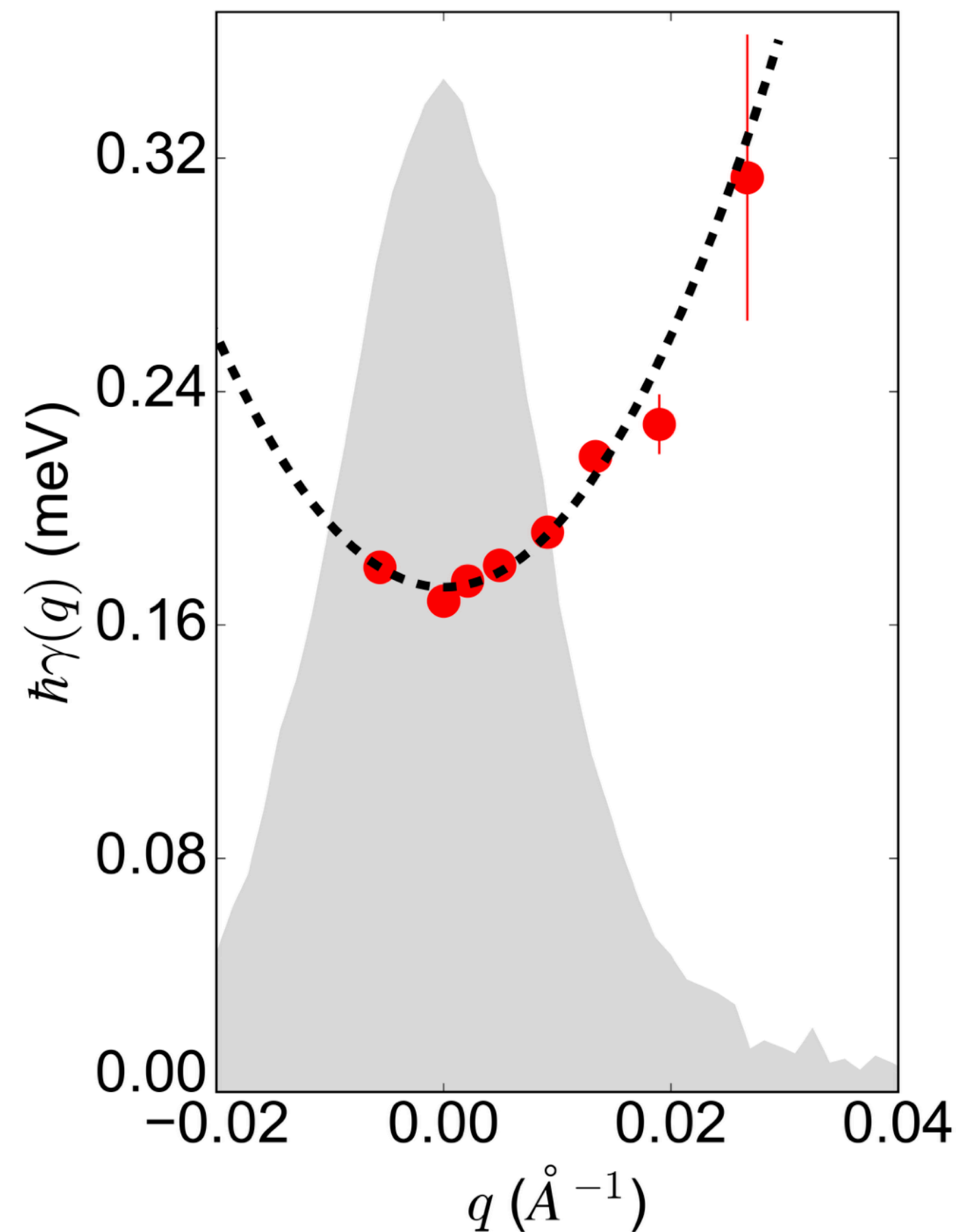
MM et al. PRB **100**, 205125 (2019)

Evidence of ultrafast charge order diffusion



- Lack of amplitude mode oscillations (unlike conventional CDW)
- Amplitude relaxation rate $\gamma(q) \propto q^2$

Evidence of ultrafast charge order diffusion



$$\frac{\delta F}{\delta \varphi} = -\frac{1}{\Gamma} \frac{\partial \varphi}{\partial t}$$

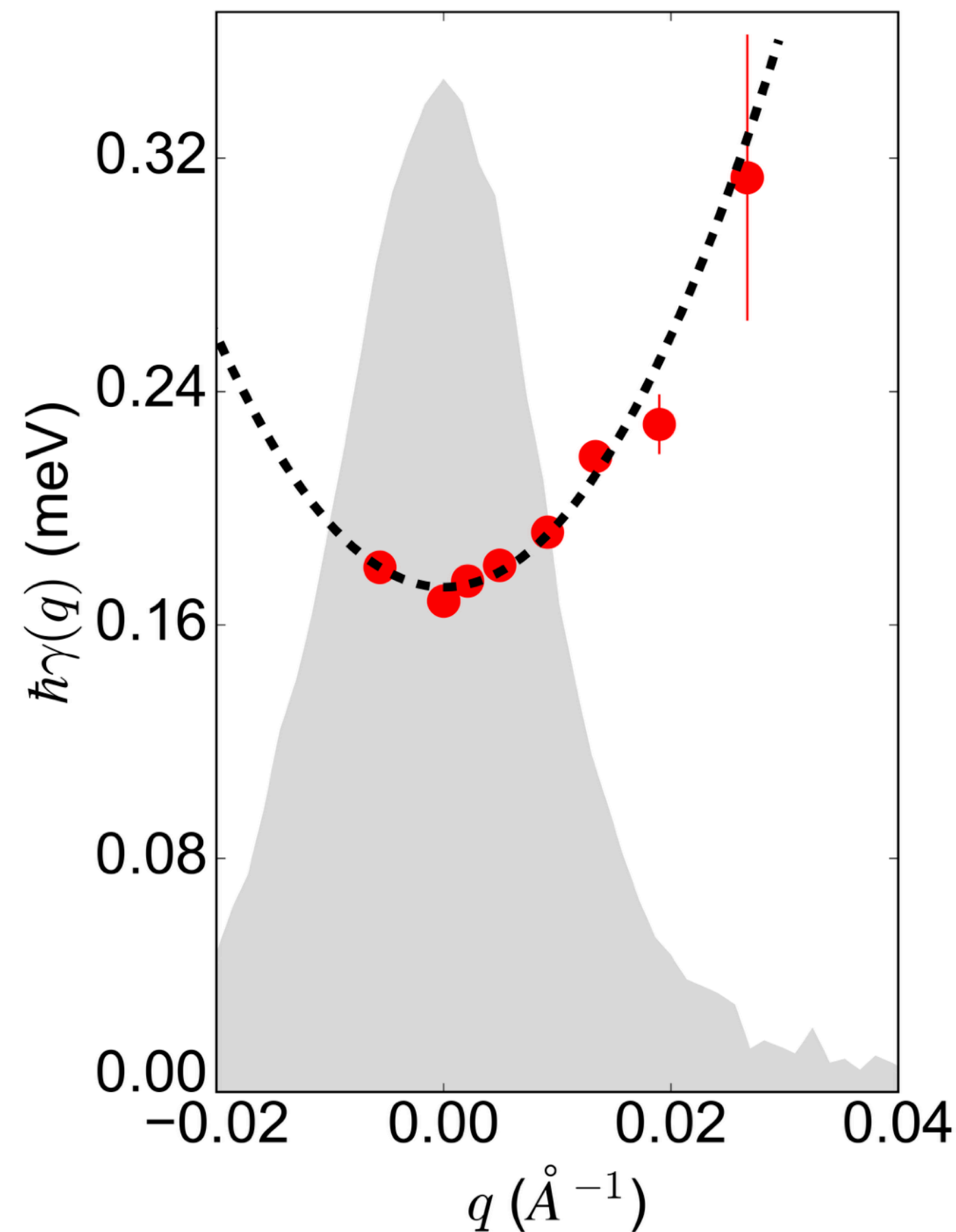
$$F[\varphi] = a\varphi^2 + b\varphi^4 + c(\nabla \varphi)^2$$

$$\frac{\partial \varphi}{\partial t} - \underbrace{\frac{1}{\tau_0} \varphi}_{\text{Relaxation}} - \underbrace{D \nabla^2 \varphi}_{\text{Diffusion}} = 0$$

Transverse charge order diffusion at energy $O(k_B T_c)$

$$i\hbar\gamma(q) = \hbar(\gamma_0 + Dq^2) \sim 2 K \quad D \sim 0.03 \text{ cm}^2 / \text{s}$$

Evidence of ultrafast charge order diffusion



Quasiparticle gas

$$D_{QP} \sim 20 \text{ cm}^2 / \text{s}$$

Gedik et al. Science **300**, 1410-1412 (2003)

$$\frac{\delta F}{\delta \varphi} = -\frac{1}{\Gamma} \frac{\partial \varphi}{\partial t}$$

$$F[\varphi] = a\varphi^2 + b\varphi^4 + c(\nabla \varphi)^2$$

$$\frac{\partial \varphi}{\partial t} - \underbrace{\frac{1}{\tau_0} \varphi}_{\text{Relaxation}} - \underbrace{D \nabla^2 \varphi}_{\text{Diffusion}} = 0$$

Charge order

$$D \sim 0.03 \text{ cm}^2 / \text{s}$$

MM et al. Science Advances **5**, eaax3346 (2019)

MM et al. PRB **100**, 205125 (2019)

The future of trRIXS

Towards quantitative trRIXS on light-induced superconductivity

Future experiments must:

- Increase count rates

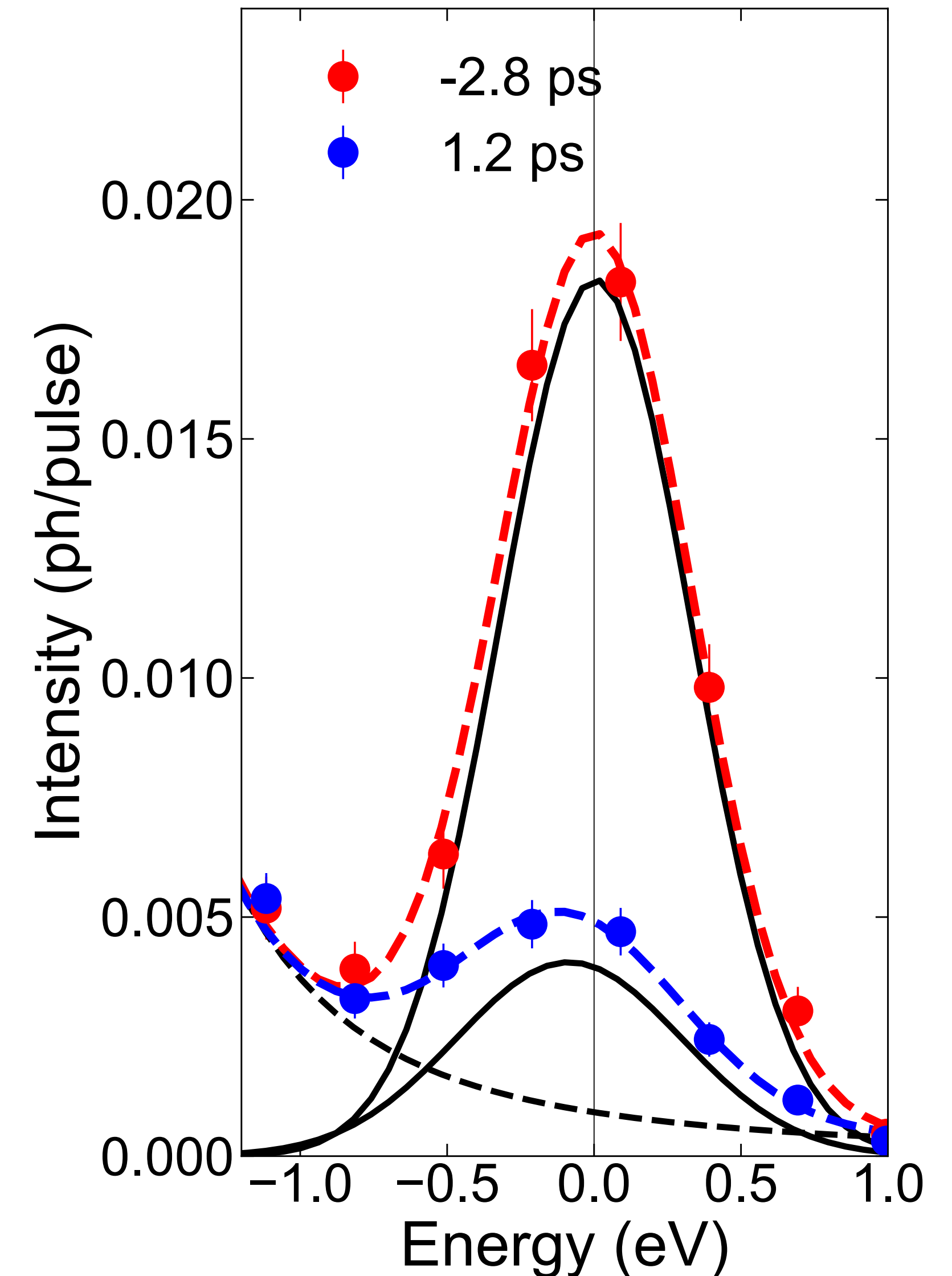
LCLS @ 120 Hz (today)

10^{10} ph/pulse at the sample yield 0.02

ph/pulse on LBCO charge order

LCLS II: 10 KHz or more

EuXFEL: 2 KHz or more



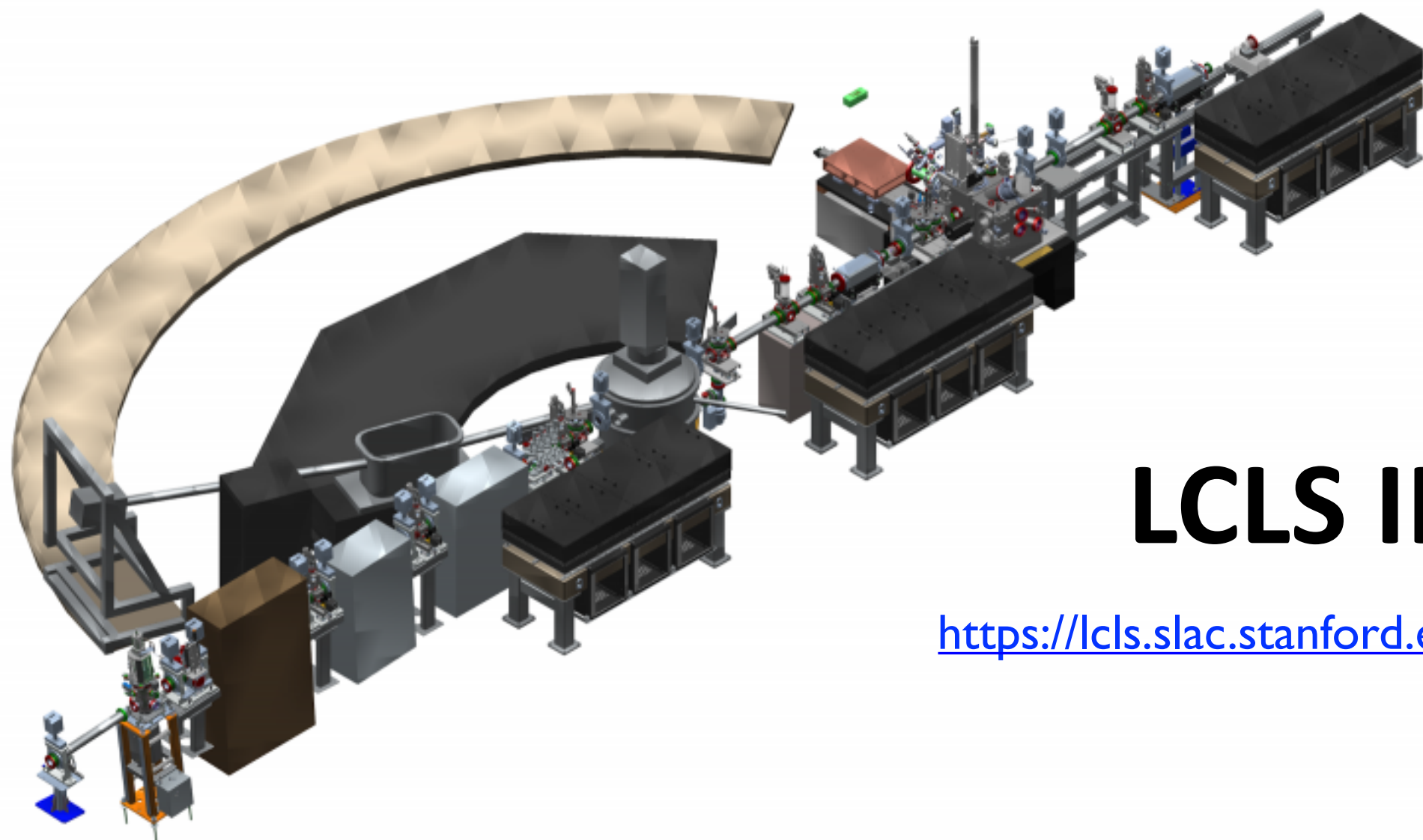
Towards quantitative trRIXS on light-induced superconductivity

Future experiments must:

- Increase count rates
- Improve 20x energy resolution (30 meV)

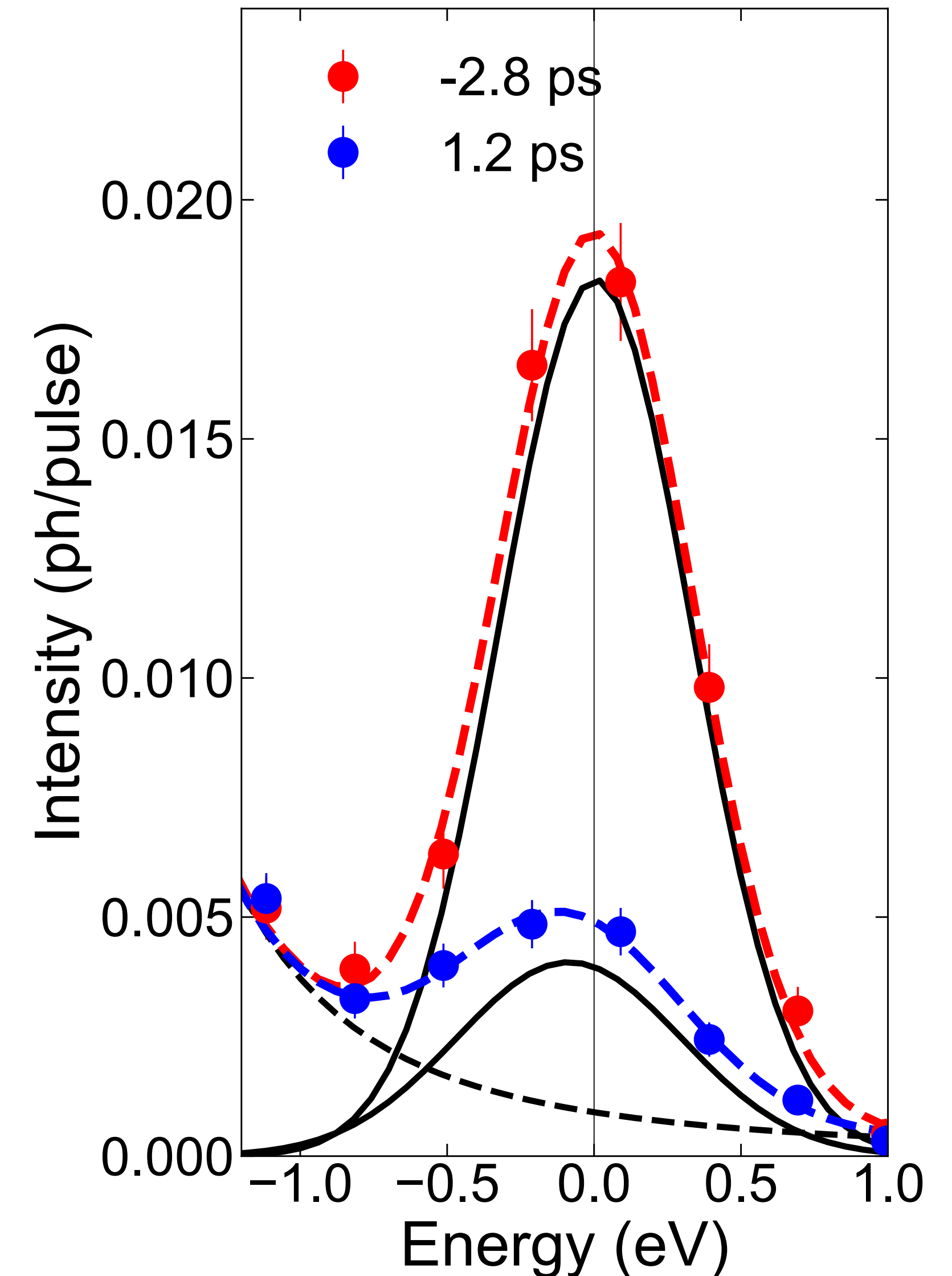
LCLS II: RP up to 50000 (NEH2.2, qRIXS)

EuXFEL: RP up to 25000 (hRIXS consortium)



LCLS II qRIXS

<https://lcls.slac.stanford.edu/instruments/neh-2-2>



To be continued...