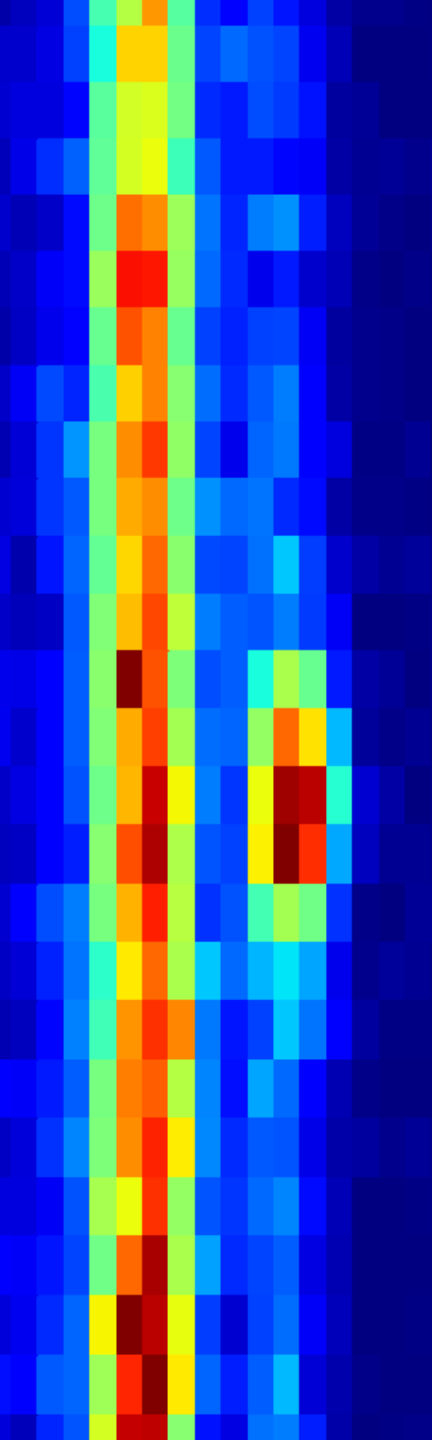




Ultrafast diffusive dynamics in a charge-ordered cuprate superconductor

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U.S. DEPARTMENT OF
ENERGY

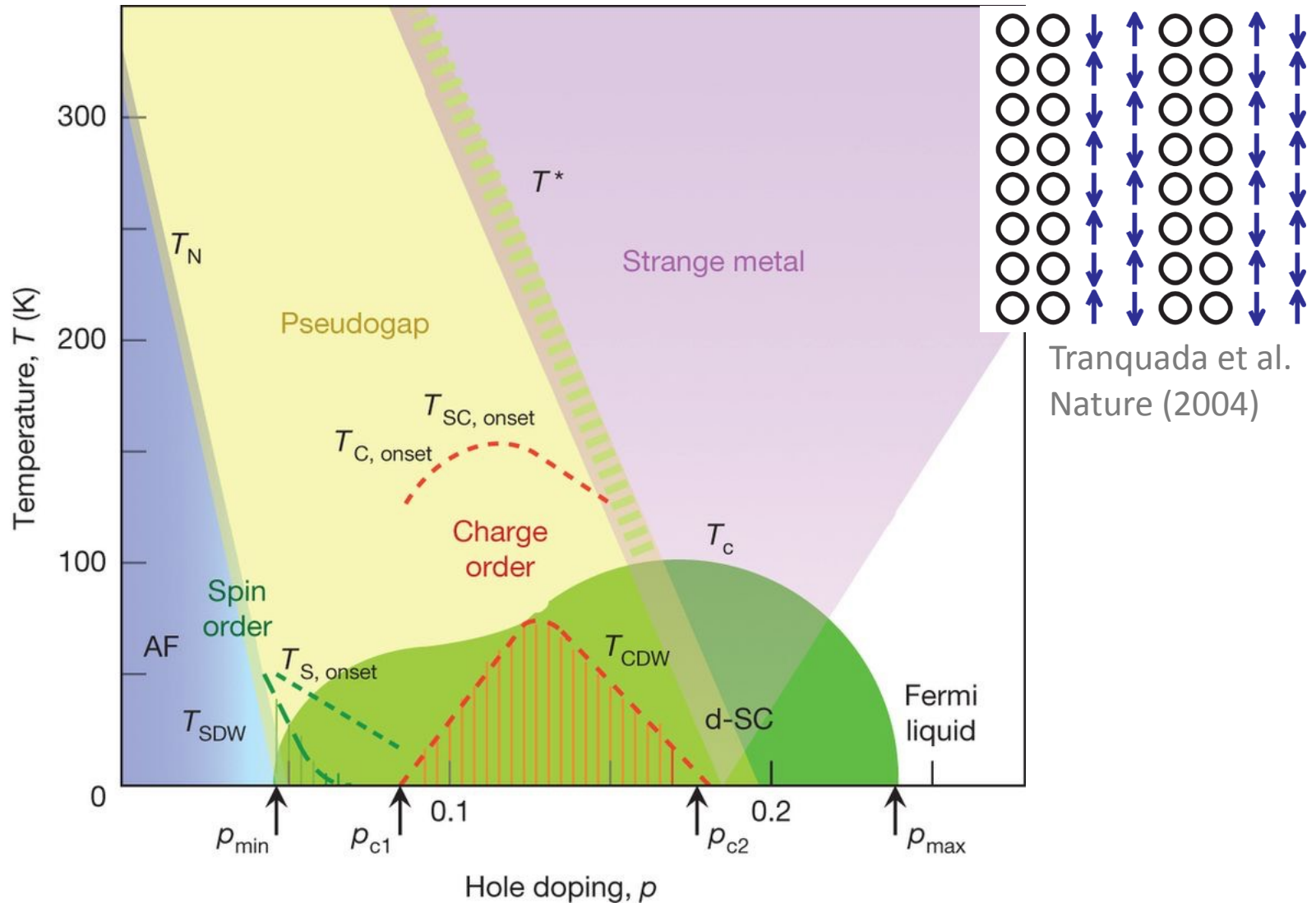
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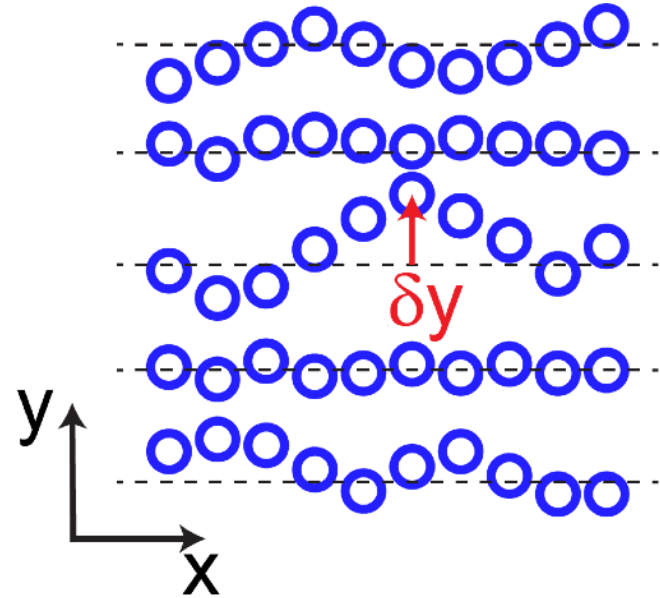
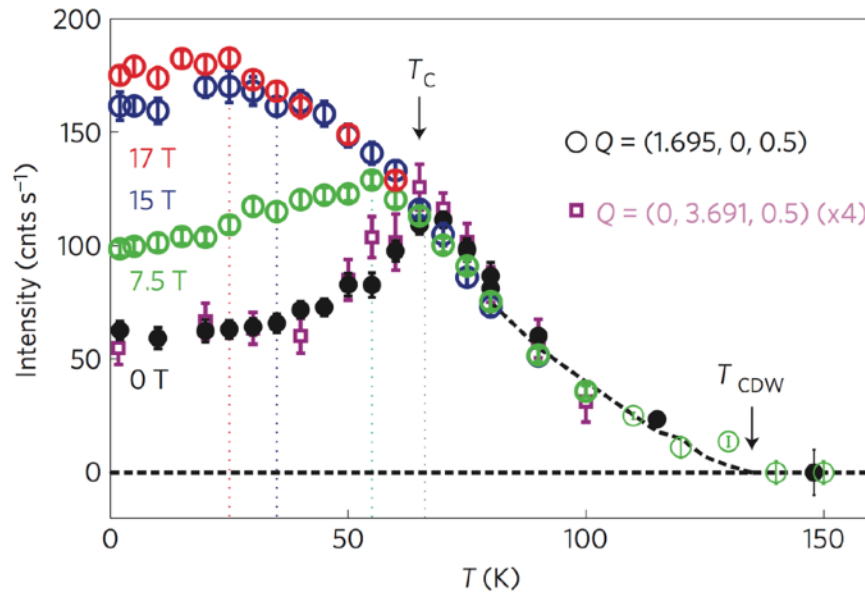


Alexander von Humboldt
Stiftung / Foundation

Charge order in the cuprates



Superconductivity and charge order

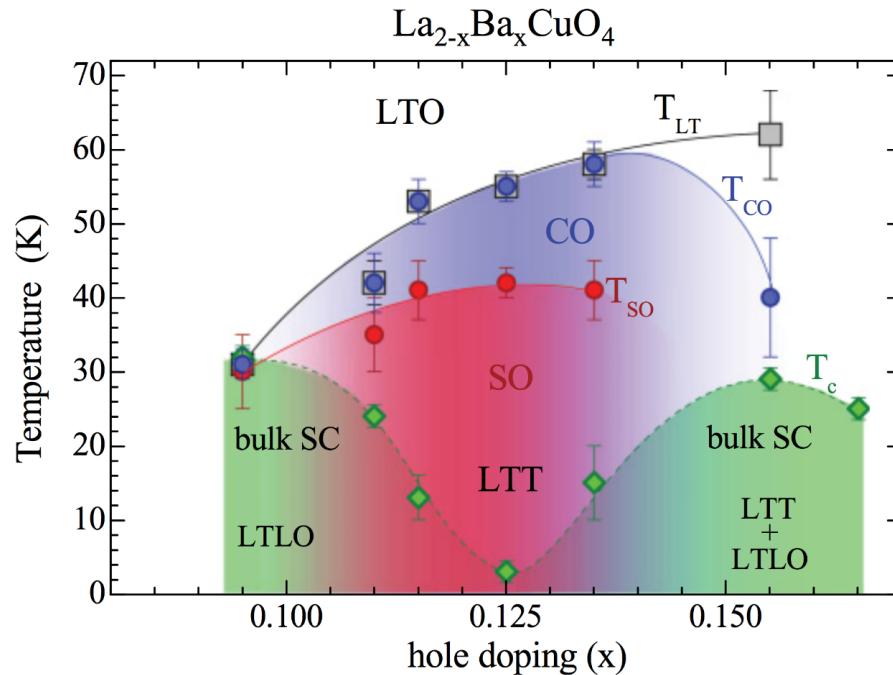


e.g. Chiang et al. Nature Phys. 8, 871–876 (2012),
Gerber et al. Science 350, 949-952 (2015), etc.

e.g. Kivelson et al. Nature 393, 550 (1998)
C. Castellani PRL 75, 4650 (1995)

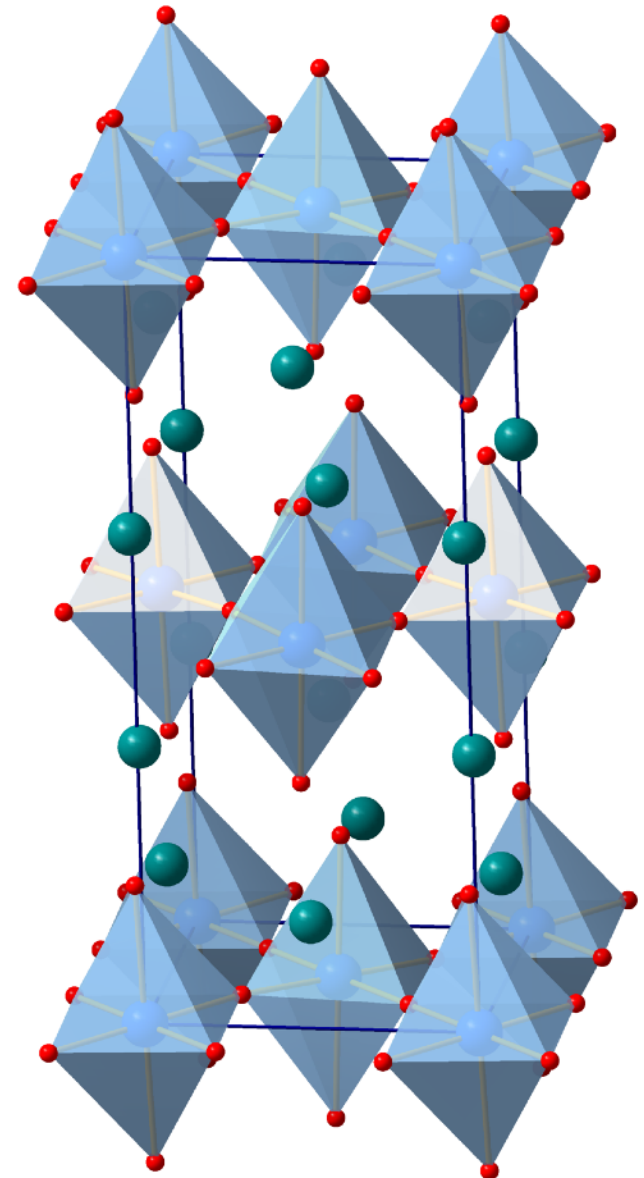
- Static CO competes with SC for states at E_F
- CO fluctuations may cooperate or intertwine with SC

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

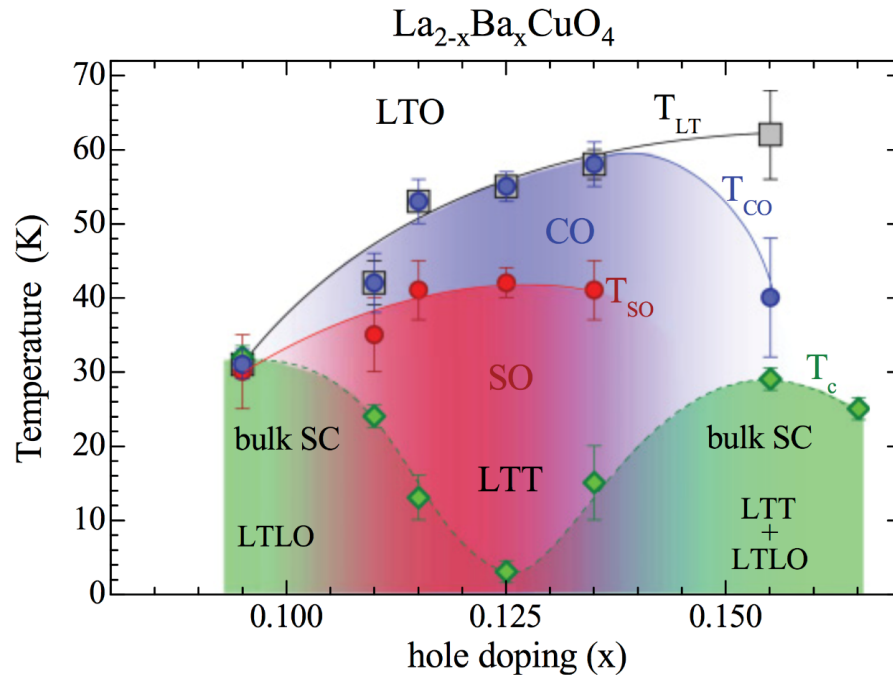


Hücker et al. PRB 83, 104506 (2011)

- Striped single layer cuprate
- Strong 1/8 anomaly

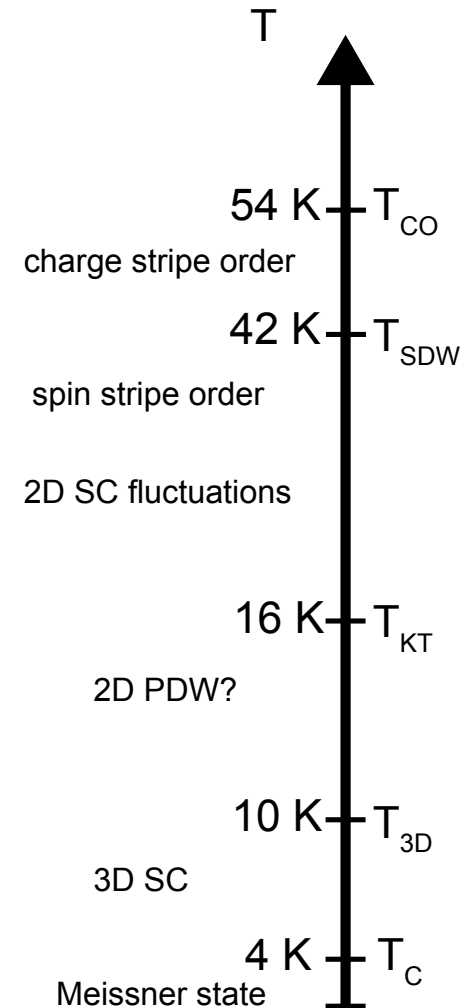


Charge order in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$



Hücker et al. PRB 83, 104506 (2011)

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

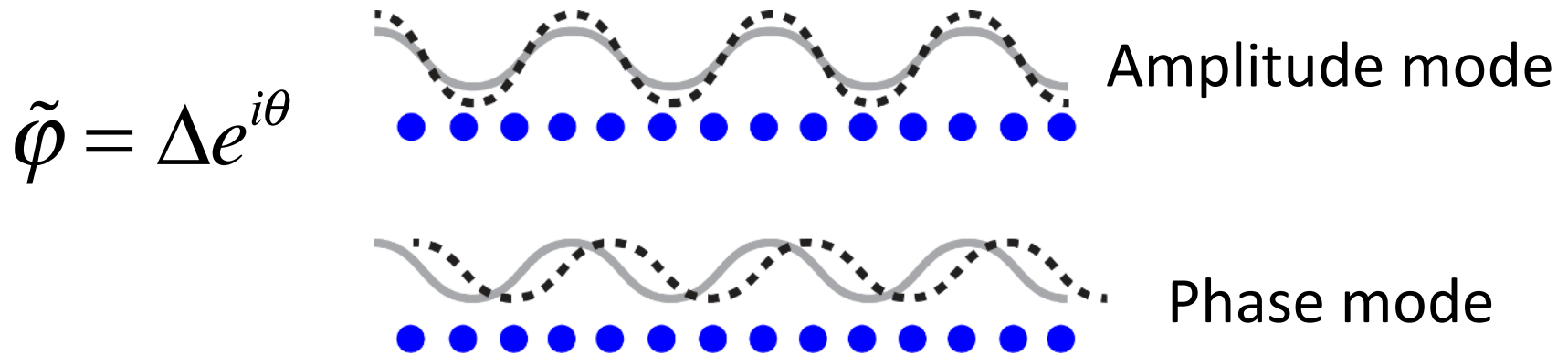


Fradkin et al. RMP 457, 104506 (2015)

Need to measure charge order fluctuations

What kind of fluctuations?

Electronic liquid crystals fluctuate



But hydrodynamics also requires:

Diffusive modes (vorticity, thermal diffusion,...)

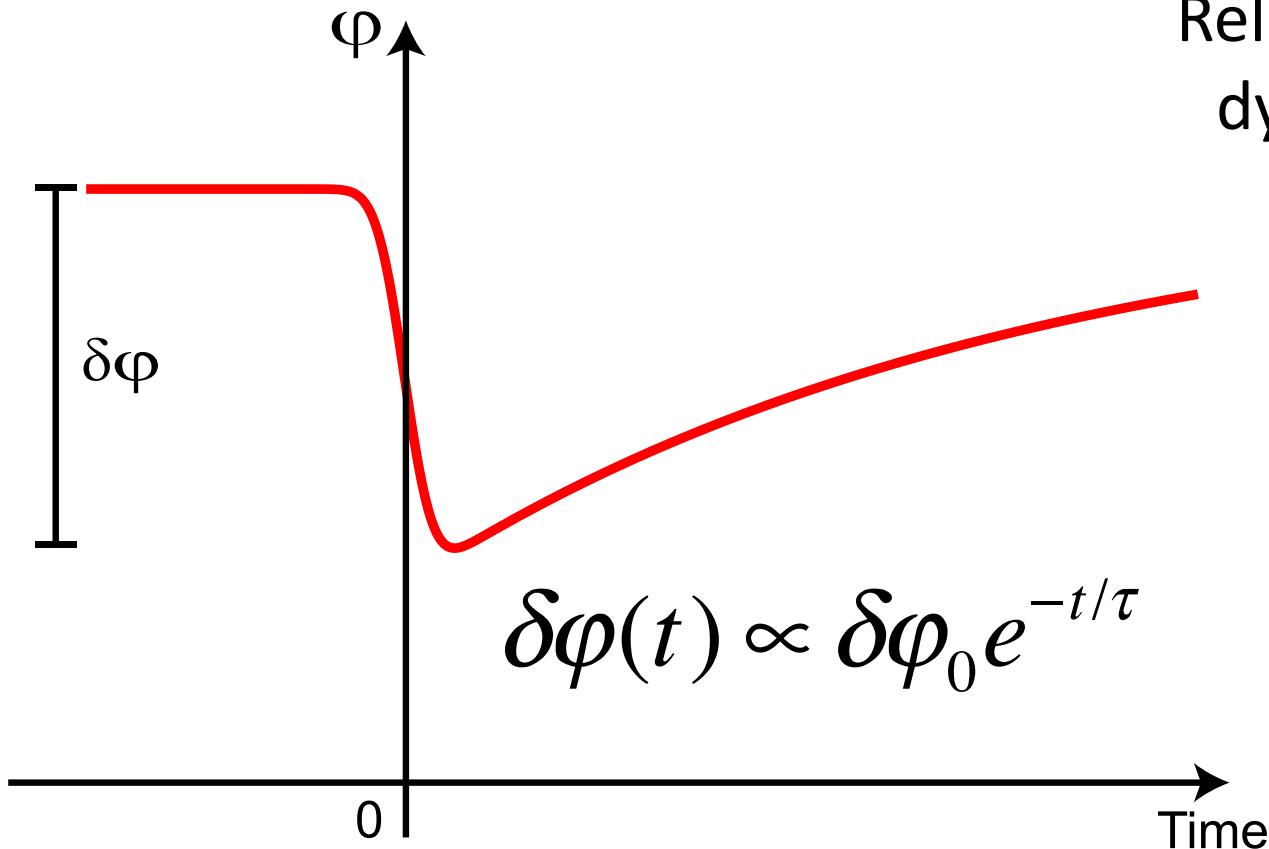
Relevant energy scale is $O(k_B T_c) \sim 1-10$ meV

Measuring fluctuations in time-domain

Evolution of a perturbed order parameter

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

Relaxational dynamics



Measuring fluctuations in time-domain

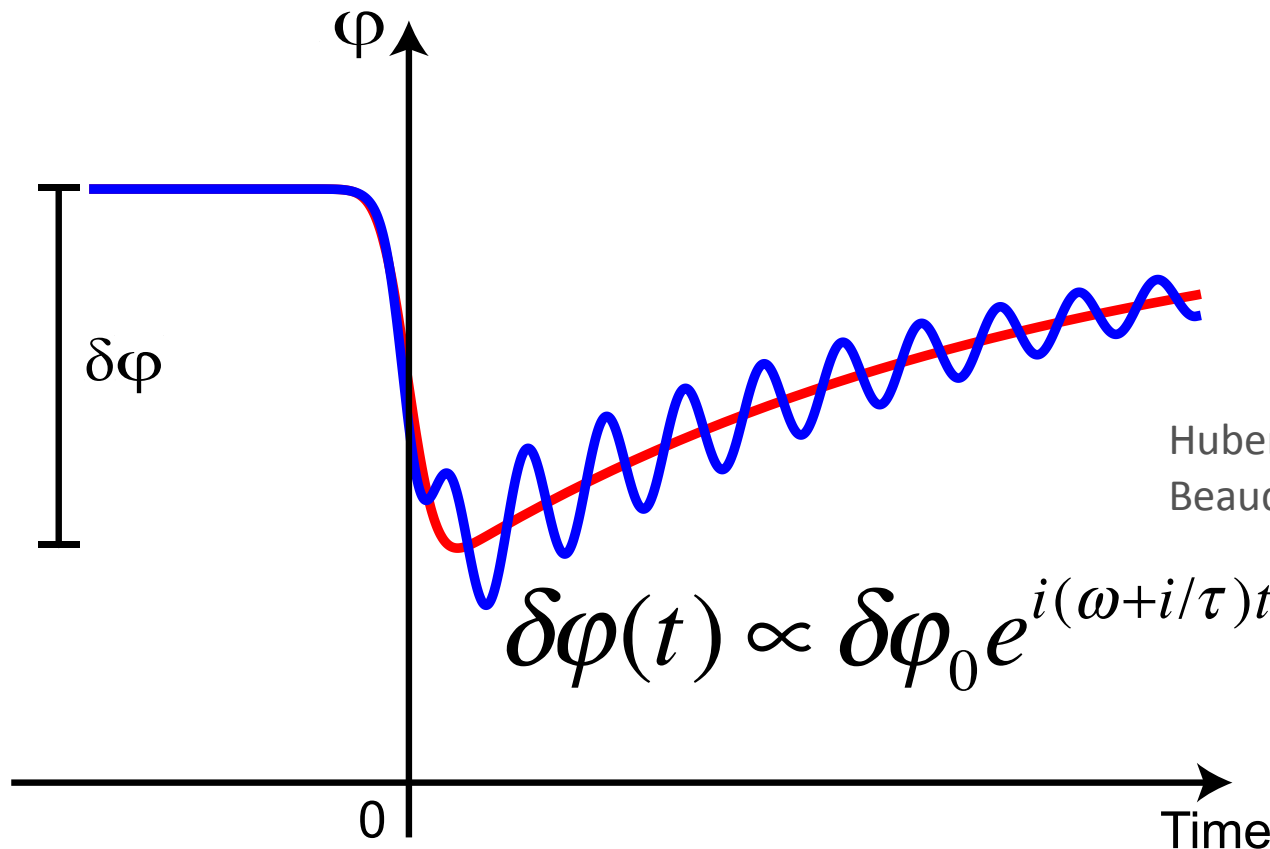
Evolution of a perturbed order parameter

$$\frac{\delta F}{\delta \varphi} = -\frac{1}{\Gamma} \frac{\partial \varphi}{\partial t} - \mu \frac{\partial^2 \varphi}{\partial t^2}$$

Inertial dynamics

Conventional CDW systems

Huber et al. PRL 113, 026401 (2014)
Beaud et al. Nat. Mat. 13, 923 (2014)



Need for a scattering probe

Evidence of sub-20 meV dynamics

Torchinsky et al. Nature Materials 12, 387 (2013)

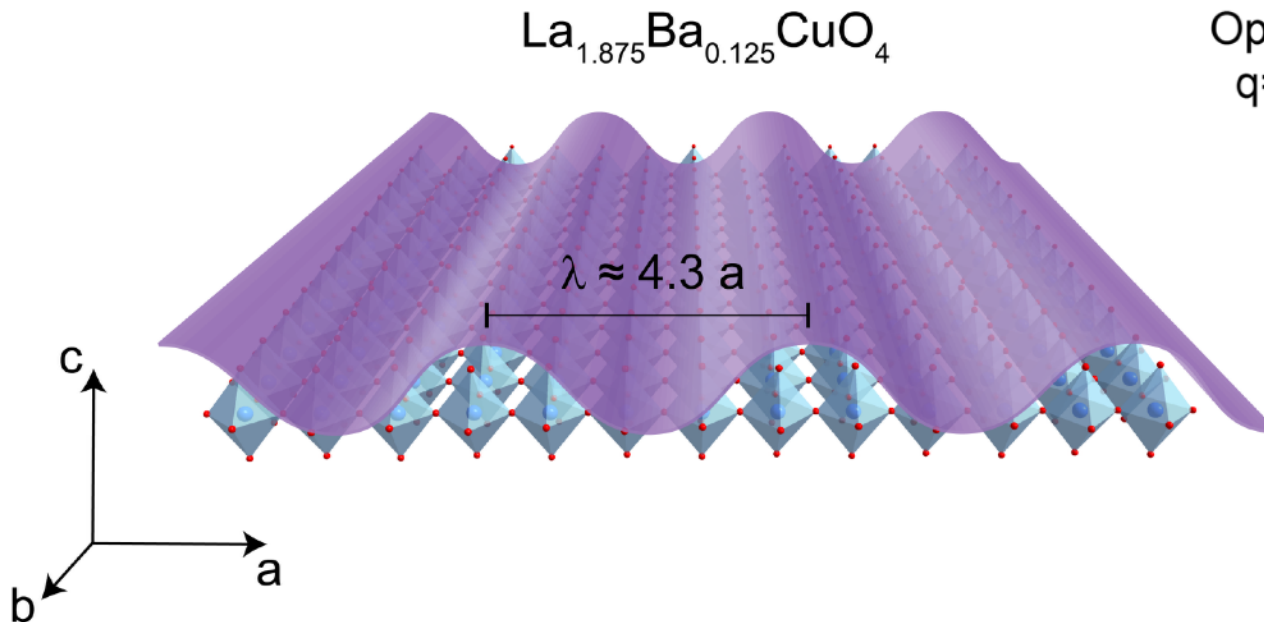
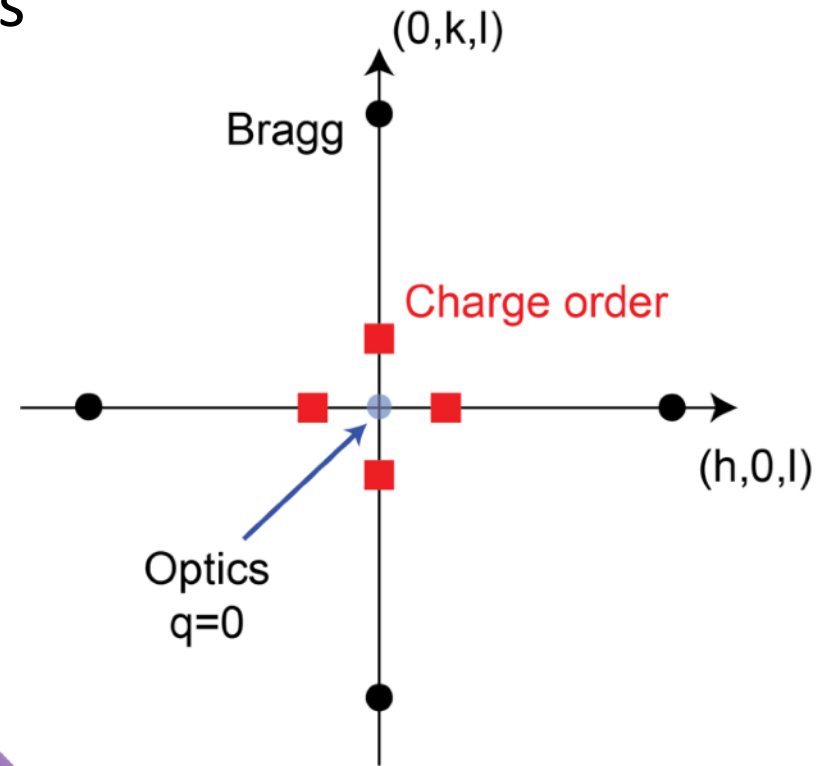
Hinton et al. PRB 88, 060508 (2013)

Dakovski et al. PRB 91, 220506 (2015)

Ergecen et al. forthcoming (2019)

Arpaia et al. arXiv: 1809.04949 (2018)

But order parameter at finite q



Need for a scattering probe

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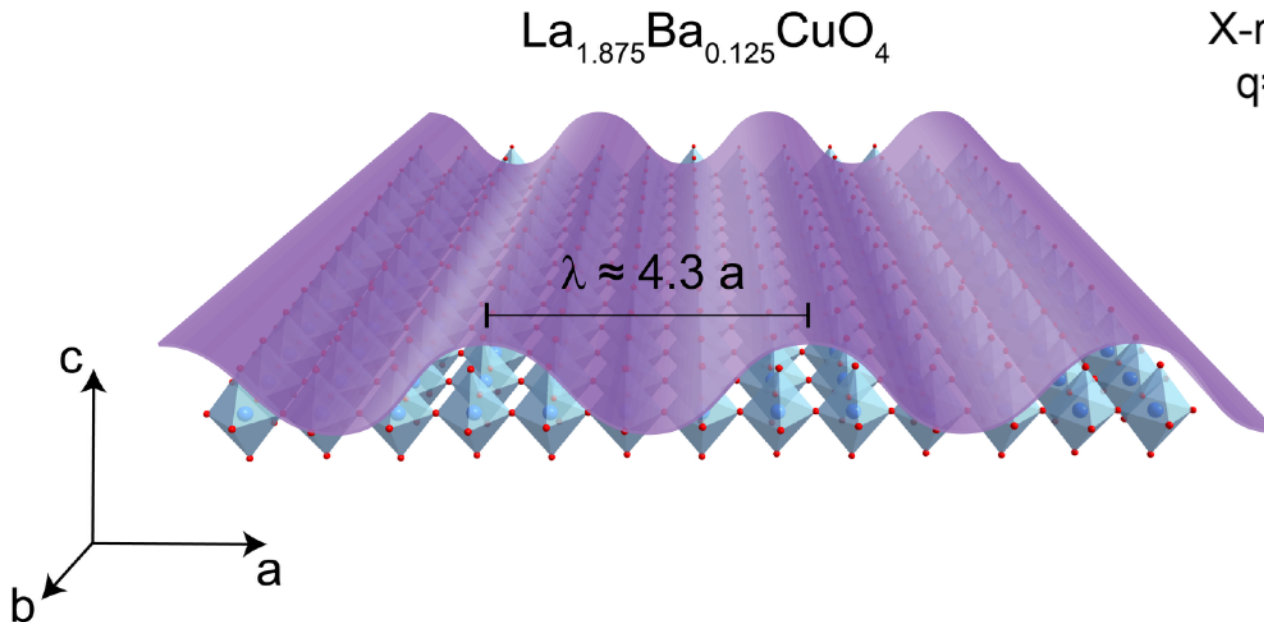
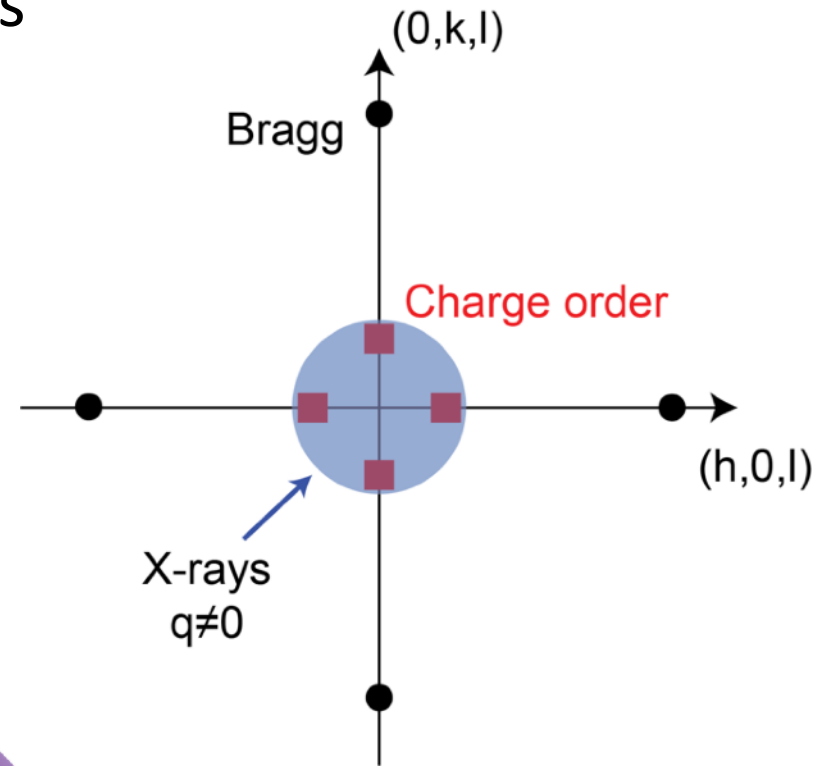
Hinton et al. PRB 88, 060508 (2013)

Dakovski et al. PRB 91, 220506 (2015)

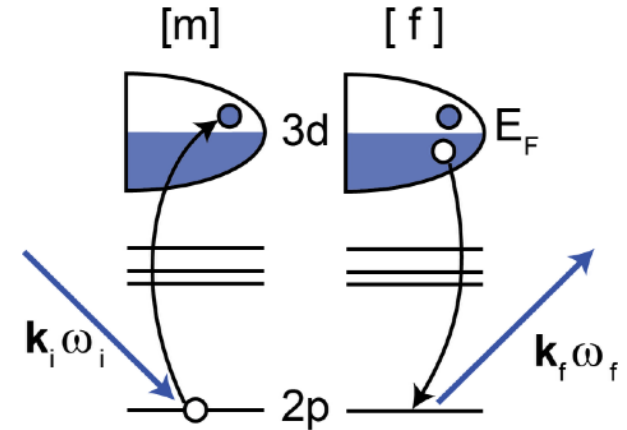
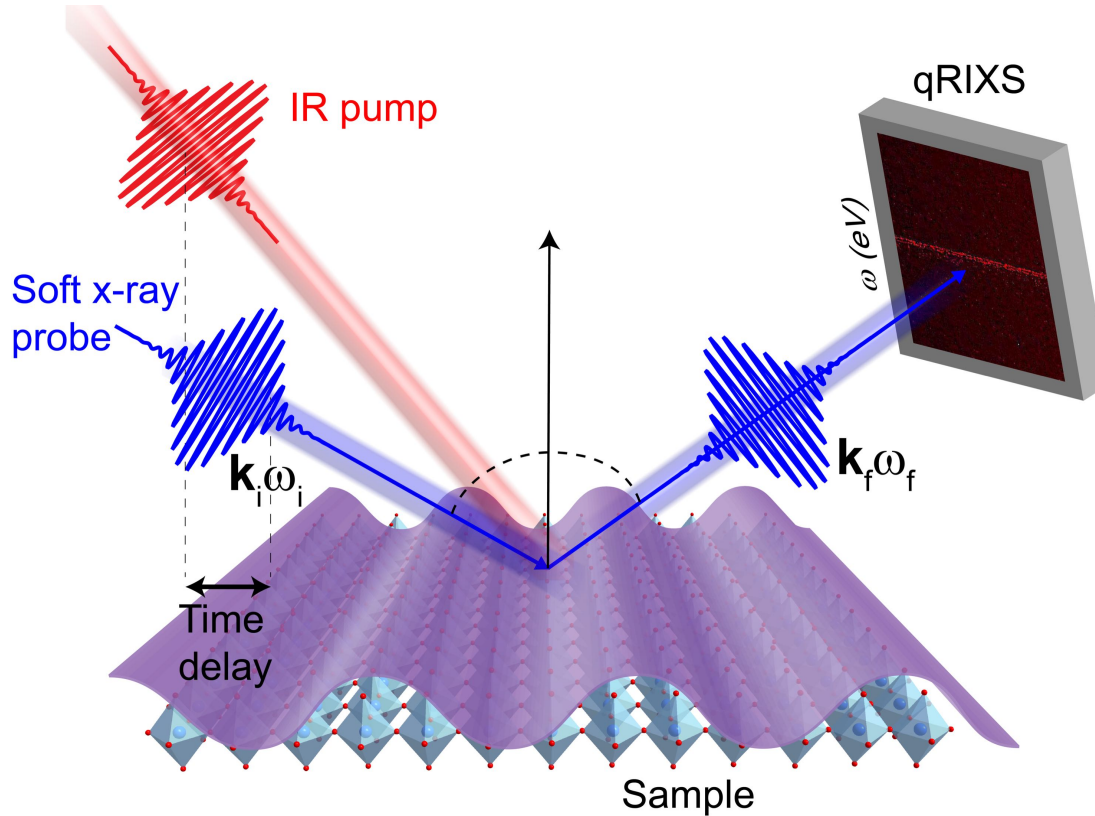
Ergecen et al. forthcoming (2019)

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But order parameter at finite q



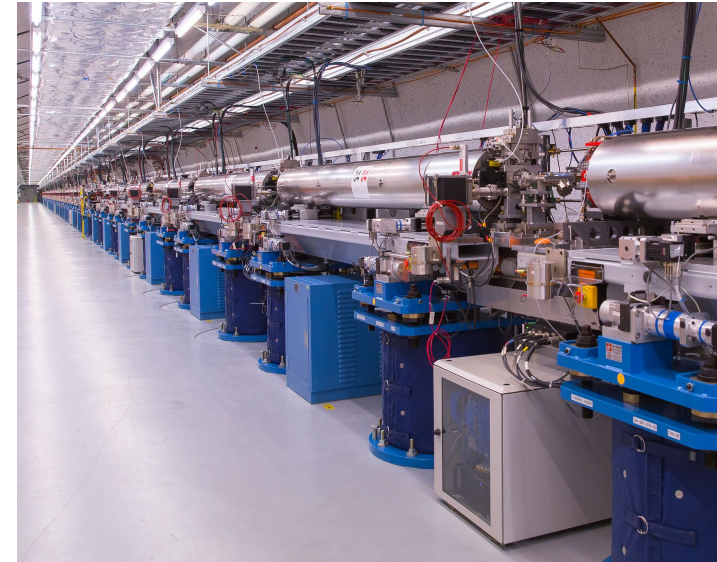
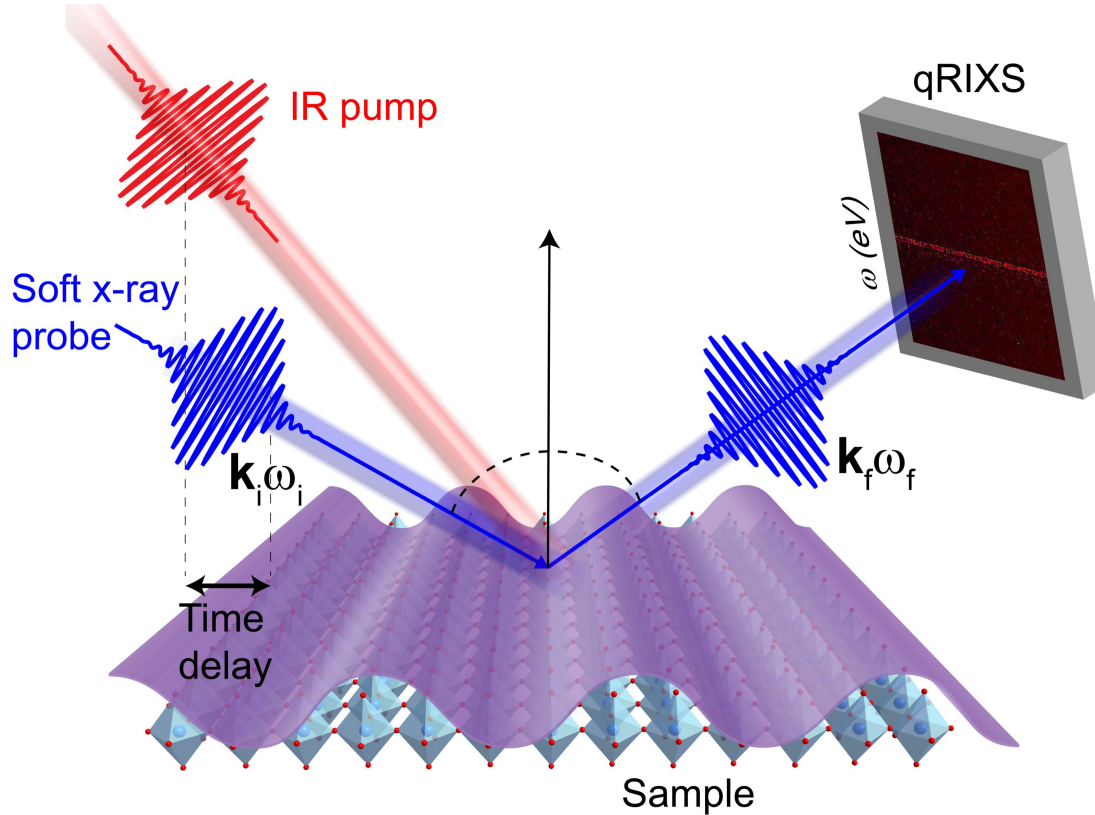
Probing charge order dynamics with trRSXS



Direct RSXS at
Cu L edge
(931.5 eV)

- Resonant process sensitive to valence holes
- trRSXS probes finite-q order parameter dynamics

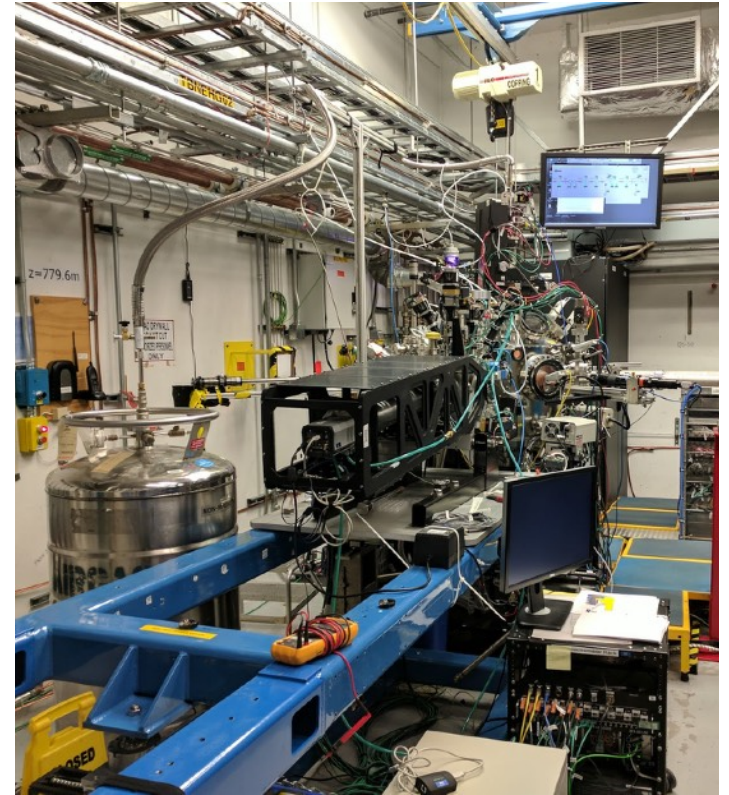
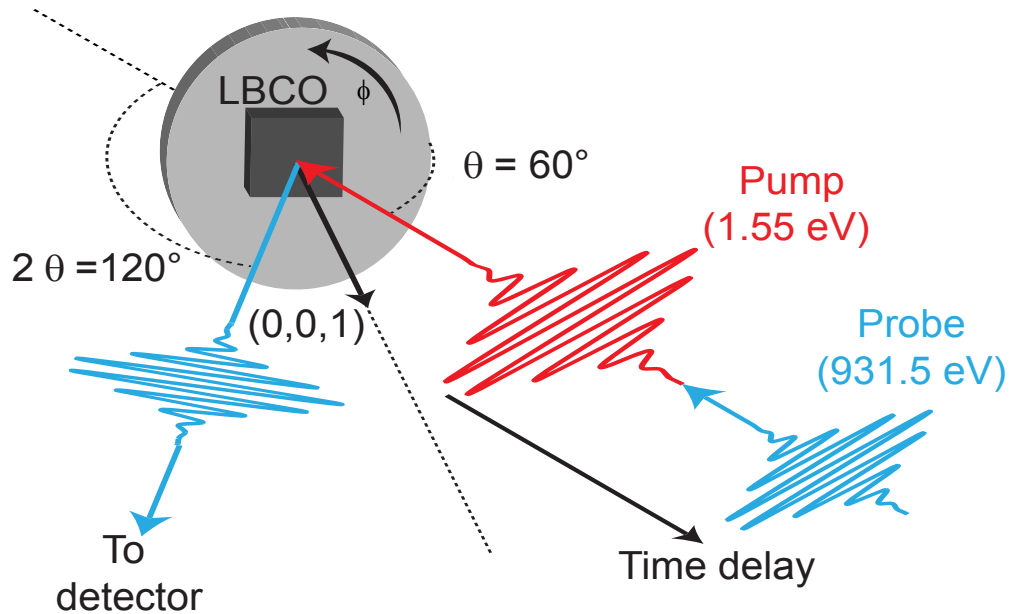
Probing charge order dynamics with trRSXS



**LCLS Free
Electron Laser**

- Resonant process sensitive to valence holes
- trRSXS probes finite-q order parameter dynamics

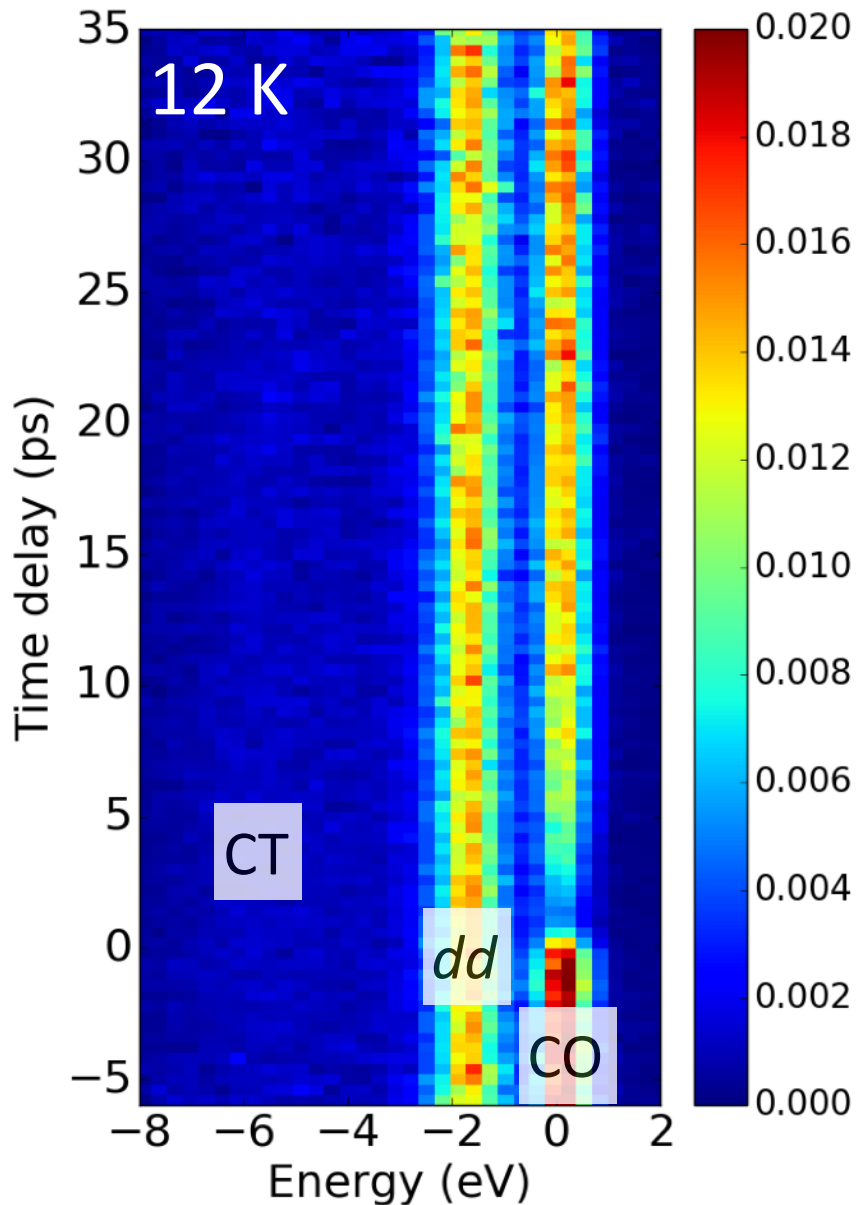
The tr-RSXS experiment



- Both energy-resolved and energy-integrated
- High-temporal resolution (130 fs)

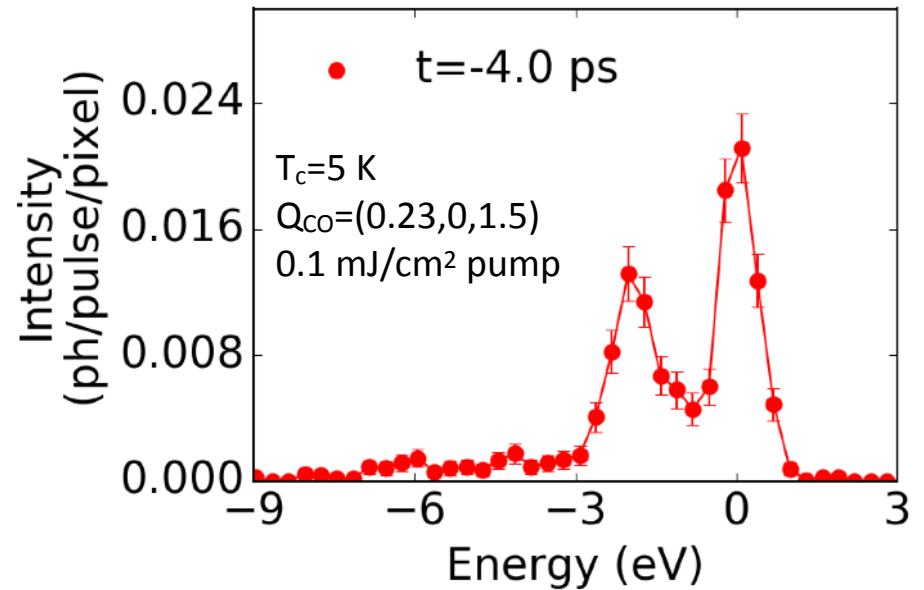
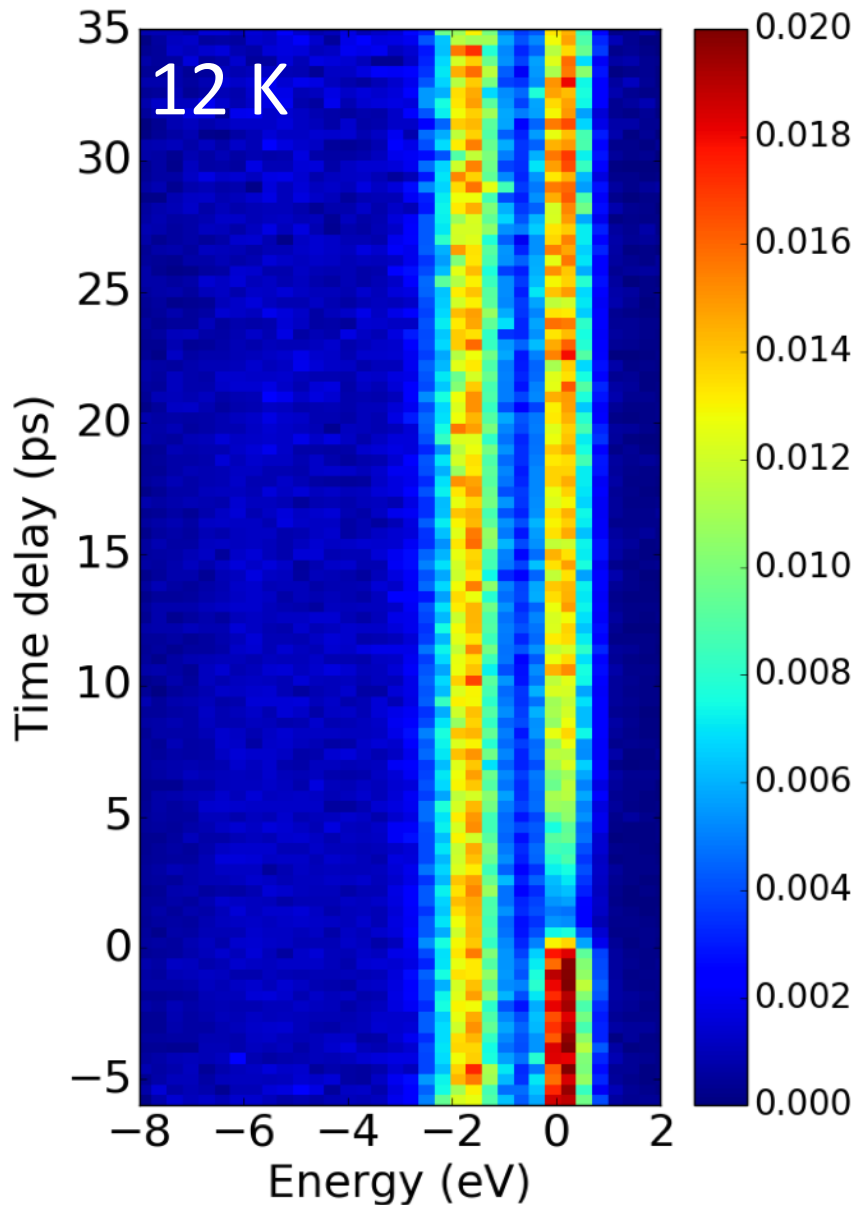
Revealing the CO dynamics of LBCO-1/8

Pump selectively targets charge order



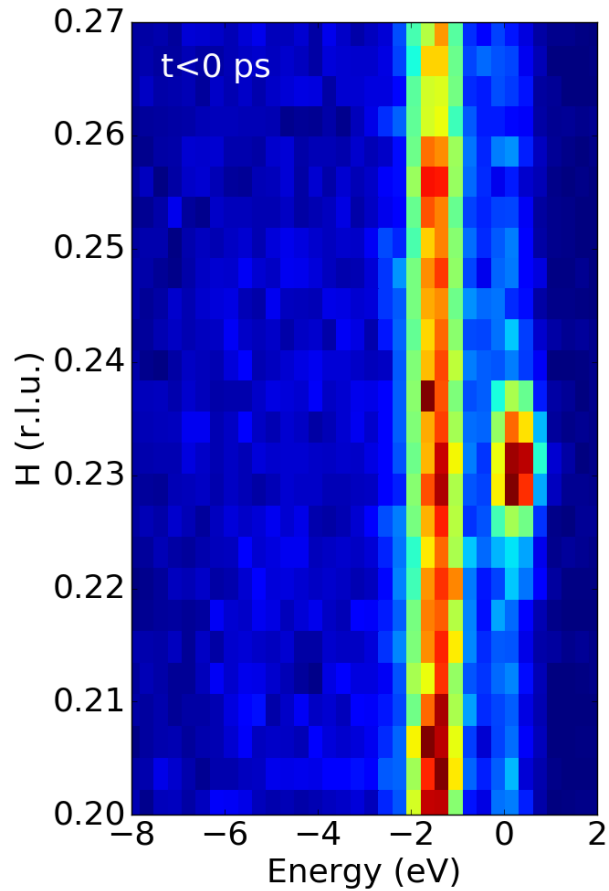
- Only CO responds
- Intensity not conserved
- $\Delta E = 0.65$ eV @ Cu L₃ edge

Pump selectively targets charge order

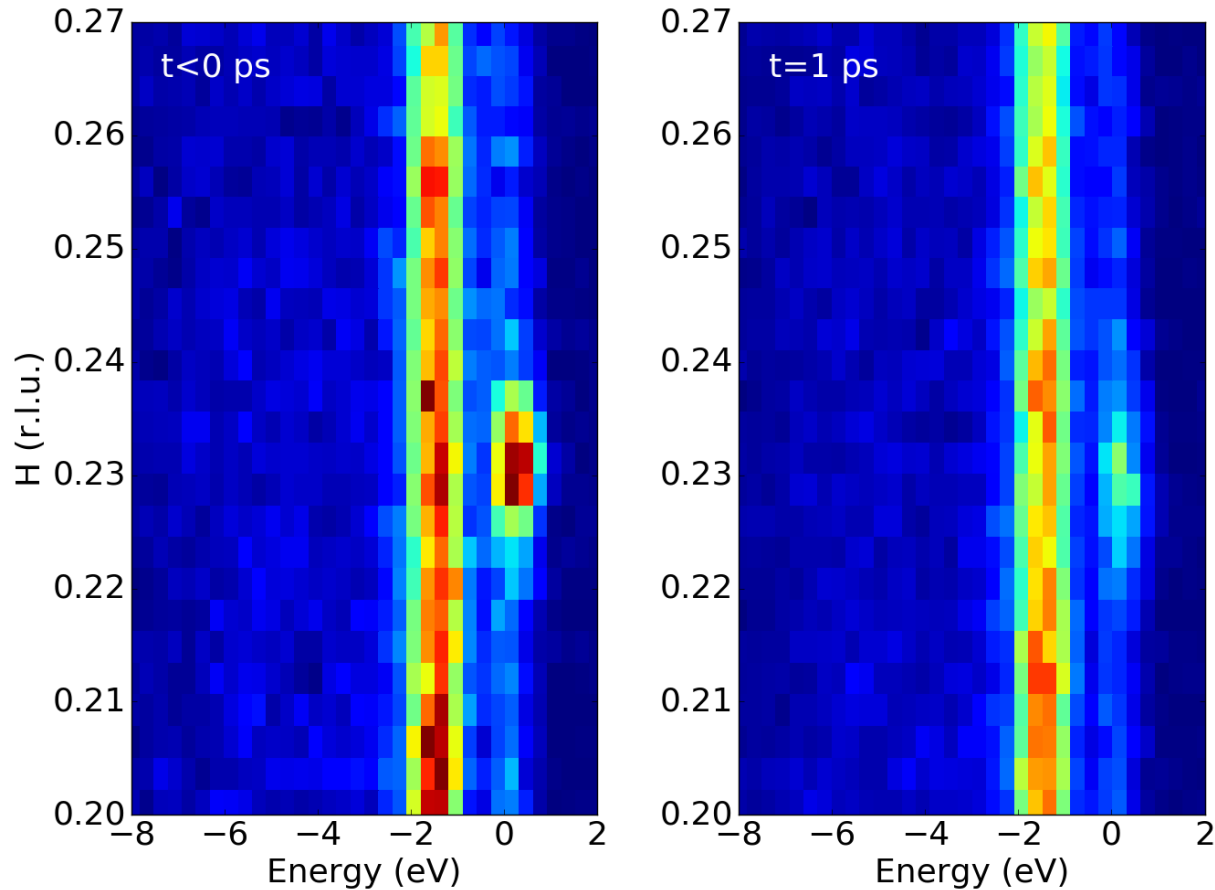


- Only CO responds
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Momentum-resolved order parameter quench

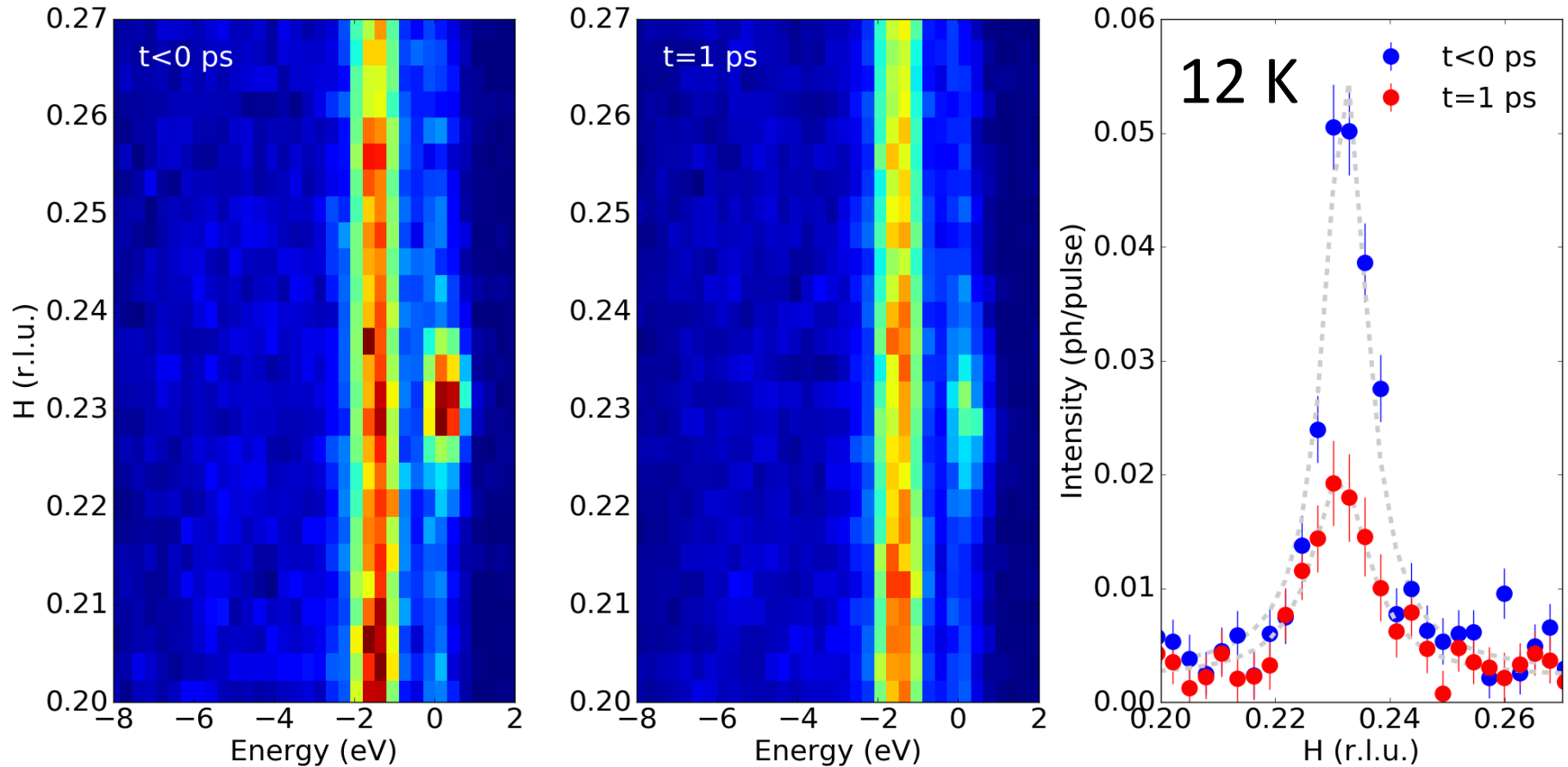


Momentum-resolved order parameter quench



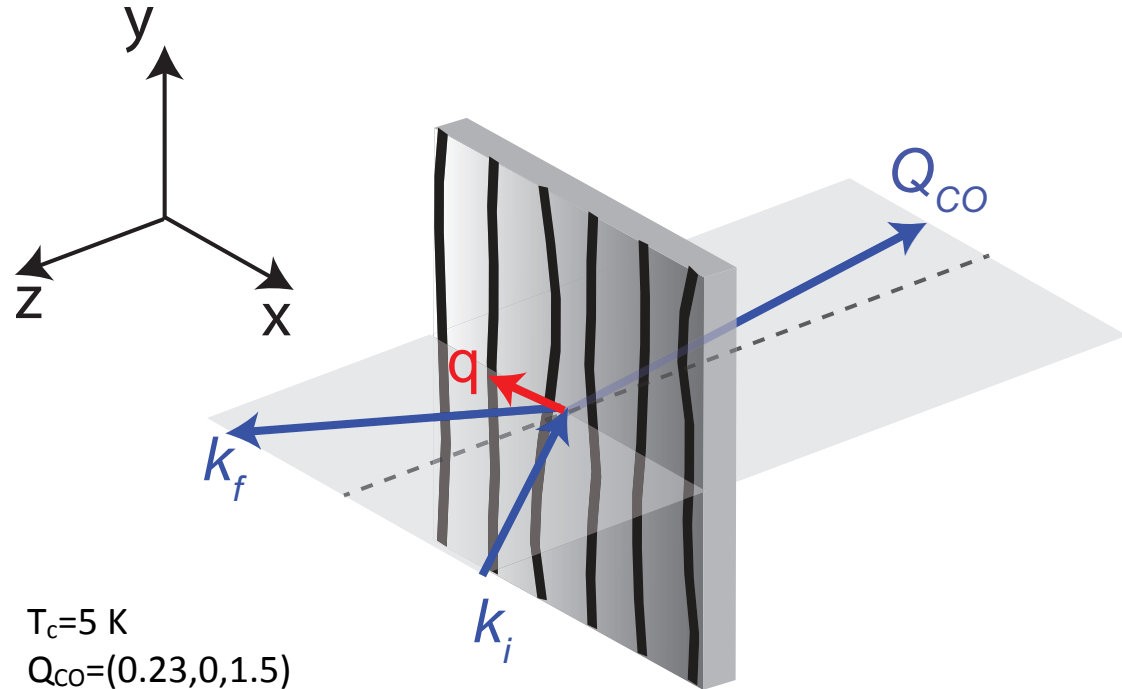
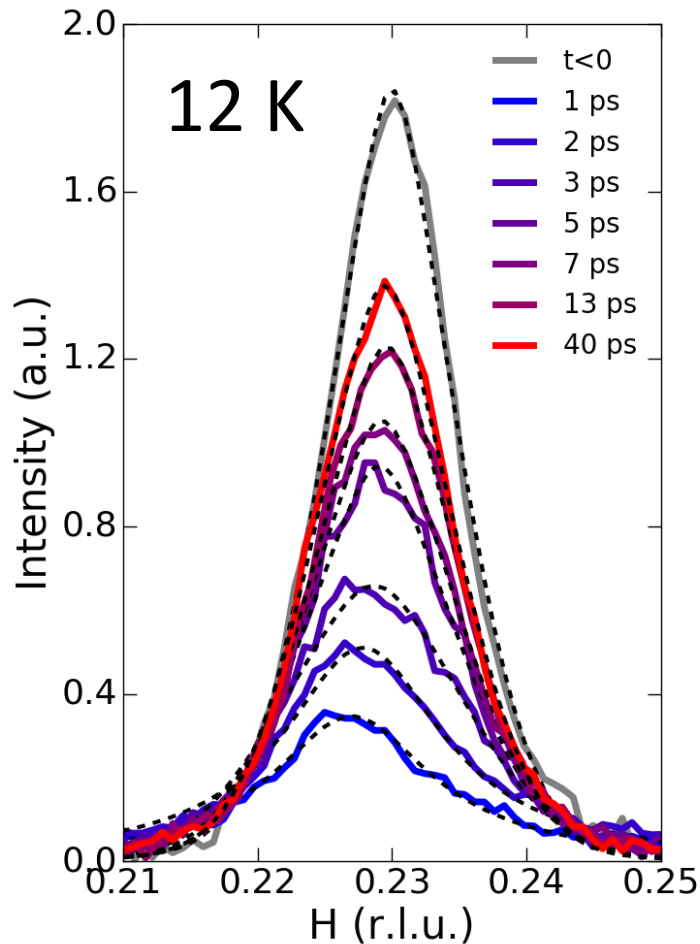
- CO peak drops 75% in intensity
- Momentum broadening by 45% (defects)

Momentum-resolved order parameter quench



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- Momentum broadening by 45% (defects)

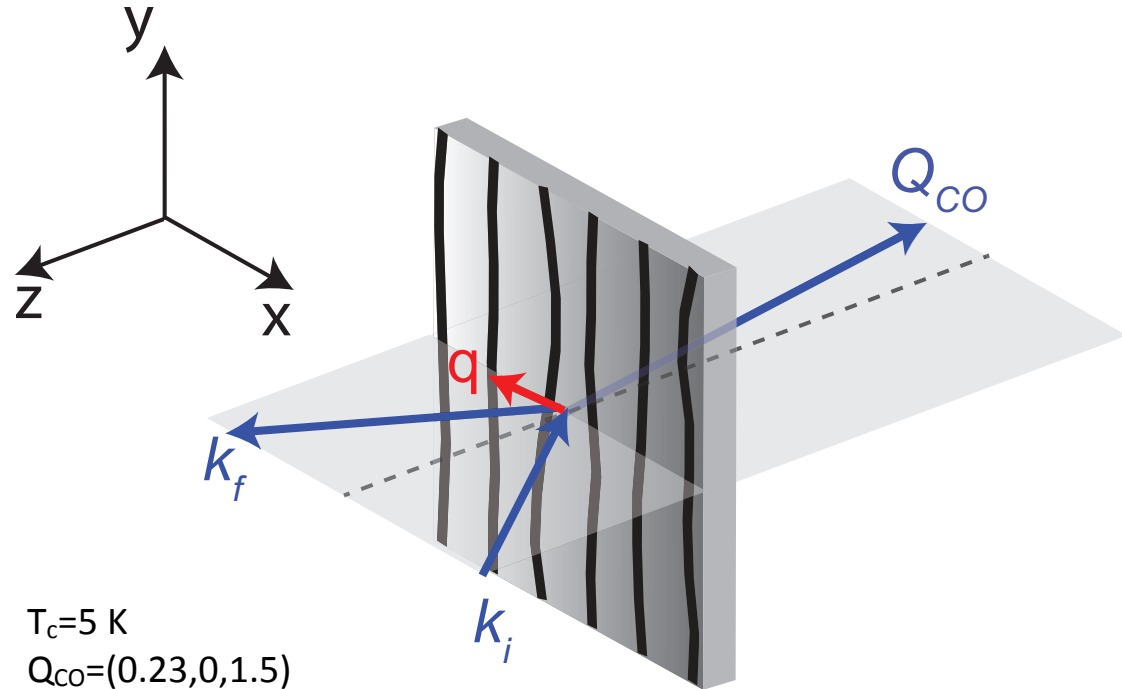
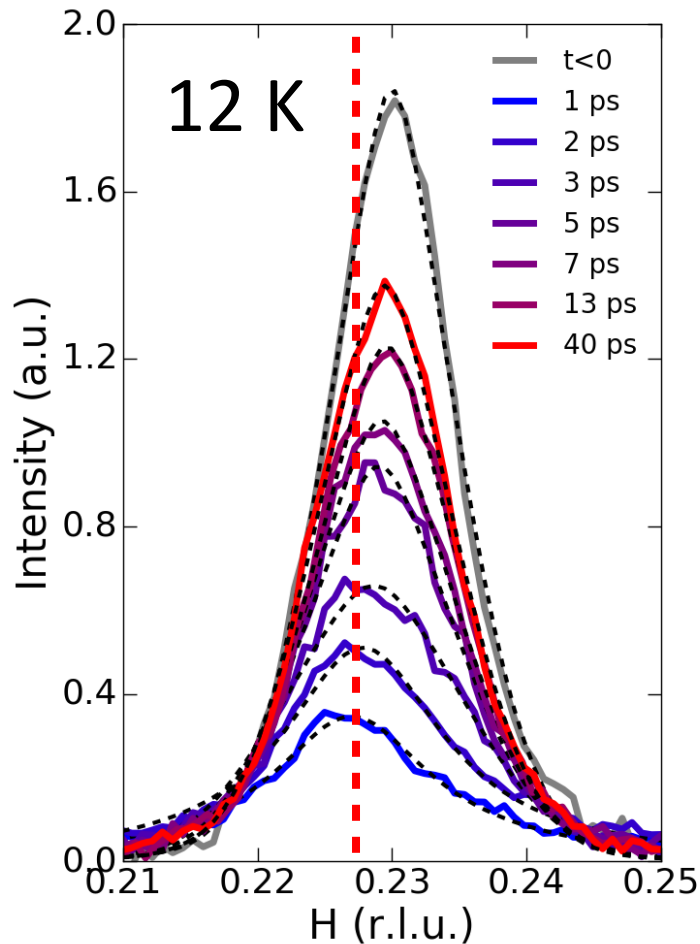
Dynamics of the charge order recovery



$T_c = 5$ K
 $Q_{CO} = (0.23, 0, 1.5)$
0.1 mJ/cm² pump

Sampling transverse charge order fluctuations in time

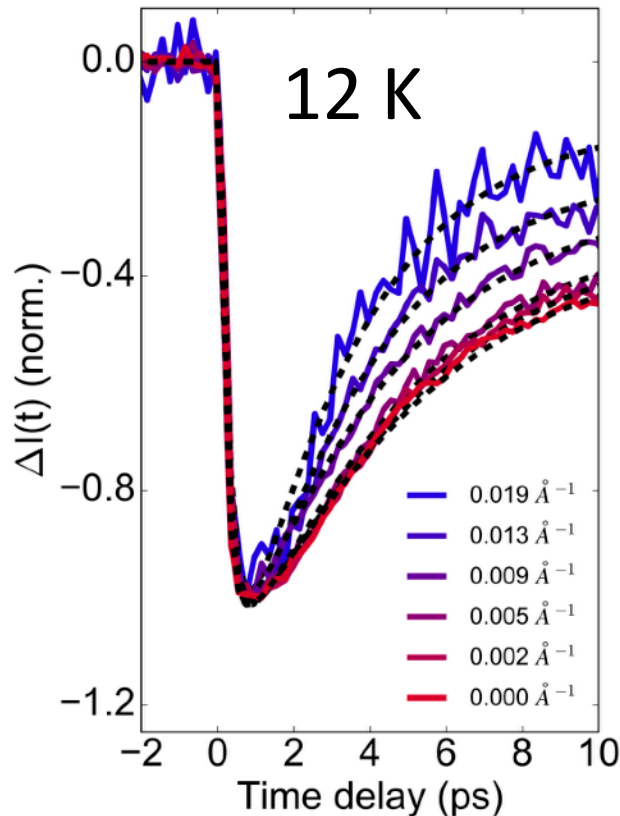
Dynamics of the charge order recovery



$T_c = 5$ K
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Sampling transverse charge order fluctuations in time

Evidence of diffusive charge order mode



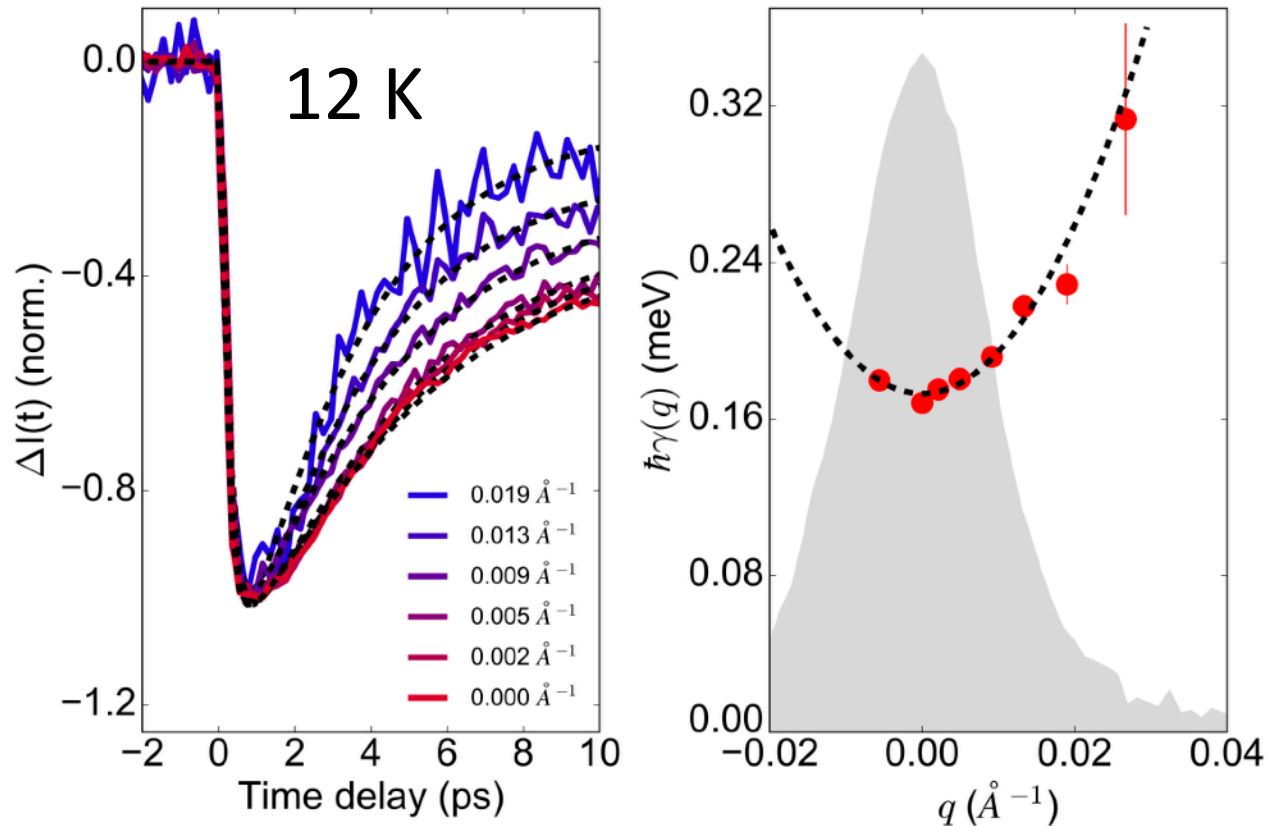
Relaxational
dynamics

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

$$\delta \varphi(t) \propto \delta \varphi_0 e^{-t/\tau}$$

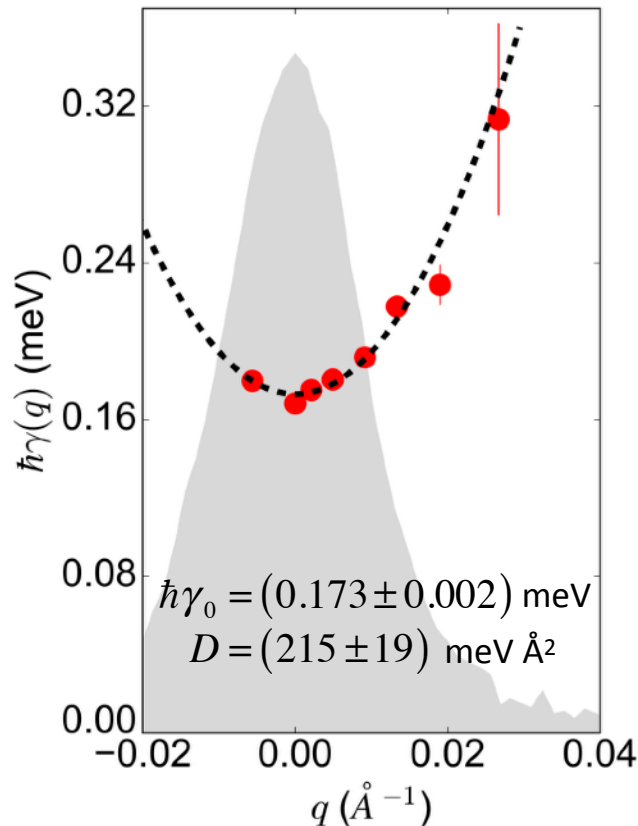
- No coherent oscillations (gapless)

Evidence of diffusive charge order mode



- No coherent oscillations (gapless)
- Recovery scales with q^2

Evidence of diffusive charge order mode



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

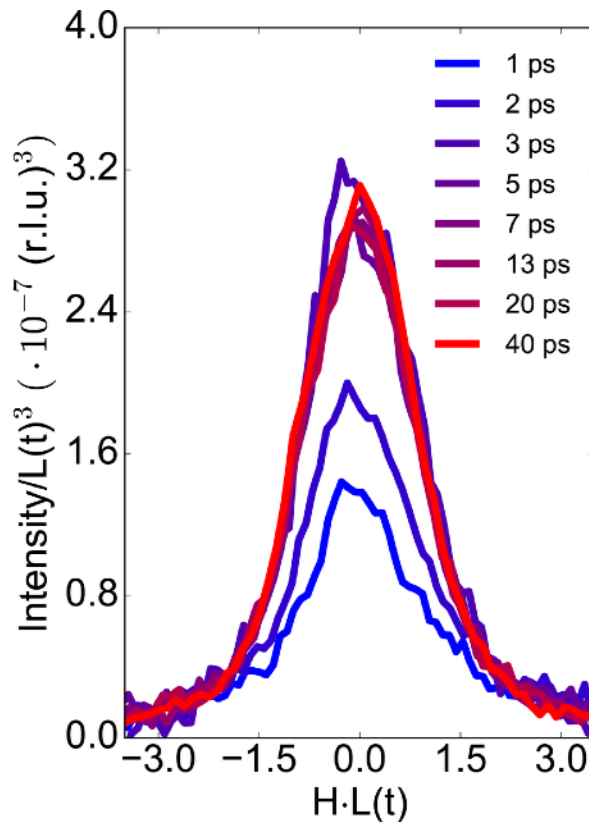
$$F[\varphi] = a\varphi^2 + b\varphi^4 + c(\nabla \varphi)^2$$

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

Transverse CO diffusion at energy $O(k_B T_c)$

$$i\hbar\gamma(q) = \hbar(\gamma_0 + Dq^2) \sim 2K$$

Dynamic critical scaling and defects



Previously observed in

- block copolymers
- binary alloys

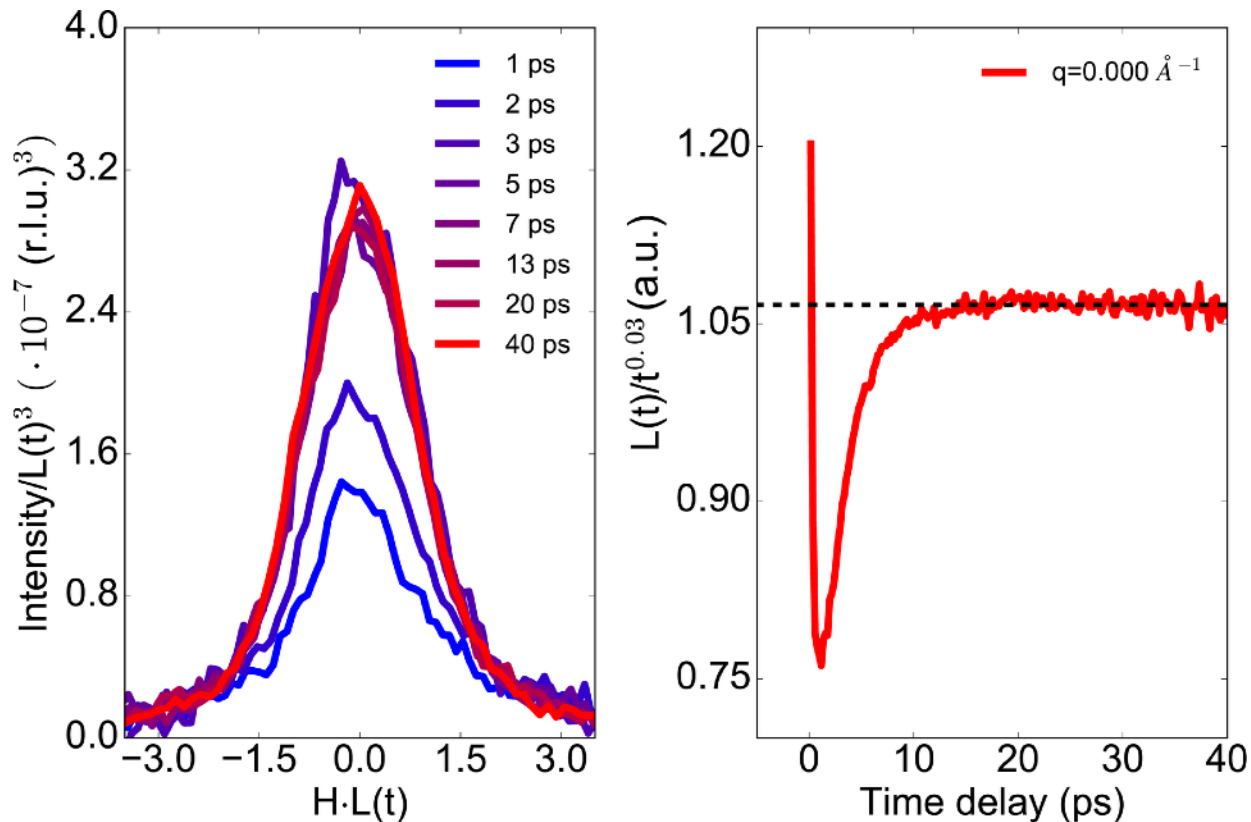
e.g. Gaulin et al. PRL 59, 668 (1987)

Harrison et al. Science 290, 1558 (2000)

- CO obeys dynamic critical scaling towards equilibrium

$$S(q,t) = L(t)^3 F(qL(t))$$

Dynamic critical scaling and defects

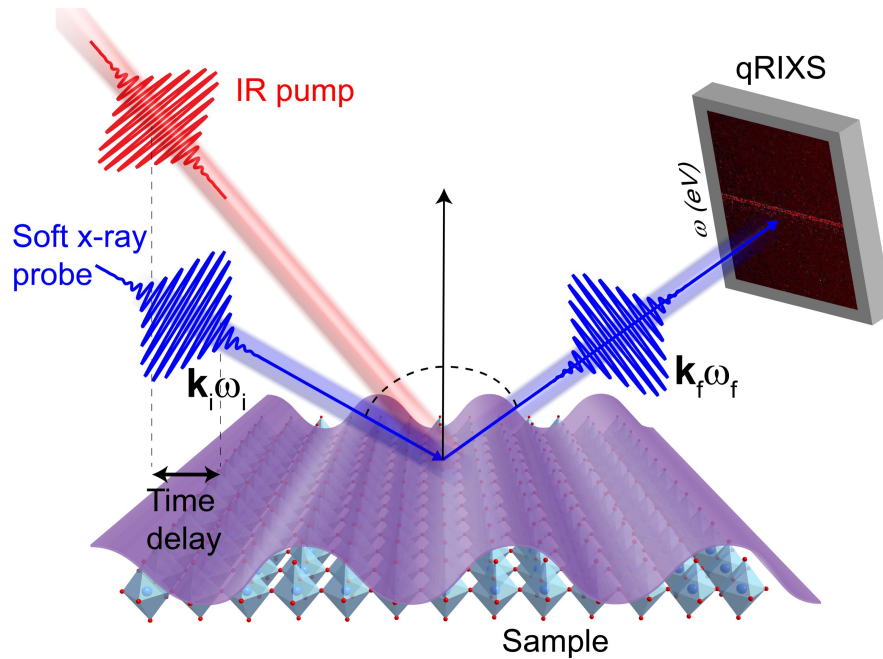


- CO obeys dynamic critical scaling towards equilibrium

$$S(q, t) = L(t)^3 F(qL(t))$$

- Propagation of topological defects $L(t) \sim \ln(t)$

Conclusions



- LBCO charge order is *dynamic*
- Gapless diffusive excitations
- Possible relevance to interstripe tunneling

