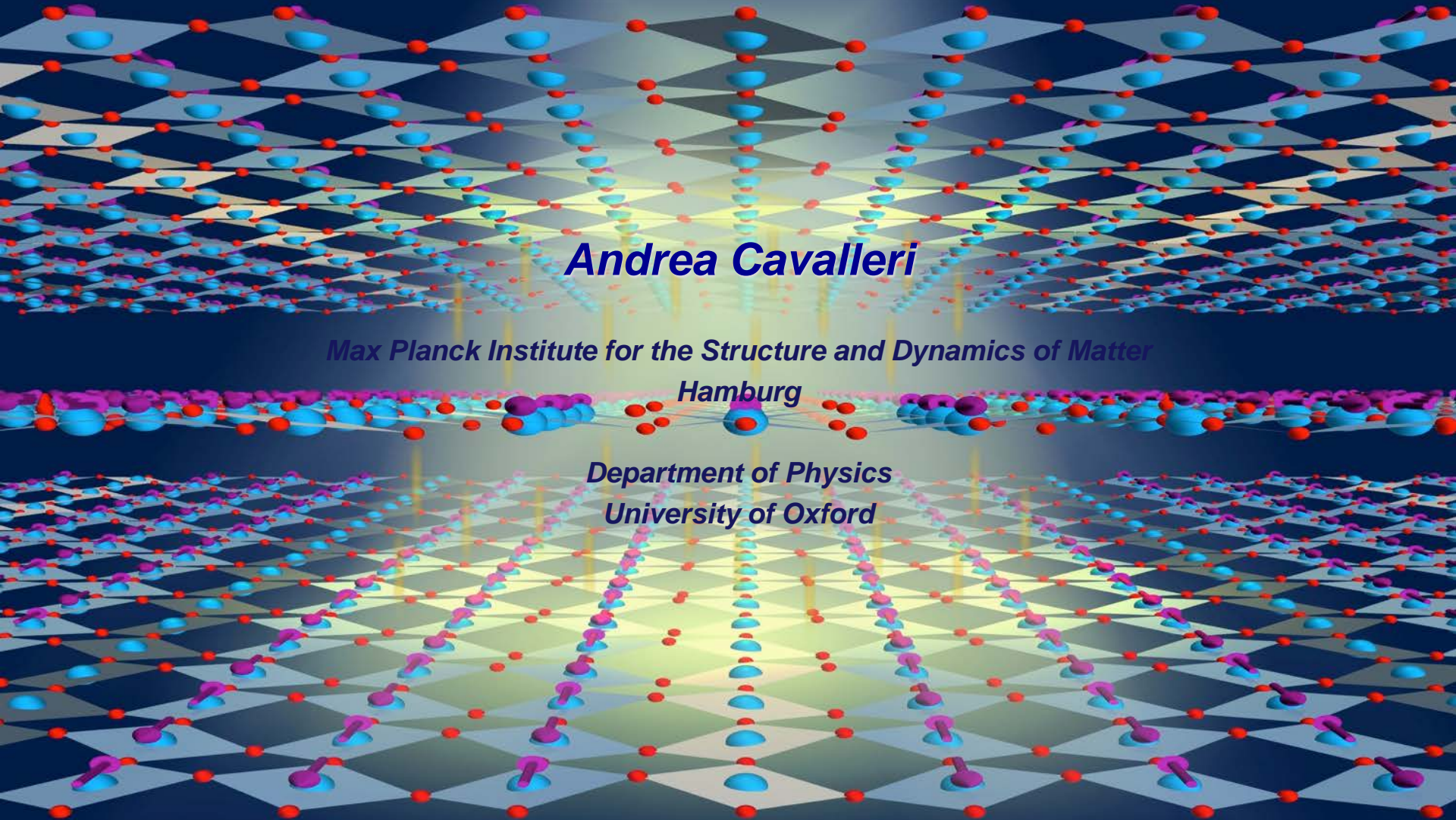


Light-control of quantum solids

Andrea Cavalleri

*Max Planck Institute for the Structure and Dynamics of Matter
Hamburg*

*Department of Physics
University of Oxford*

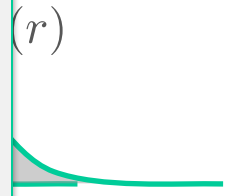
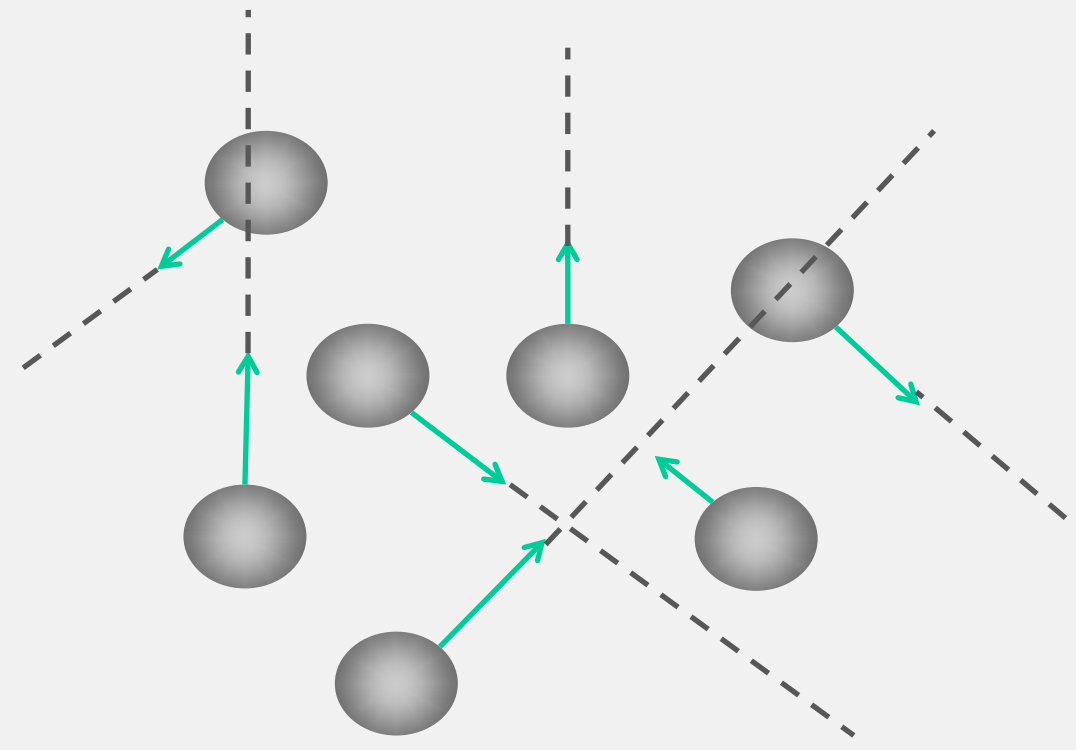
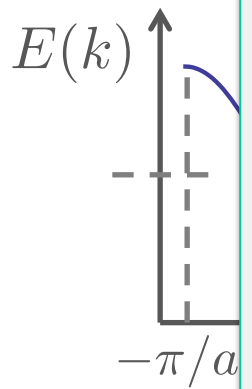


Conventional view of a solid

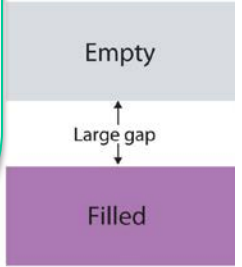
Many

are free

weak effective interactions
 can extrapolate *en masse* behaviour from one
 particle



Band structure
 metals, semiconductors
 and insulators



Metals

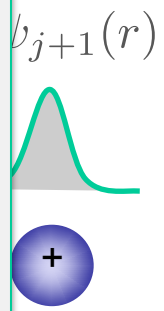
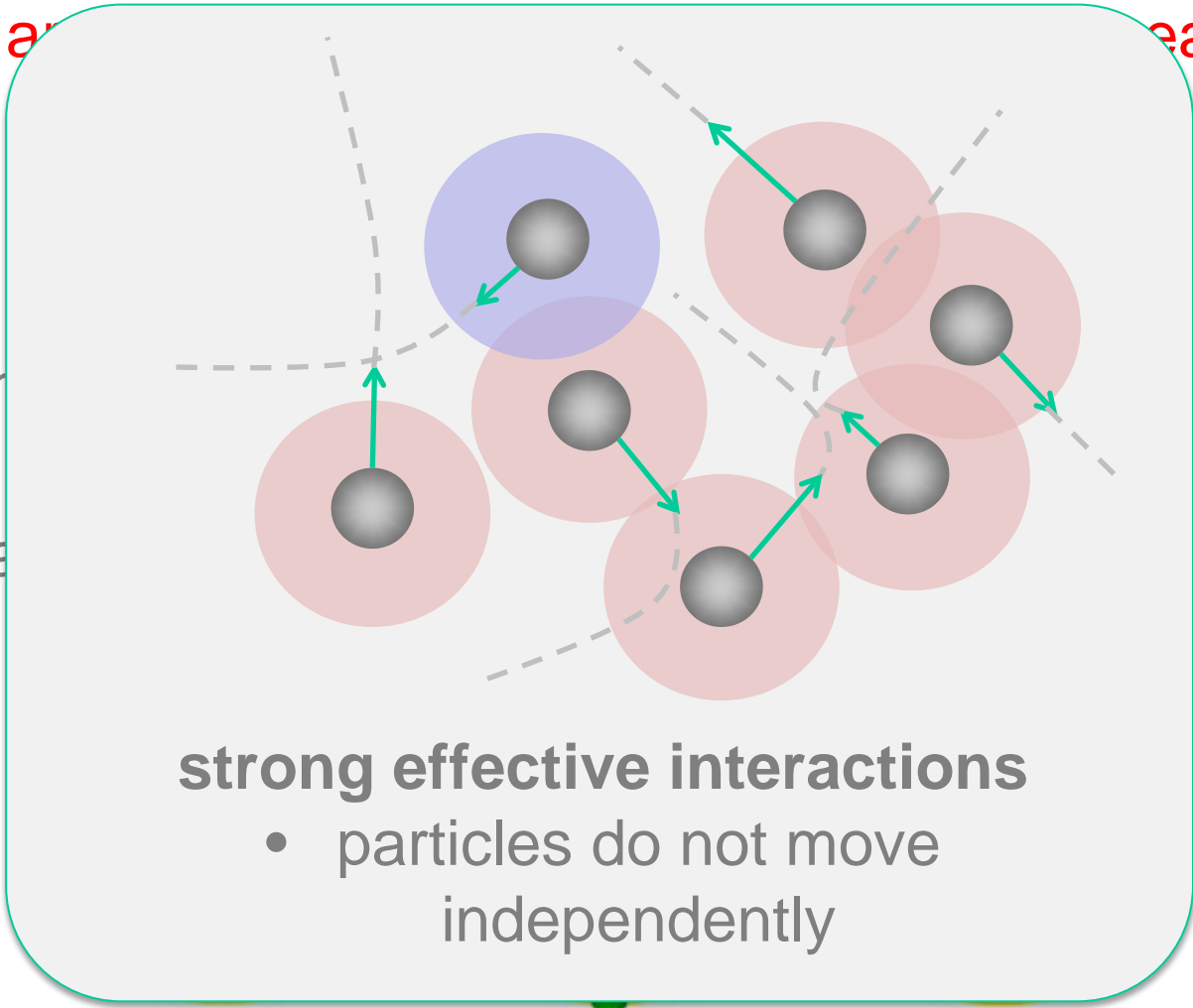
Semiconductors

Insulators

Strong correlations

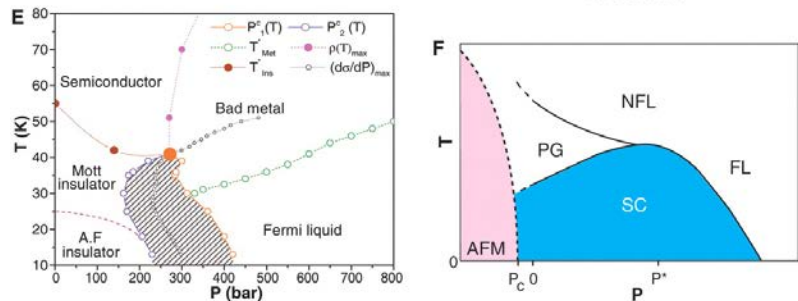
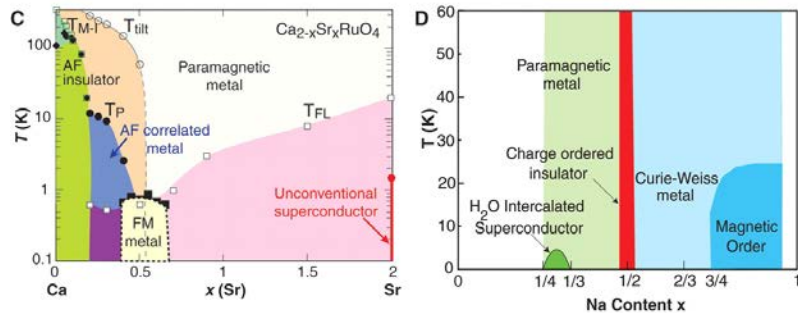
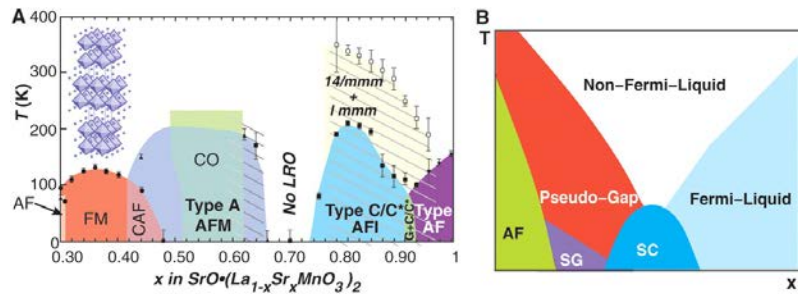
This picture fails for materials like transition metal oxides, which are Mott insulators. The reason is:

“core-l
small
confir
Mott insula



Big consequences

Materials with strong correlations possess a wide variety of competing phases with different and unconventional properties:



Metal-insulator transitions

Colossal magnetoresistance

High-temperature superconductivity

Complex materials

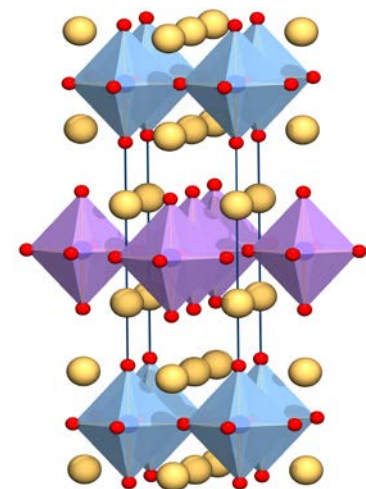
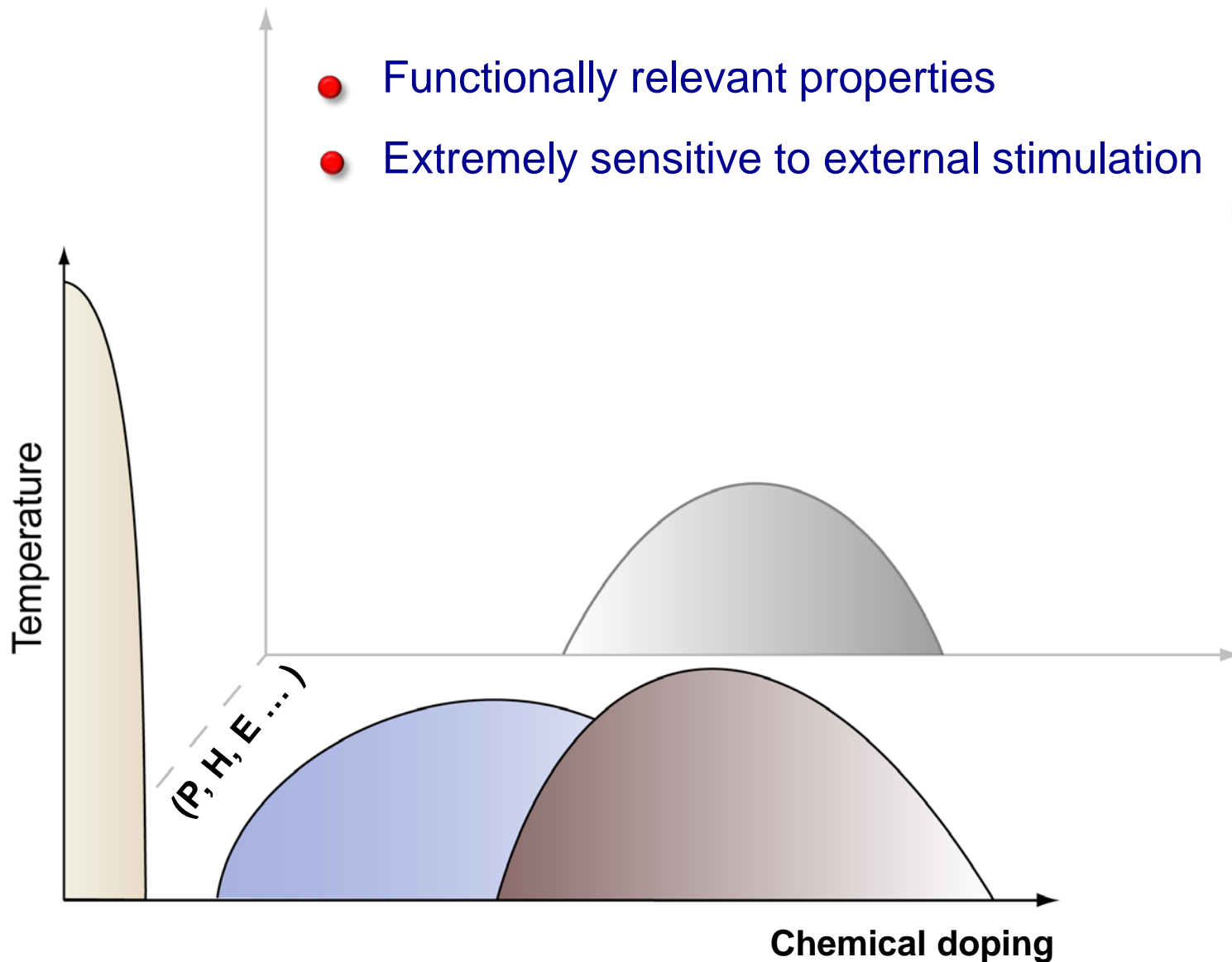
Strong correlations produce **collective giant responses** to **small** external perturbations.

Such responses are often *functionally* relevant.

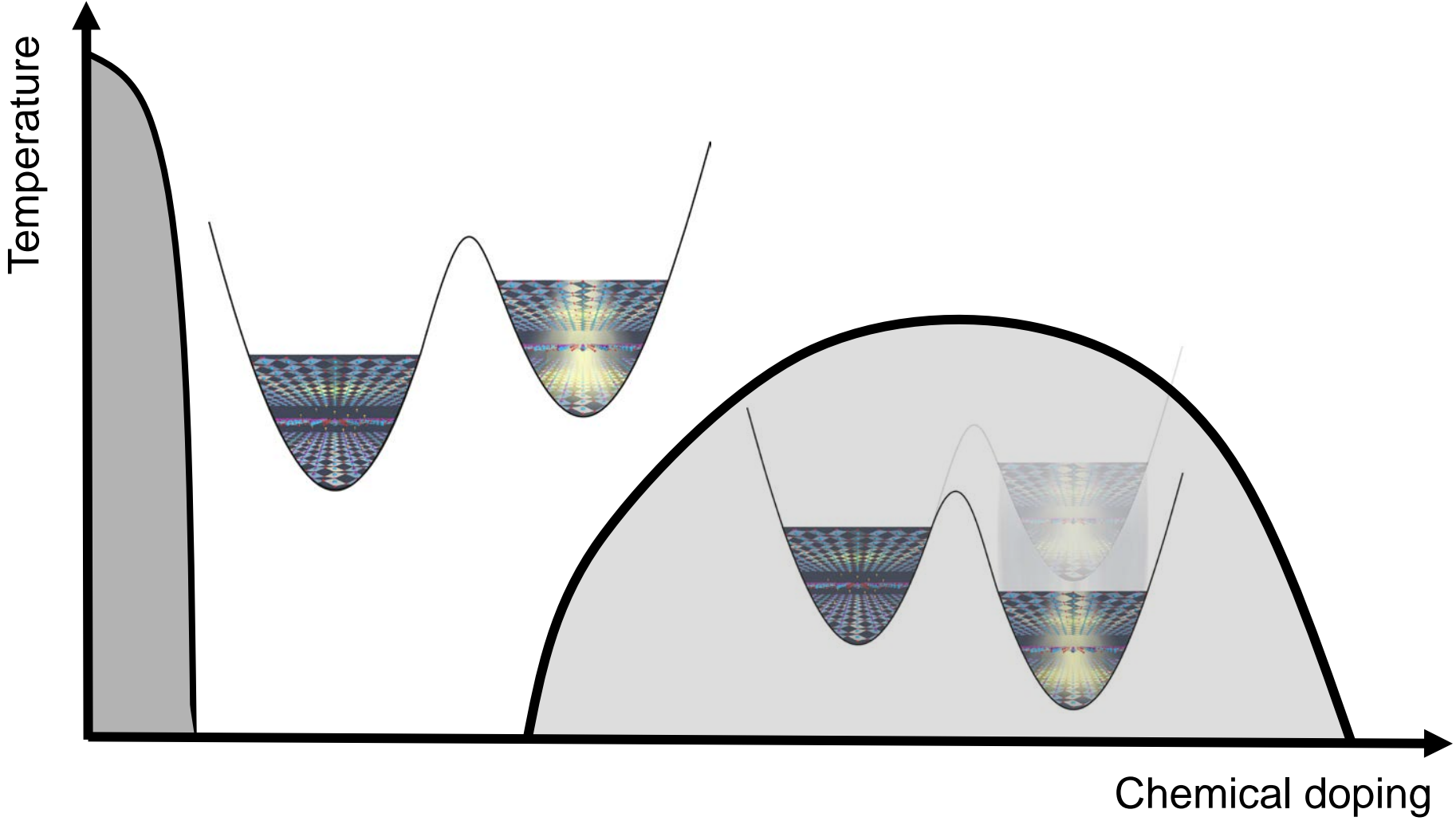
Our goal is to **CONTROL** materials, induce these phenomena at higher temperatures or modulate amplify their responses

Complex solids: many competing phases

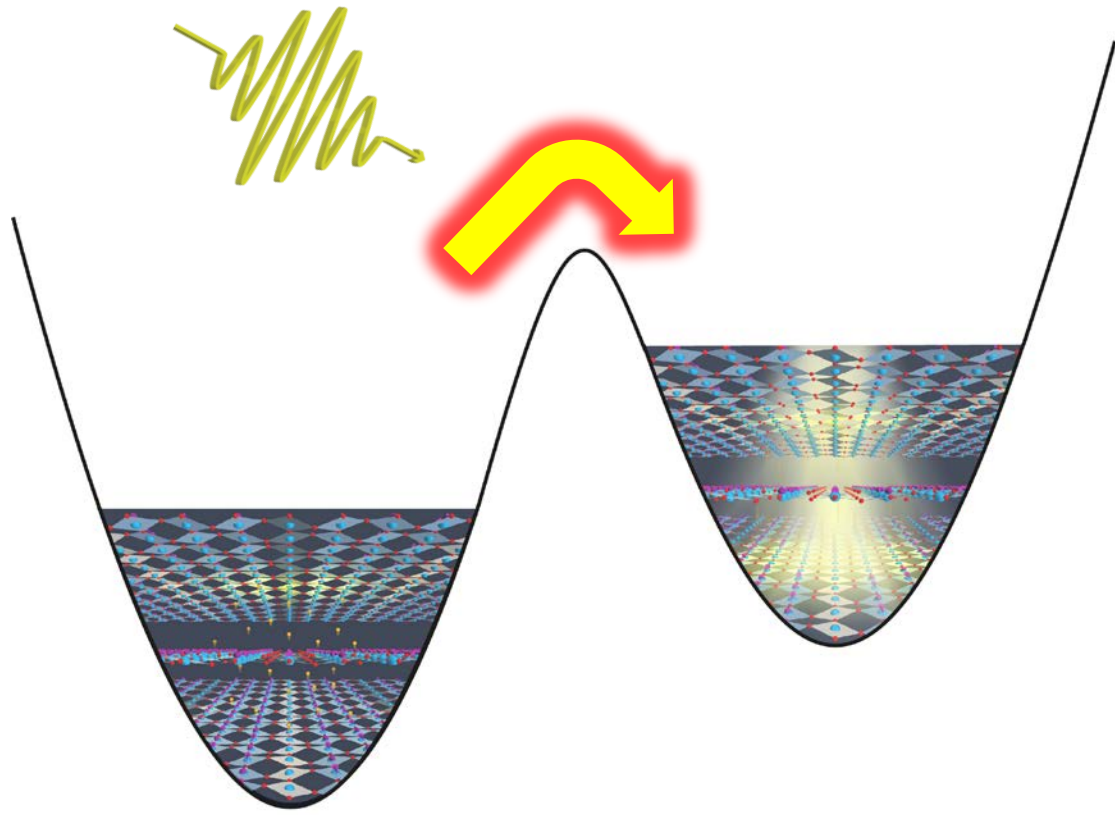
- Functionally relevant properties
- Extremely sensitive to external stimulation



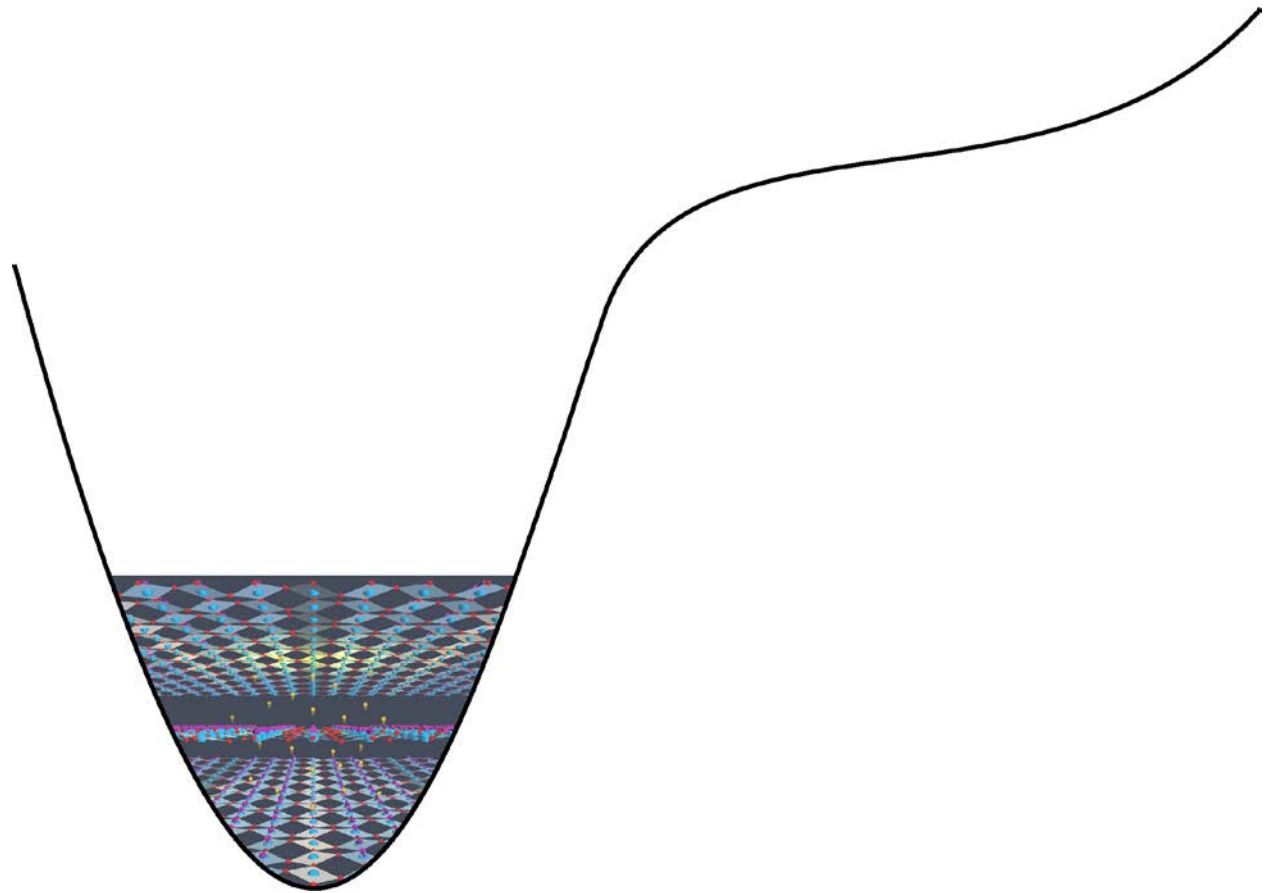
Ground states and hidden phases



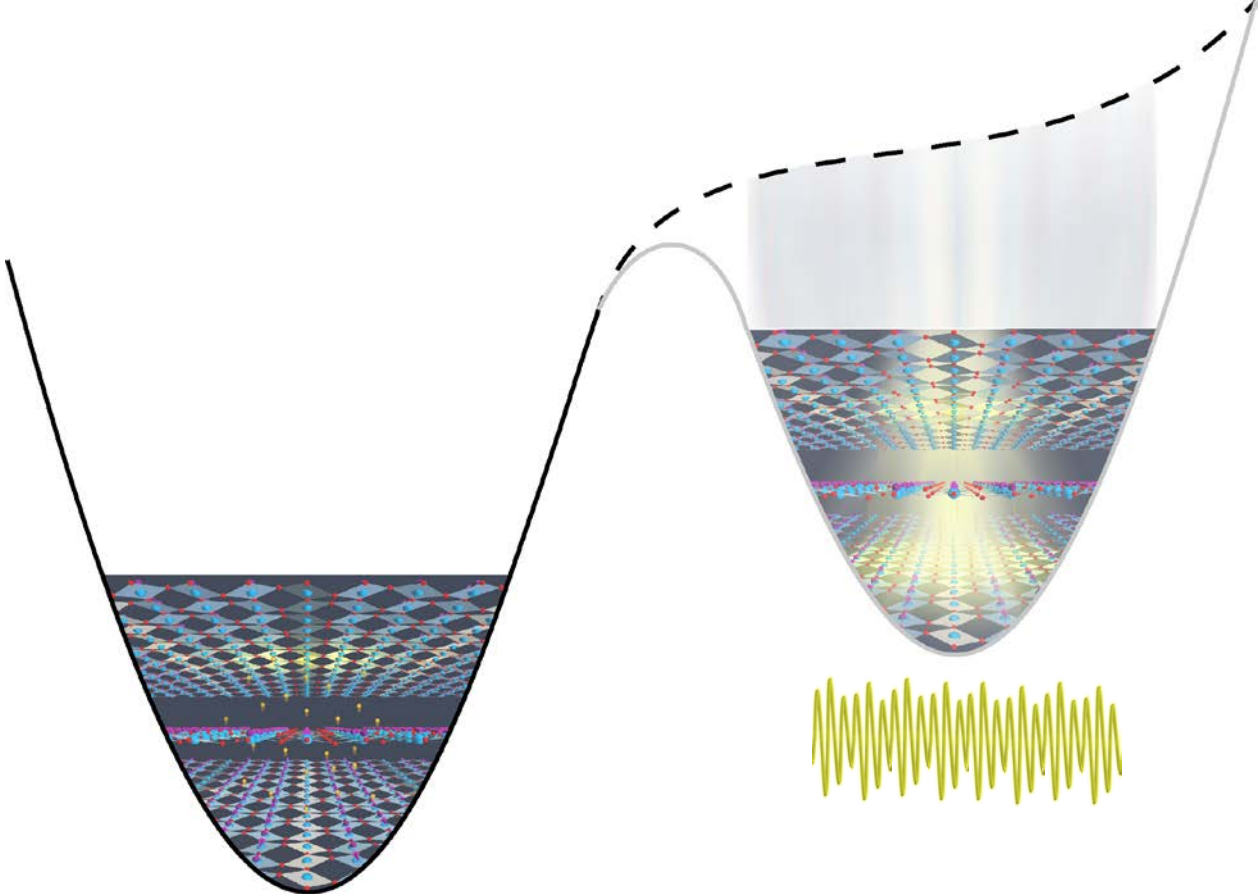
Dynamical control (1): switching into hidden phases



Dynamical control (2): creating new order by driving

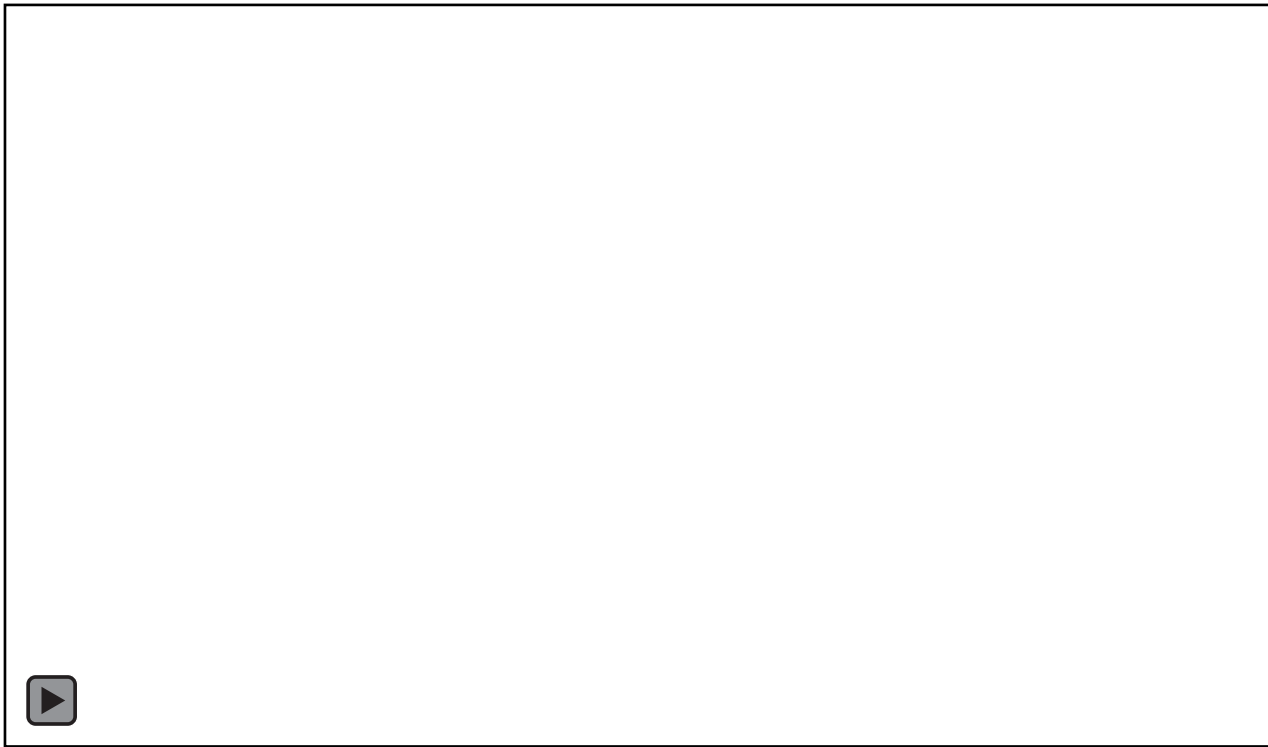


Dynamical control (2): creating new order by driving



Periodically driven systems ARE different

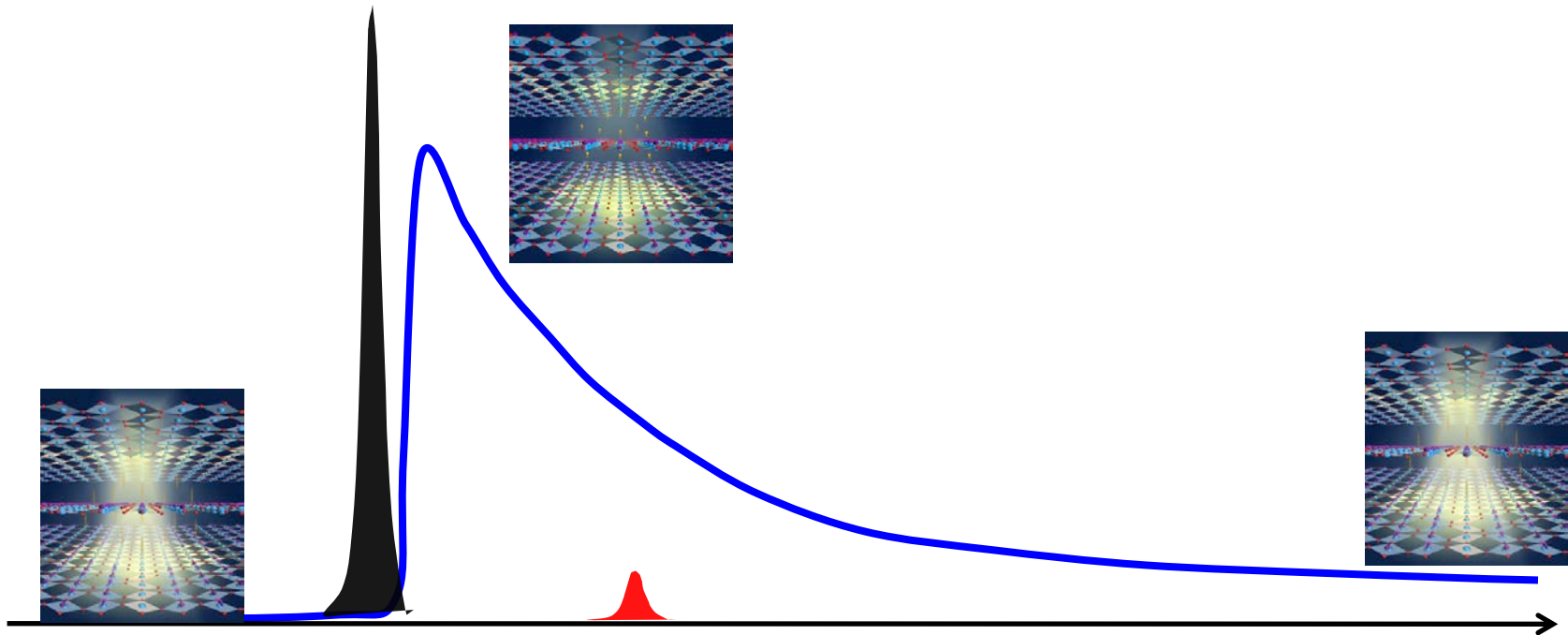
Driven systems often exhibit new regions of stability. A famous example – take a pendulum and vibrate its pivot point:



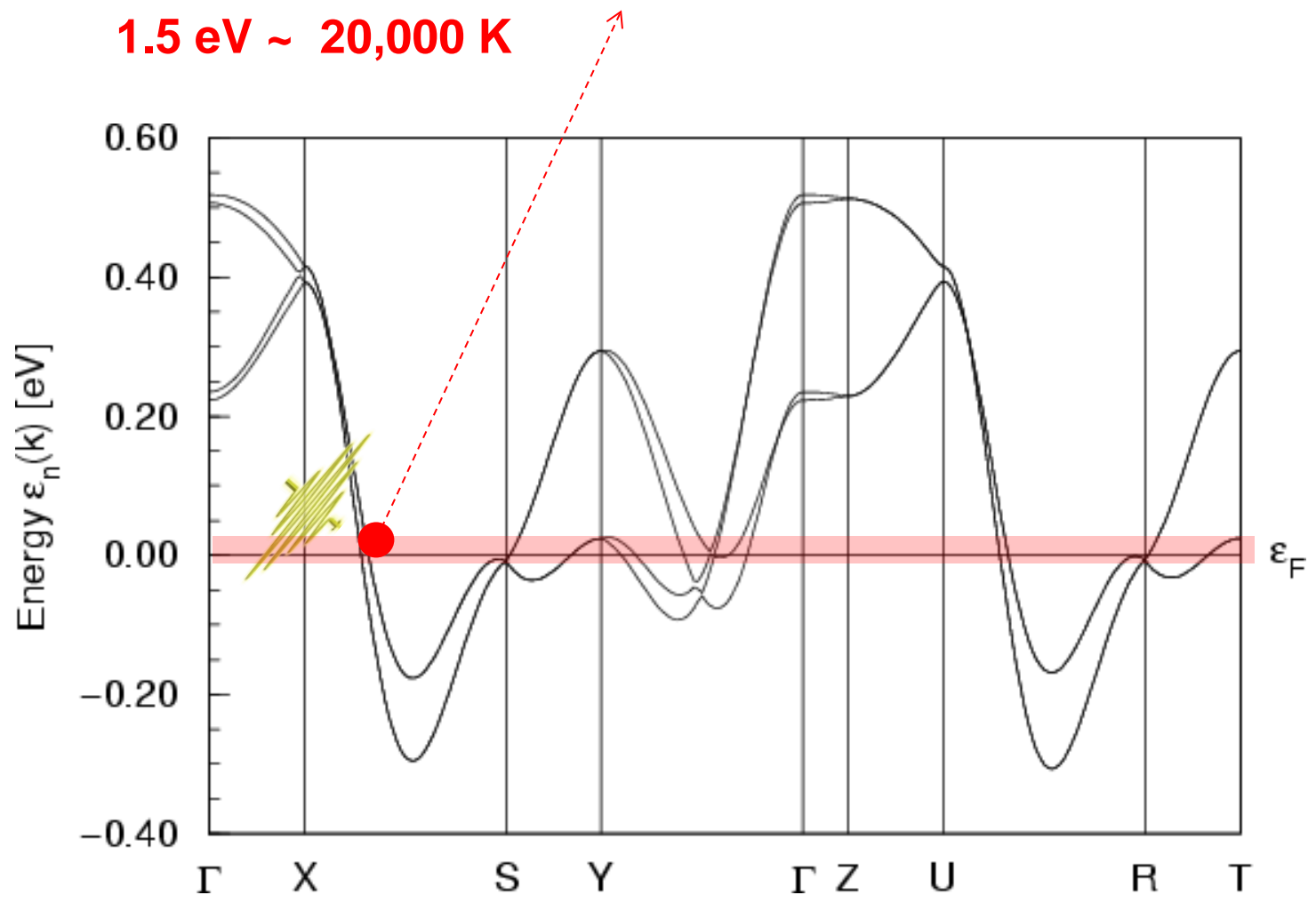
P.L. Kapitza, "Dynamic stability of a pendulum with an oscillating point of suspension,"
Zh. Eksp. Teor. Fiz. 21, 588 (1951)

L.D. Landau and E.M. Lifschitz *Mechanics* (Pergamon, Oxford 1976)

Dynamical phenomena: experiments



Stay away from visible light !

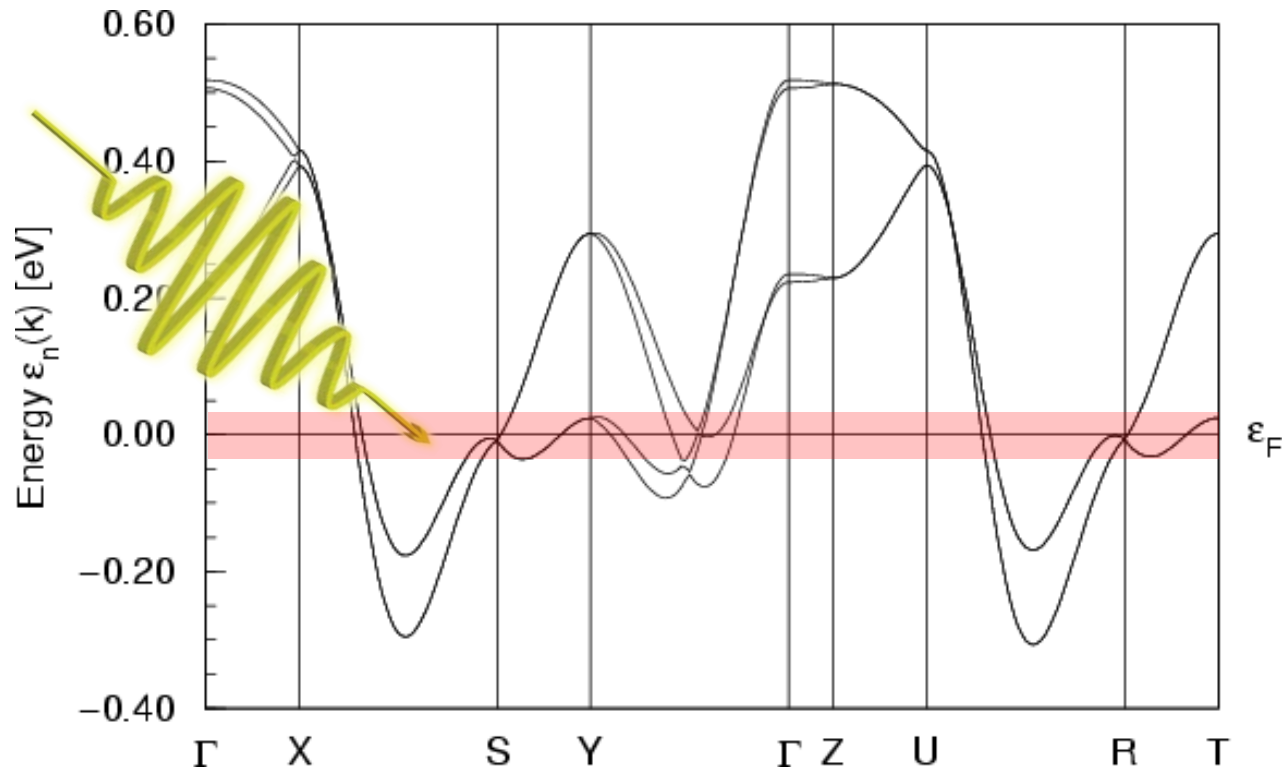


High energy scales

Incoherent

Entropy

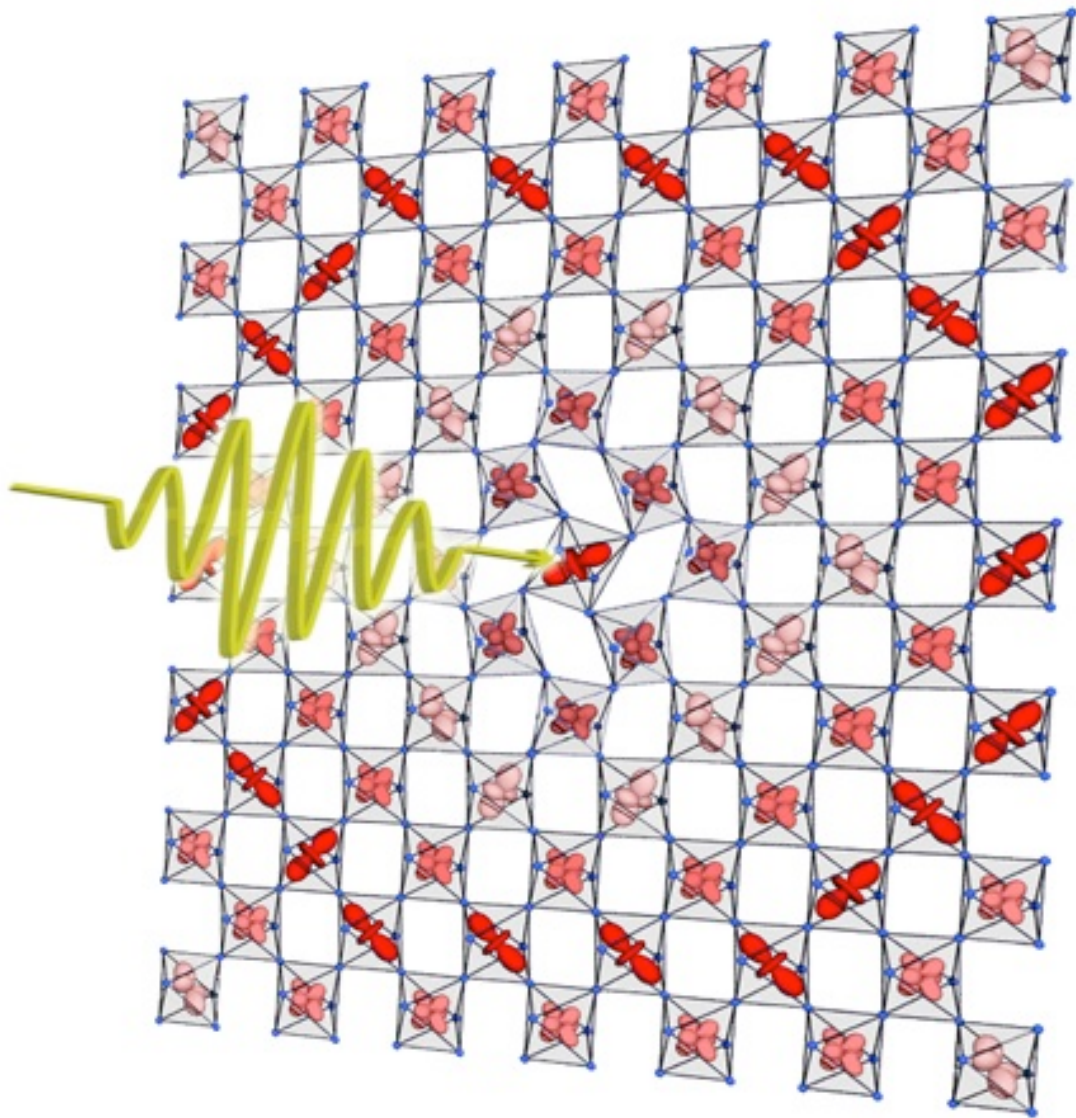
1 THz ~ 50 K ~ 4 meV



Low energy scales

Long coherence times

Coherent control of the lattice

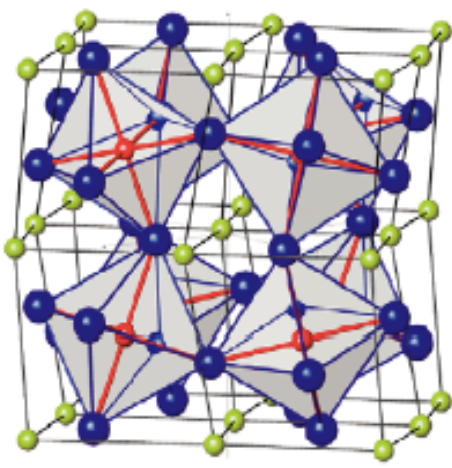
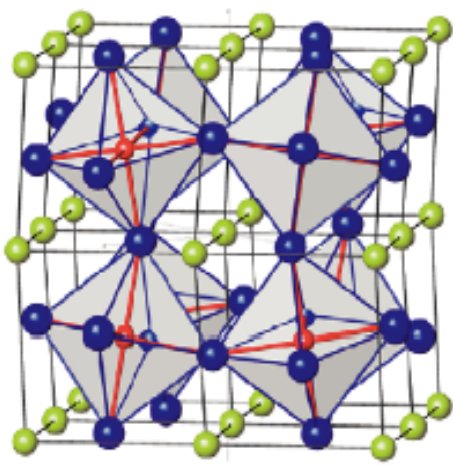
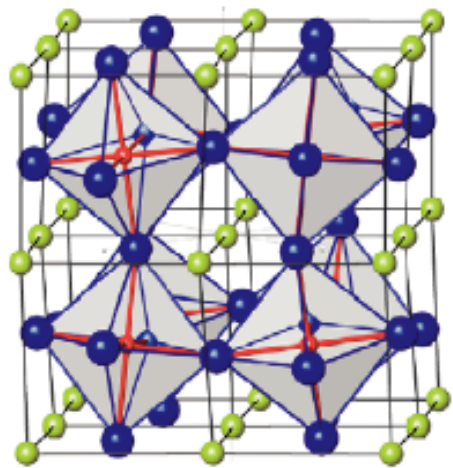
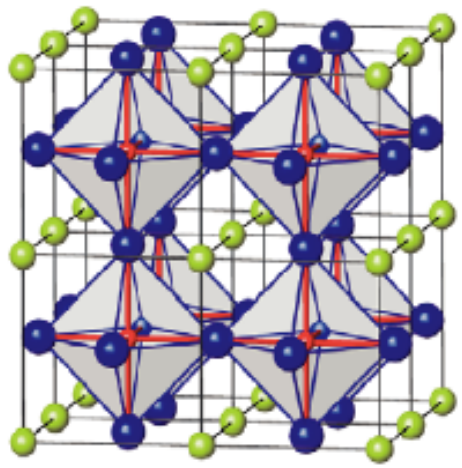


Why? e.g. controlling bond angles in oxides

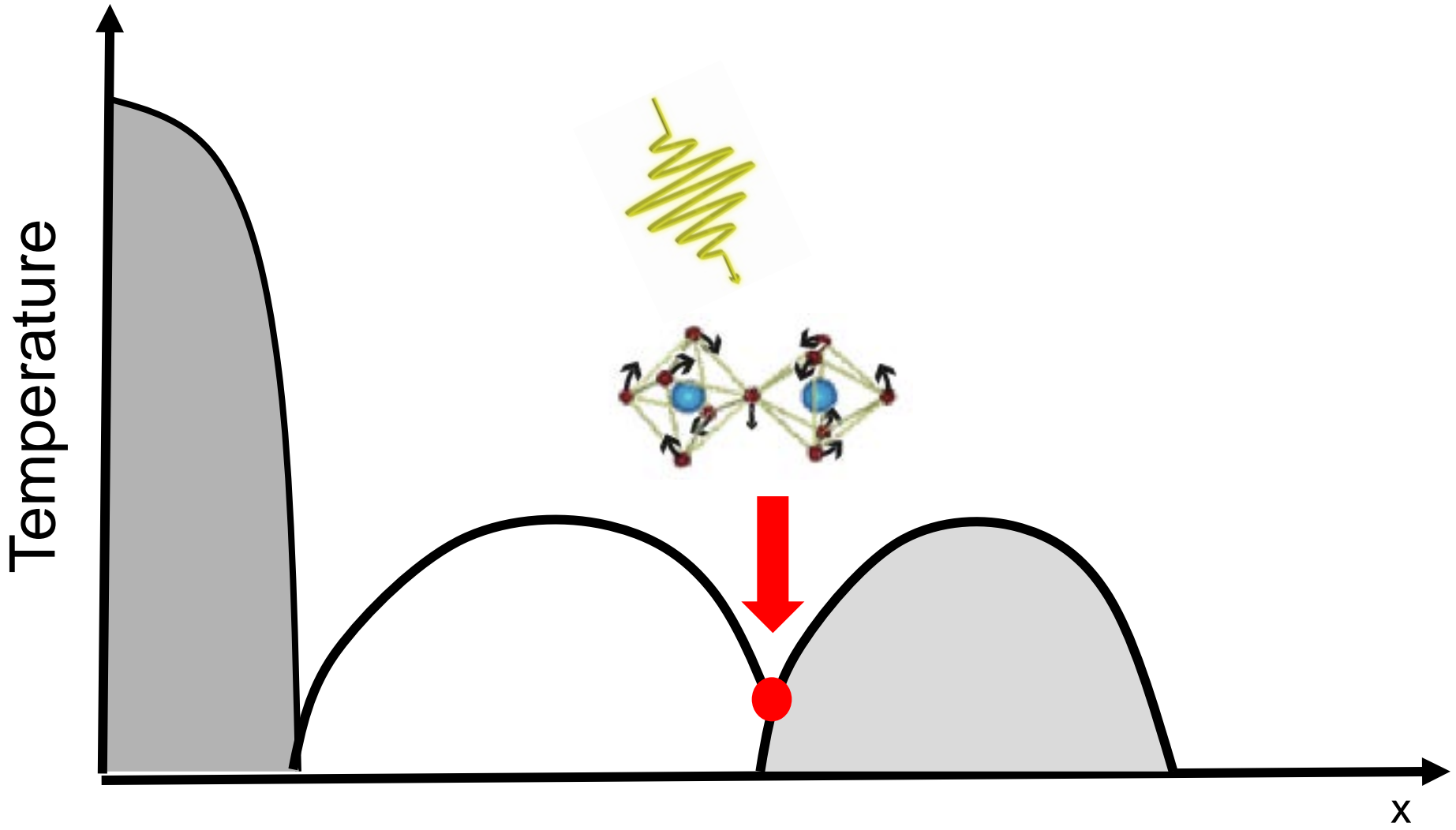
Metal



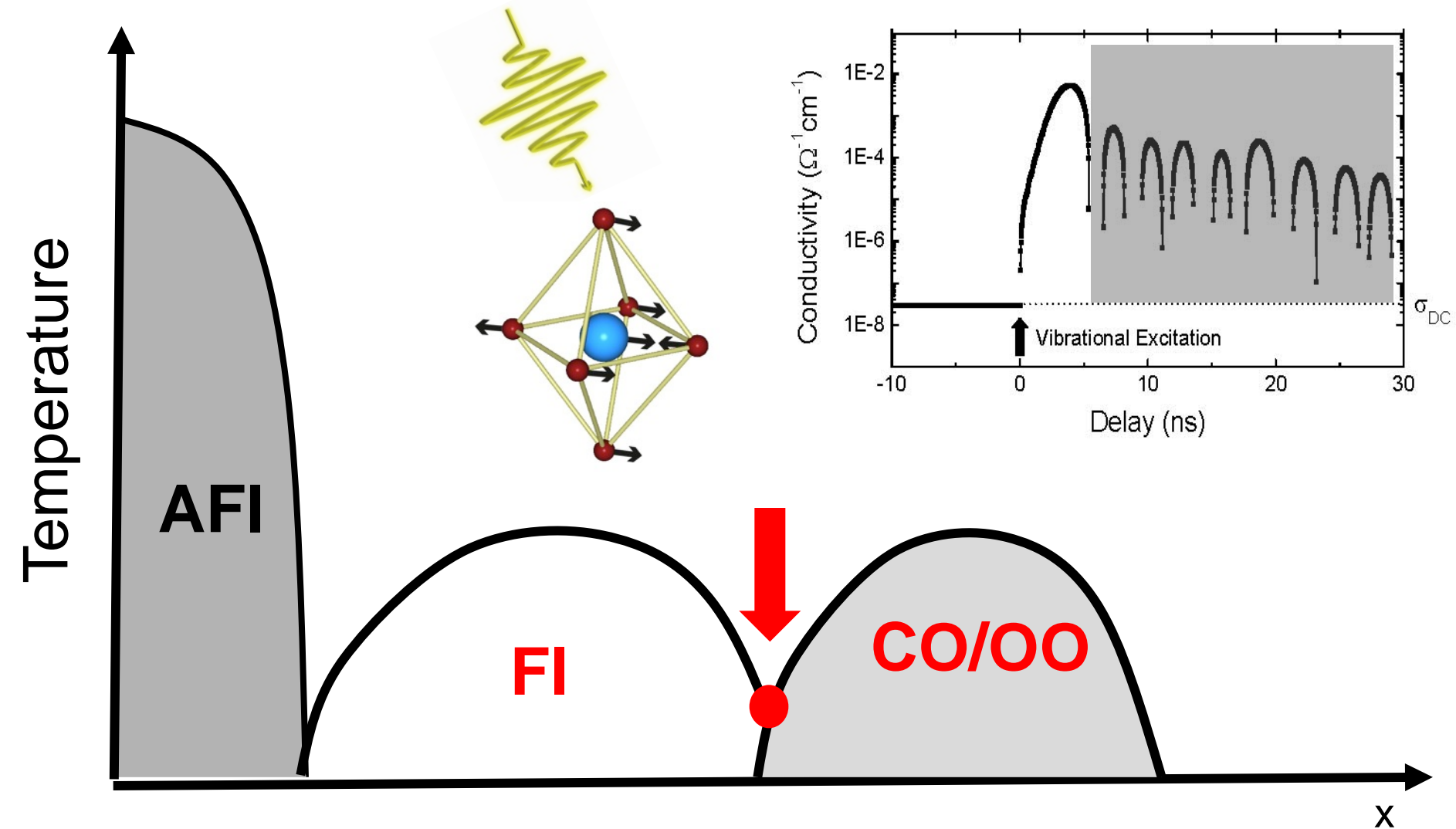
Insulator



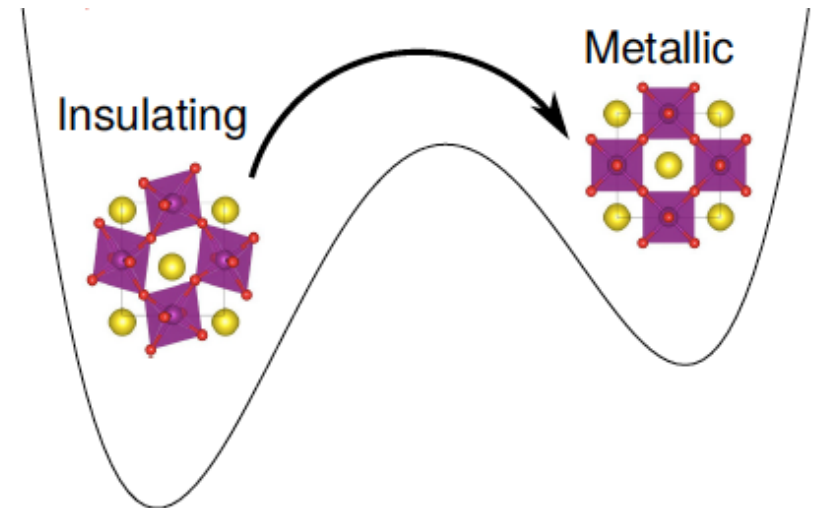
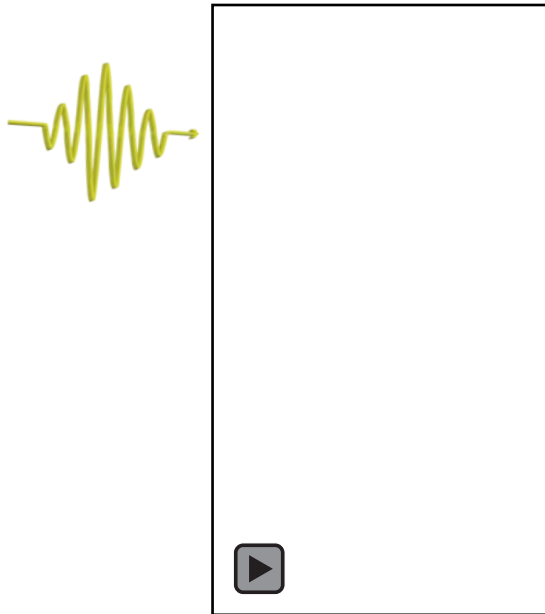
Can I control a bond angle with light



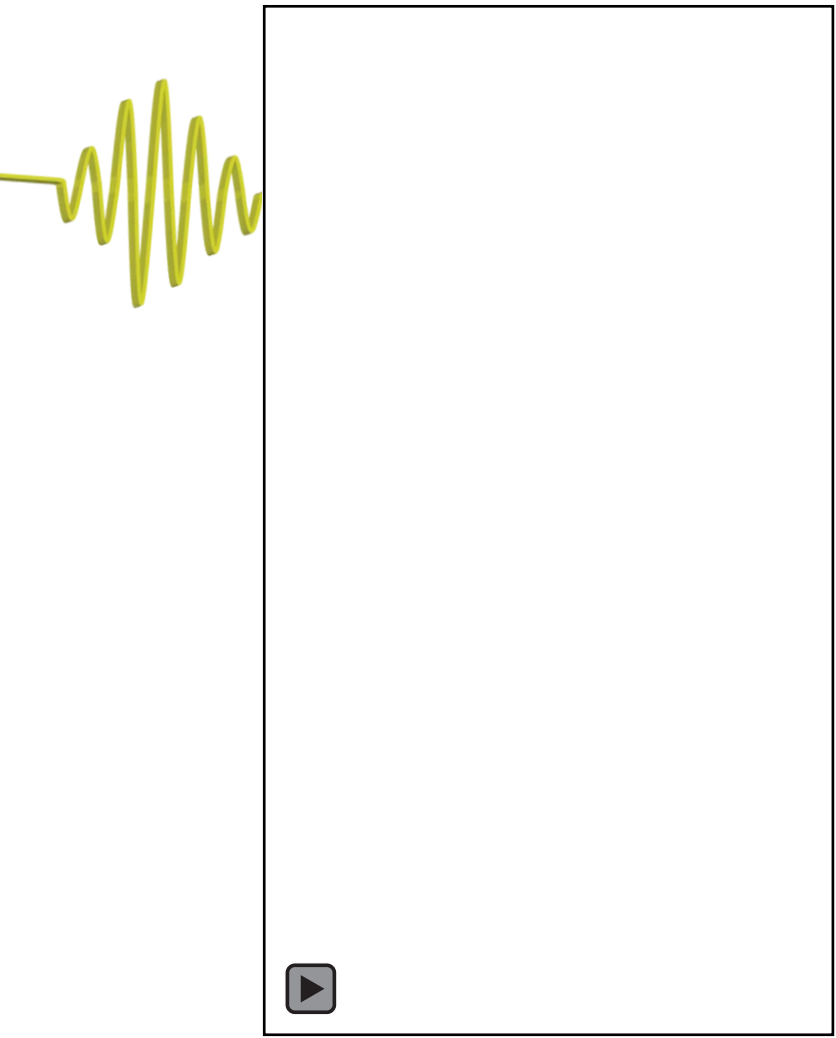
$\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$: Phonon Driven I-M Transition



How can optical excitation
displace the crystal bond angles?

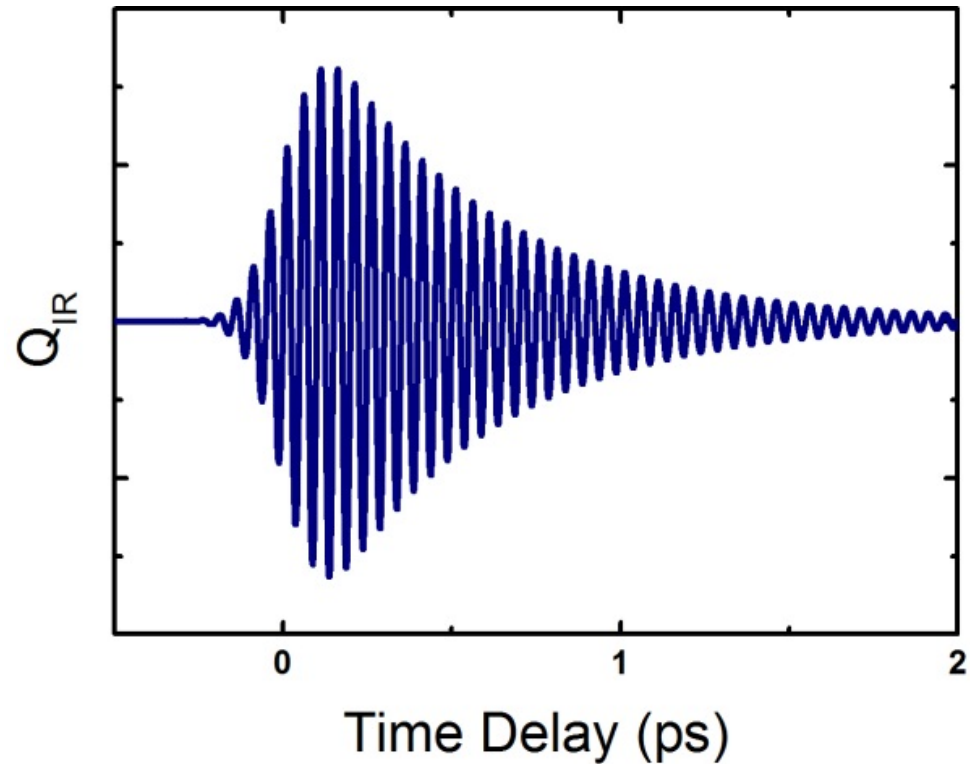


Linear response: no average displacement



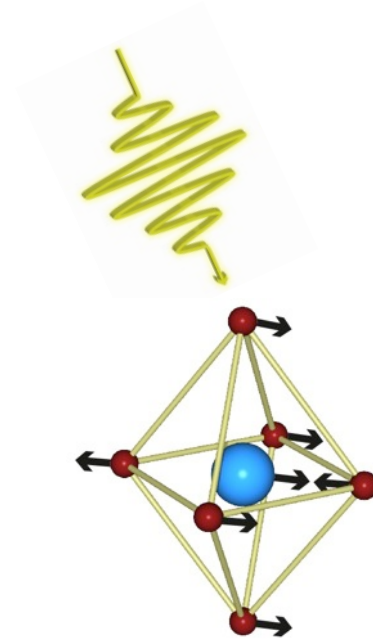
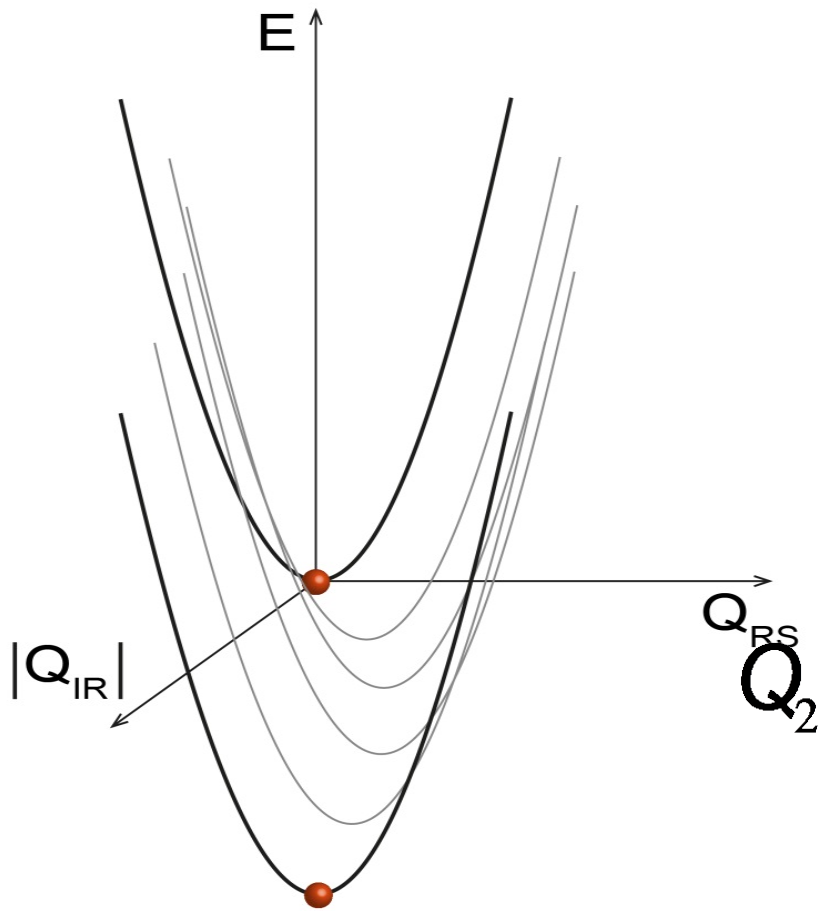
$$V = \frac{1}{2} \mu_{IR} \omega_{IR}^2 Q_{IR}^2$$

$$\ddot{Q}_{IR} + 2\gamma \dot{Q}_{IR} + \omega_{IR}^2 Q_{IR} = A \exp(i\omega t)$$



Anharmonic coupling to a second mode

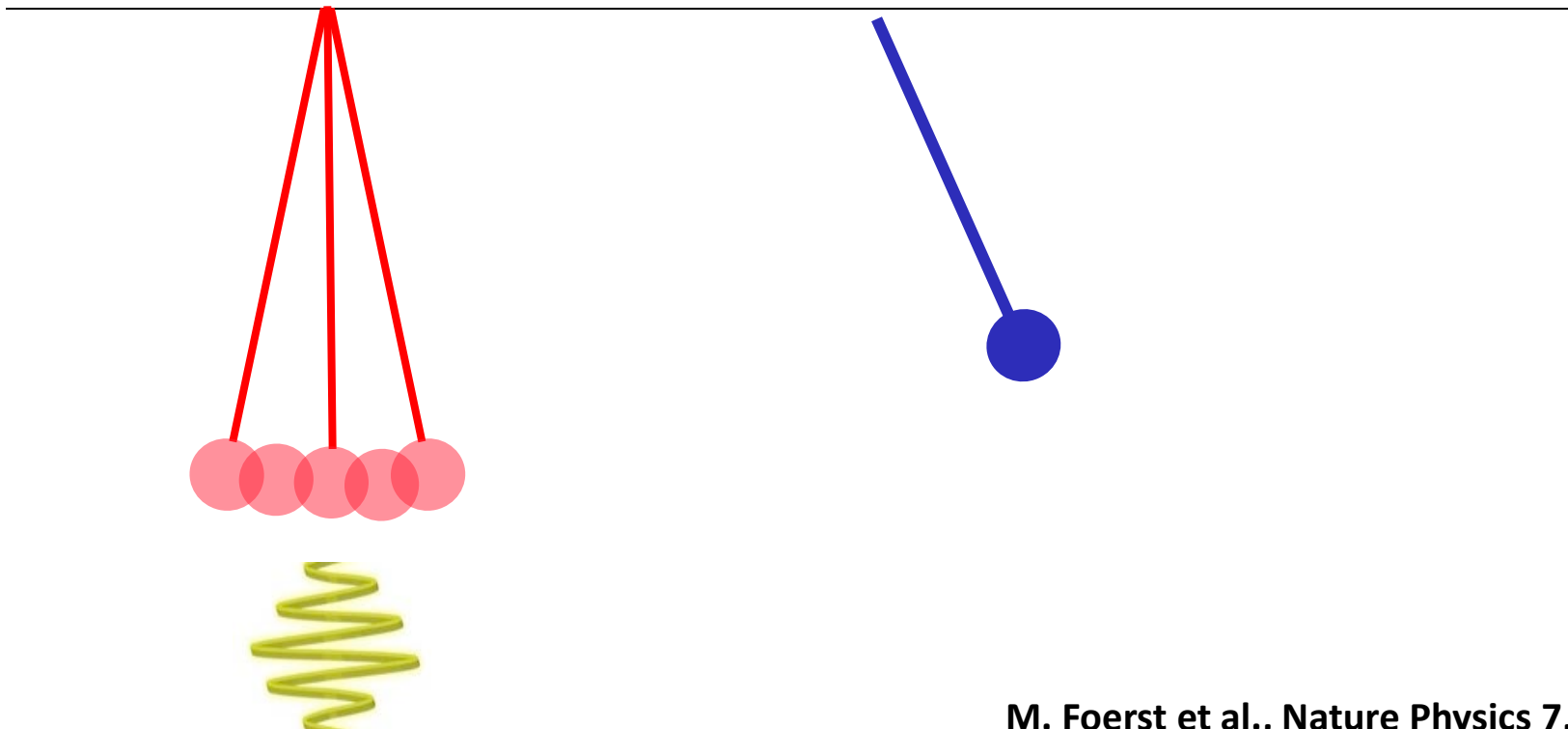
$$V = \frac{1}{2} \mu_{IR} \omega_{IR}^2 Q_{IR}^2 + NAQ_{IR}^2 Q_2$$

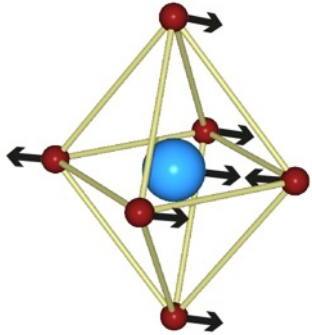


Equations of Motion: Two coupled oscillators

$$(\ddot{Q}_{IR} + 2\gamma\dot{Q}_{IR} + \omega_{IR}^2 Q_{IR}) = A \exp(i\omega t)$$

$$(\ddot{Q}_2 + 2\gamma\dot{Q}_2 + \omega_2^2 Q_2) = BQ_{IR}^2$$





Q_{IR} of B_{1u} symmetry

As in: $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$

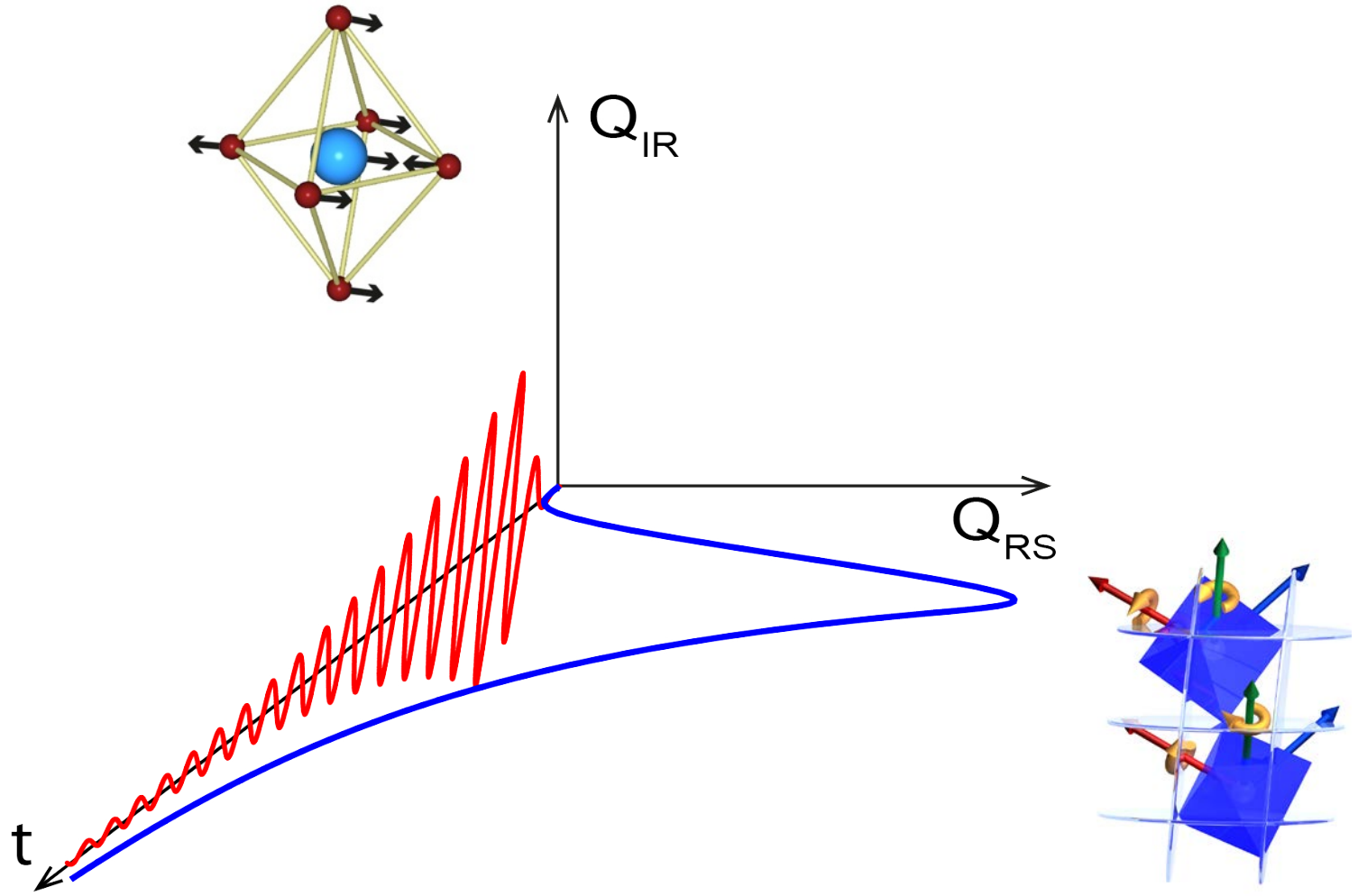
$\text{La}_{1.5}\text{Ca}_{0.5}\text{MnO}_4$

$\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$

$$Q_{\text{IR}}^2 Q_2 \neq 0$$

only if Q_2 is a Raman mode of A_g symmetry

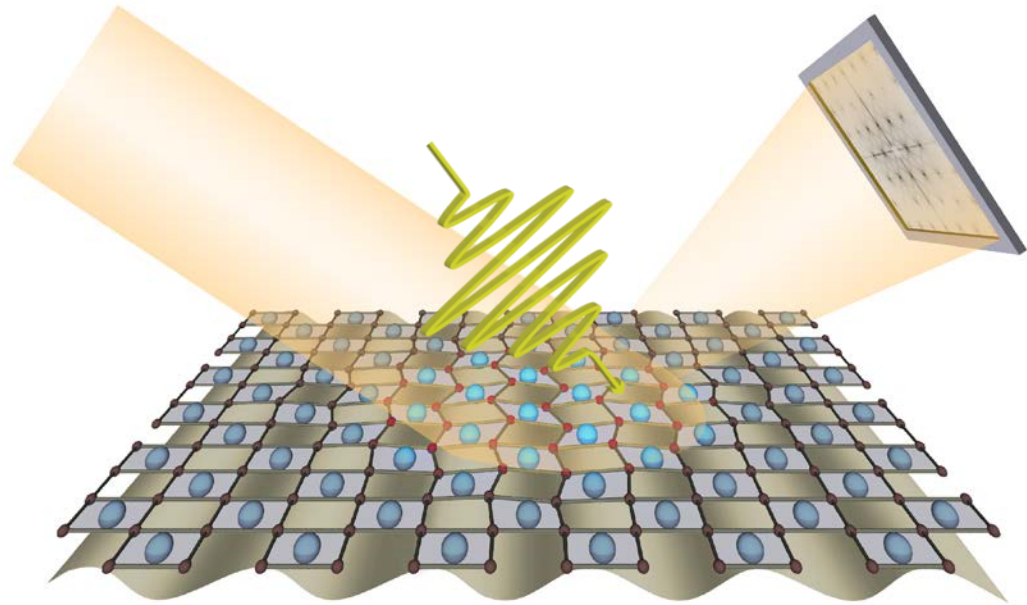
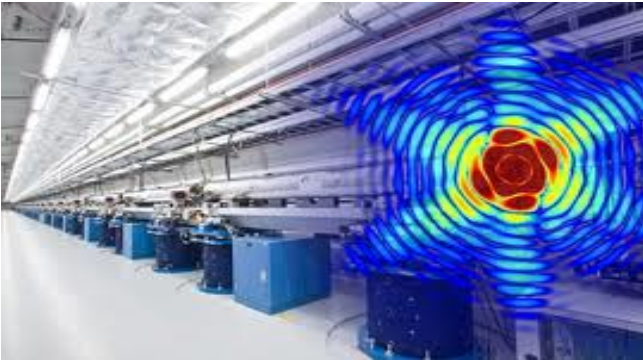
Rectified stretching leads to bending



Is there a nonzero average displacement ?

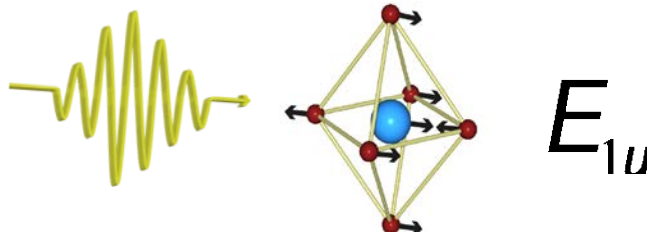
How far are the atoms being displaced ?

THz pump - Ultrafast x-ray diffraction probe

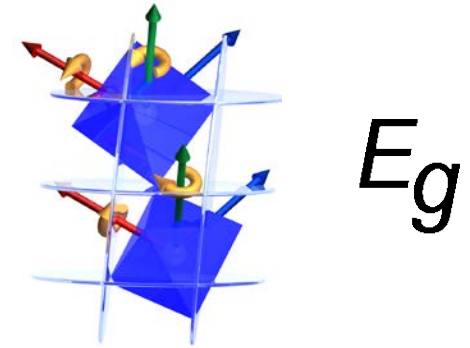


Femtosecond x-rays: quantify displacement

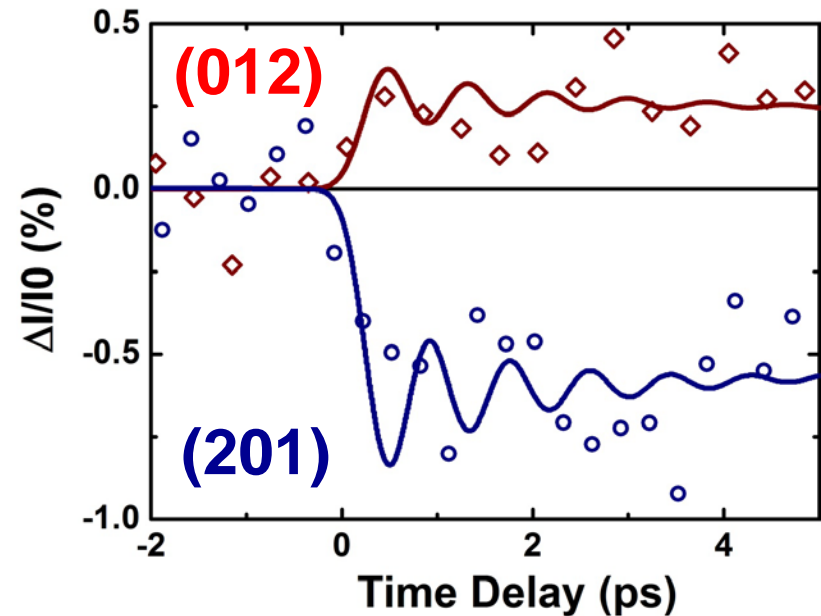
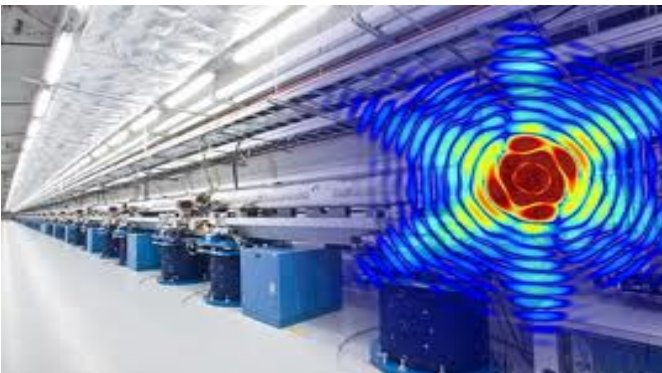
Mid-IR pump (E_{1u} mode)



Displacive field (E_g mode)

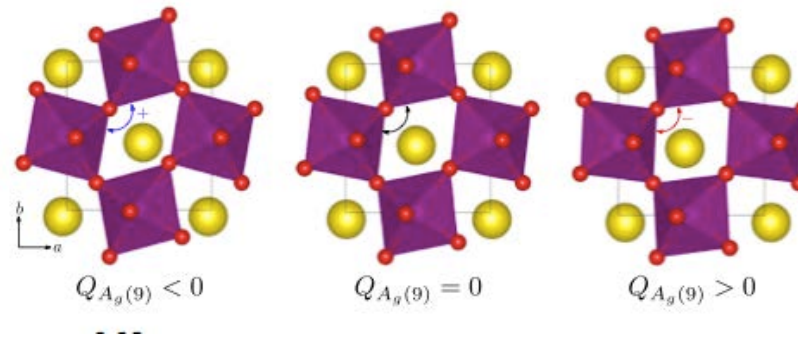
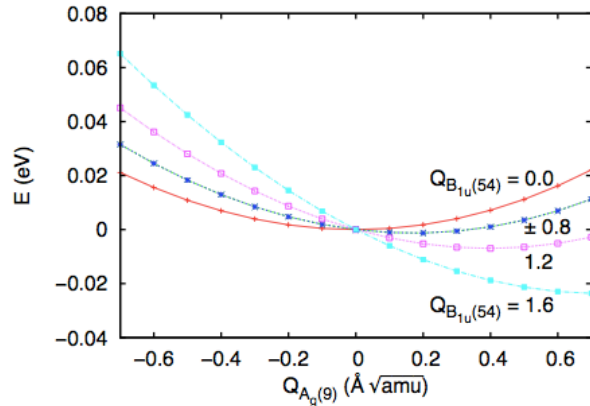


X-ray probe

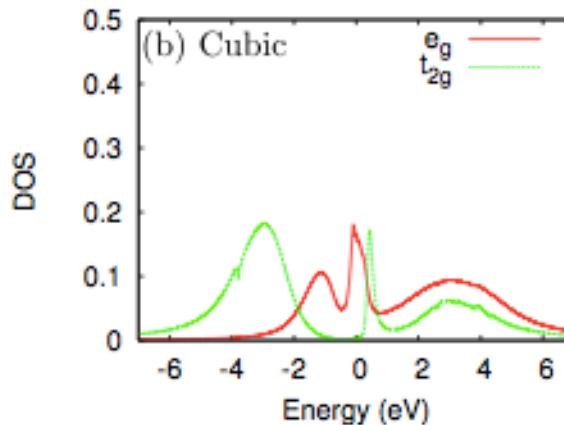
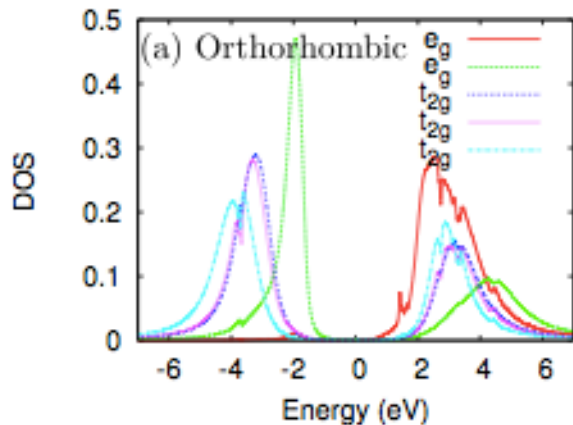


Theory: octahedral rotations make a metal

Frozen Phonon

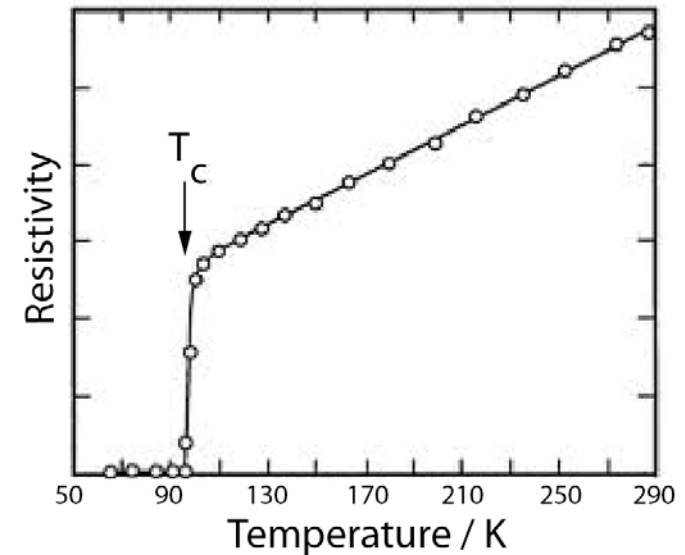
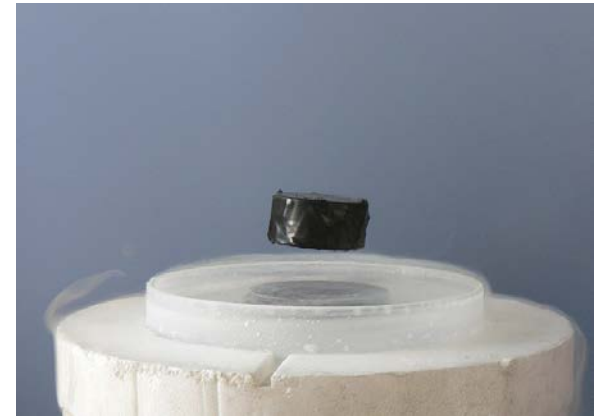
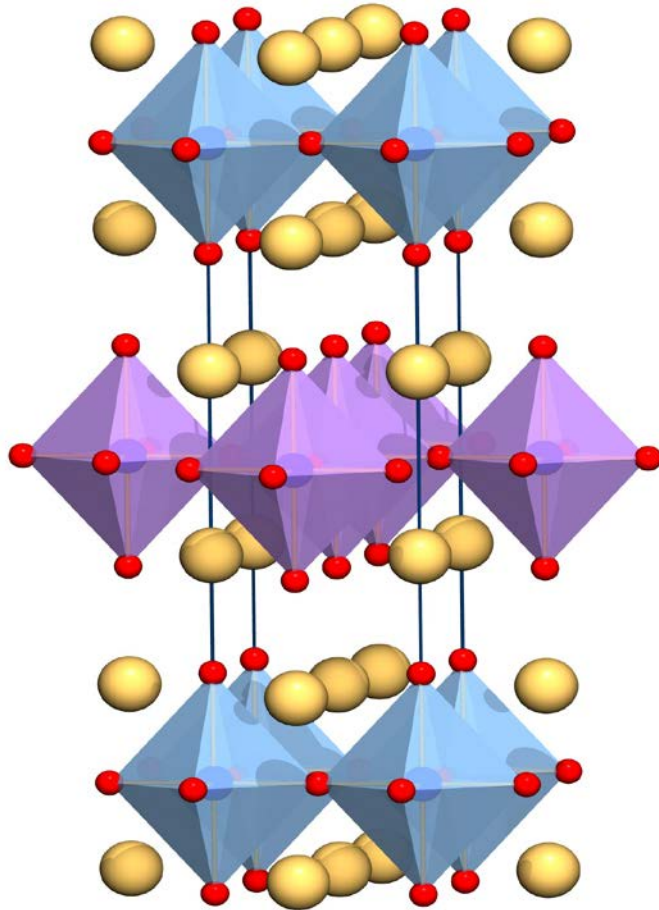


Electronic Structure in the distorted state -> metallic

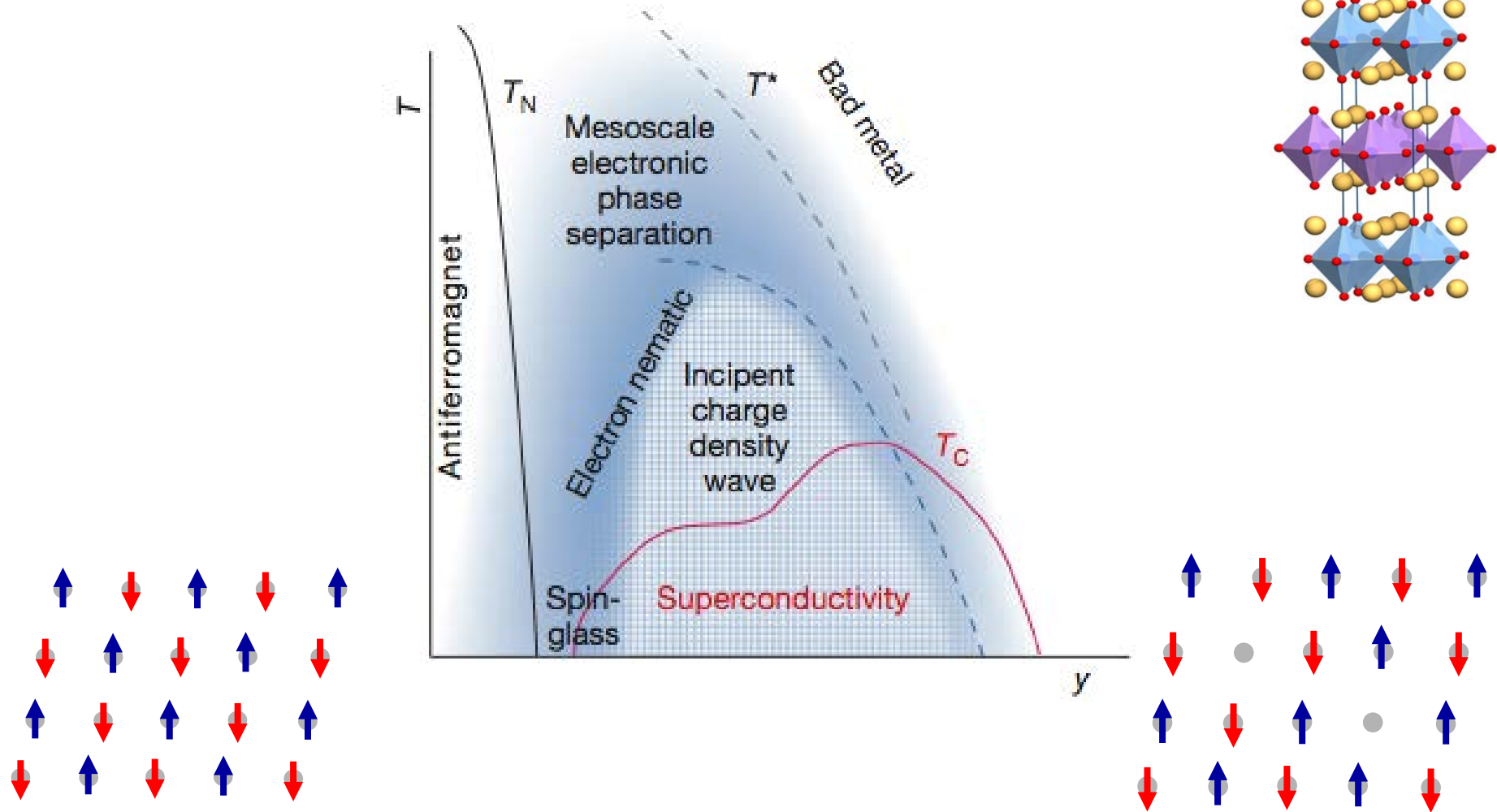


What else can I control ?

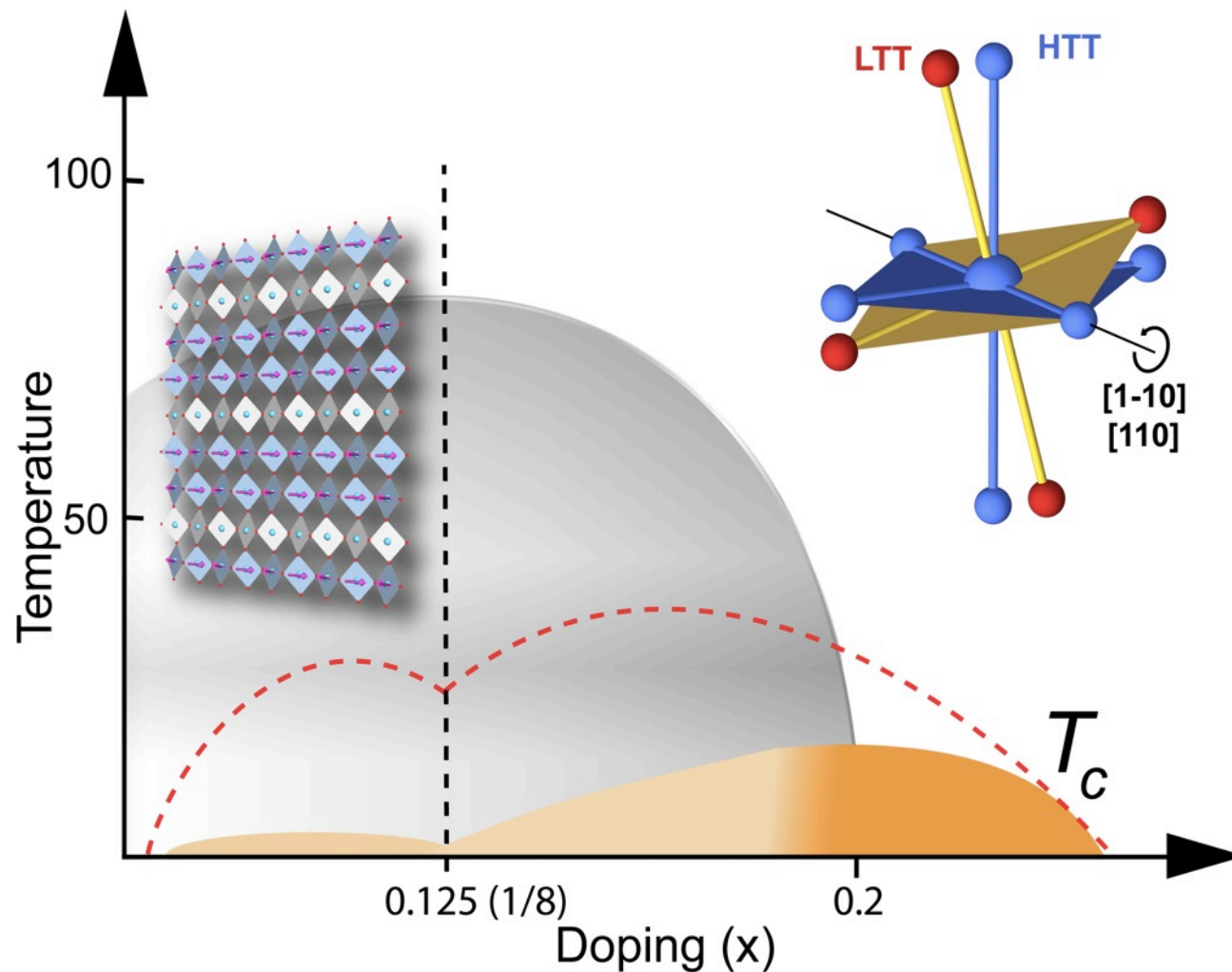
Below a critical temperature T_c resistivity vanishes



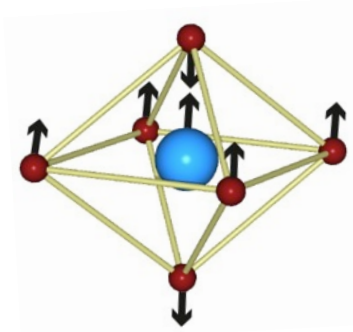
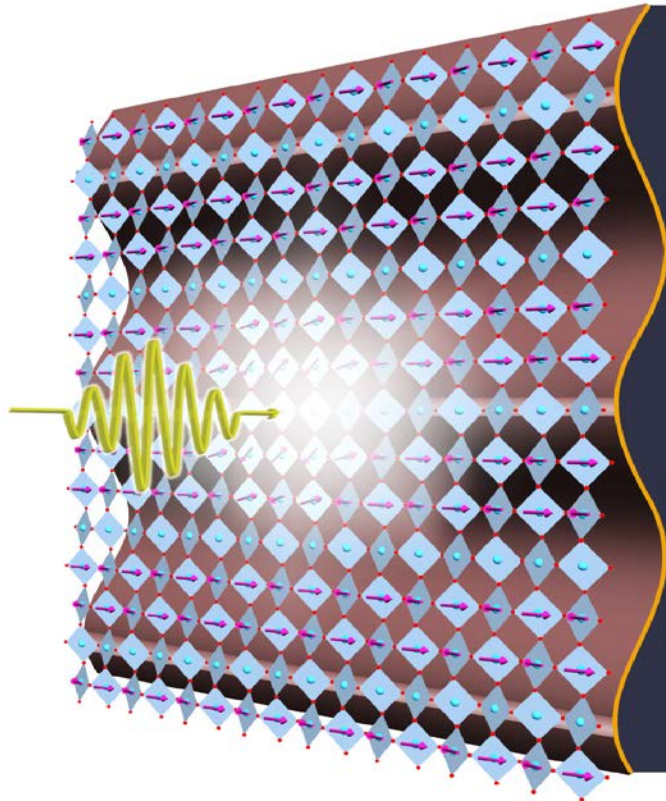
Competing orders can quench T_c



Eu:LSCO_{1/8} stripe charge order



Excitation of in plane Cu-O stretch

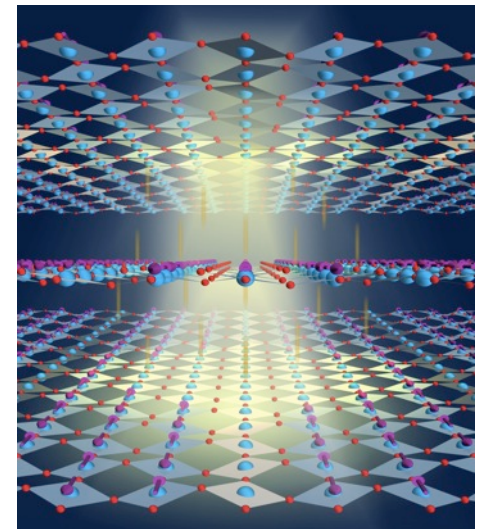
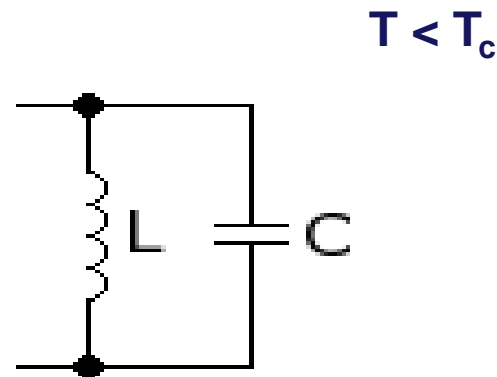
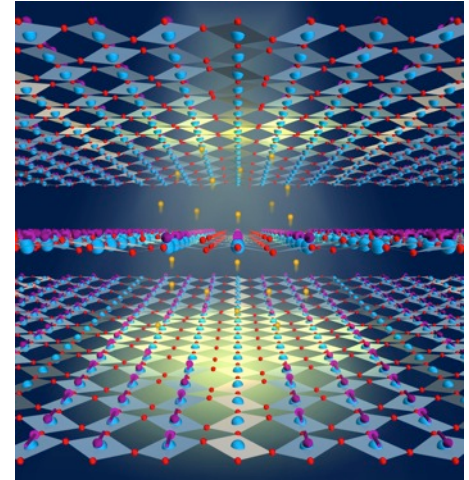
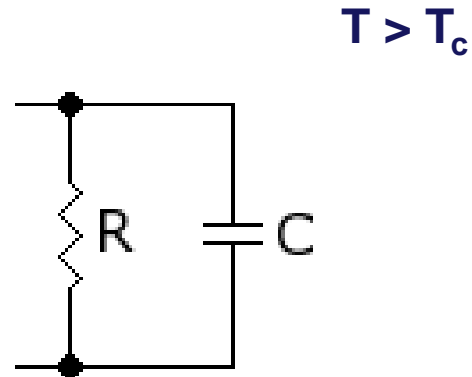
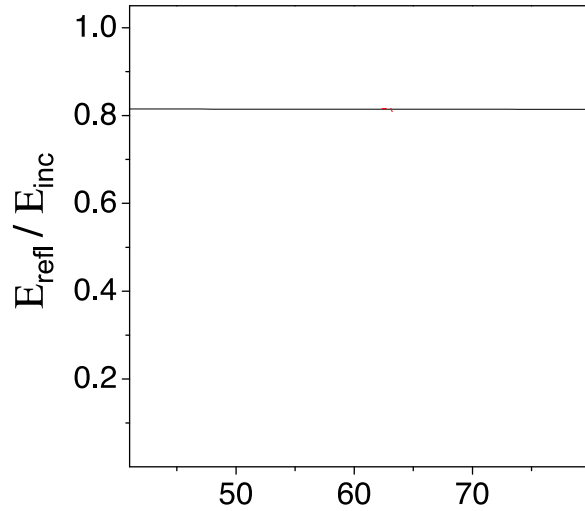


600 cm⁻¹

**16 μm wavelength
μJ pulses
MV/cm fields**

How do I recognize a transient superconductor ?

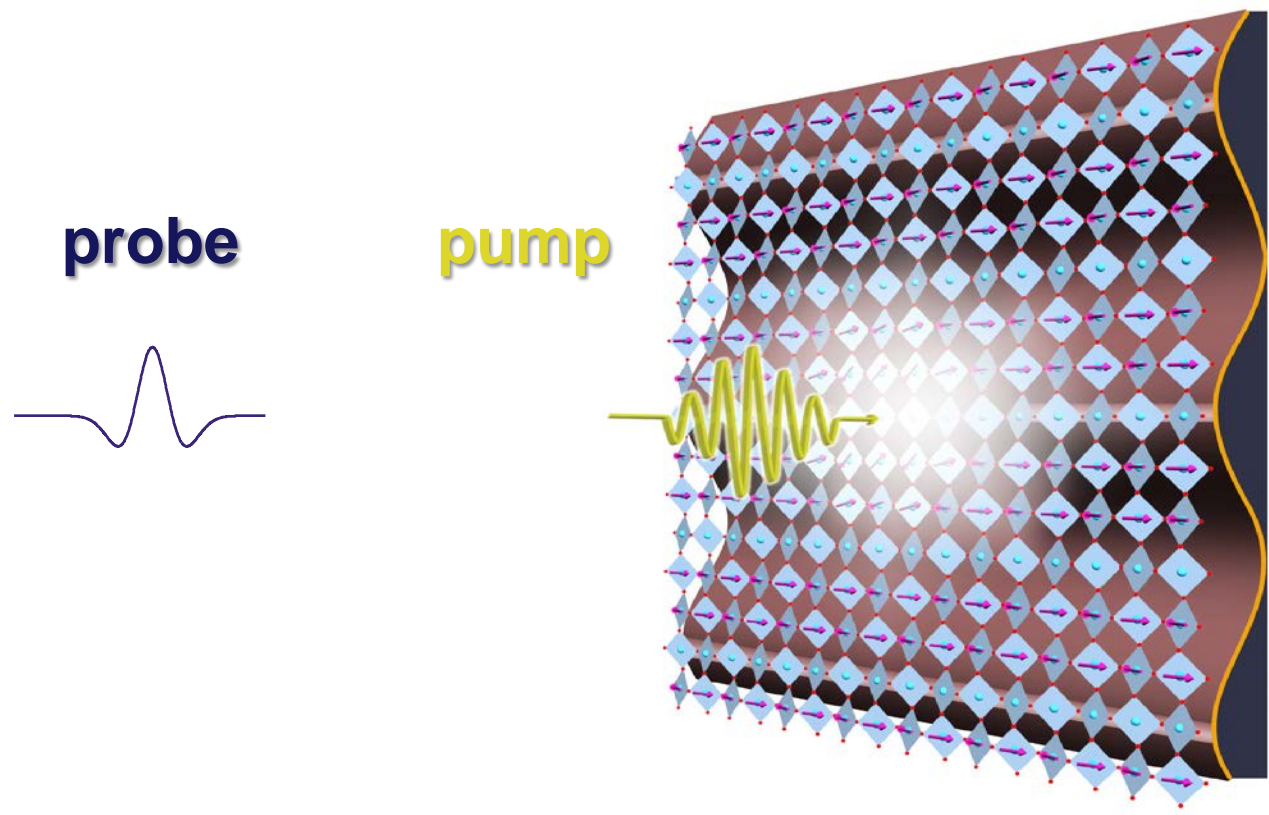
Josephson Plasmon



Kresin and Morawitz PRB (1988)

van der Marel and A. A. Tsvetkov Czech. J. Phys. (1996)

Mid-IR pump / THz Probe Spectroscopy



A light Induced Josephson plasma edge

Equilibrium LSCO

Superconducting (eq.)

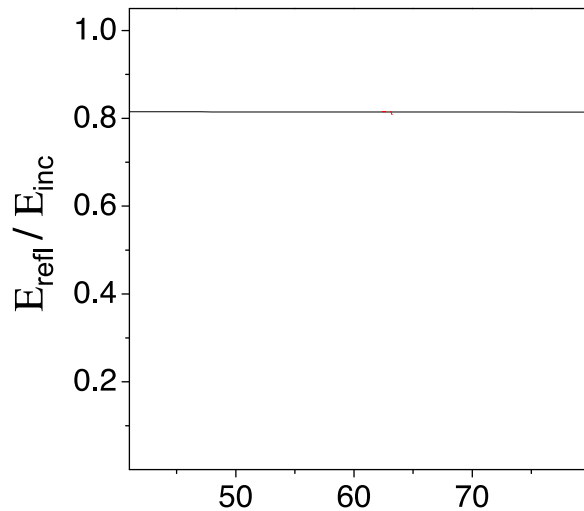
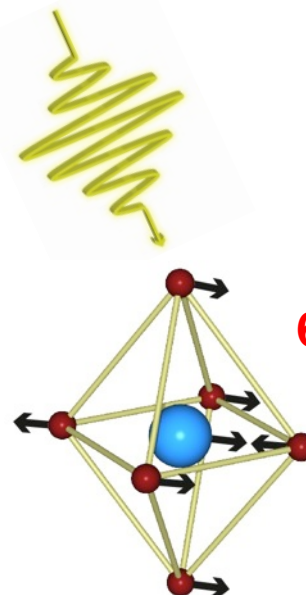
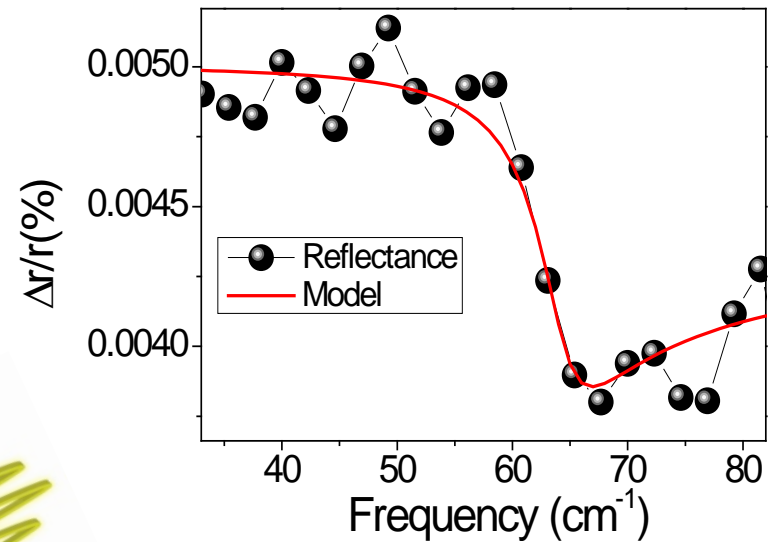


Photo-induced LESCO

Superconducting (non eq.)

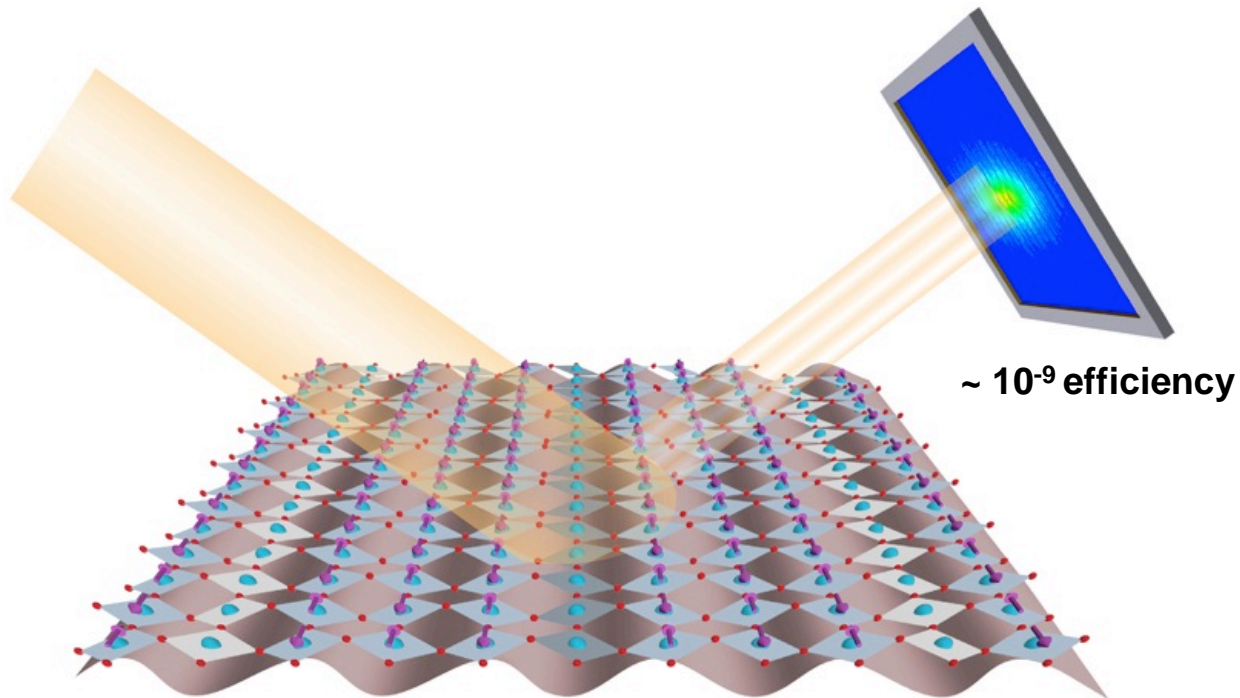


Am I melting charge stripes with light ?

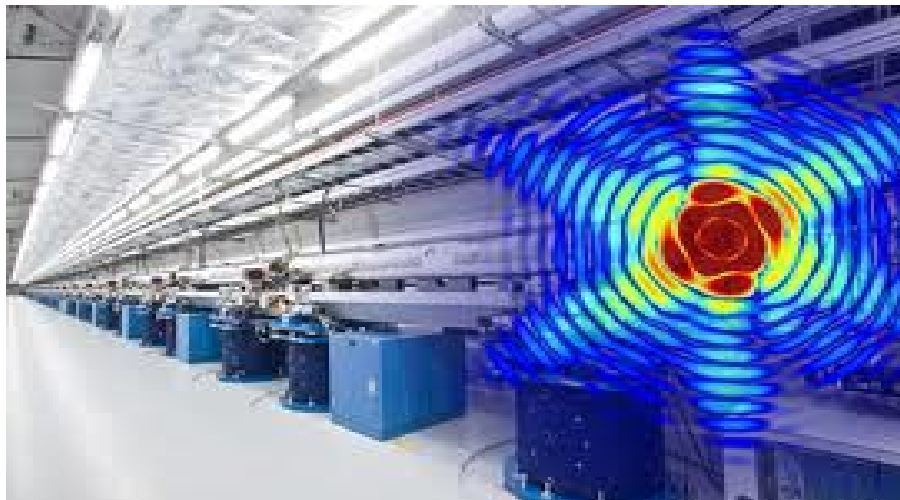
Charge stripes are seen by soft x-ray scattering

O Kedge

(0.25, 0, 0.65)

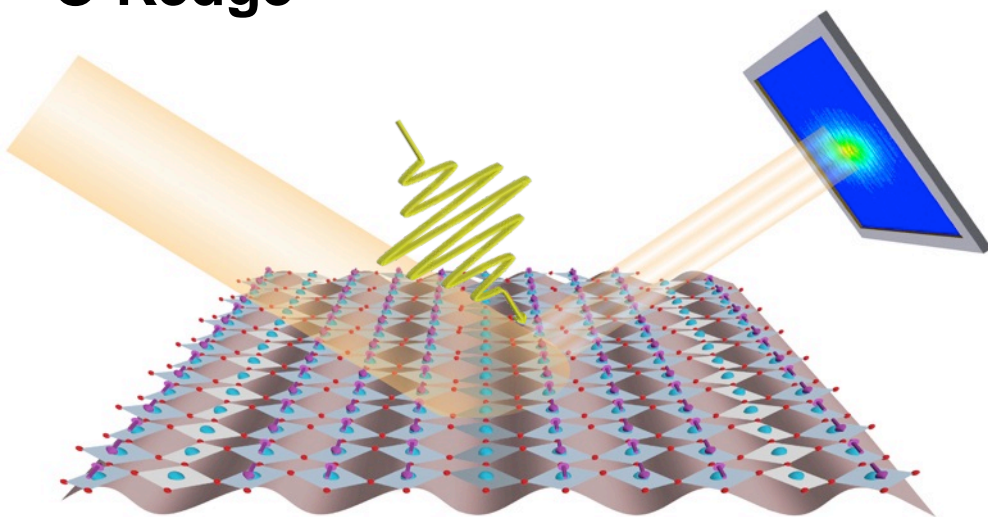


Ultrafast soft X-ray diffraction

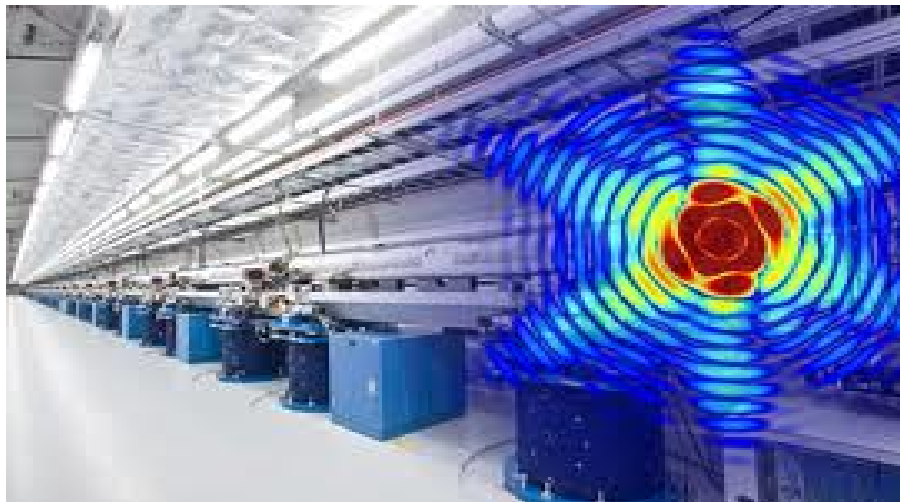


O Kedge

(0.25, 0, 0.65)

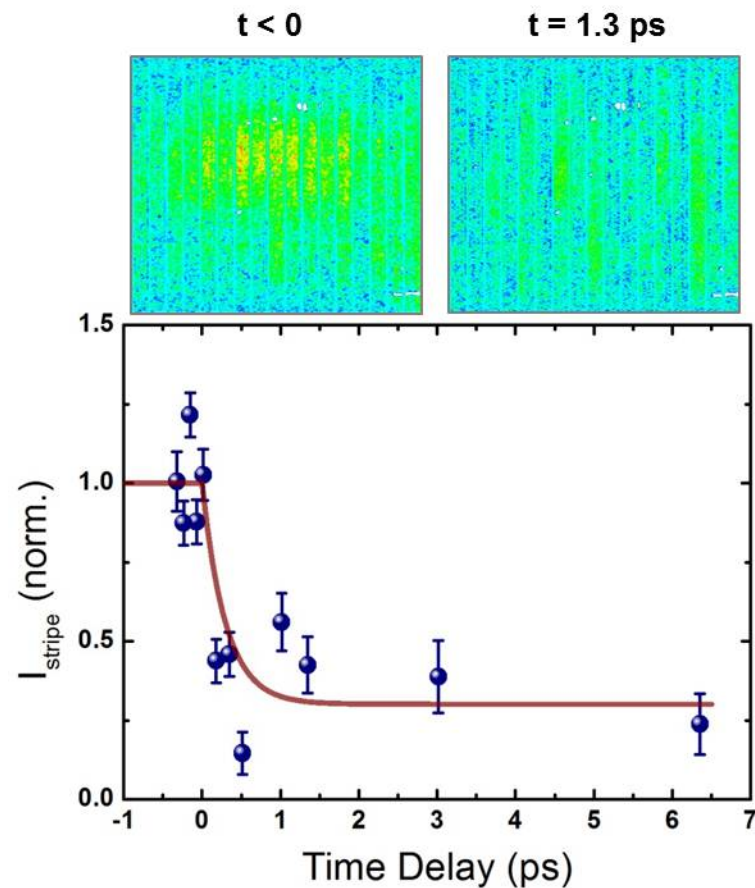
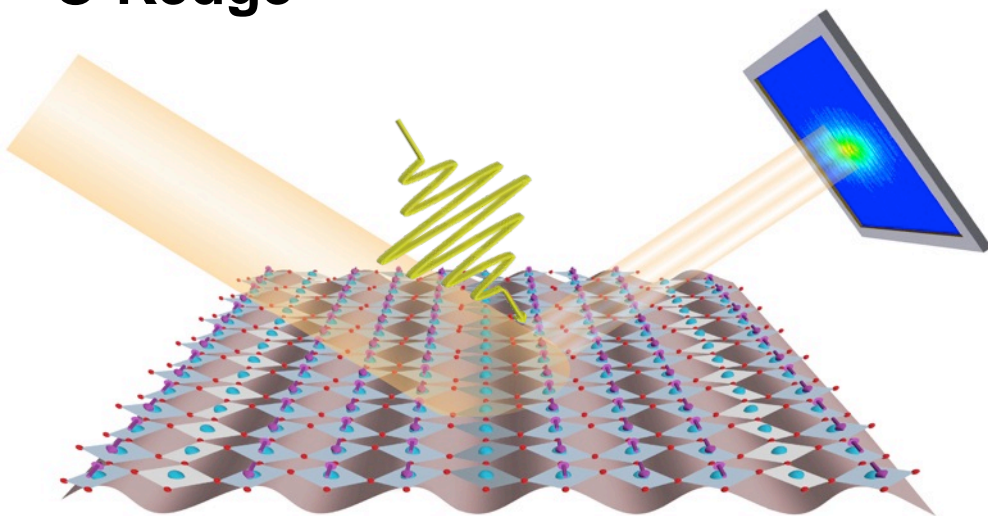


Ultrafast soft X-ray diffraction



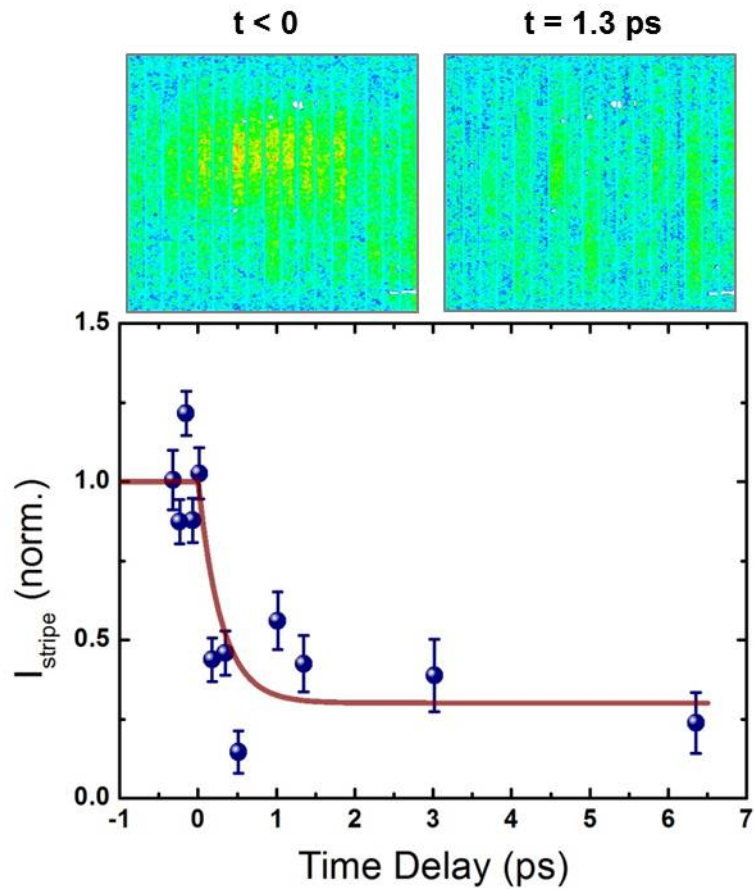
O Kedge

(0.25, 0, 0.65)

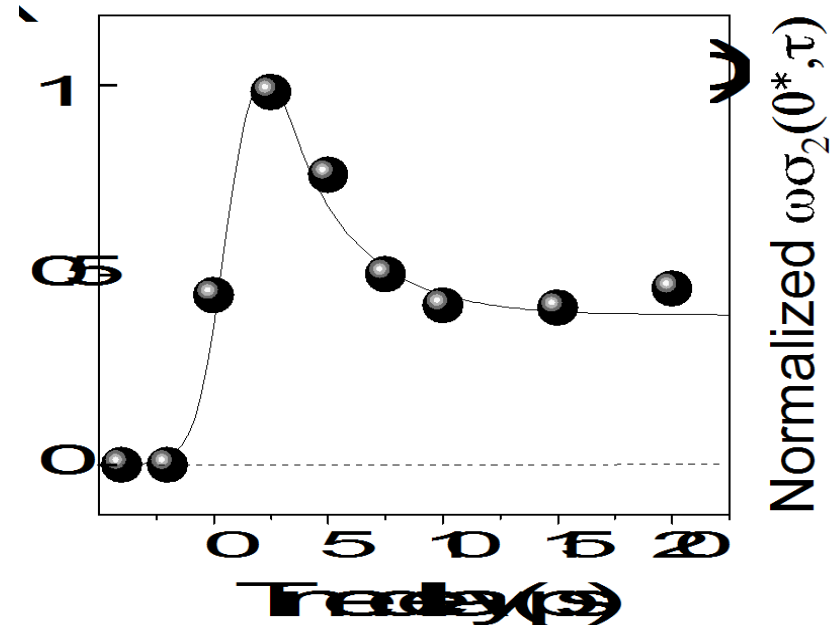


Charge stripe melting - superconductivity

- Charge Stripes melt concomitantly with the formation of the SC

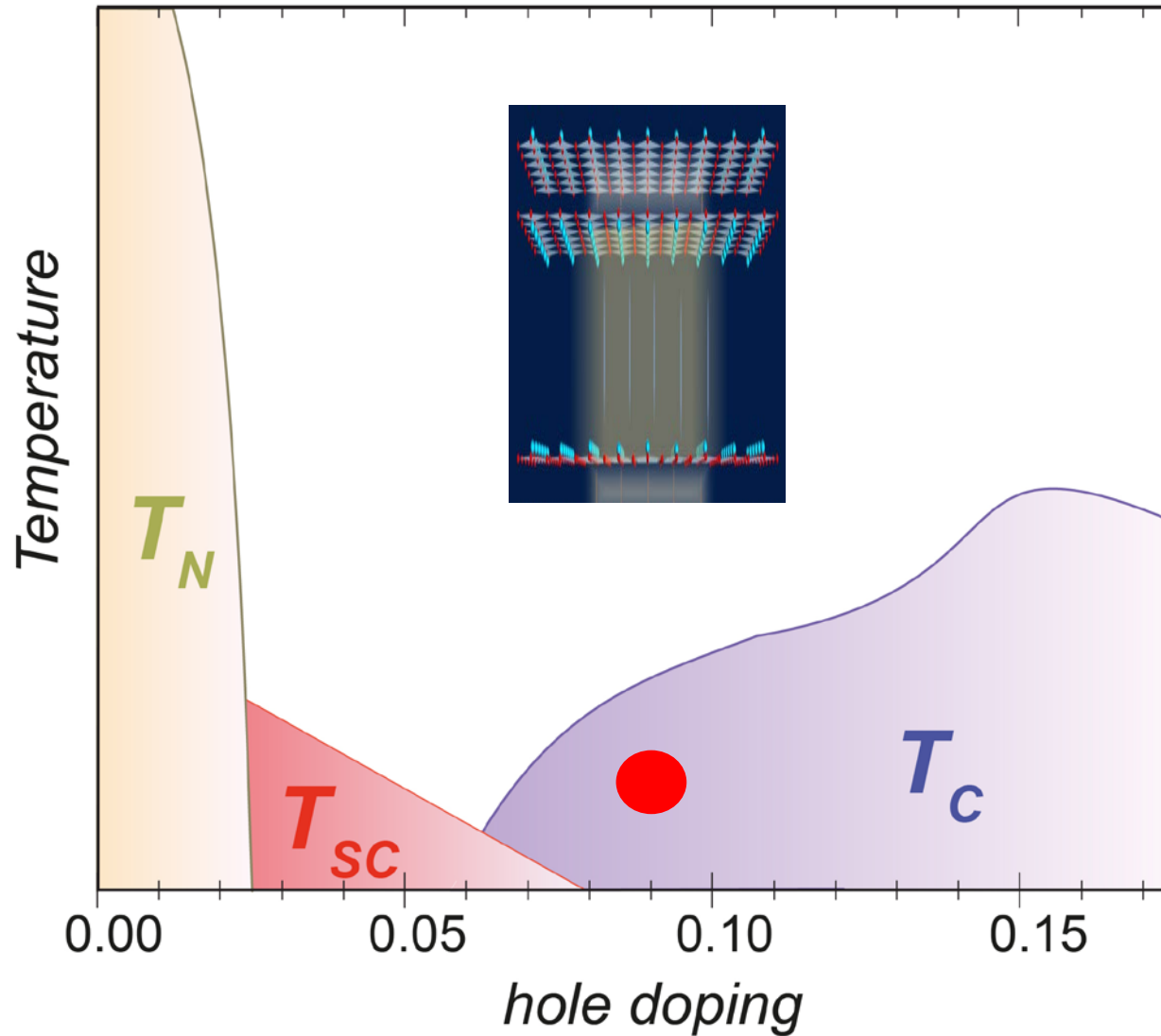


superconductivity

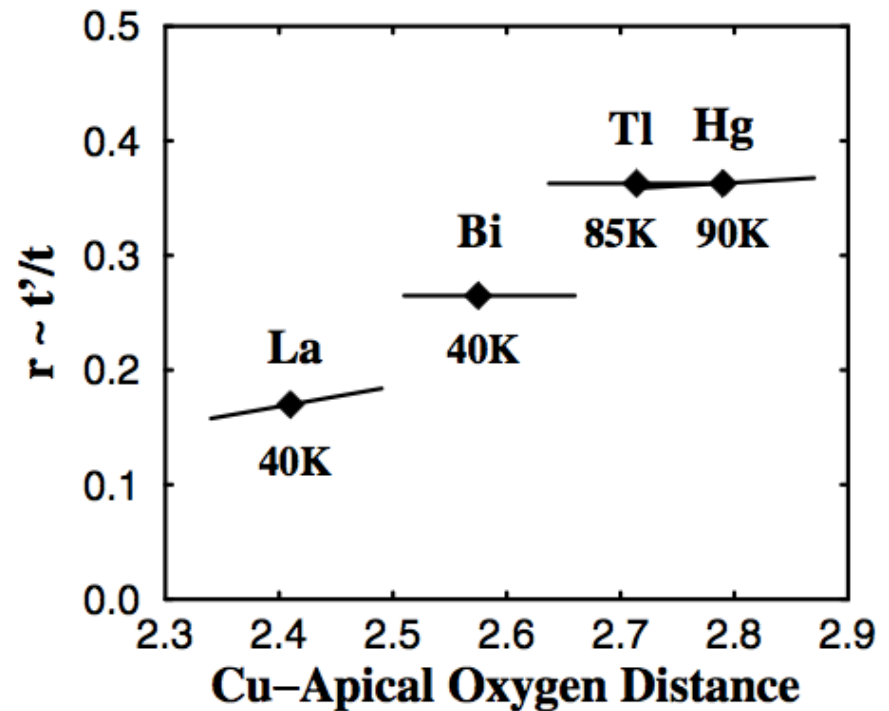
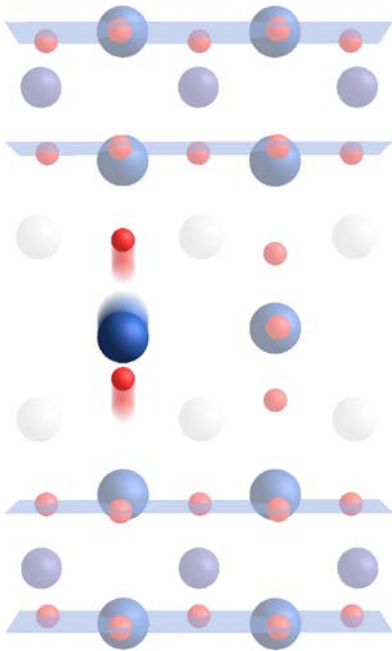


Can I do this in other cuprates ?

Bilayer cuprates: YBCO



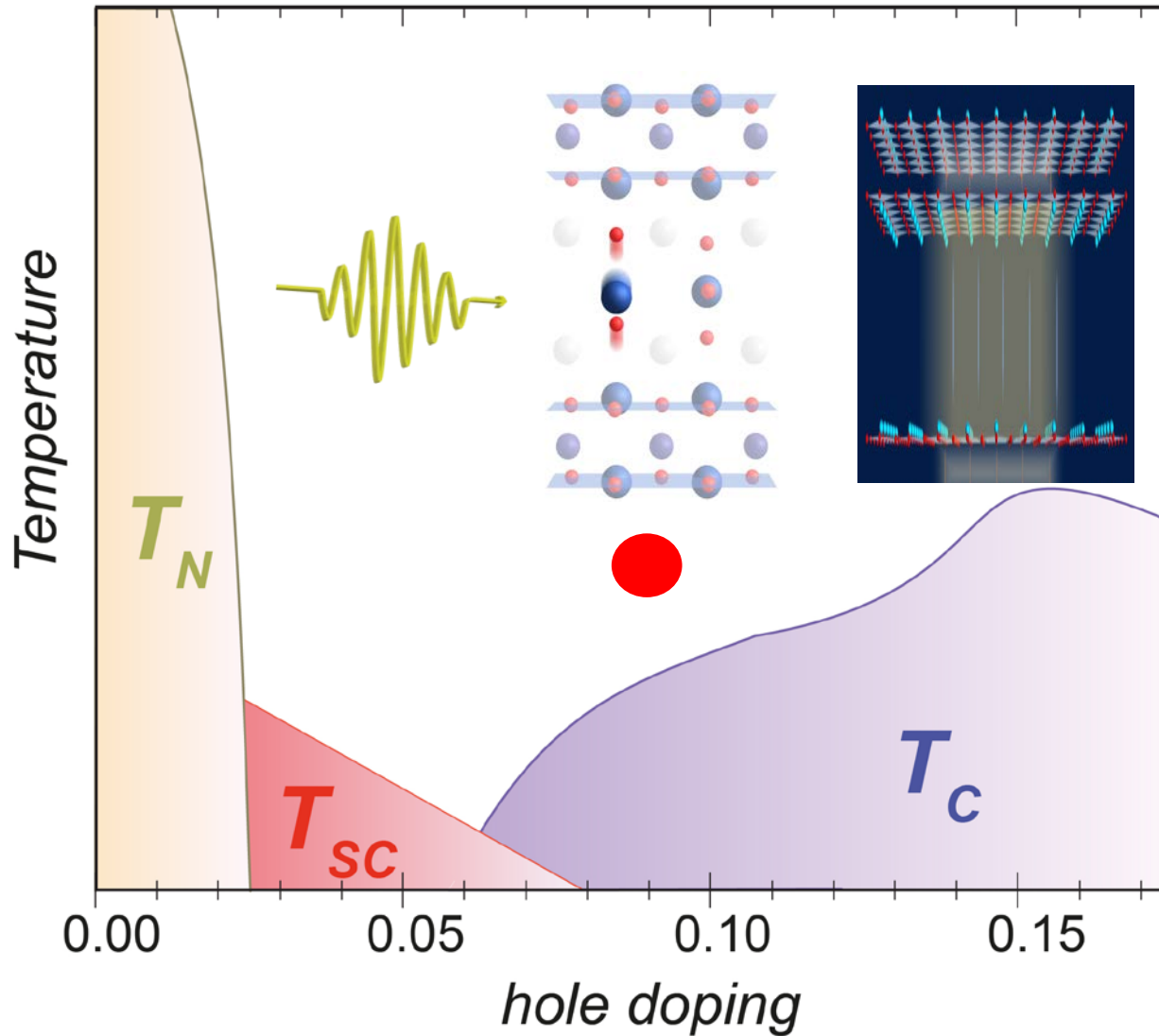
Apical oxygen correlates with T_c at equilibrium



E. Pavarini et al., *PRL* 87, 047003 (2001)

C. Weber et al. *Phys. Rev. B* 82, 125107 (2010).

Can I induce coherence above T_c ?

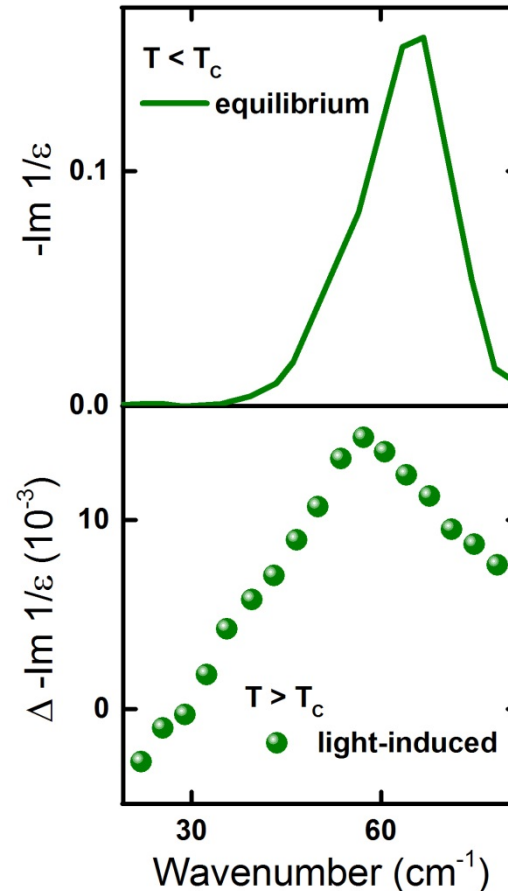
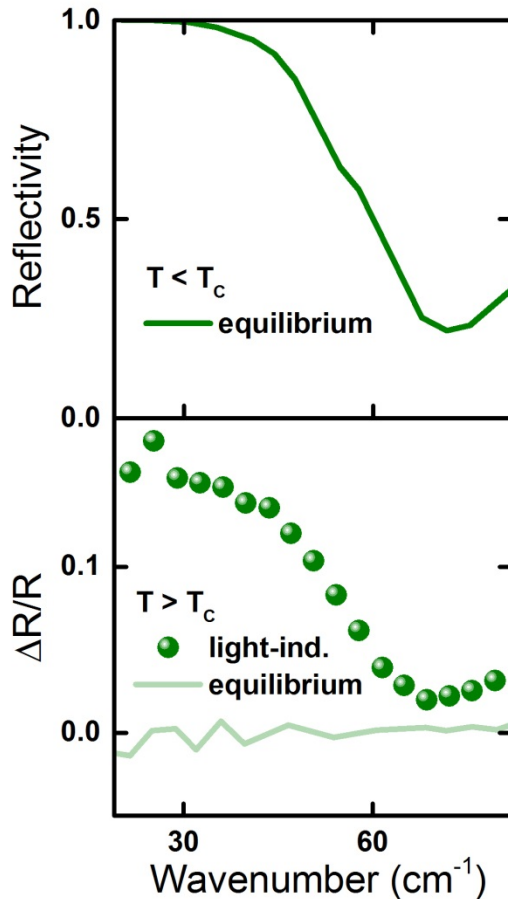


Light induced Josephson Coupling – 2 X T_c

Plasma edge

$$\epsilon_1(\omega_{JPR}) = 0$$

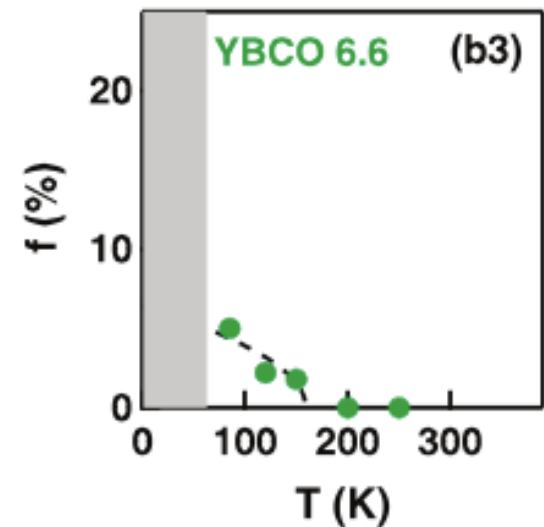
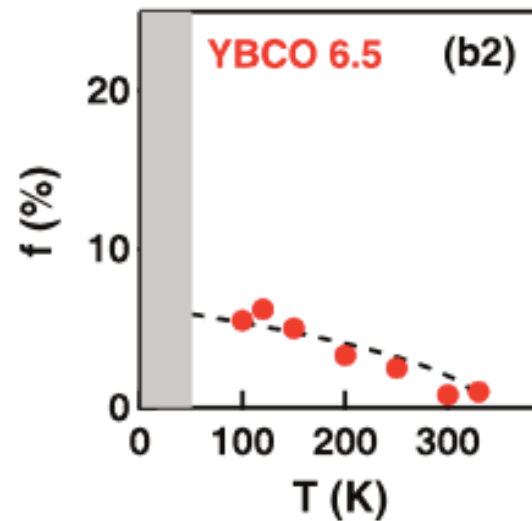
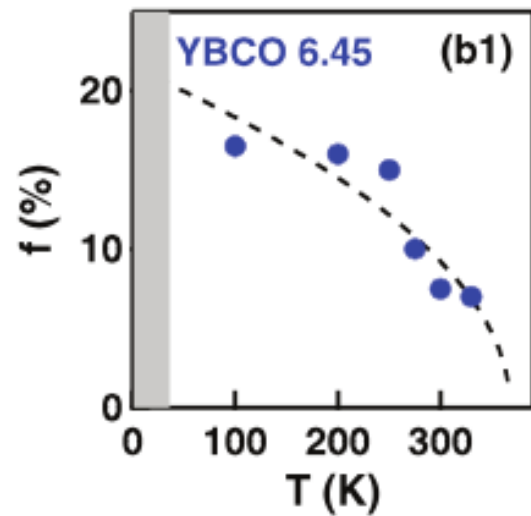
YBCO_{6.6} – 100 K



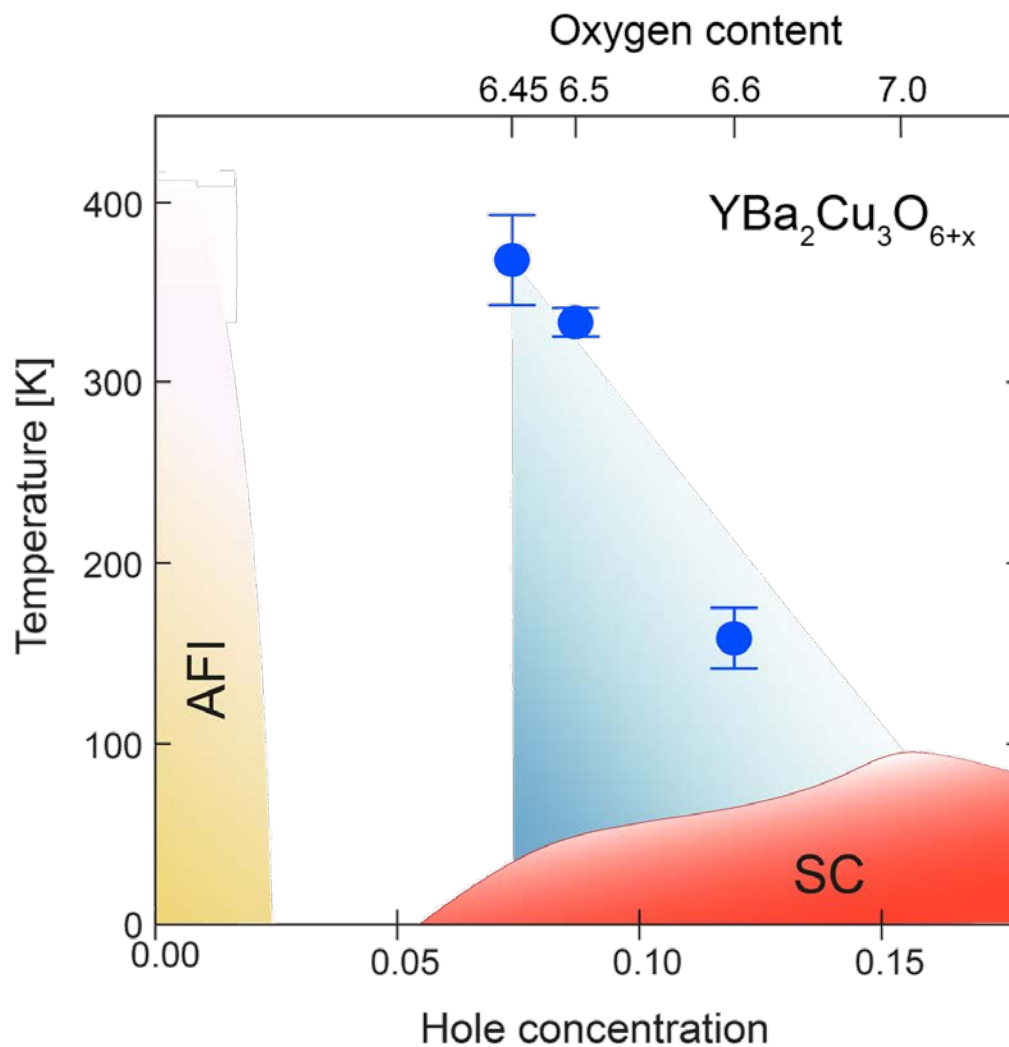
Equilibrium $T < T_c$

Light induced $T > T_c$

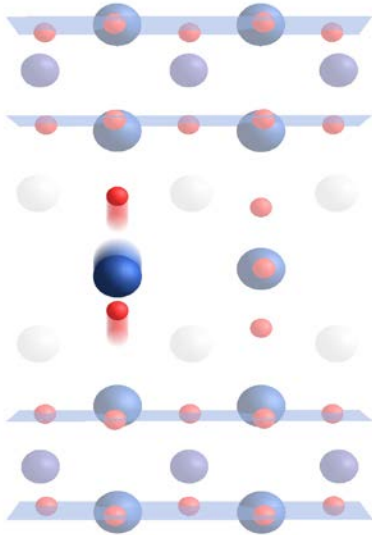
Surprise..... Up to room temperature



Throughout the pseudogap phase



What is the lattice doing ?

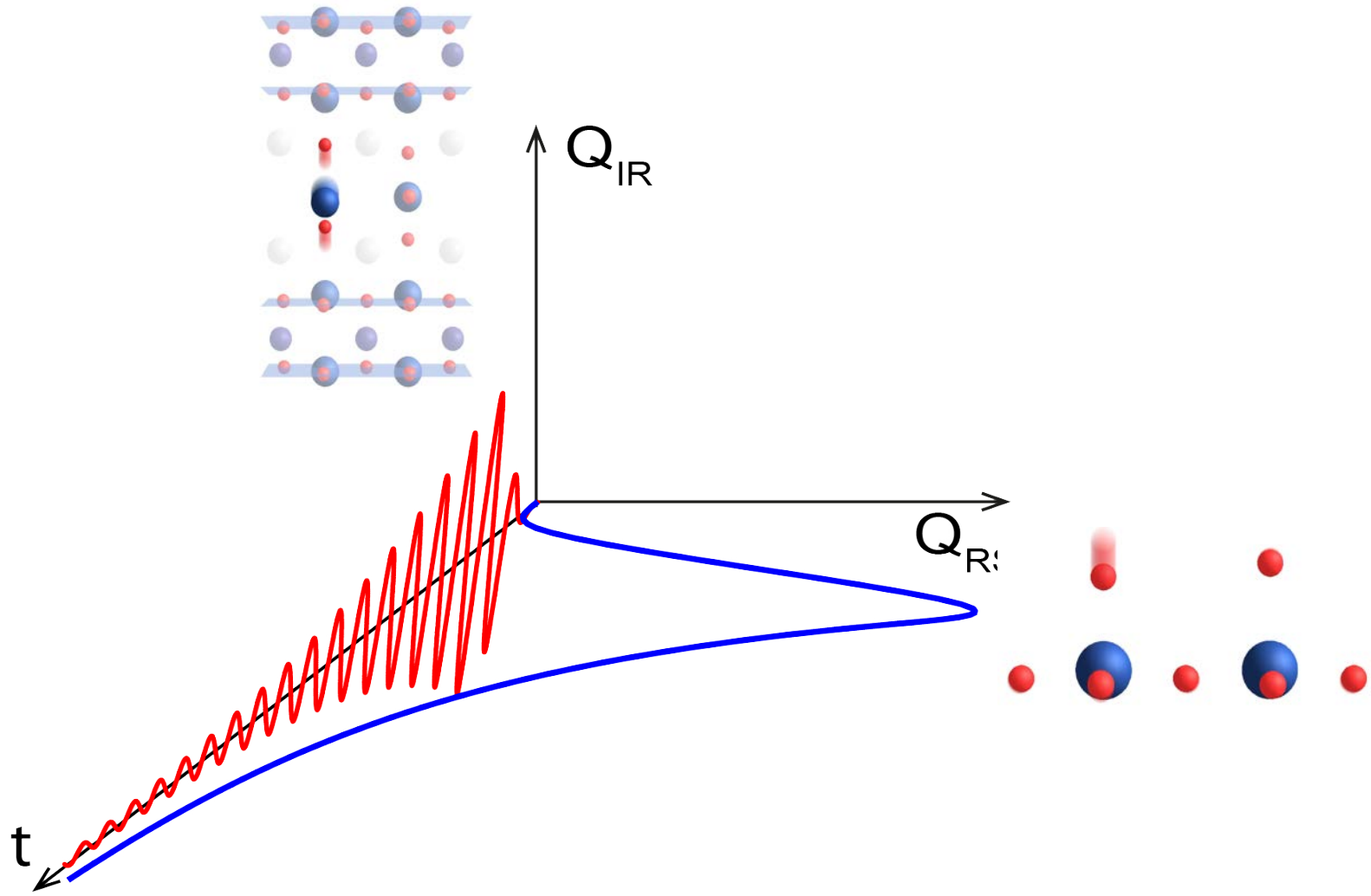


Q_{IR} of B_{1u} symmetry

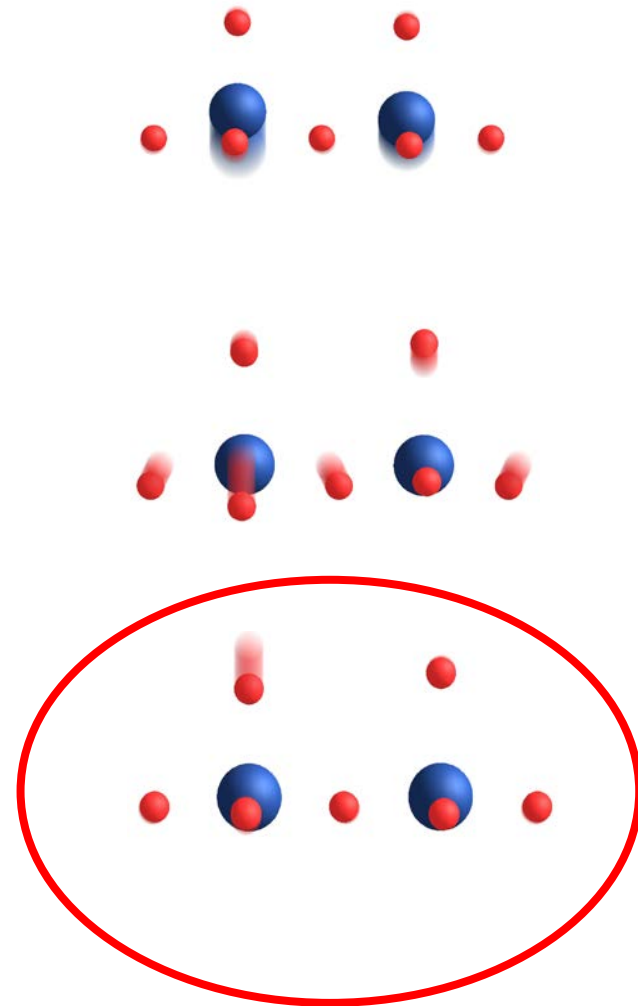
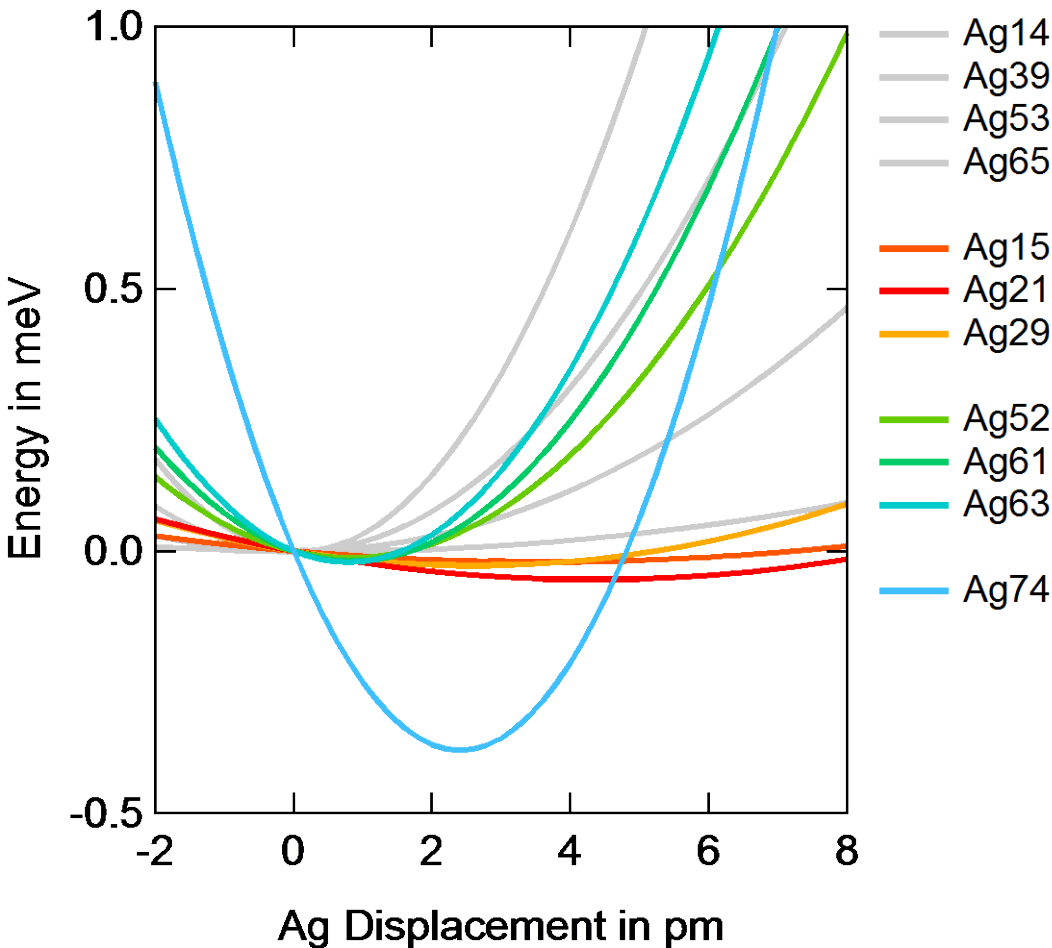
$$Q_{\text{IR}}^2 Q_2 \neq 0$$

only if Q_2 is a Raman mode of A_g symmetry

Excite B_{1u} and displace along A_g

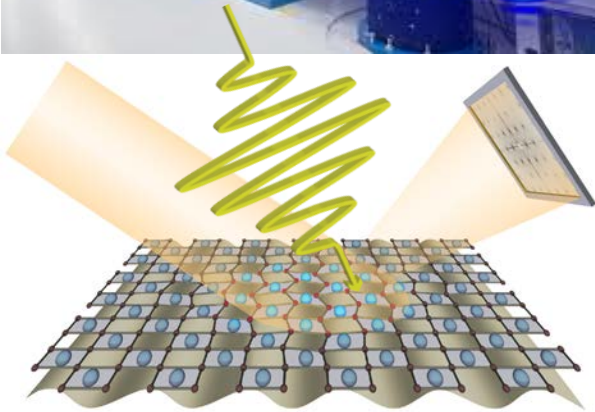
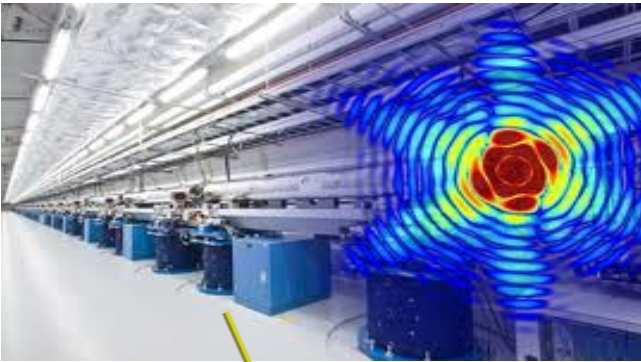


Doped YBCO: 11 A_g Raman modes

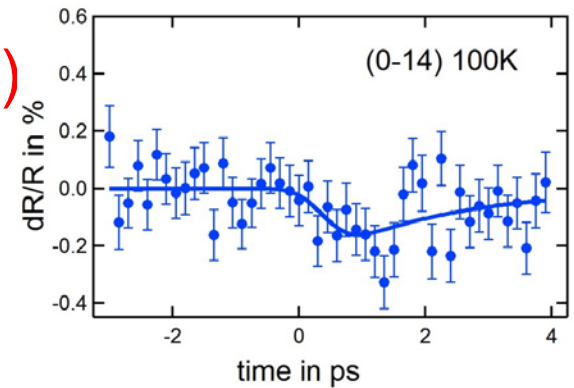


Only three A_g modes are coupled strongly with B_{1u}

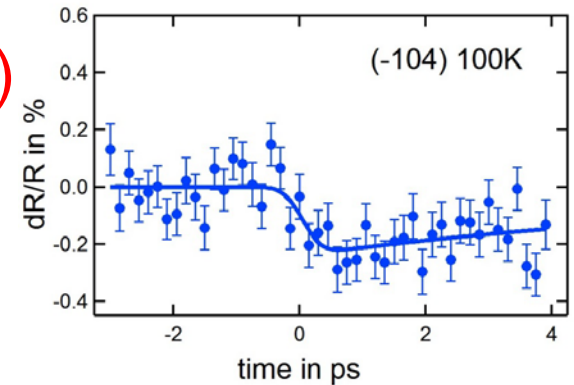
Femtosecond X-ray Scattering



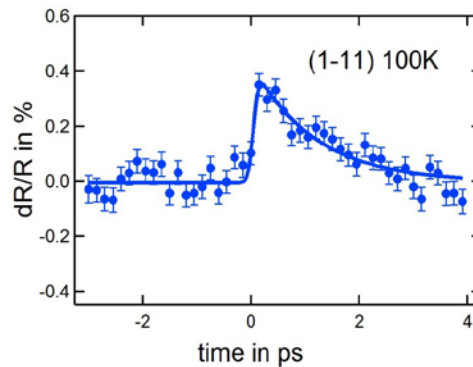
(0,-1,4)



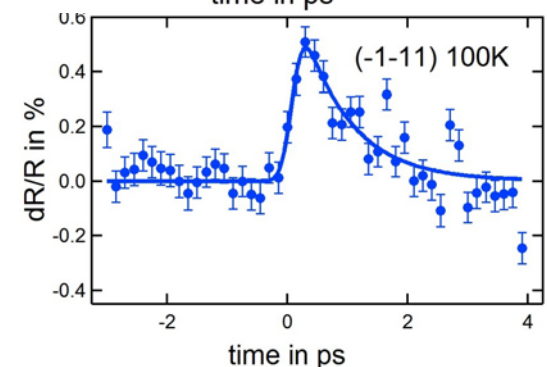
(-1,0,4)



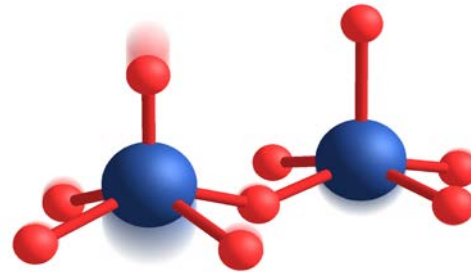
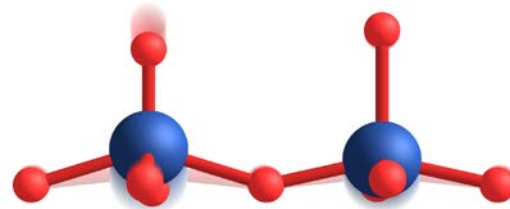
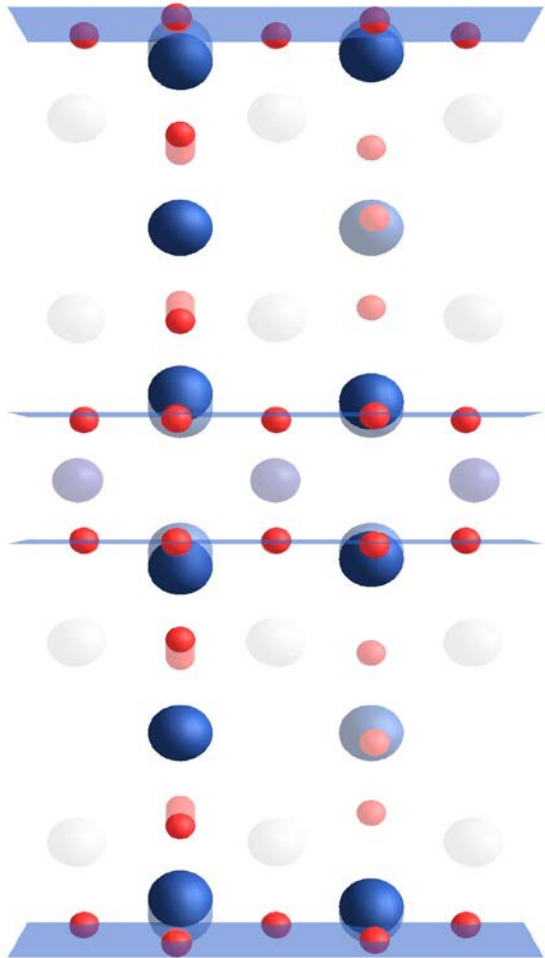
(-1,1,1)



(1,-1,1)

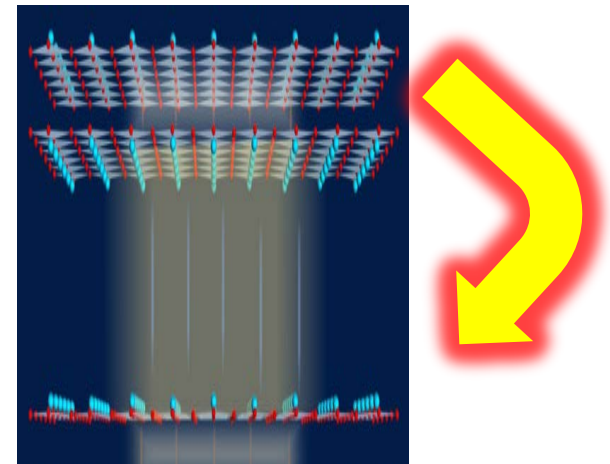
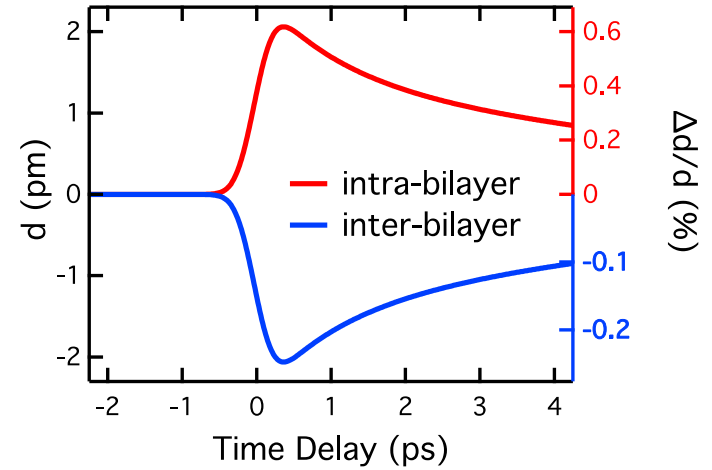
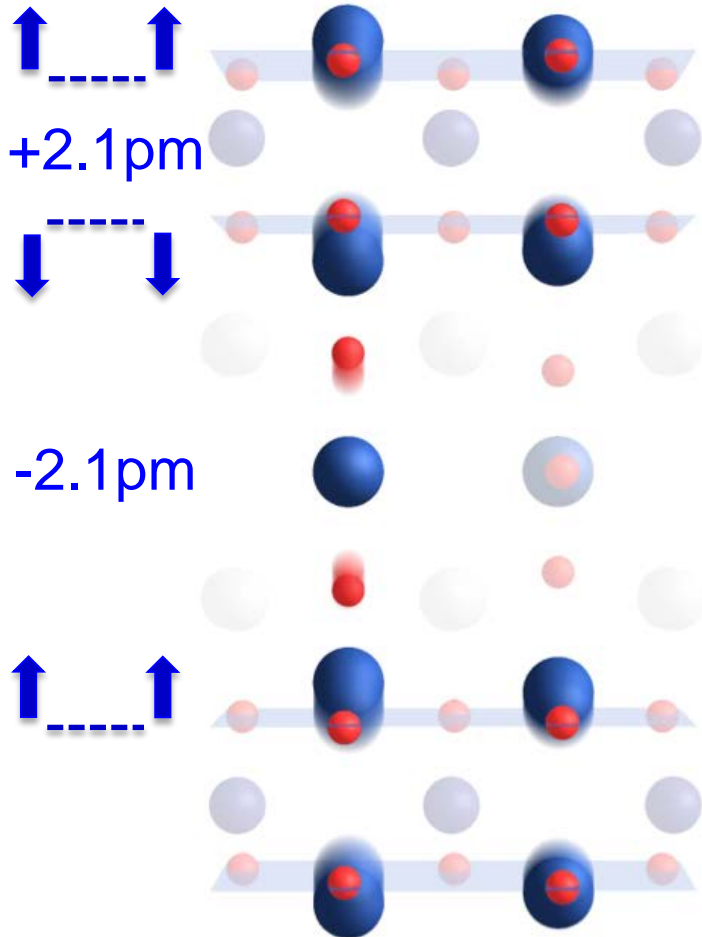


A new, transient crystal structure

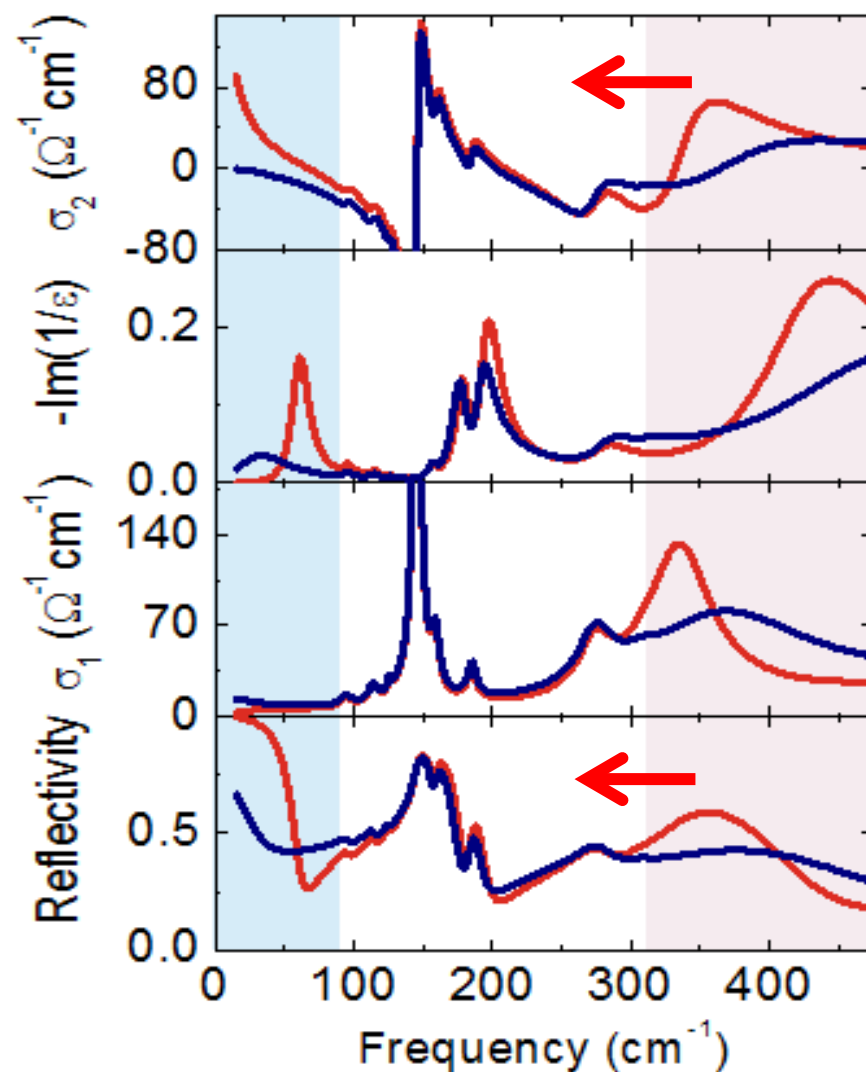
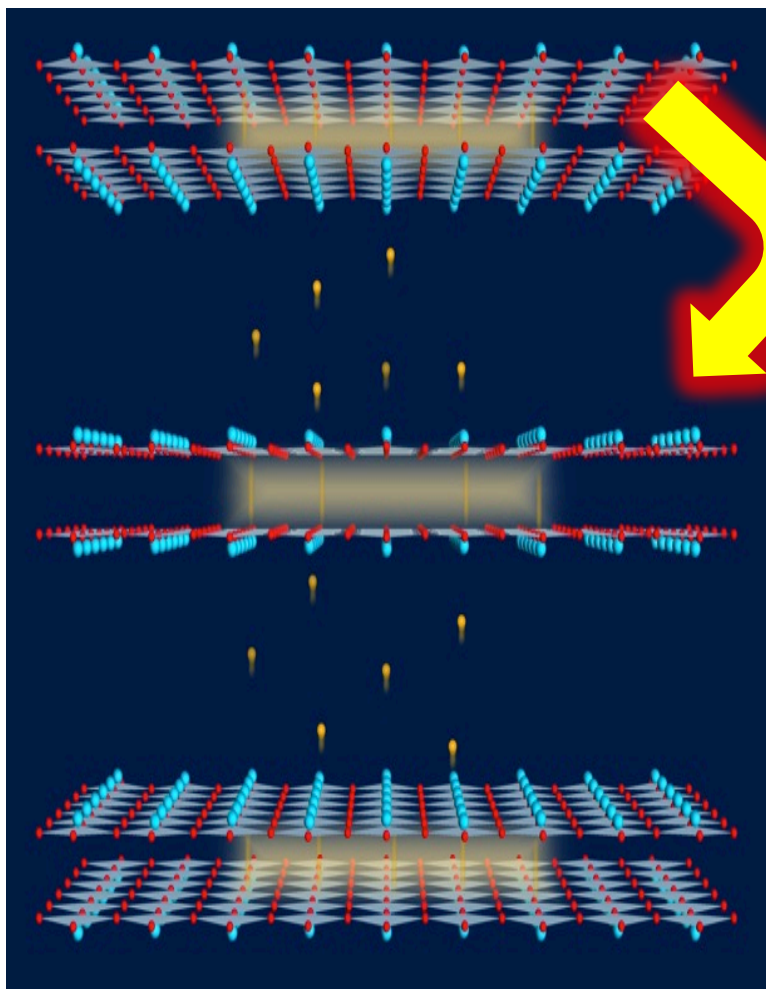


**Is this the structure of a room temperature
superconductor ?**

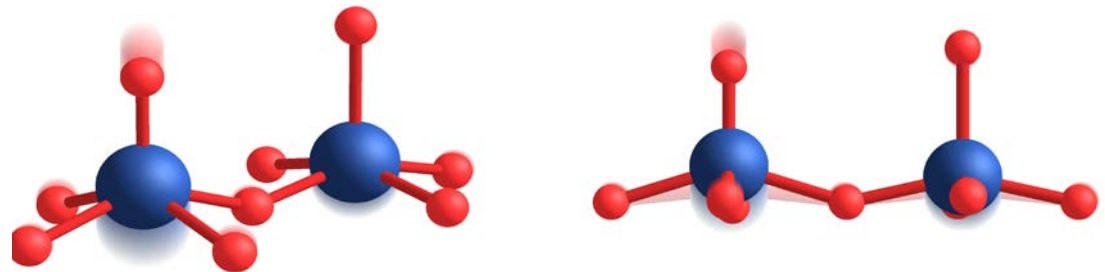
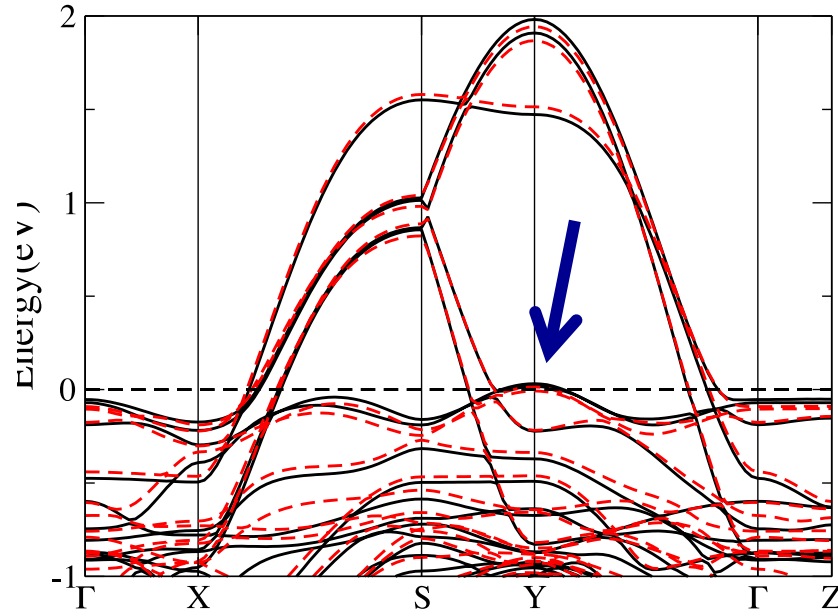
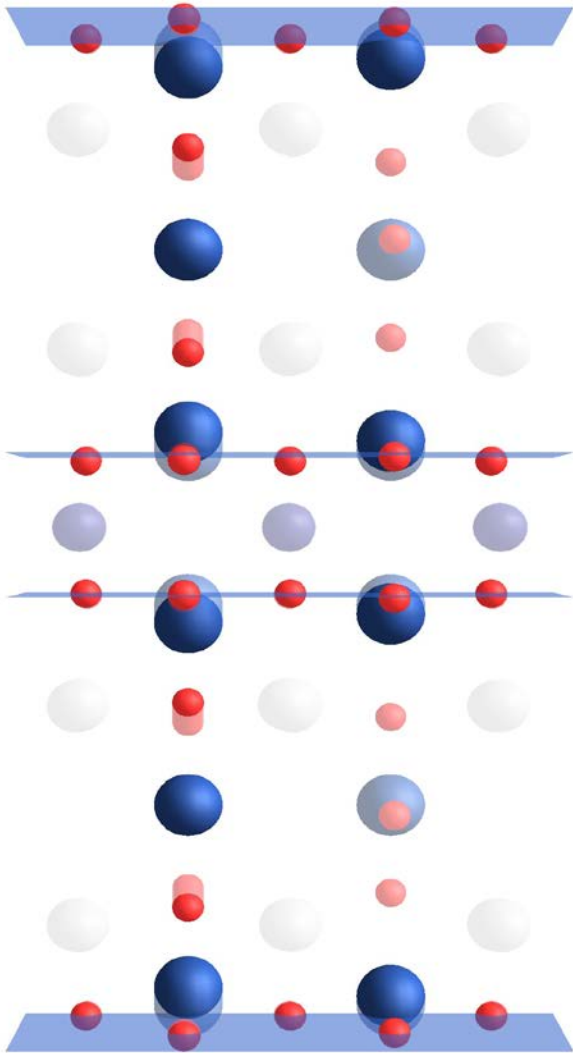
1) Staggered motion of the planes



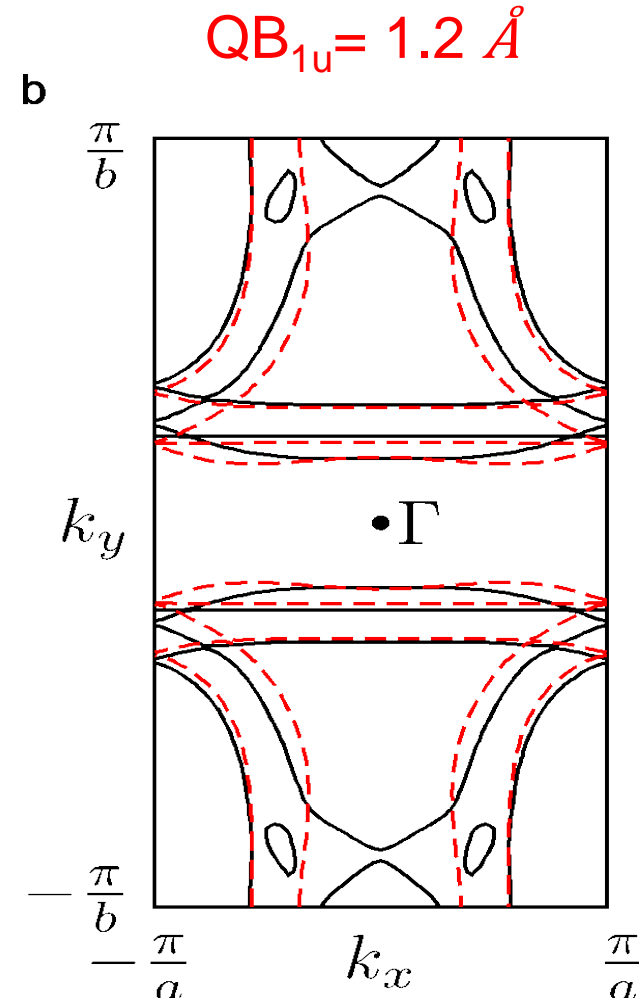
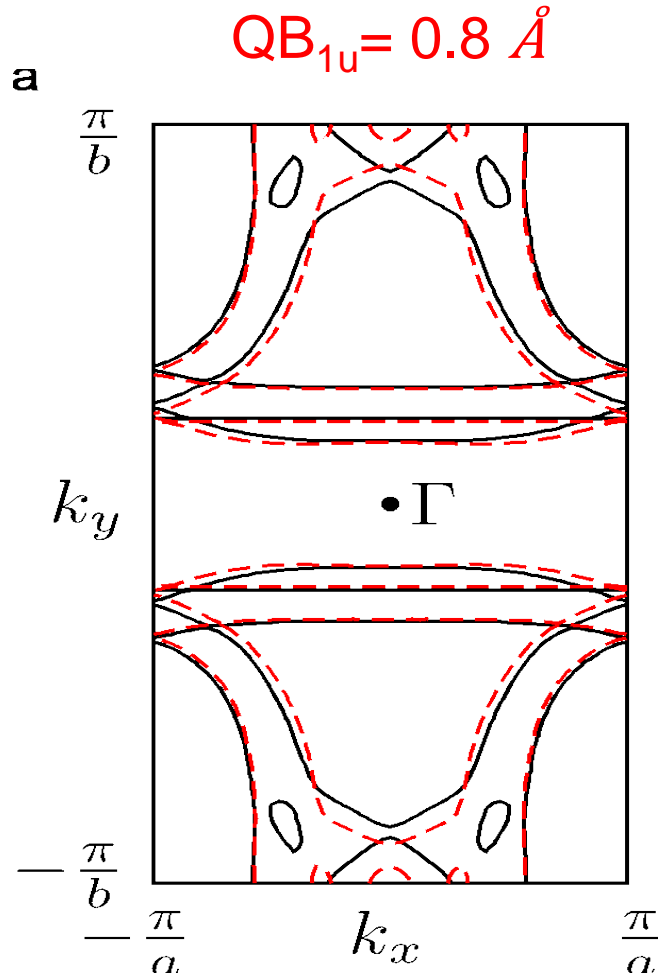
Spectral weight from high frequency



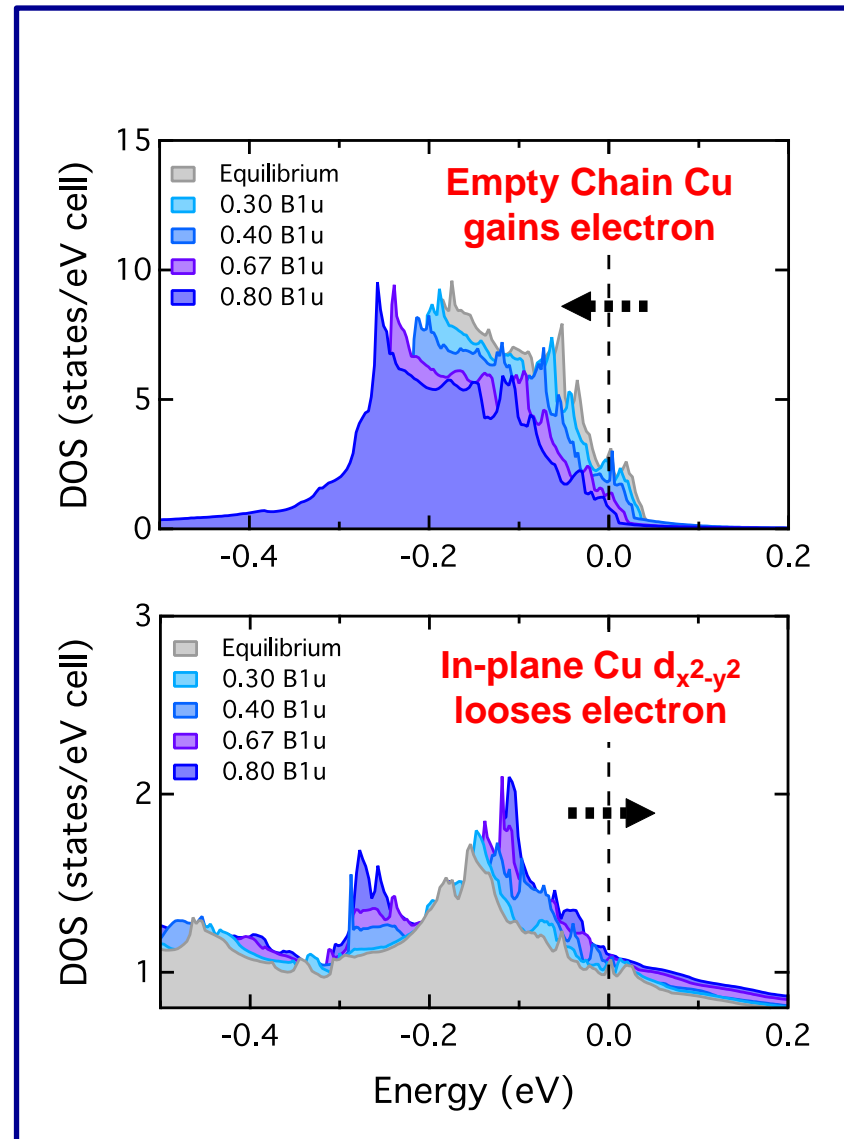
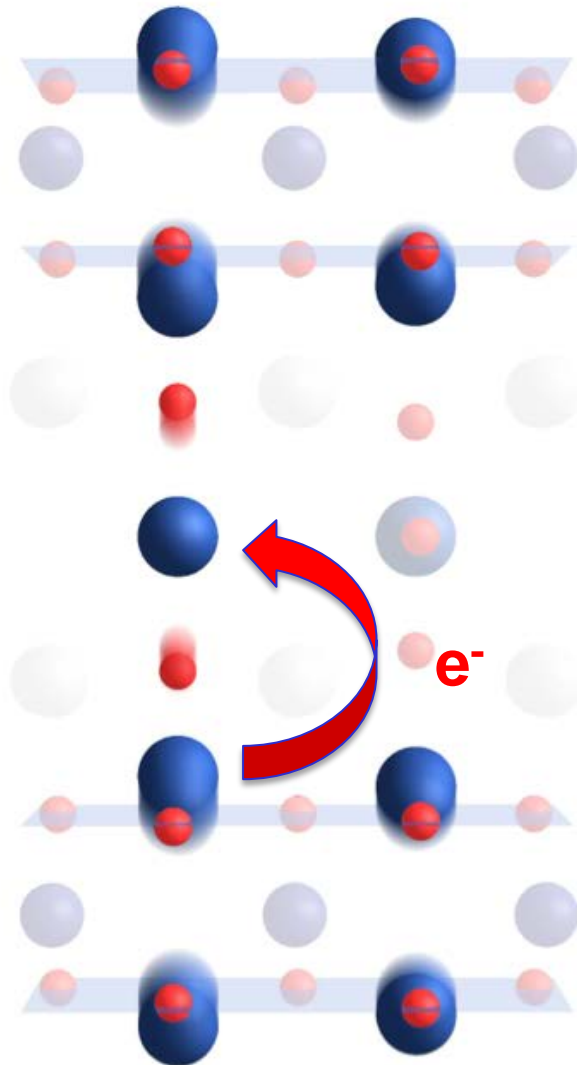
2) Empty chain band moves down in energy



3) A “cleaner” LDA electronic structure

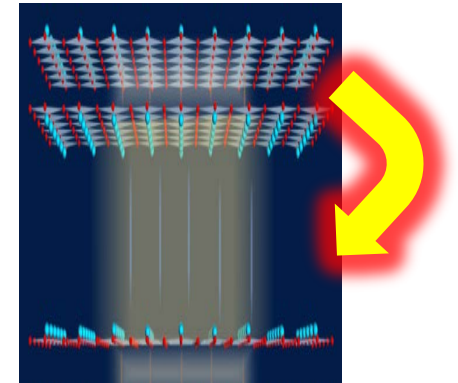


3) Charge transfer from the planes to the chains

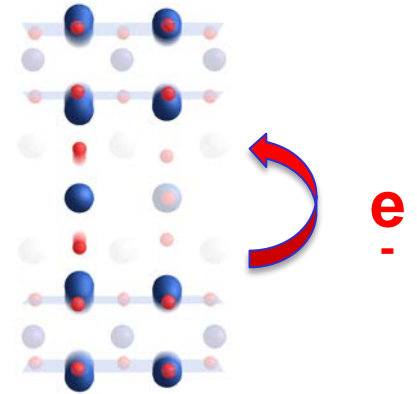


Summary: three good things

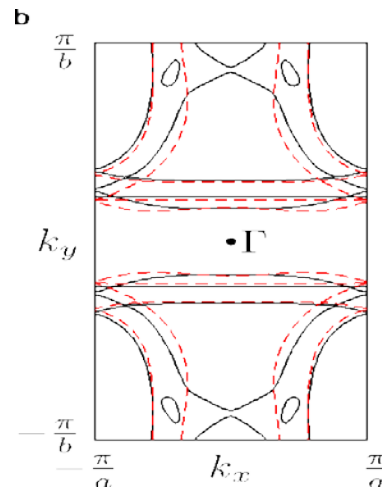
1) Staggered motion of the layers



2) Charge transfer from to chains

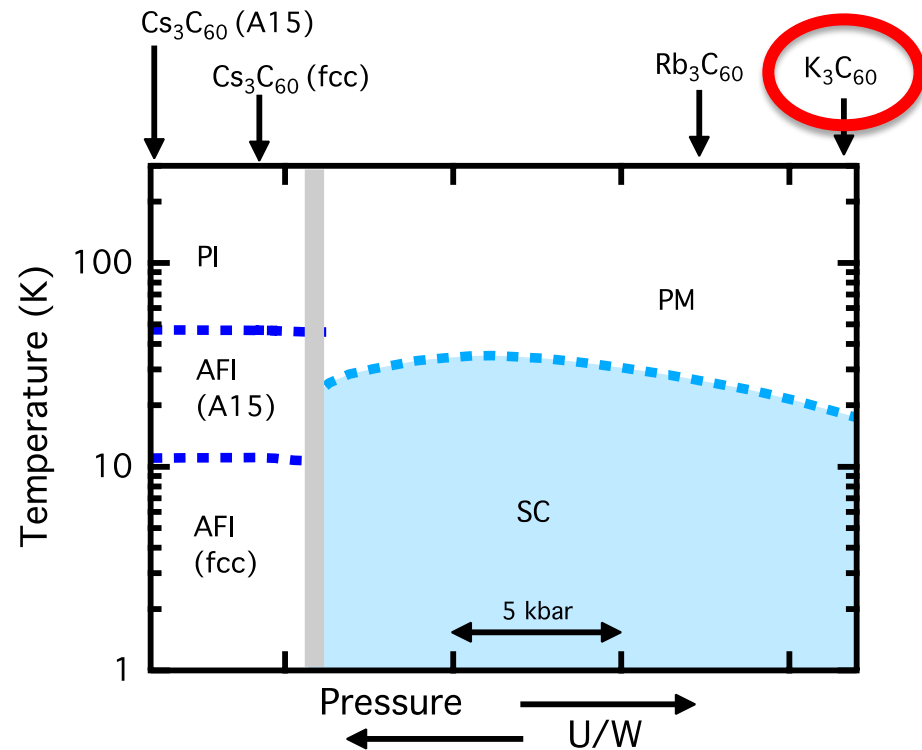
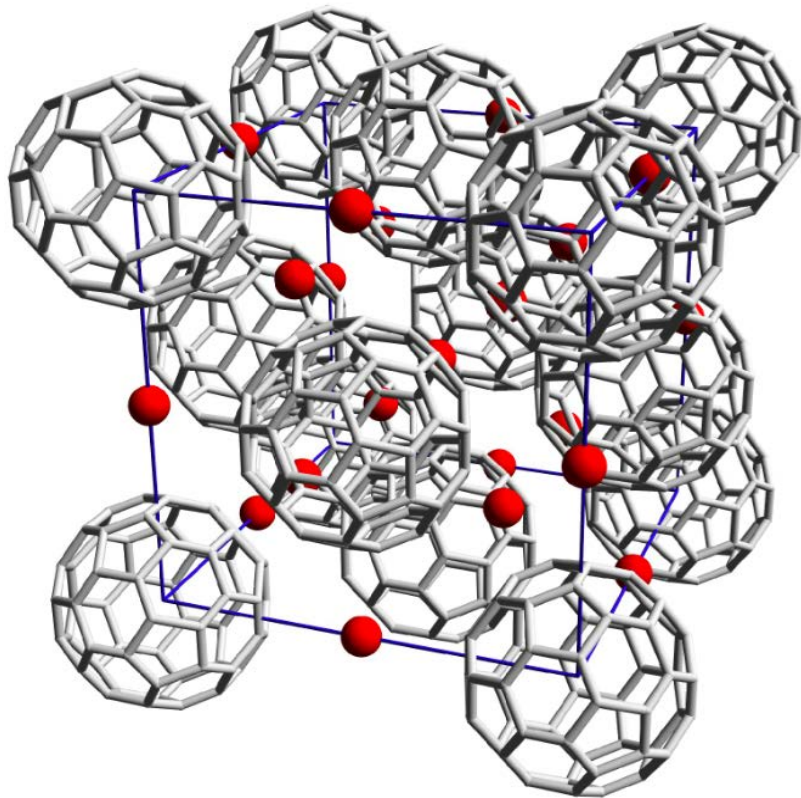


3) dx^2-y^2 Fermi surface



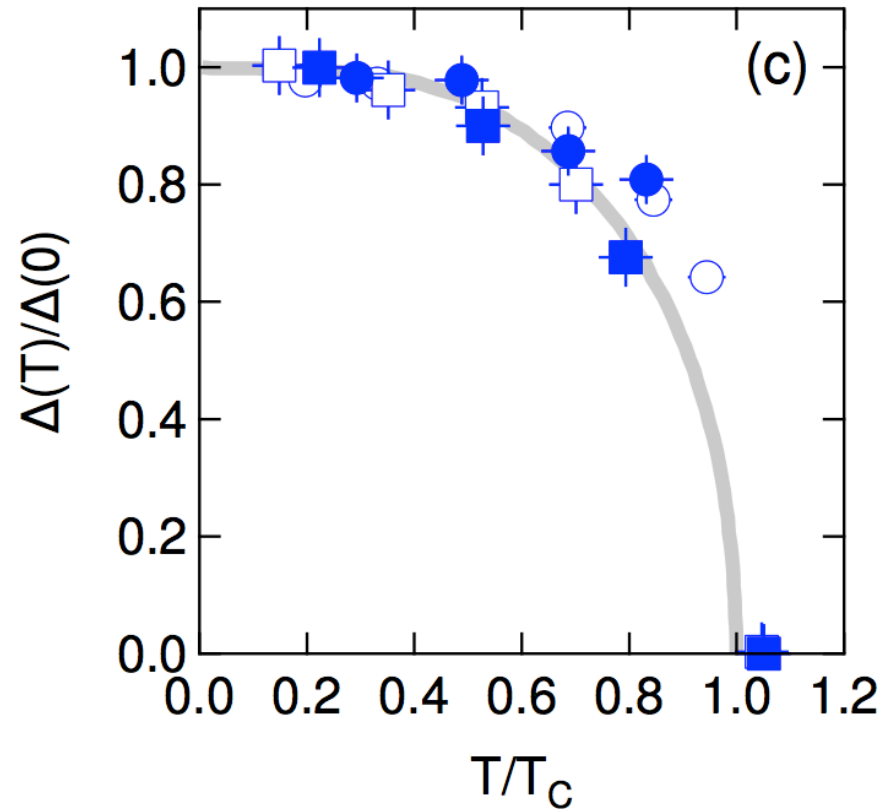
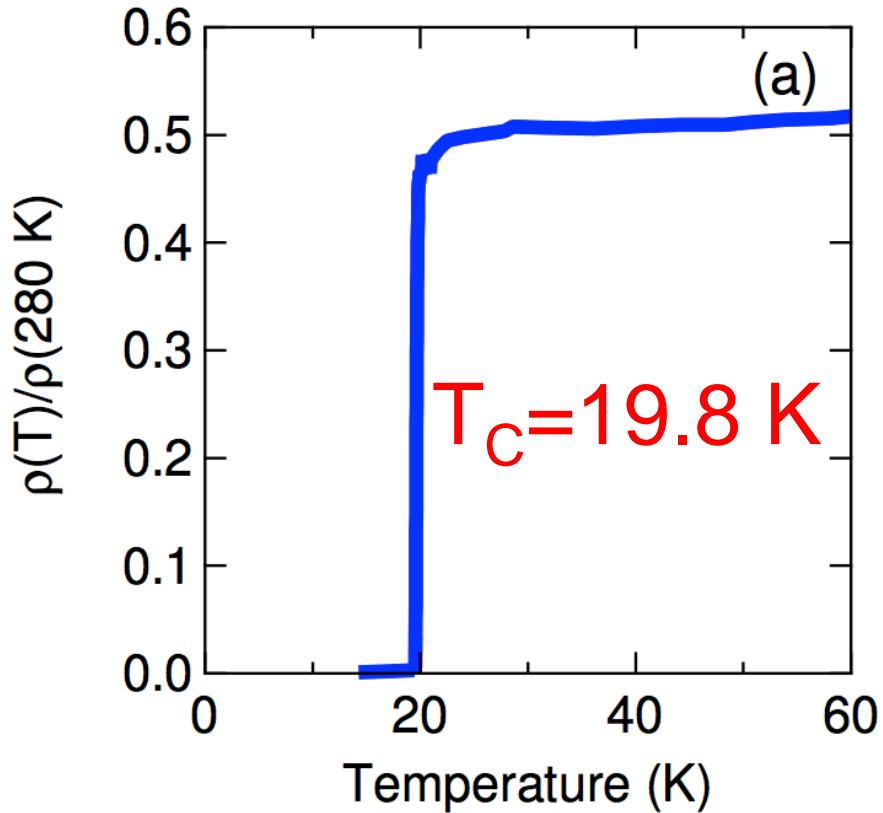
**Is this a phenomenon specific to cuprates
or is it more general ?**

K_3C_{60} : a 20 K superconductor



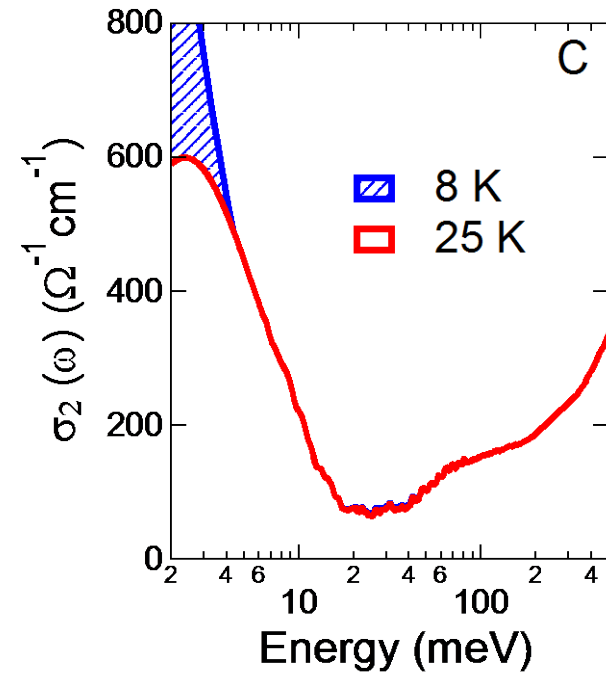
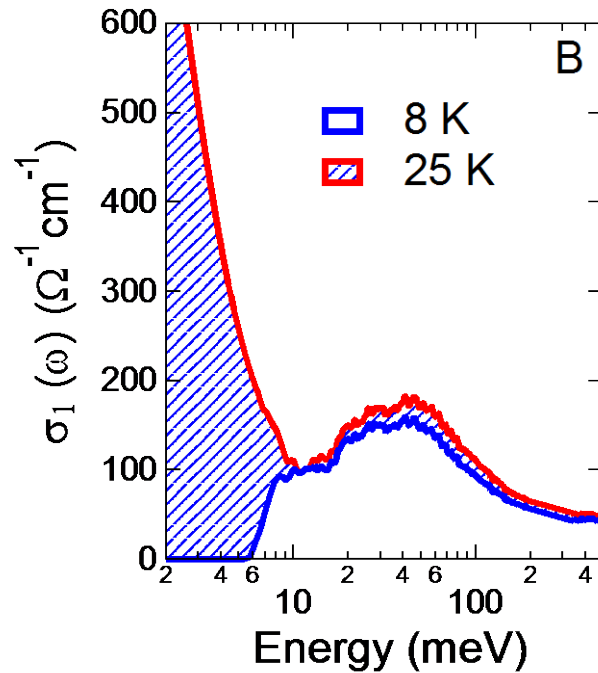
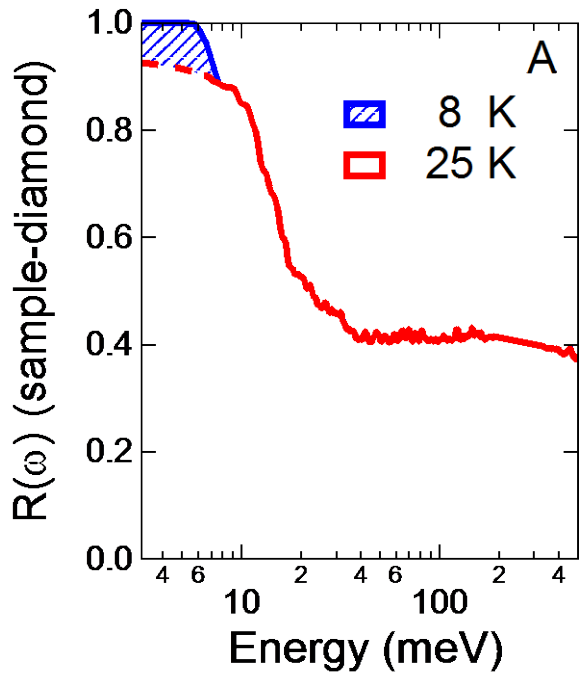
- Organic molecular solid
- High T_c (20 K)
- 3D electronic structure

Equilibrium Superconductivity in K_3C_{60}



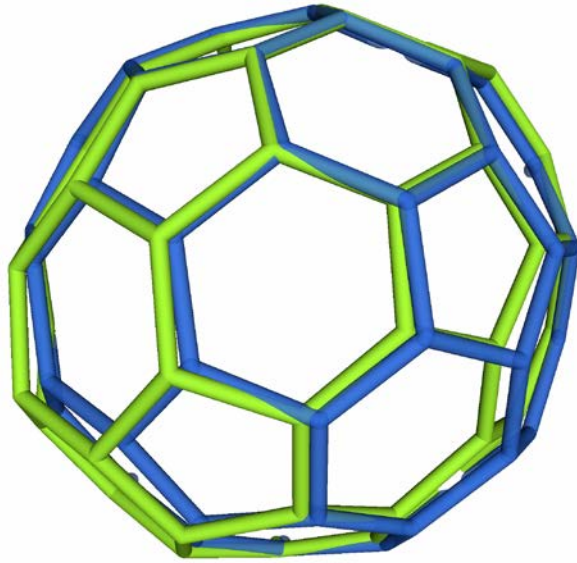
From literature data, MM PhD thesis



Equilibrium Superconducting Transition

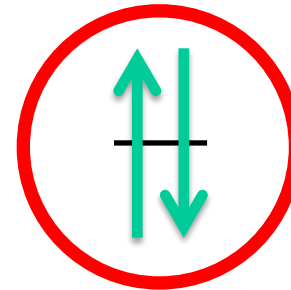


- Increase in $R(\omega)$
- Gap opening in $\sigma_1(\omega)$
- Increase in $\sigma_2(\omega)$

Pairing Interaction in K_3C_{60}



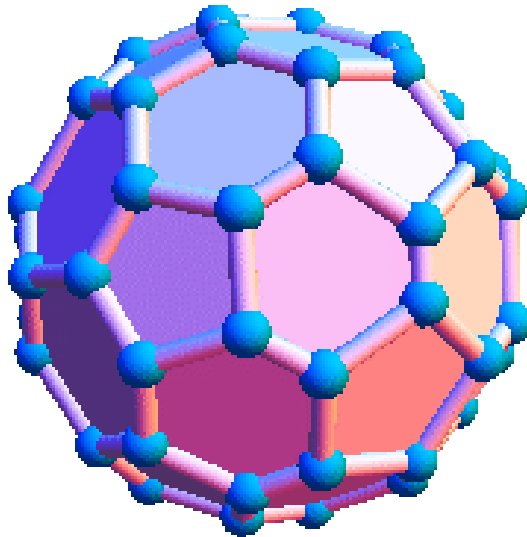
-  H_g mode (JT distortion)
-  Undistorted



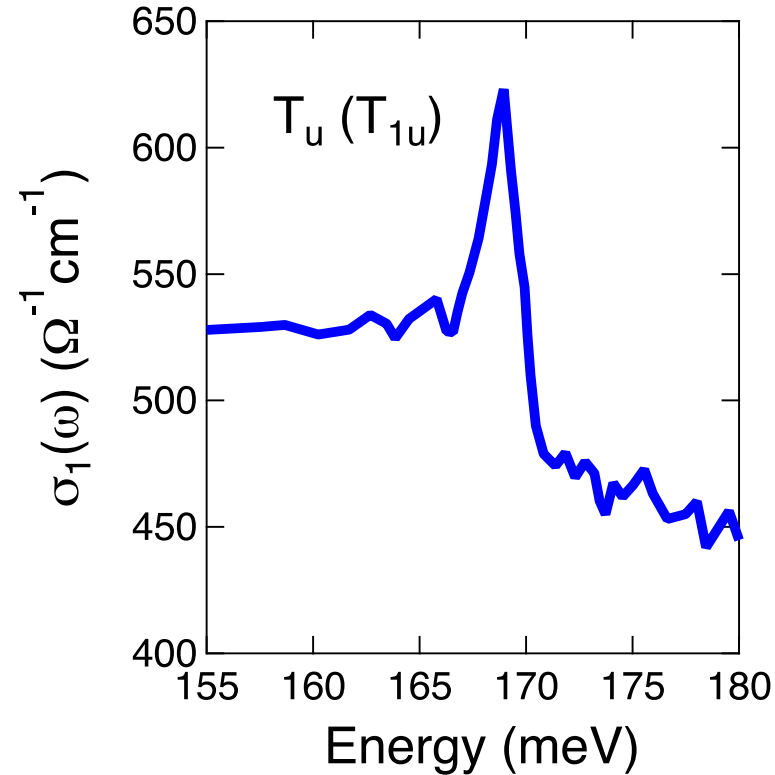
“On ball” vibrations plus correlations favor local pairing

Vibrational pump

$T_{1u}(4)$
170 meV



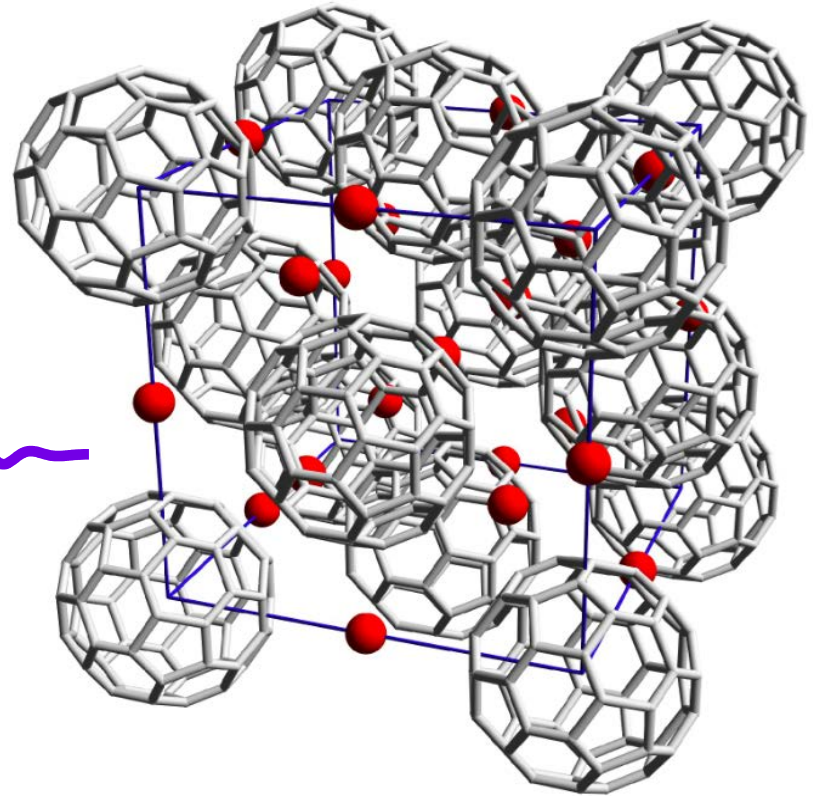
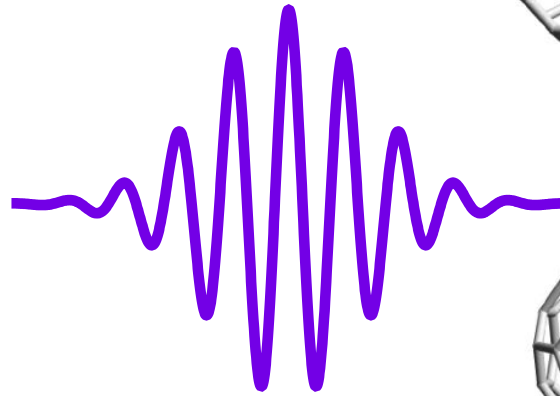
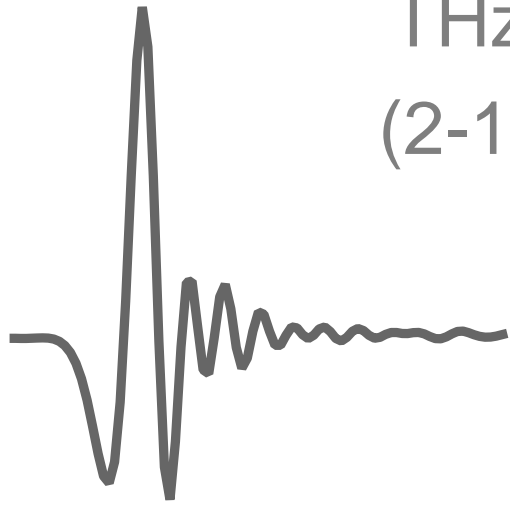
MIR pump 170 meV (7.3 μm)



Iwasa et al. PRB **51**, 3678 (1995)

Vibrational pump THz probe in K_3C_{60}

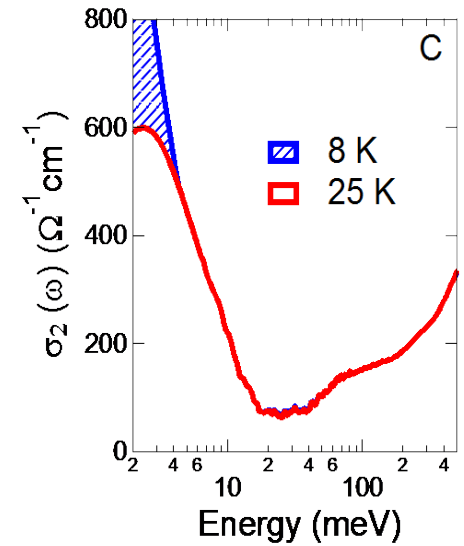
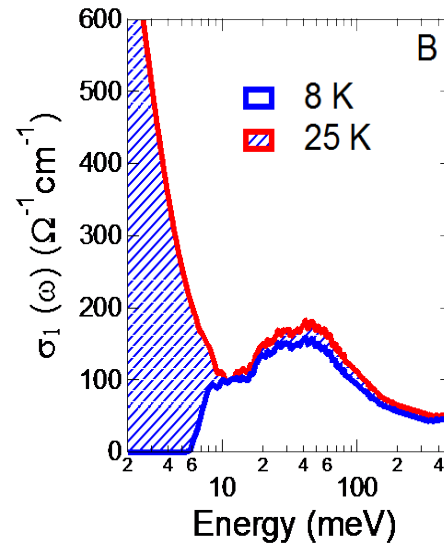
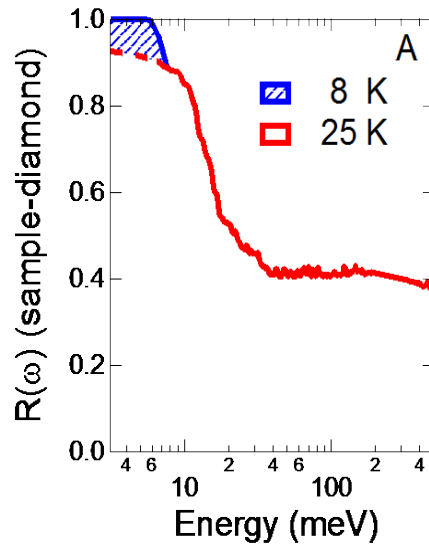
THz probe
(2-10 meV)



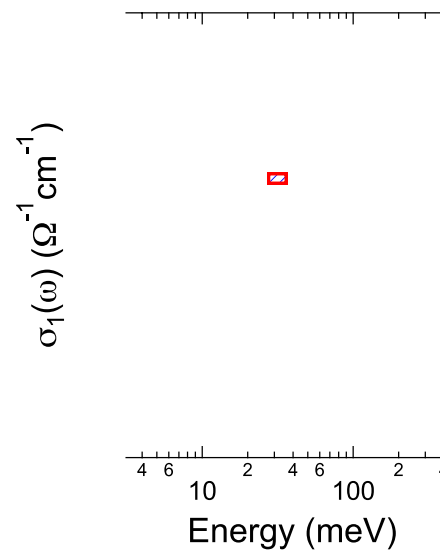
MIR pump 170 meV (7.3 μm)

Striking similarity with the low temperature SC

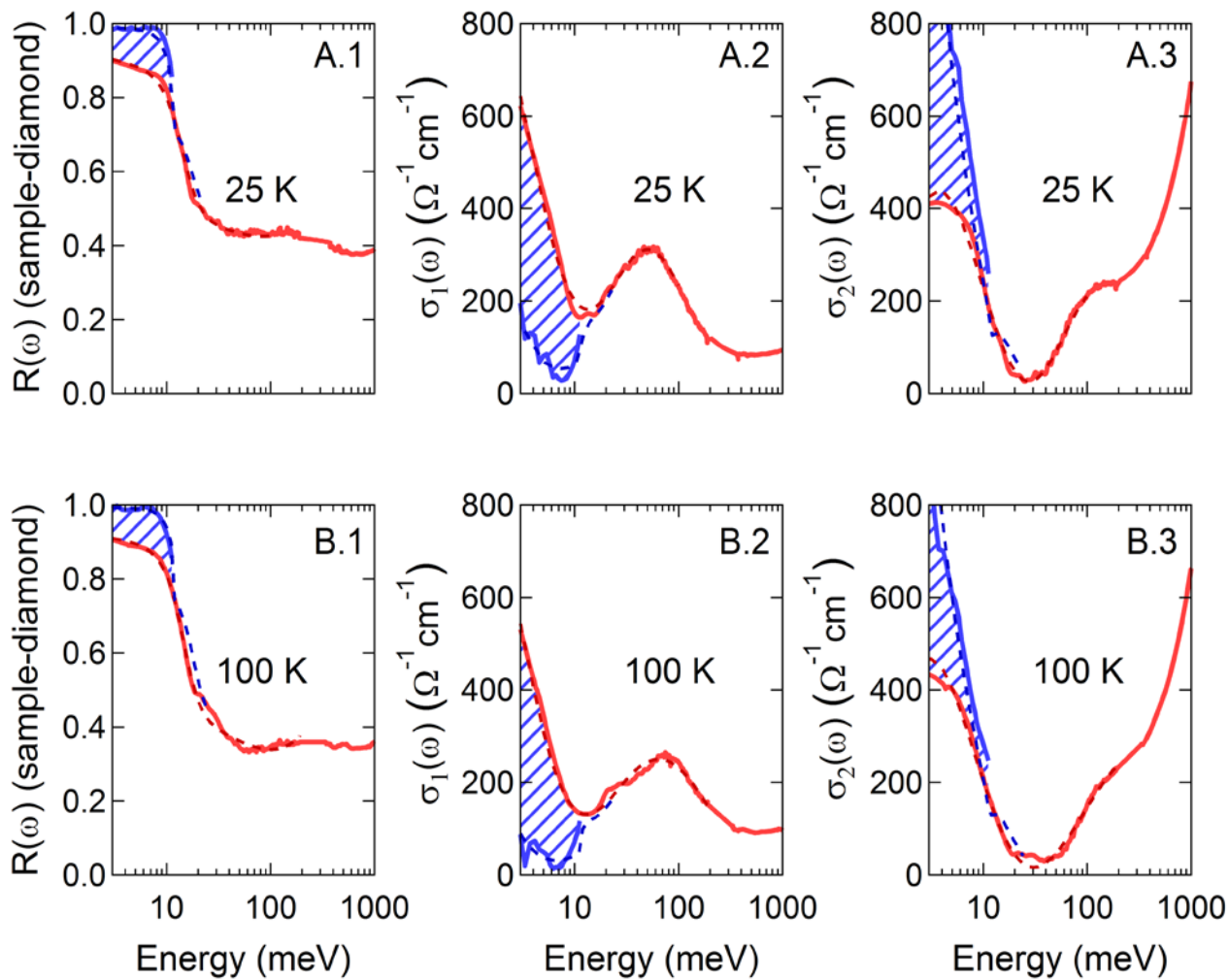
Cooling



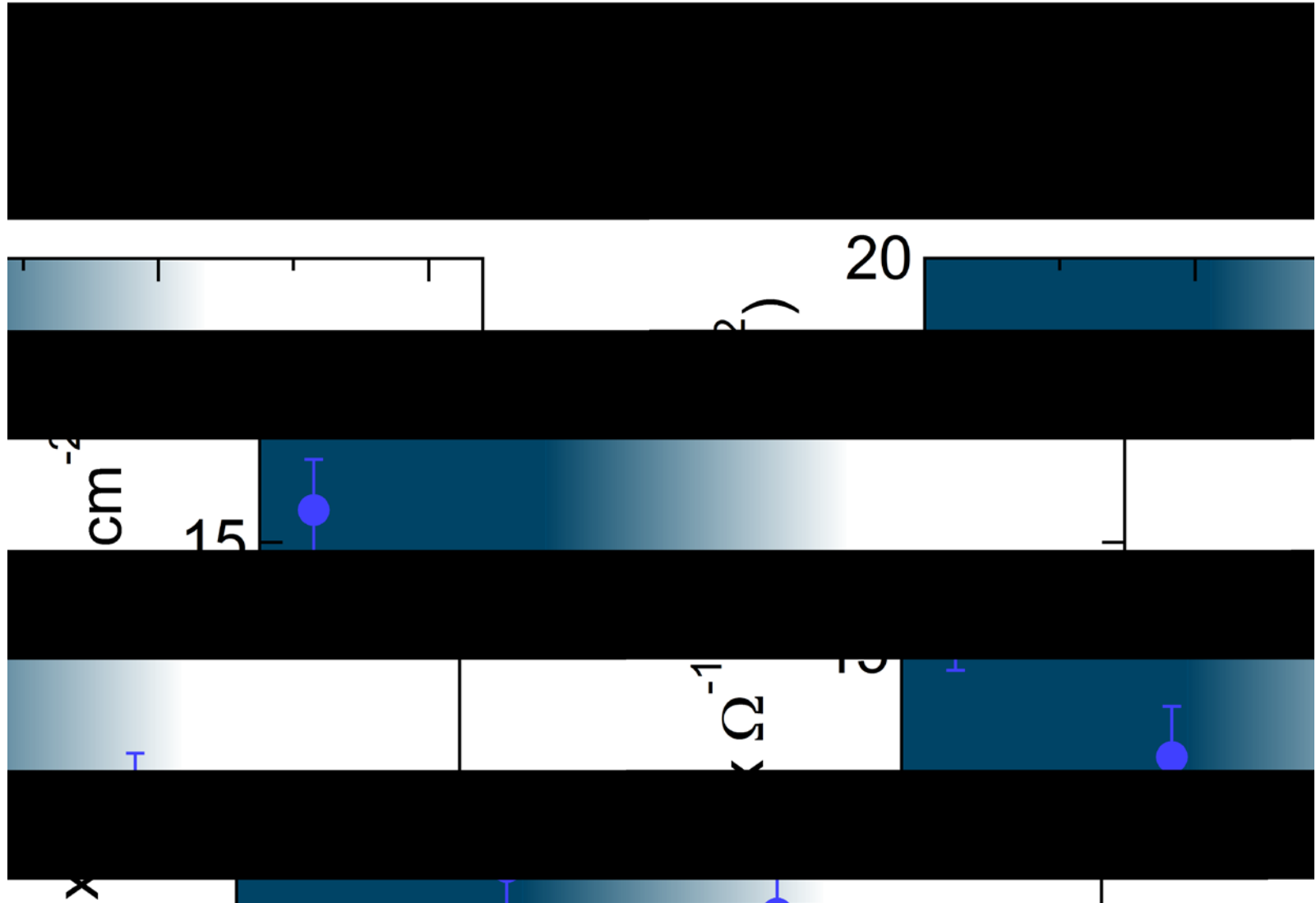
Light-induced
T=25 K



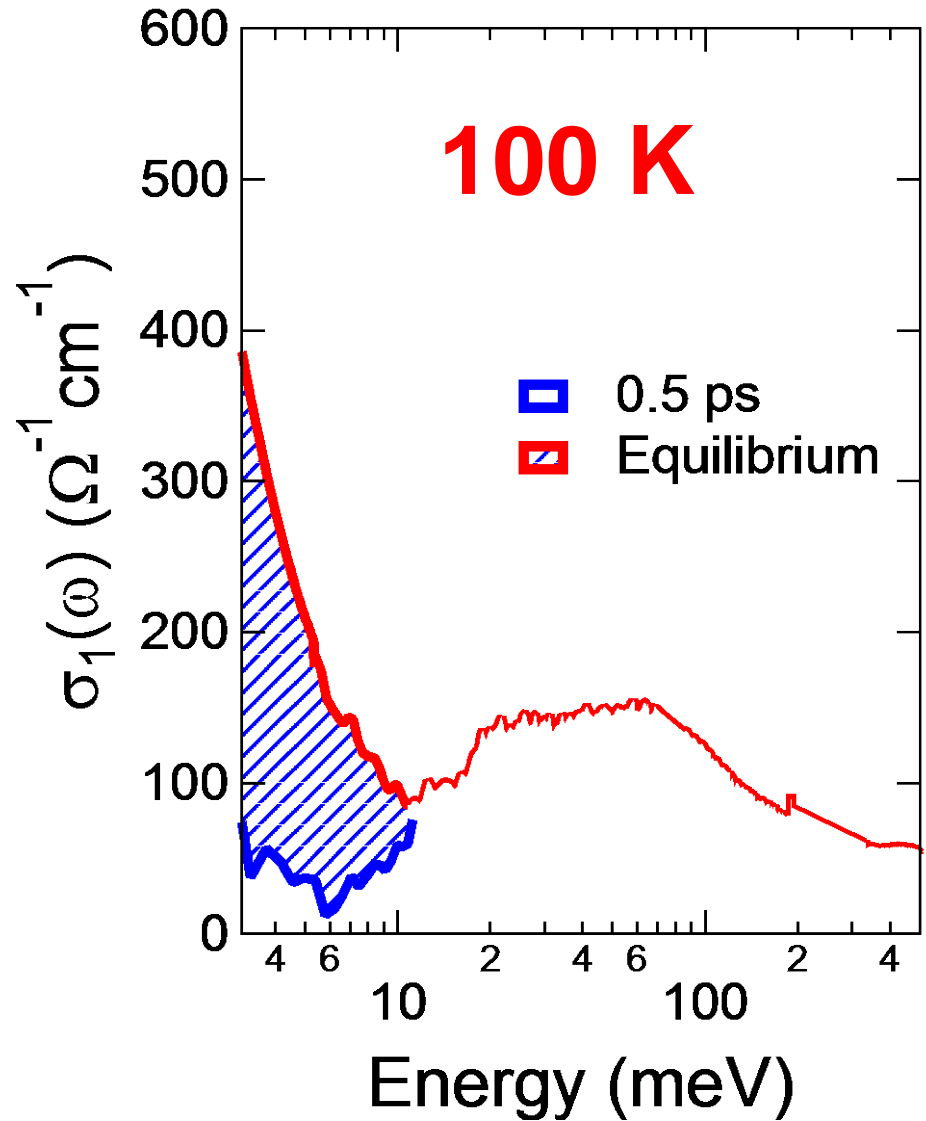
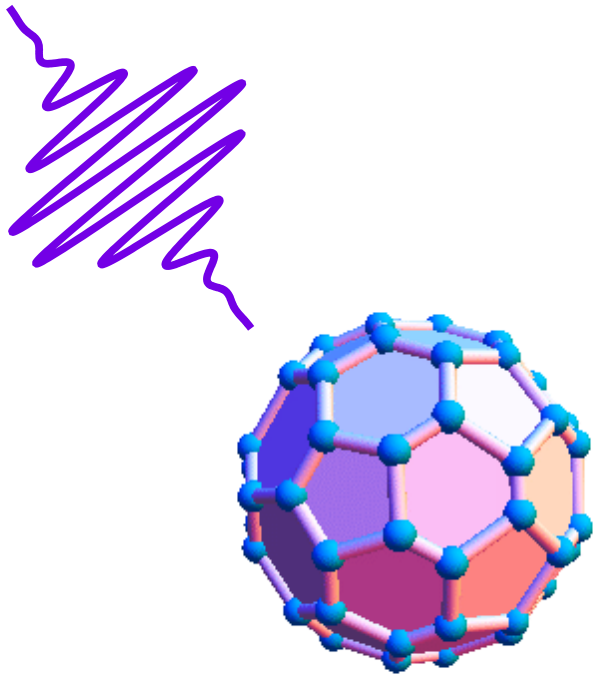
Temperature dependence



Crossover at ~ 10 times T_c

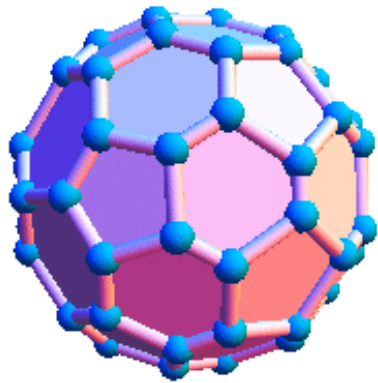


K_3C_{60} : Stimulated superconductivity ?

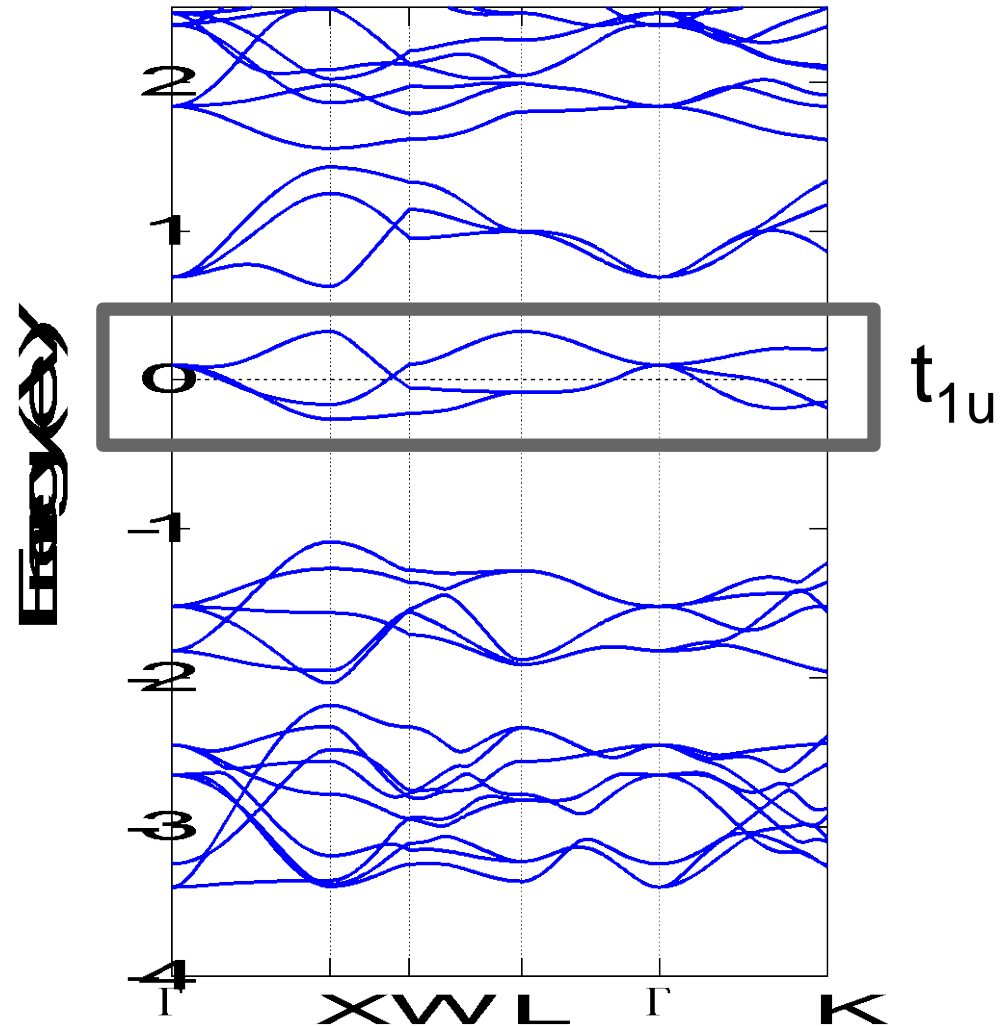


What is going on?

T_{1u} vibration: no linear e-ph coupling



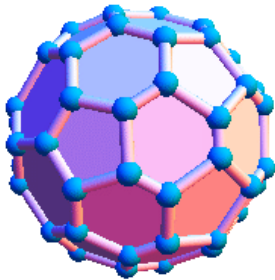
$T_{1u}(4)$
 1370 cm^{-1}



Nonlinear Coupling to Jahn Teller Phonon

$$Q^2_{T_{1u}} Q_{H_g}$$

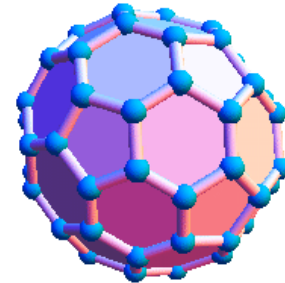
T_{1u}^2



$T_{1u}(4)$
 1370 cm^{-1}

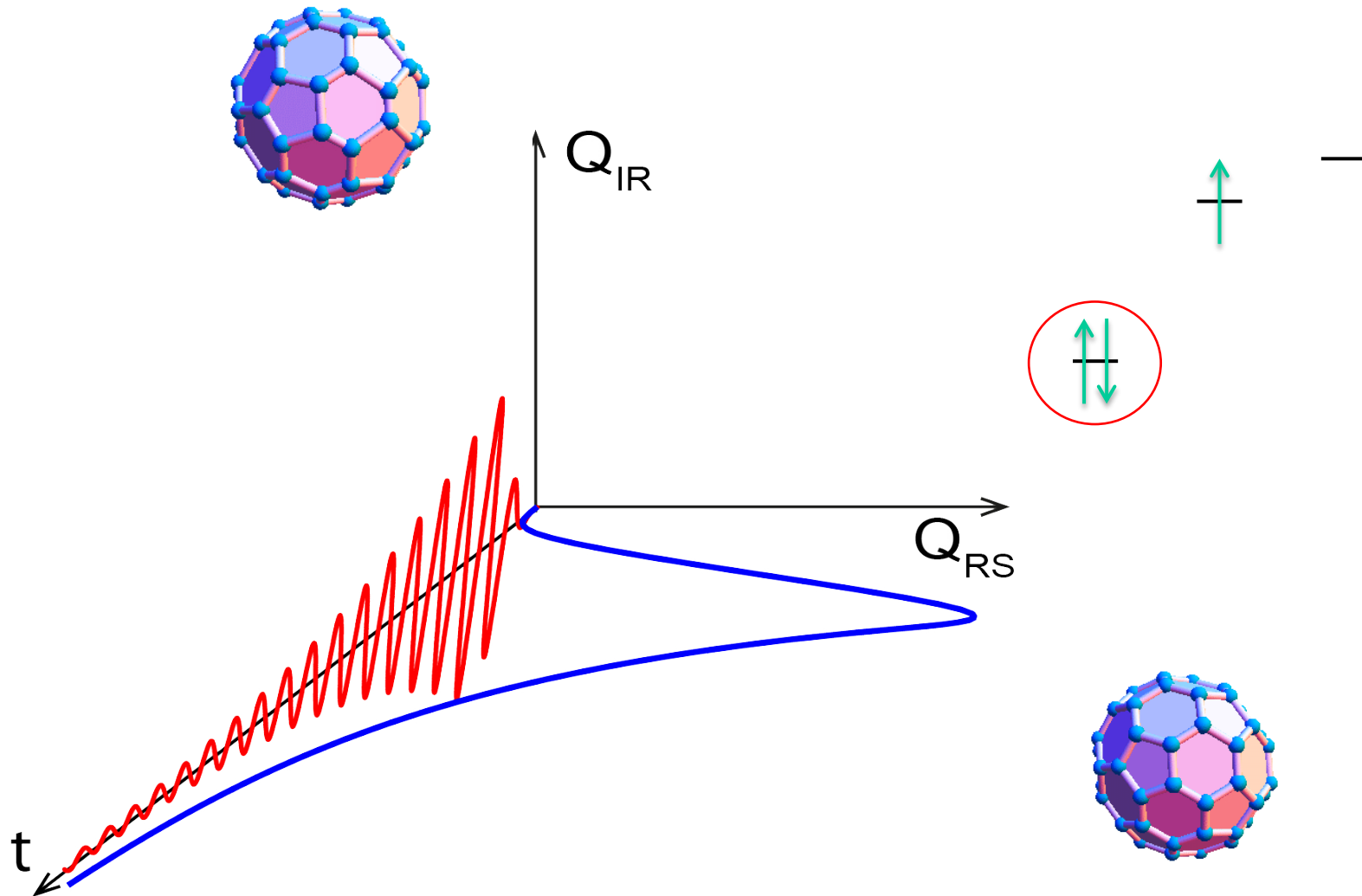


H_g



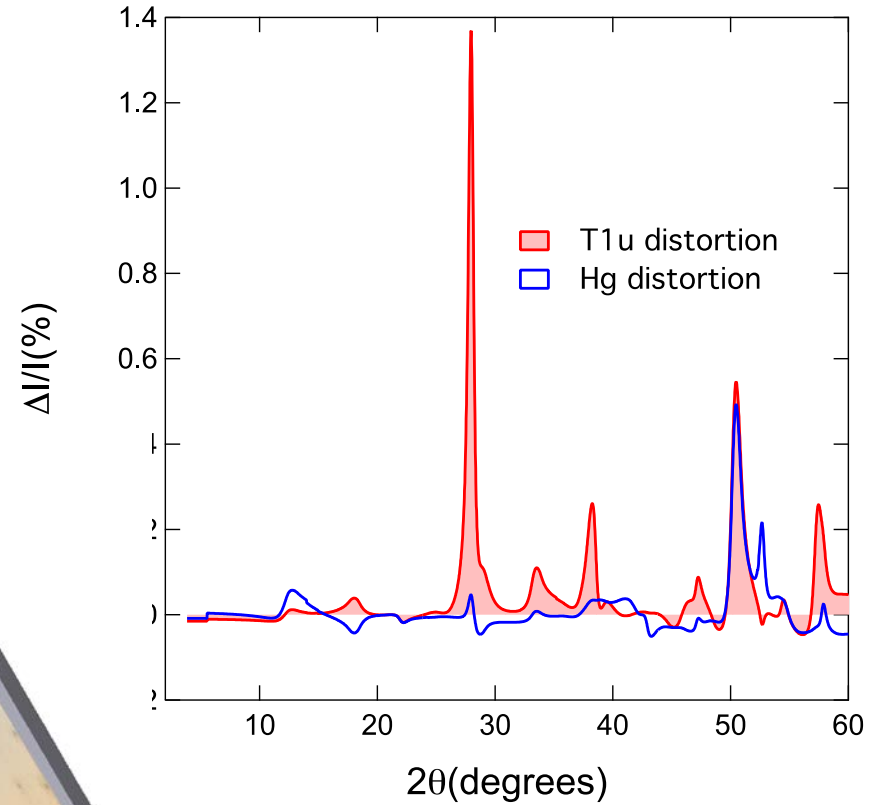
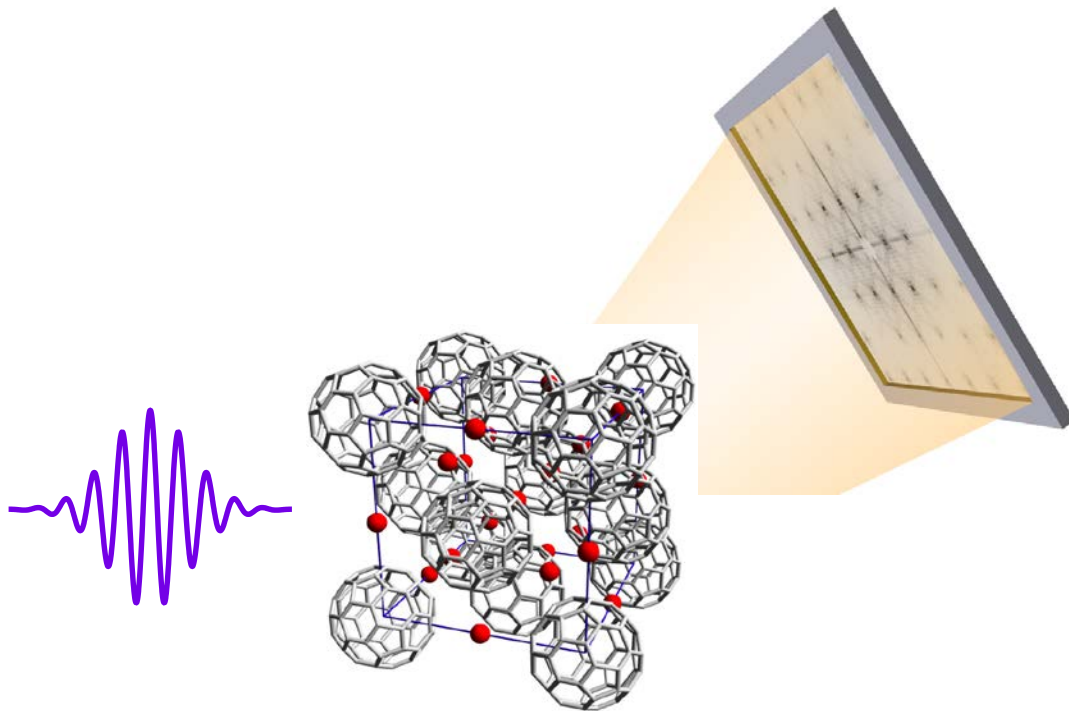
$H_g(1)$
 261 cm^{-1}

Dynamical enhancement of pairing ?



Or something else.....

Our new LCLS proposal





Matteo Mitrano



Roman Mankowski



Wanzheng Hu



Alice Cantaluppi

Theory

Stephen Clark
Dieter Jaksch
Oxford

A. Georges
Paris



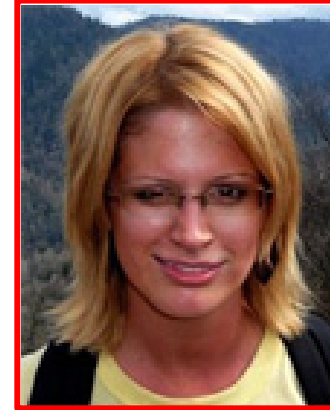
Daniele Nicoletti



Stefan Kaiser



Alaska Subedi



Cassi Hunt

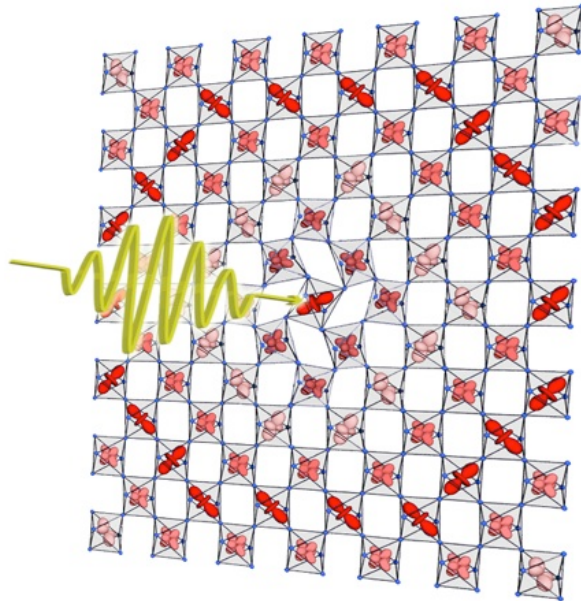
Samples

B. Keimer
Stuttgart

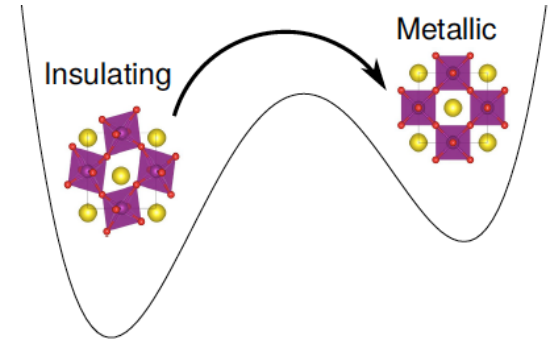
G. Gu
Brookhaven

H. Takagi
Stuttgart

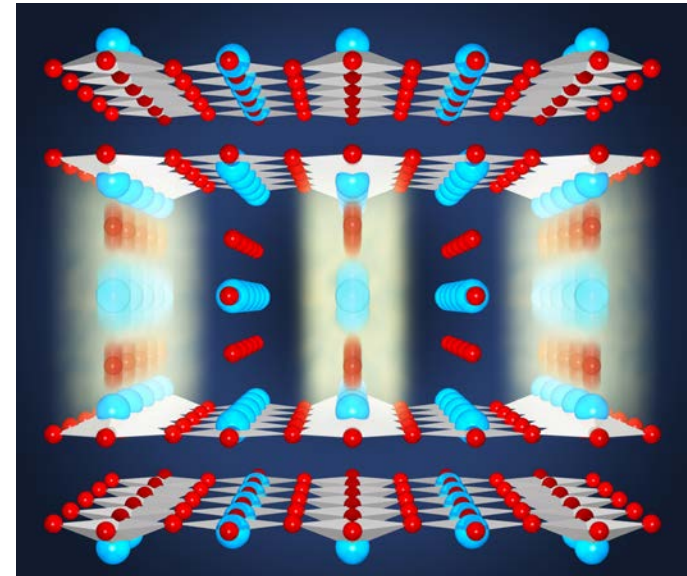
Controlling solids with light



Driving competing orders



Dynamical materials discovery



Non-equilibrium order

Non-equilibrium superconductivity

