

Quantum Condensed Matter Dynamics

Andrea Cavalleri

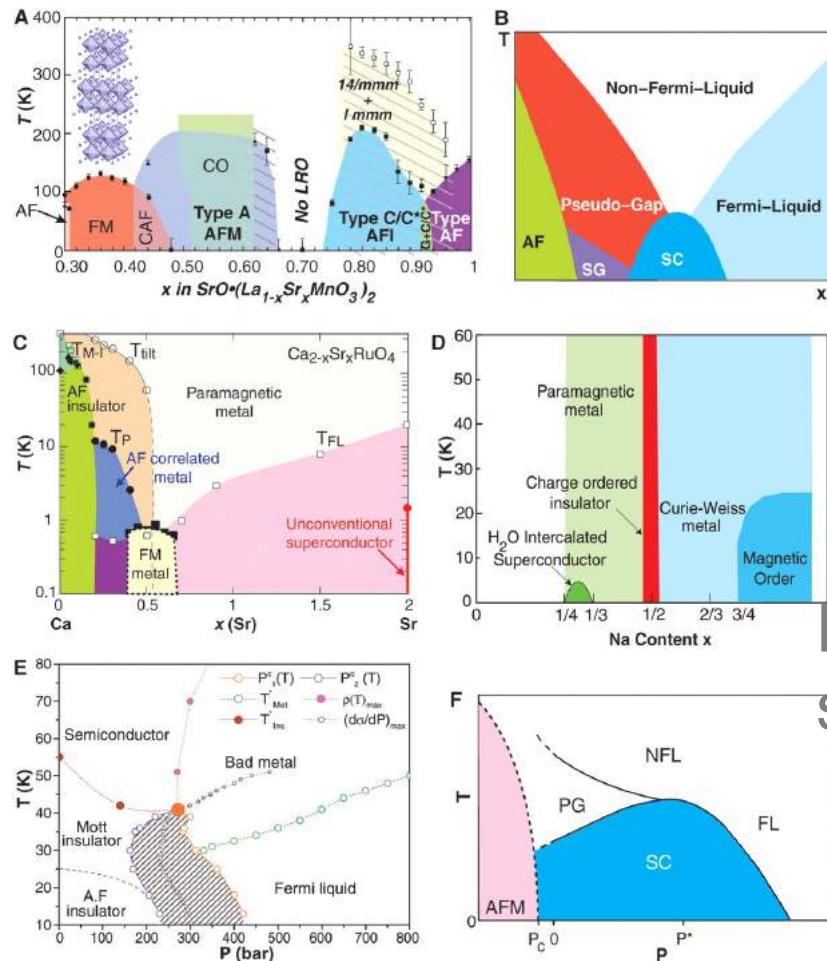
Max Planck Institut for the Structure and Dynamics of Matter

Lecture 3: Nonlinear Phononics I

Lecture 4: Nonlinear Phononics II

Quantum Materials

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

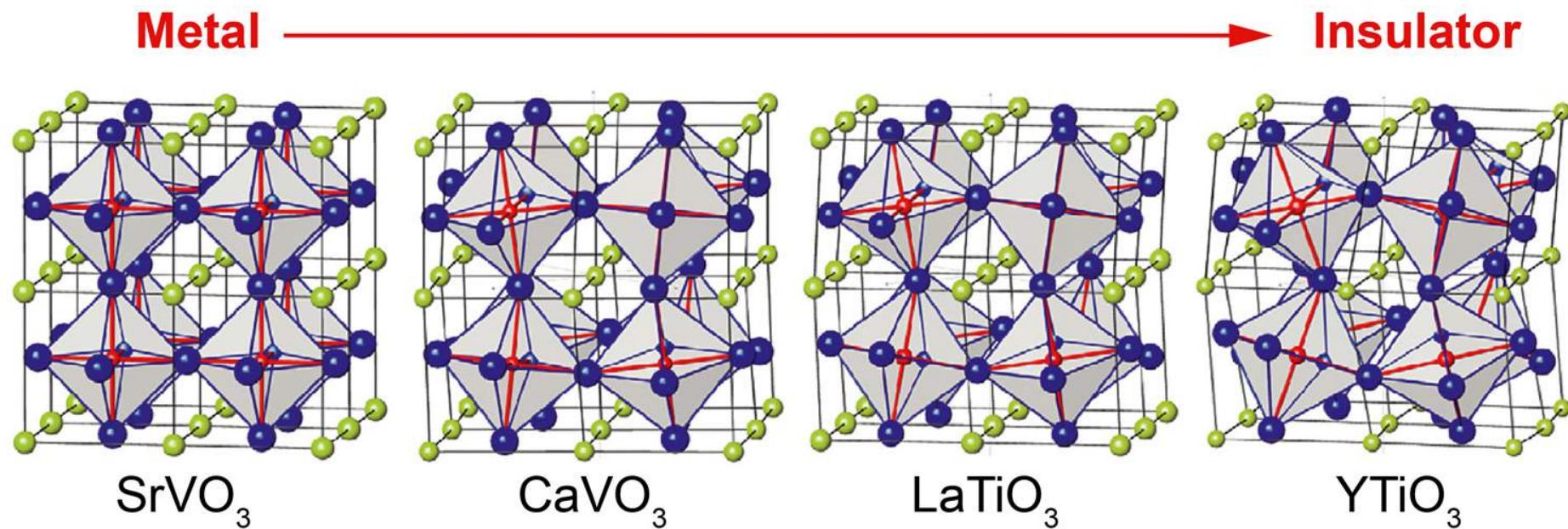


Metal-insulator transitions

Colossal magnetoresistance

High-temperature superconductivity

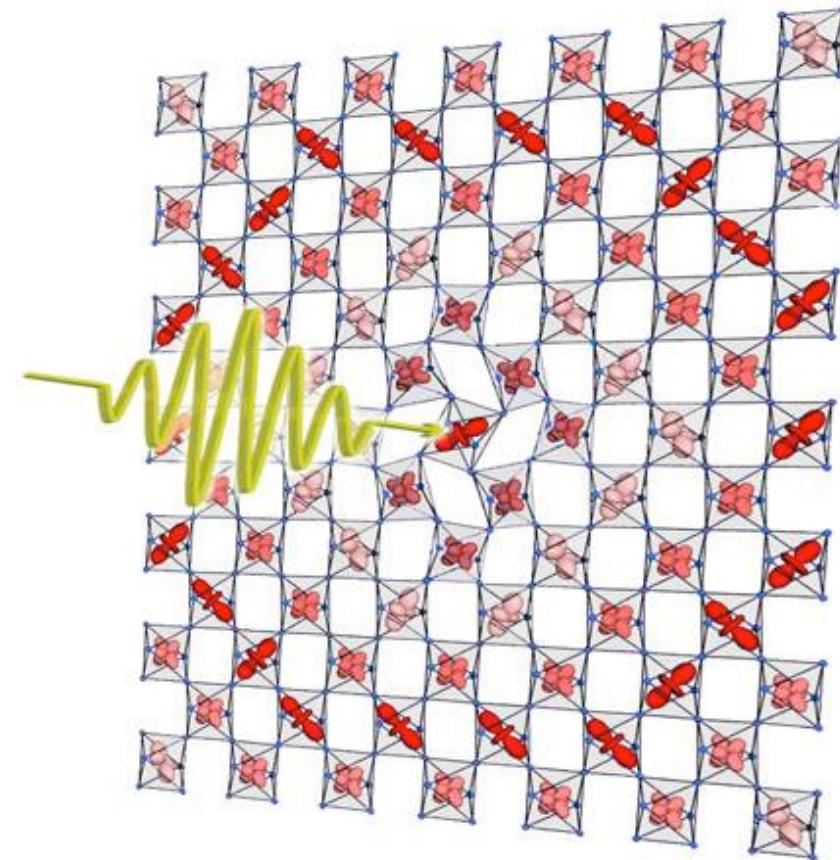
Structure-function in Quantum Materials



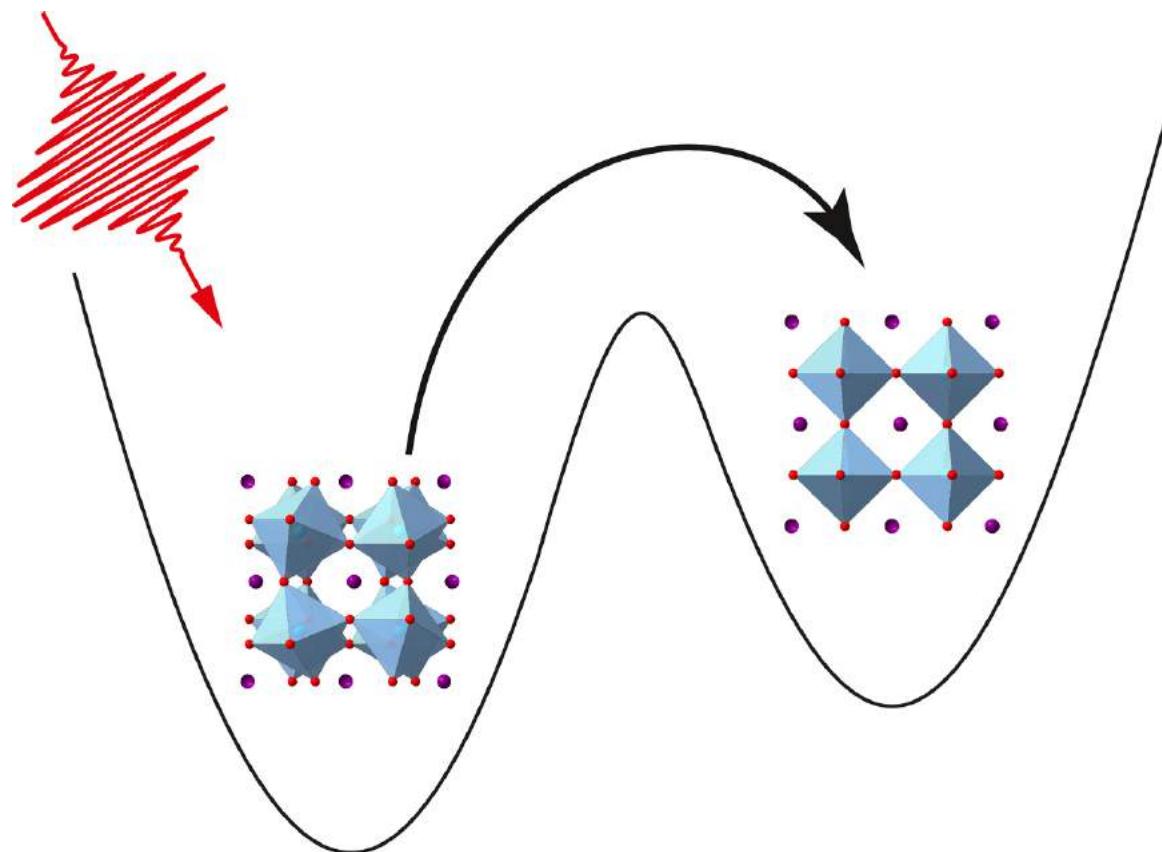
Selective control: single lattice coordinate

Mid infrared and THz light:
lattice distortions along one
(or few) normal mode
coordinates

Displacements $\sim 1\text{-}10\%$



Can one control specific bond angles with light ?

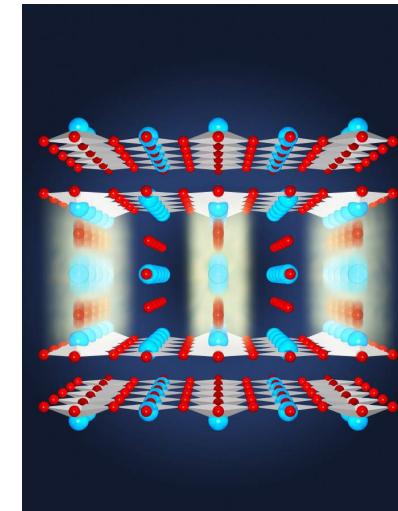
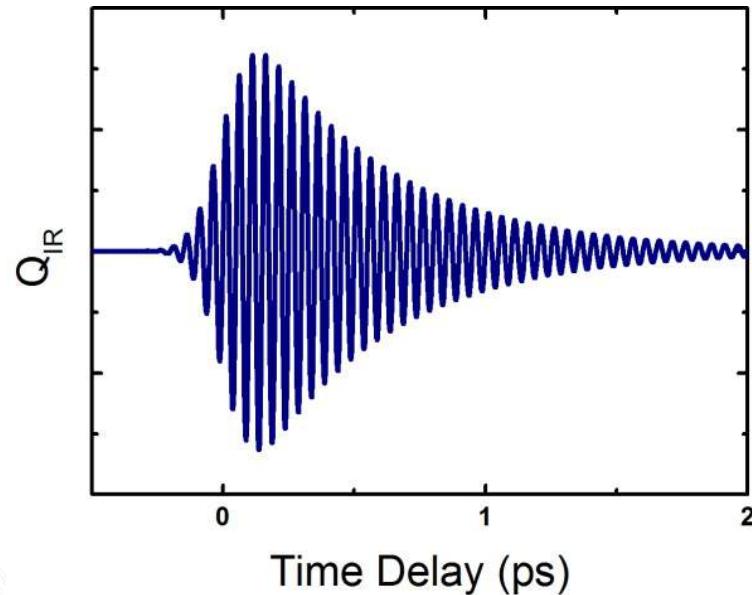


Linear coupling

Light couples to **IR active** phonons – whose coordinates that are odd against inversion

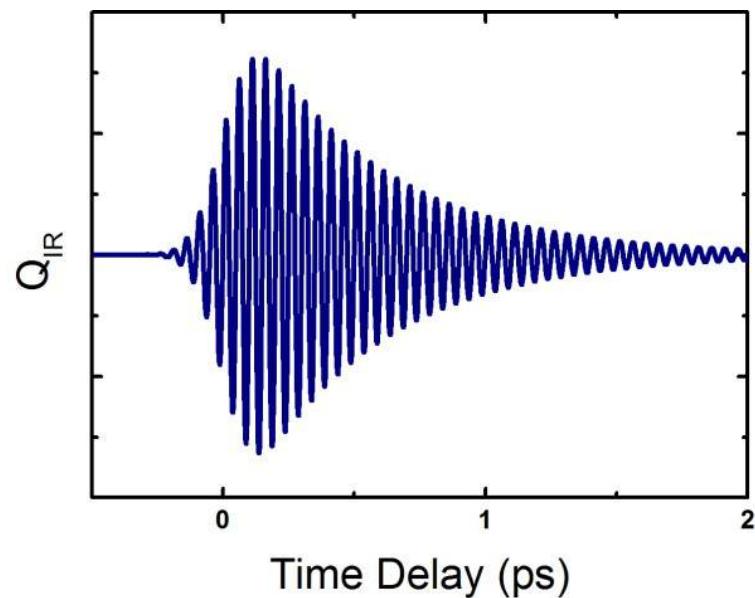
Linear optical excitation of IR-active modes does nothing on average

LINEAR - Q_{IR}

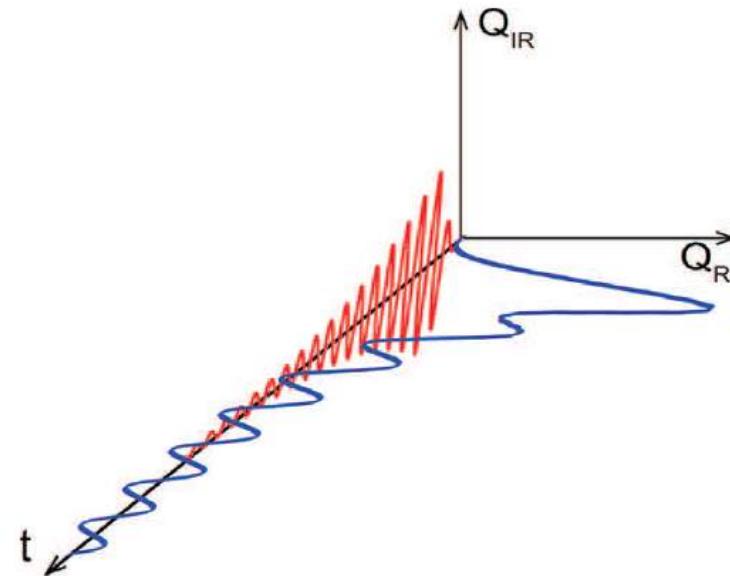


Today's lecture: beyond linear coupling

LINEAR - Q_{IR}

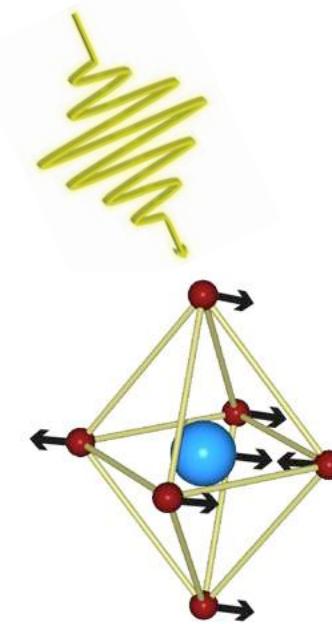
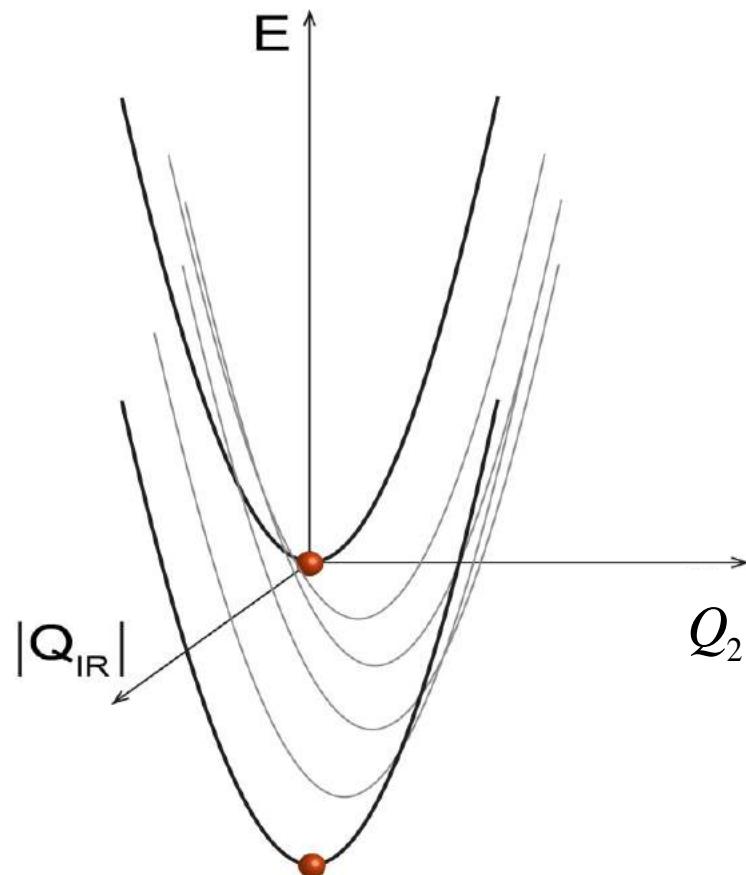


NONLINEAR - Q_2



Lowest order non-linear coupling

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



If material centrosymmetric

$$U_{\text{int}} = A Q_{ir}^2 Q_2$$

|

even

|

odd²even

Interaction is always between a
driven odd mode and an even mode

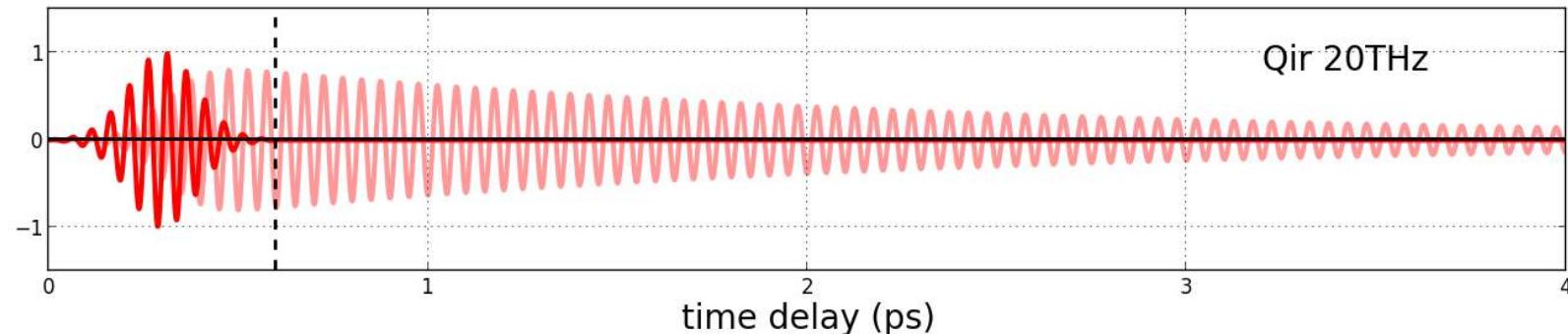


Equations of motion: oscillations in Q_{IR}

$$\ddot{Q}_{IR} + \gamma_{IR}\dot{Q}_{IR} + \omega_{IR}^2 Q_{IR} = AE_{laser}^{i\omega t}$$

harmonic oscillator

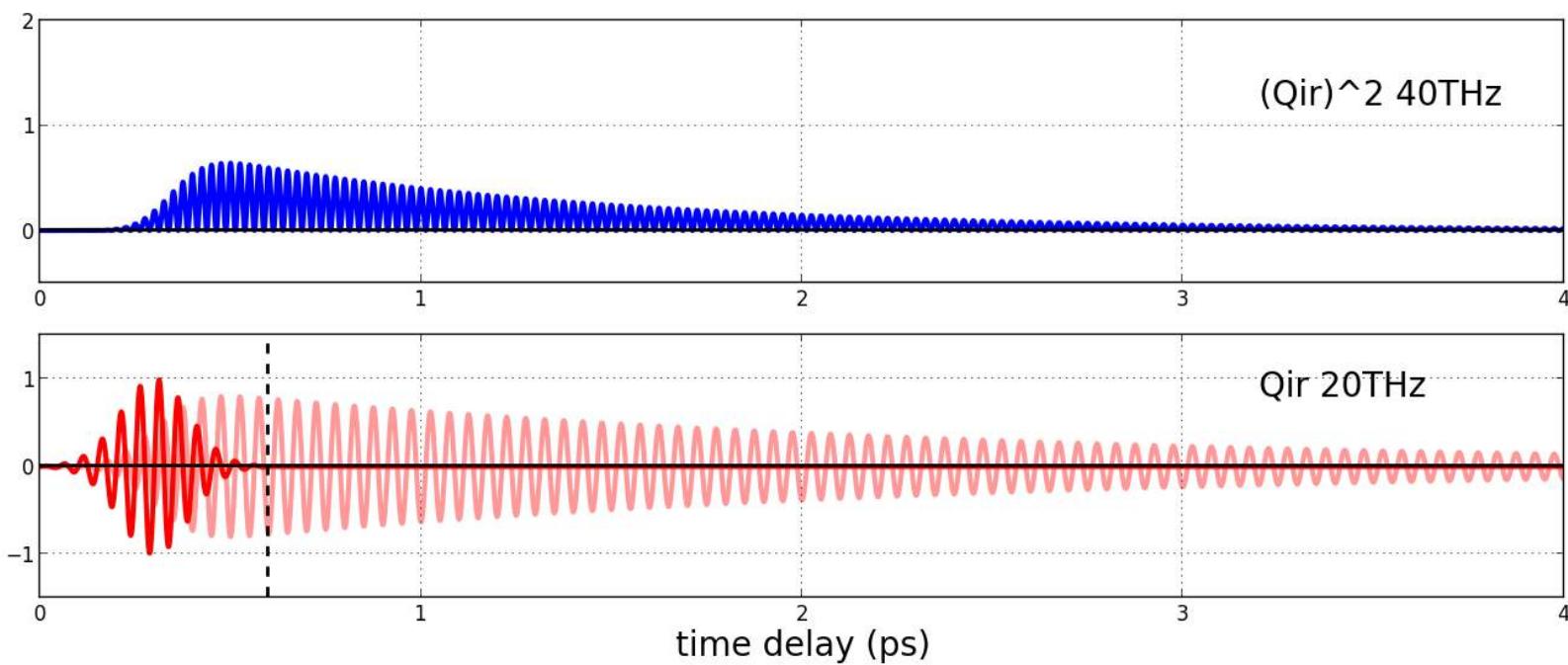
Laser field



Equations of motion: oscillations in $(Q_{IR})^2$

$$\ddot{Q}_{IR} + \gamma_{IR}\dot{Q}_{IR} + \omega_{IR}^2 Q_{IR} = AE_{laser}^{i\omega t}$$

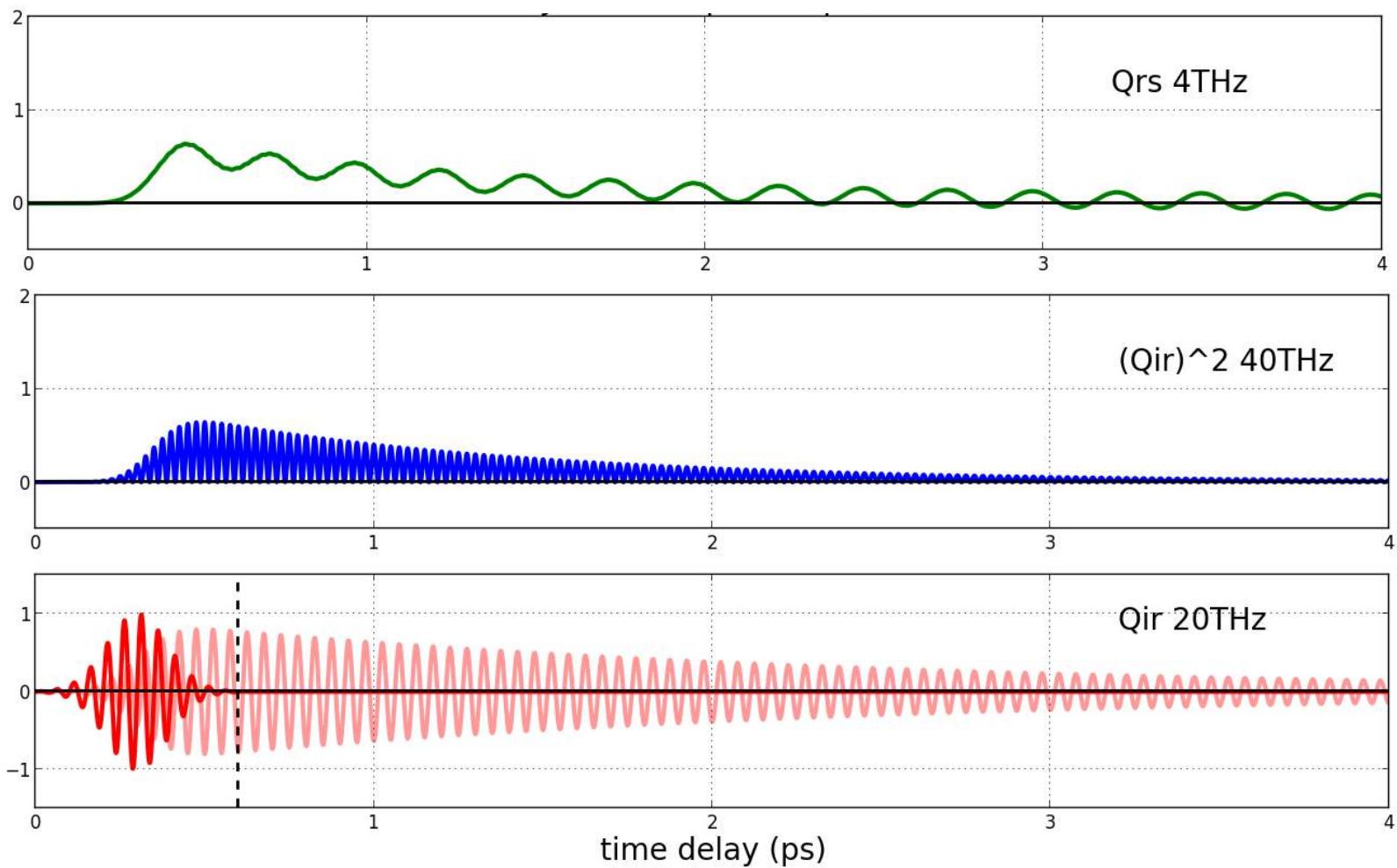
$$(\ddot{Q}_2 + \gamma\dot{Q}_2 + \omega_2^2 Q_2) = BQ_{IR}^2$$



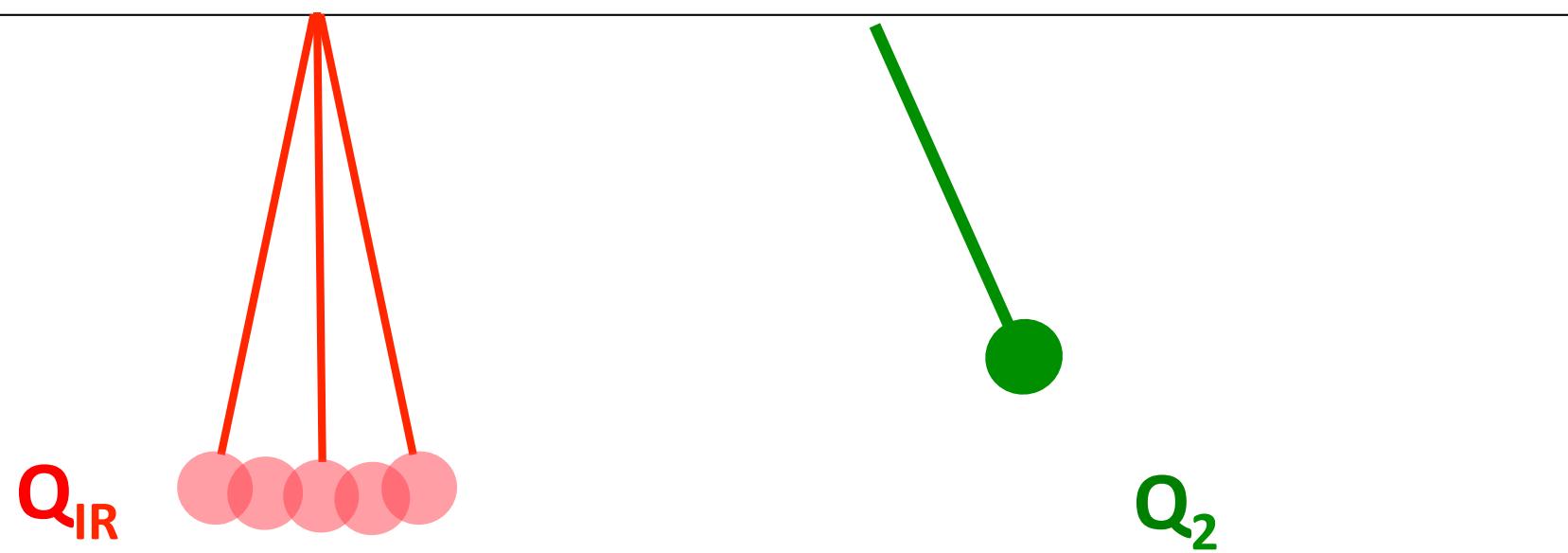
Oscillations in Q_{IR} displace Q_2

$$\ddot{Q}_{IR} + \gamma_{IR}\dot{Q}_{IR} + \omega_{IR}^2 Q_{IR} = AE_{laser}^{i\omega t}$$

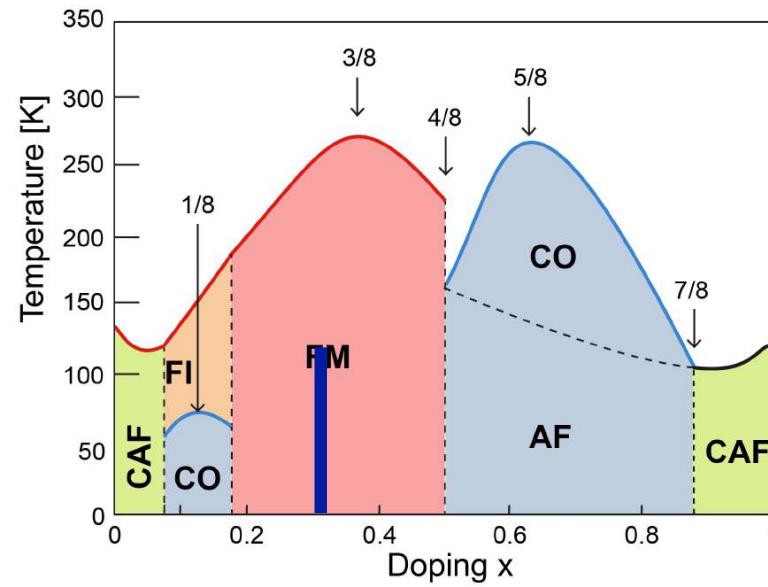
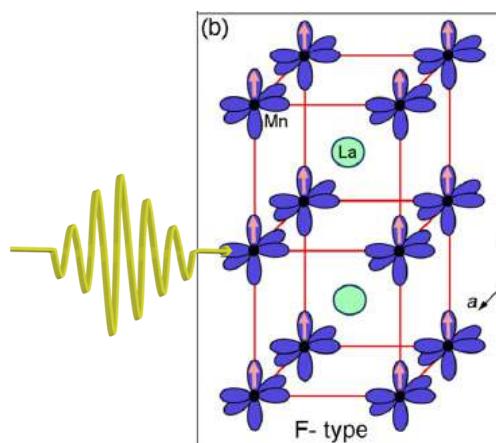
$$(\ddot{Q}_2 + \gamma\dot{Q}_2 + \omega_2^2 Q_2) = BQ_{IR}^2$$



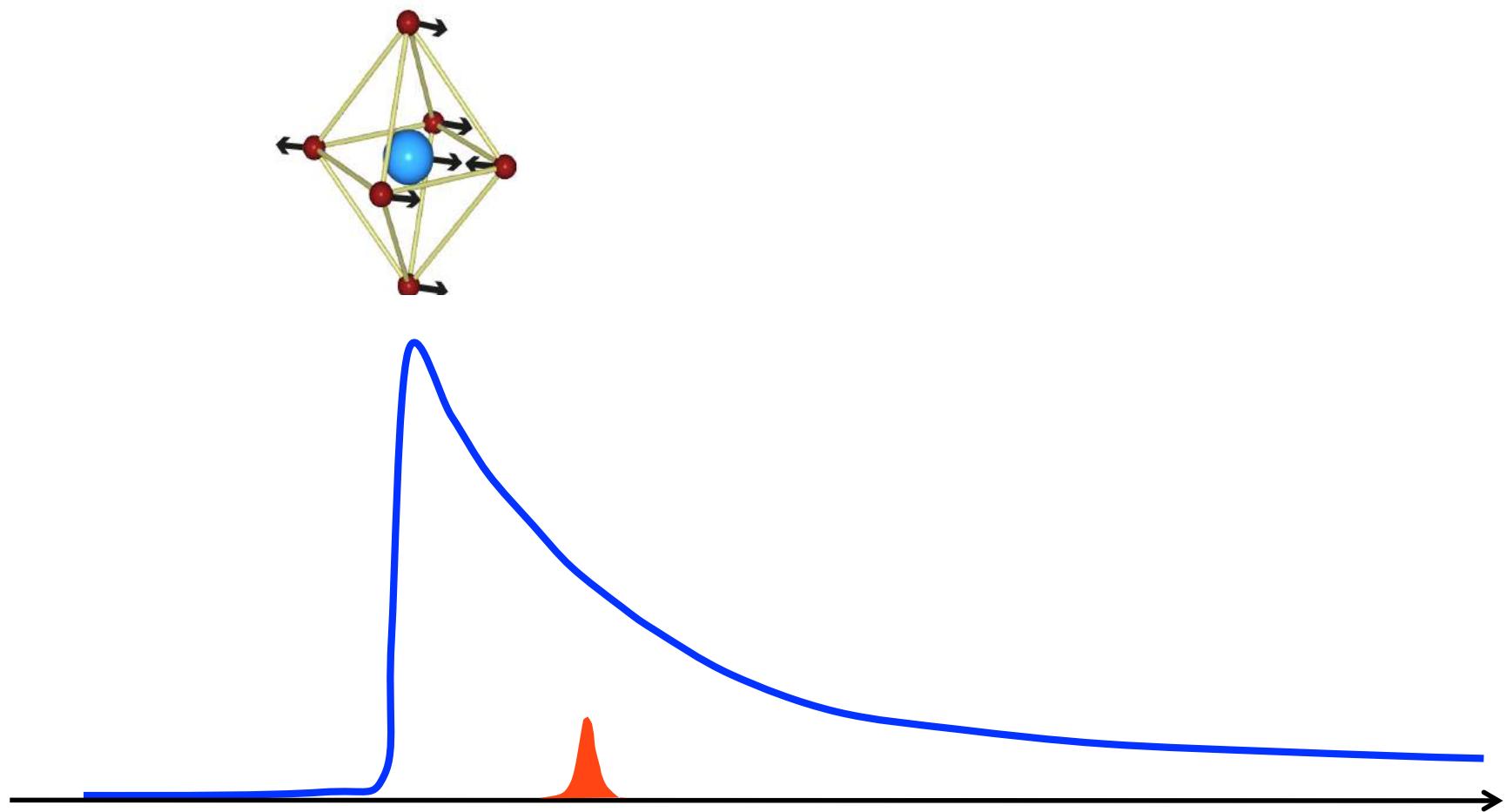
Q_{IR} Q_2 term: Oscillations in Q_{IR} displace Q_2



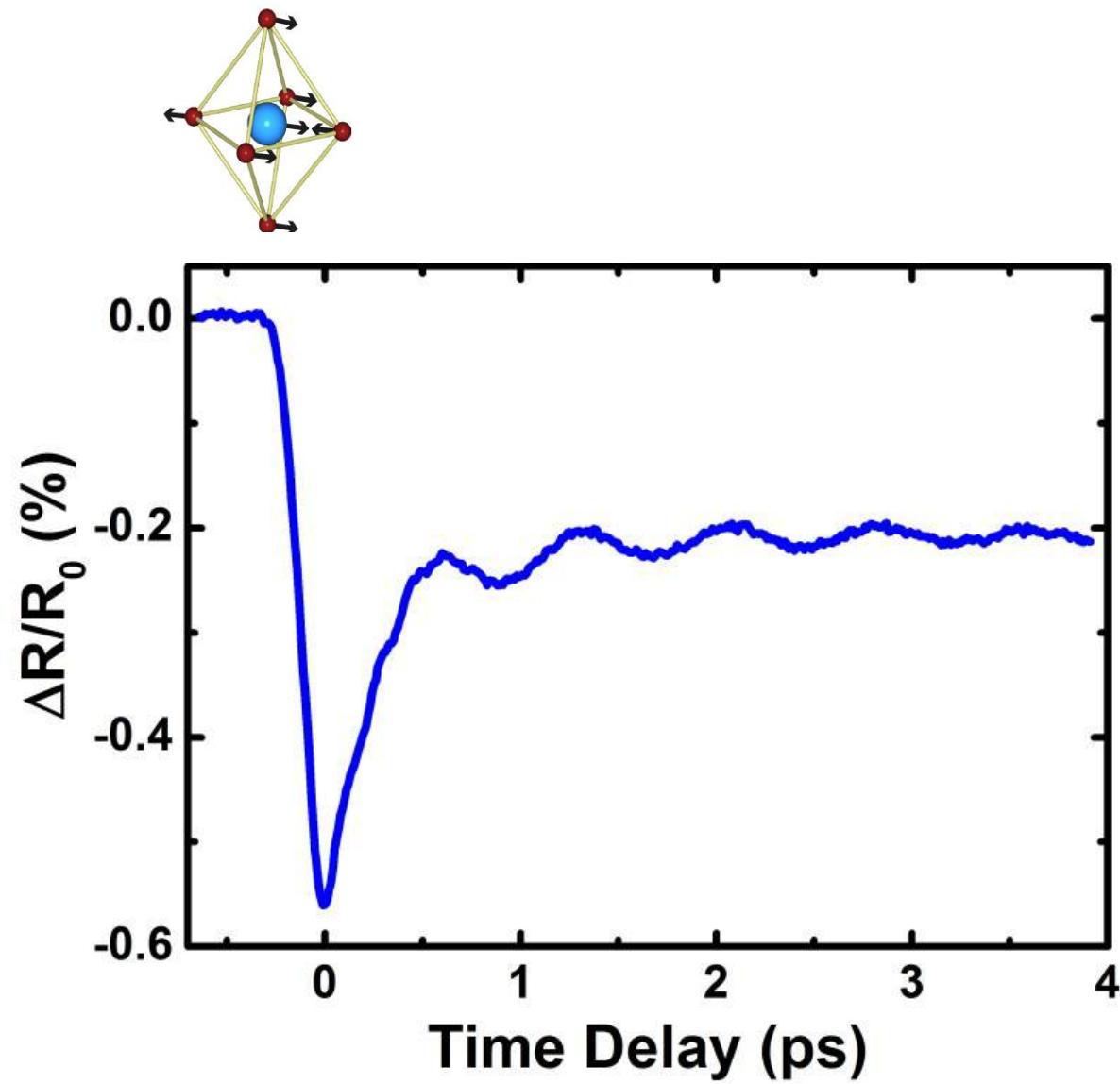
Example: $\text{La}_{0.3}\text{Sr}_{0.3}\text{MnO}_3$



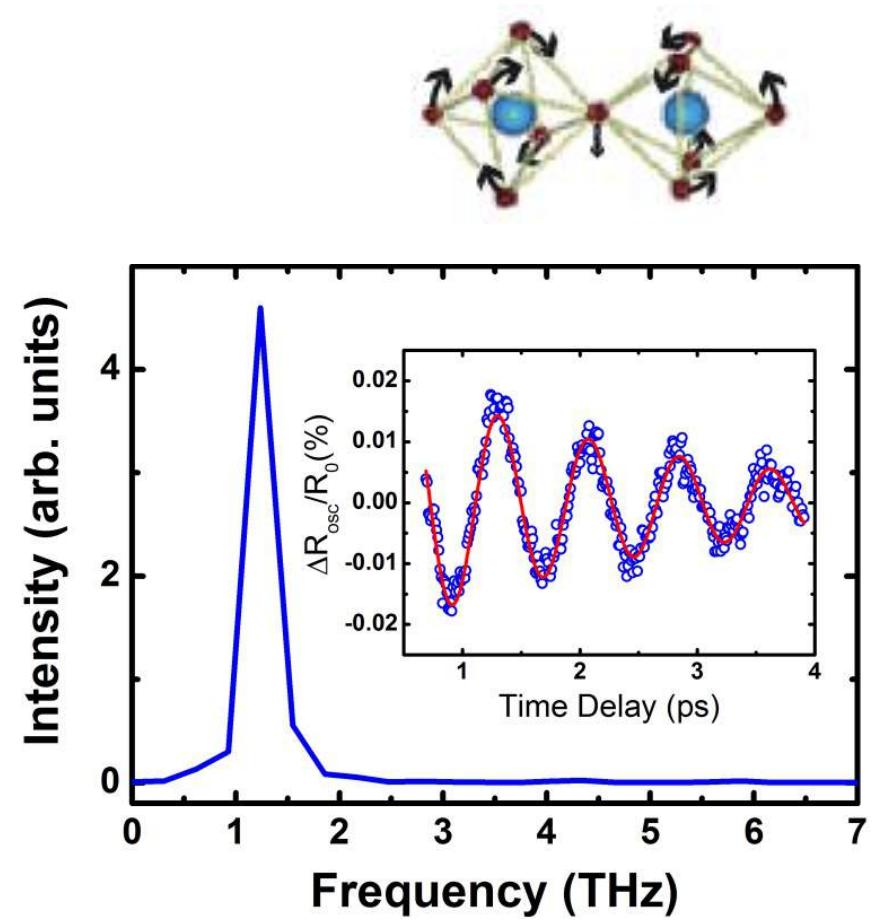
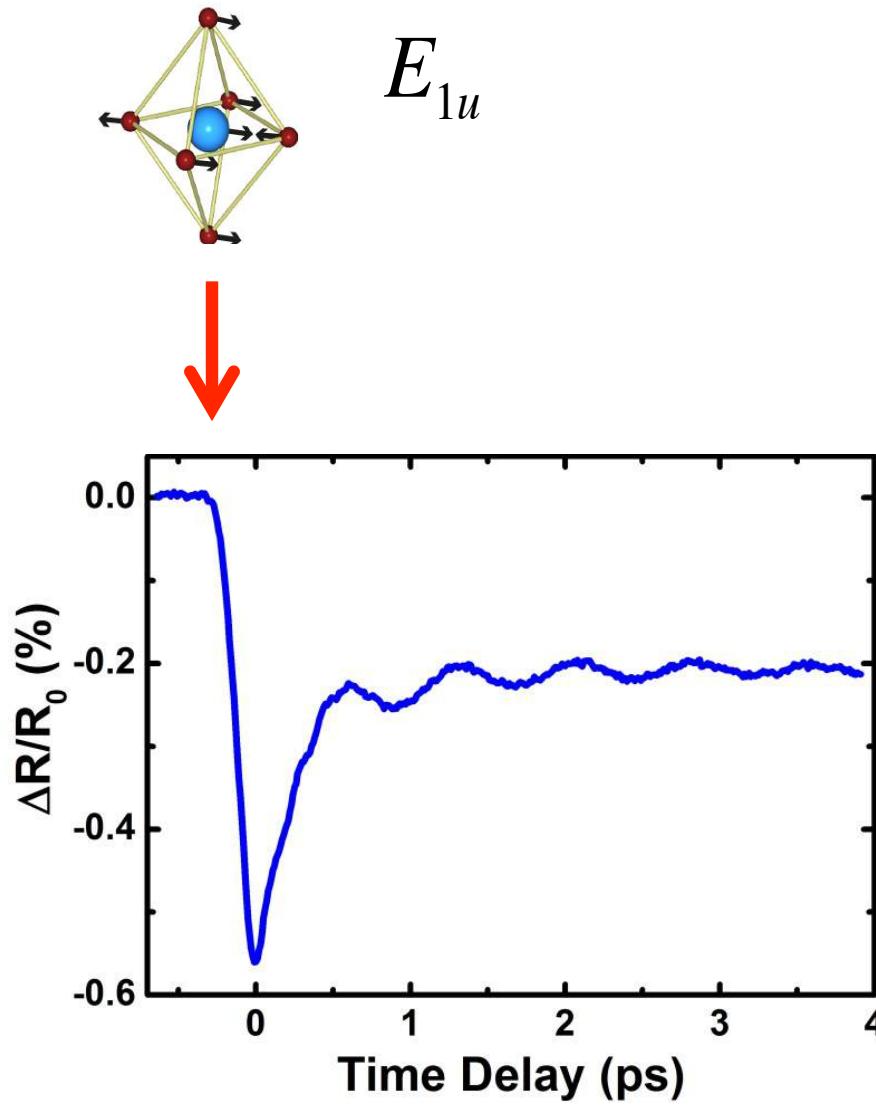
Excite Oxygen Stretch of E_u symmetry



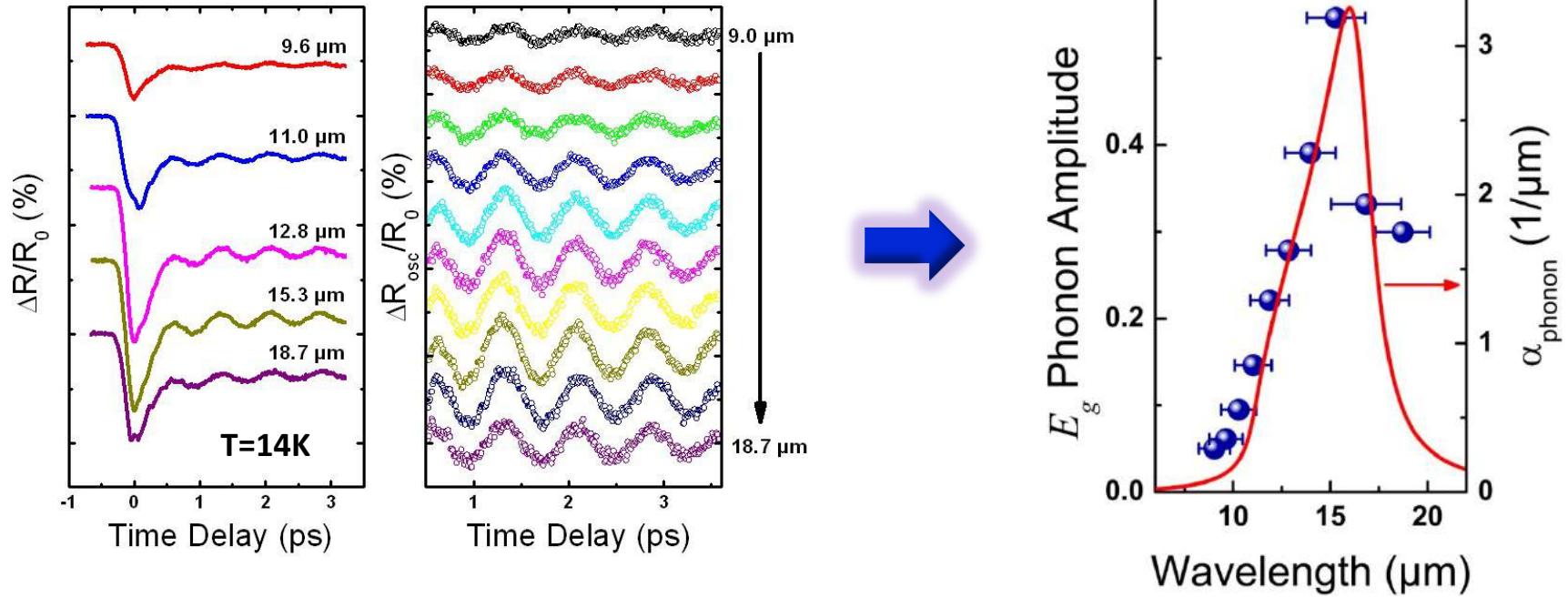
Time resolved reflectivity oscillates



Oscillations indicate excitation of E_{g} mode

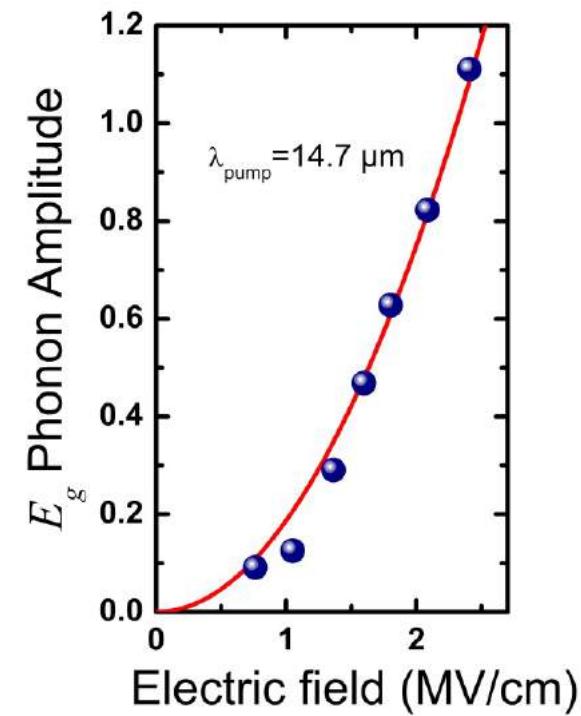
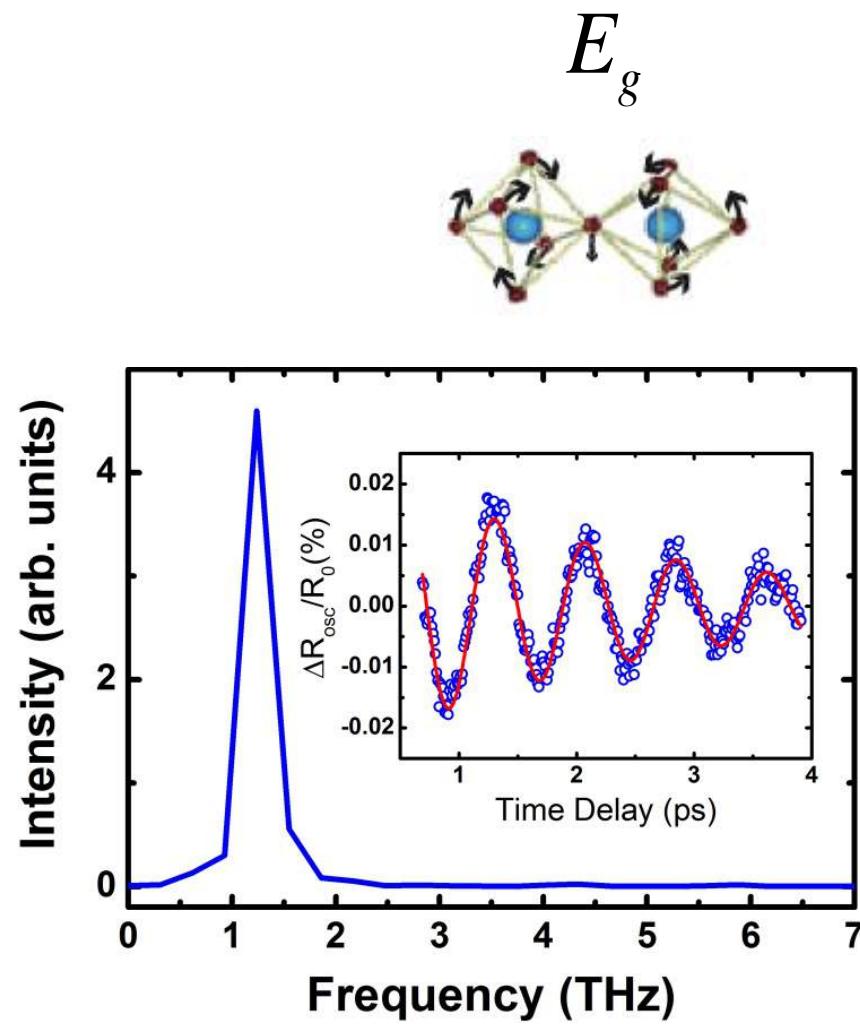


E_g only there if light resonant with E_u



M. Foerst et al., Nature Physics 7, 854 (2011)

E_g amplitude follows $(E_{\text{laser}})^2$



E_g mode has the correct symmetry

$$U_{\text{int}} = A Q_{ir}^2 Q_2$$

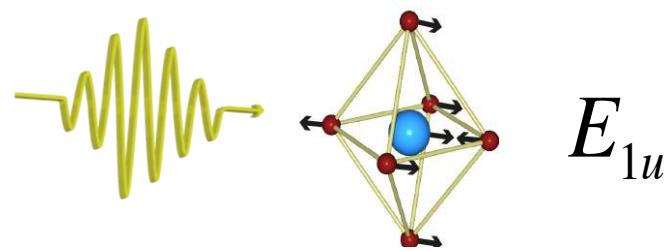


$$E_{1u}^2 E_g$$

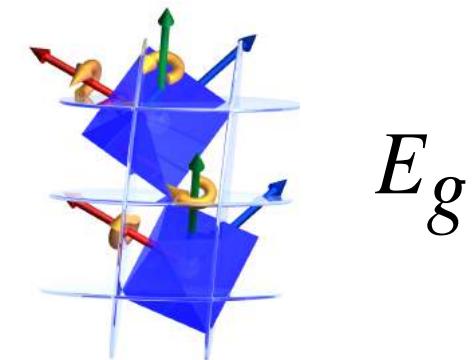


Is there an average displacement along E_g ?

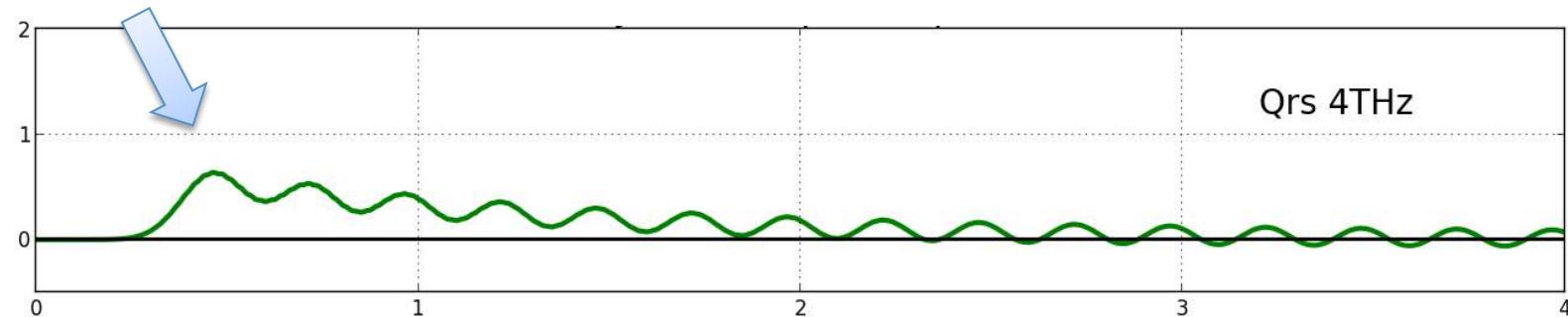
Mid-IR pump (E_{1u} mode)



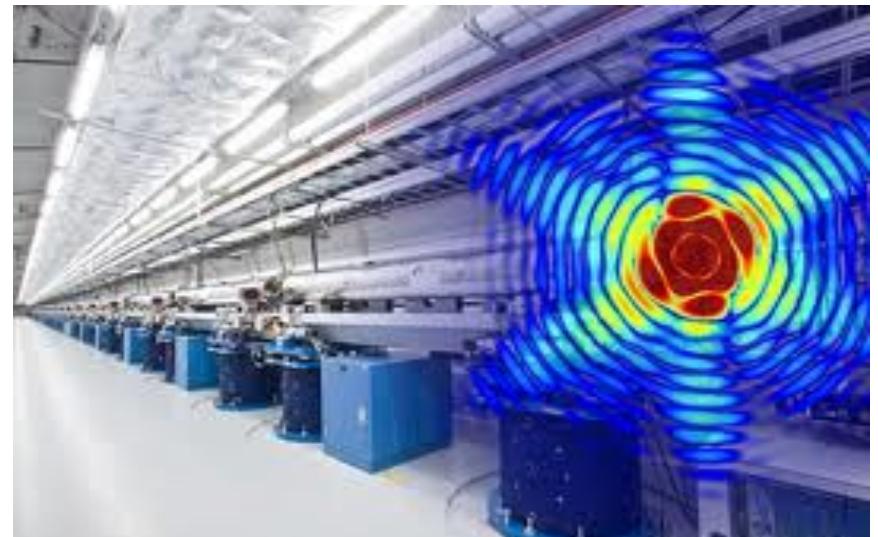
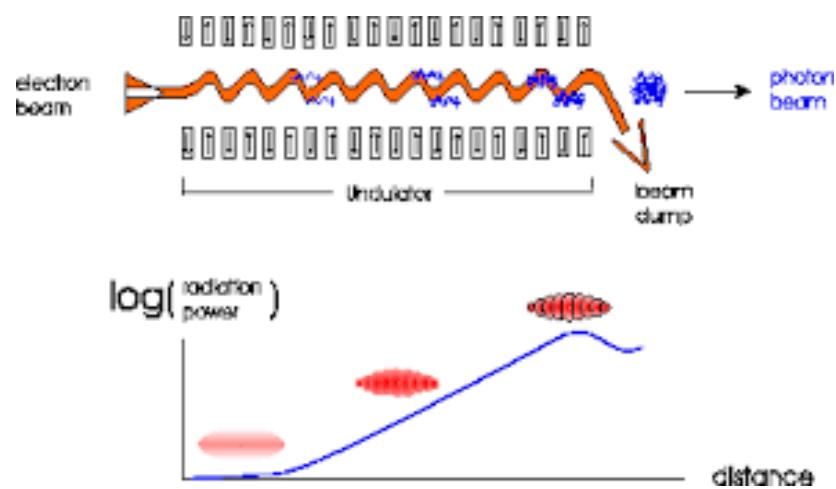
Displacive field (E_g mode)



$Q_r(t)$

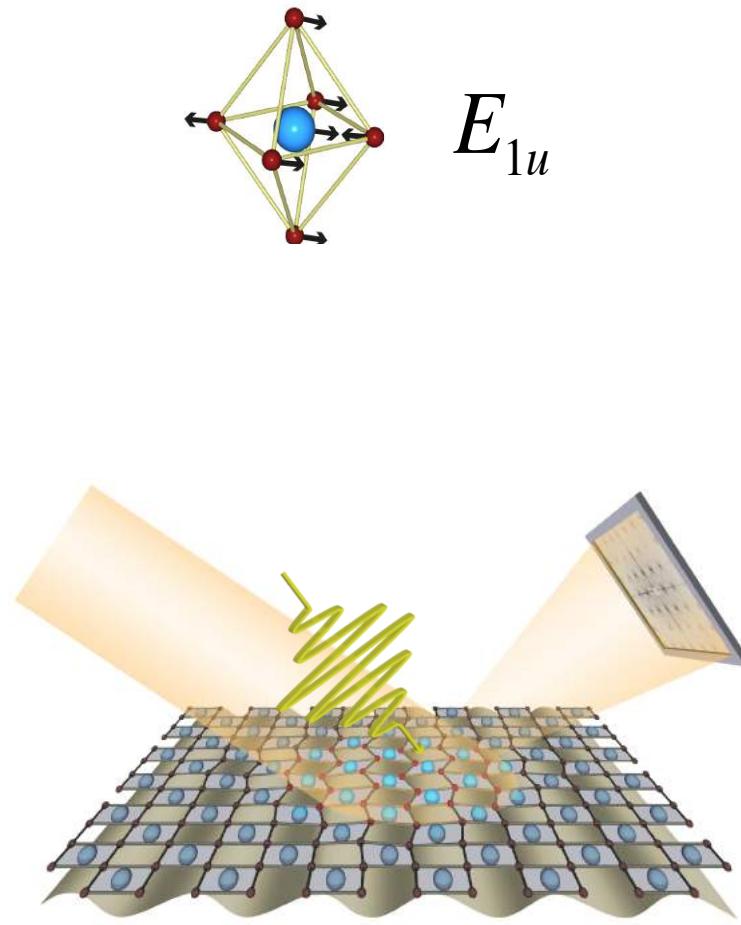


Pump probe experiment using X-ray FEL

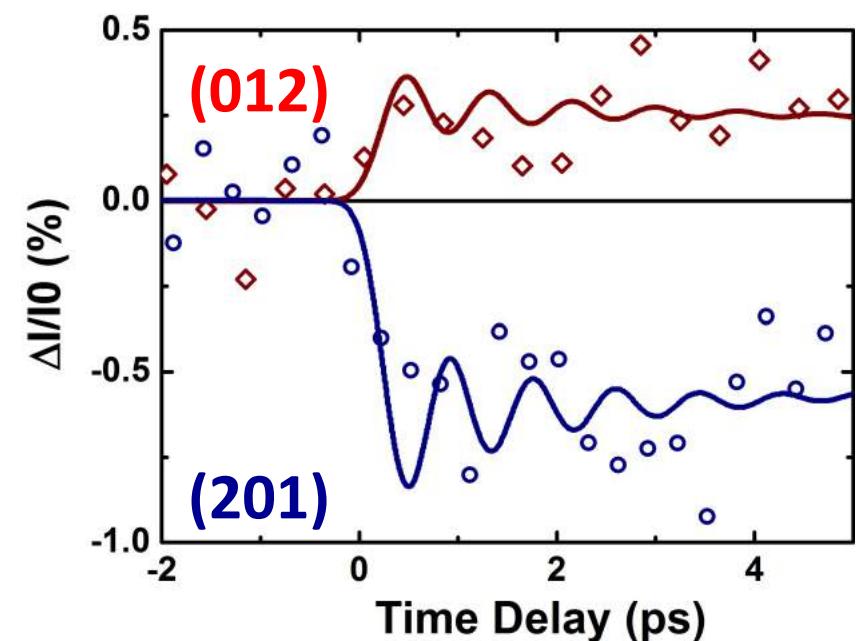
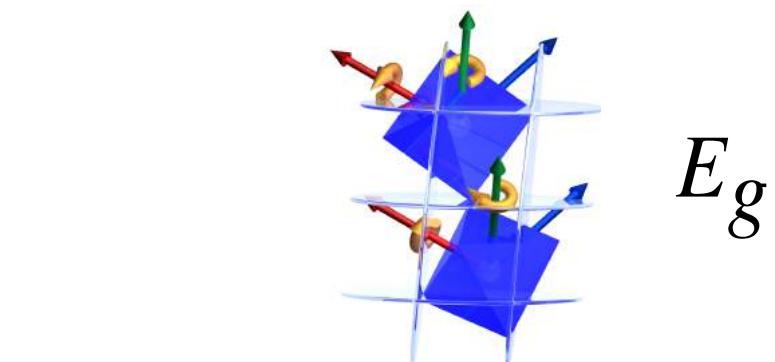


Step change in structure factor

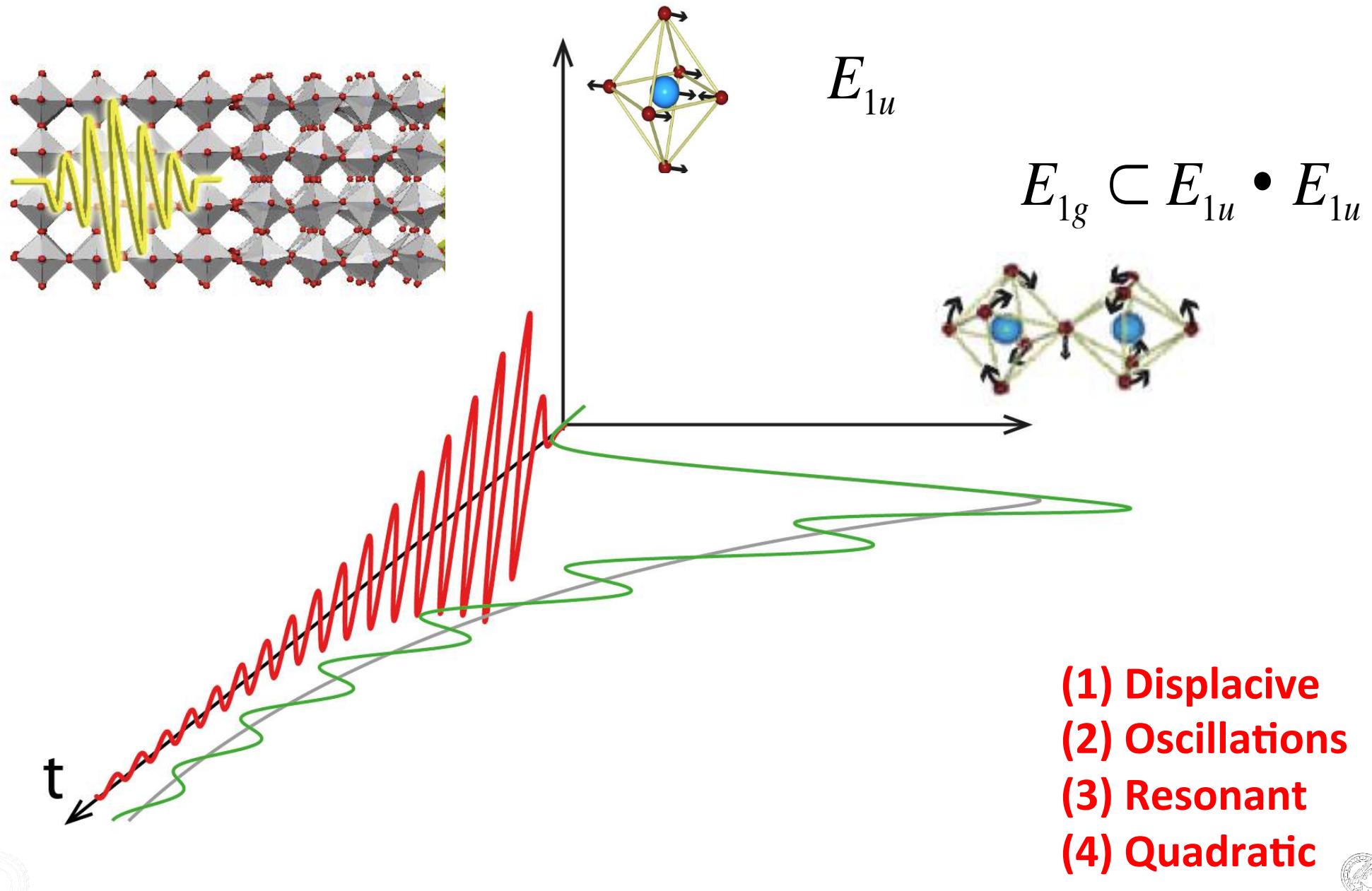
Mid-IR pump (E_{1u} mode)



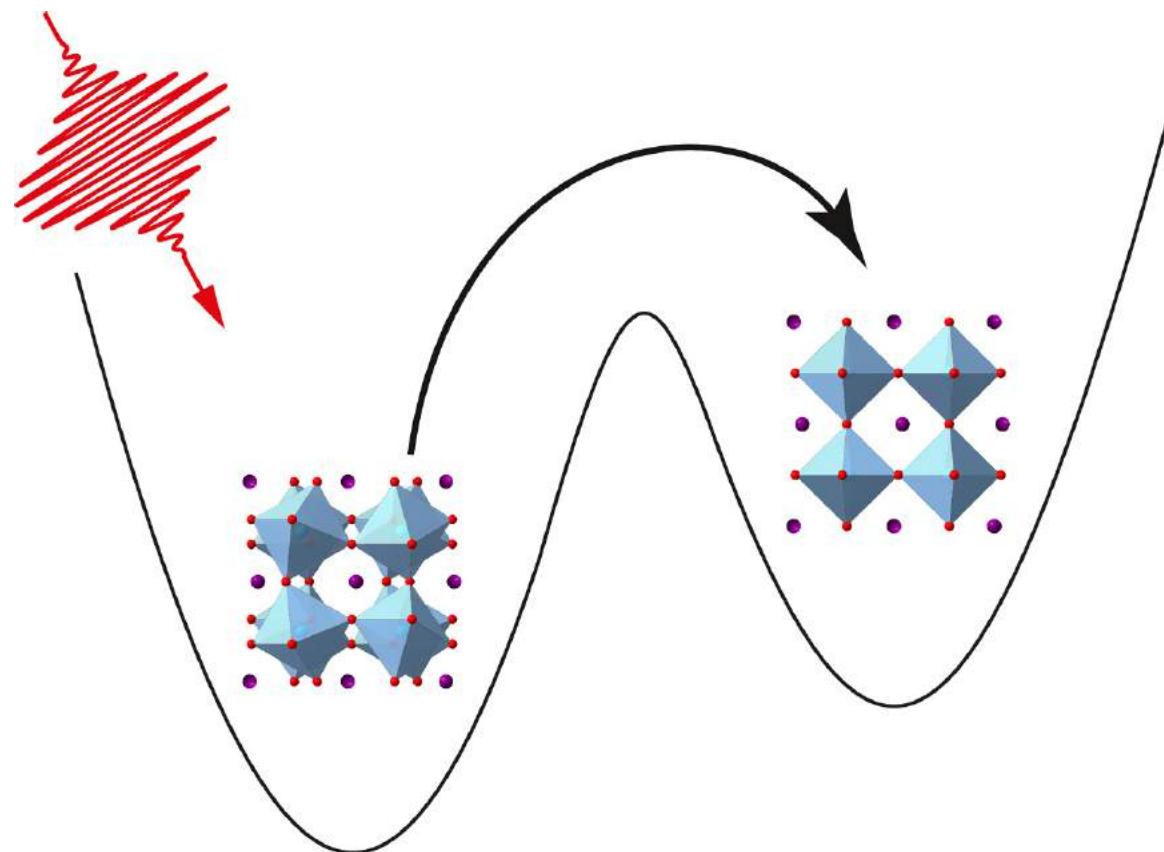
Displacive field (E_g mode)



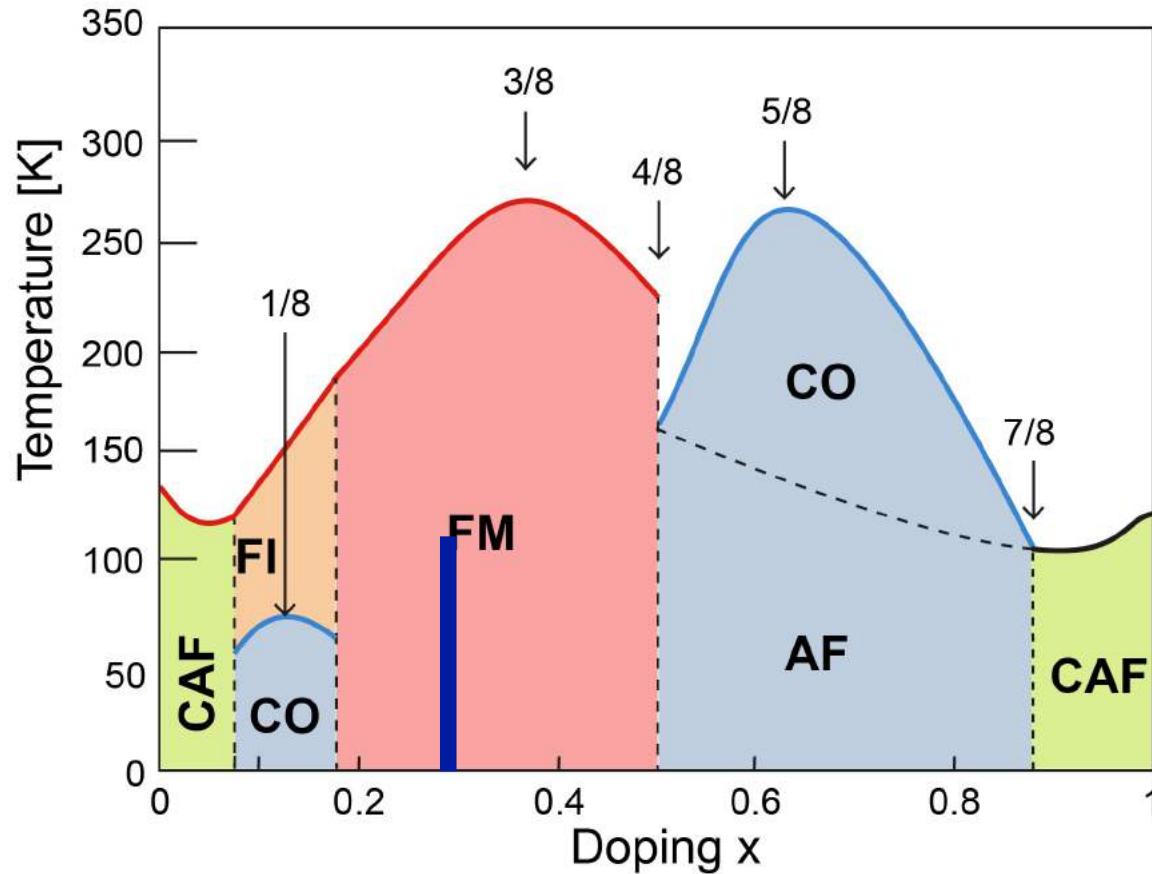
Lowest order nonlinear phononics



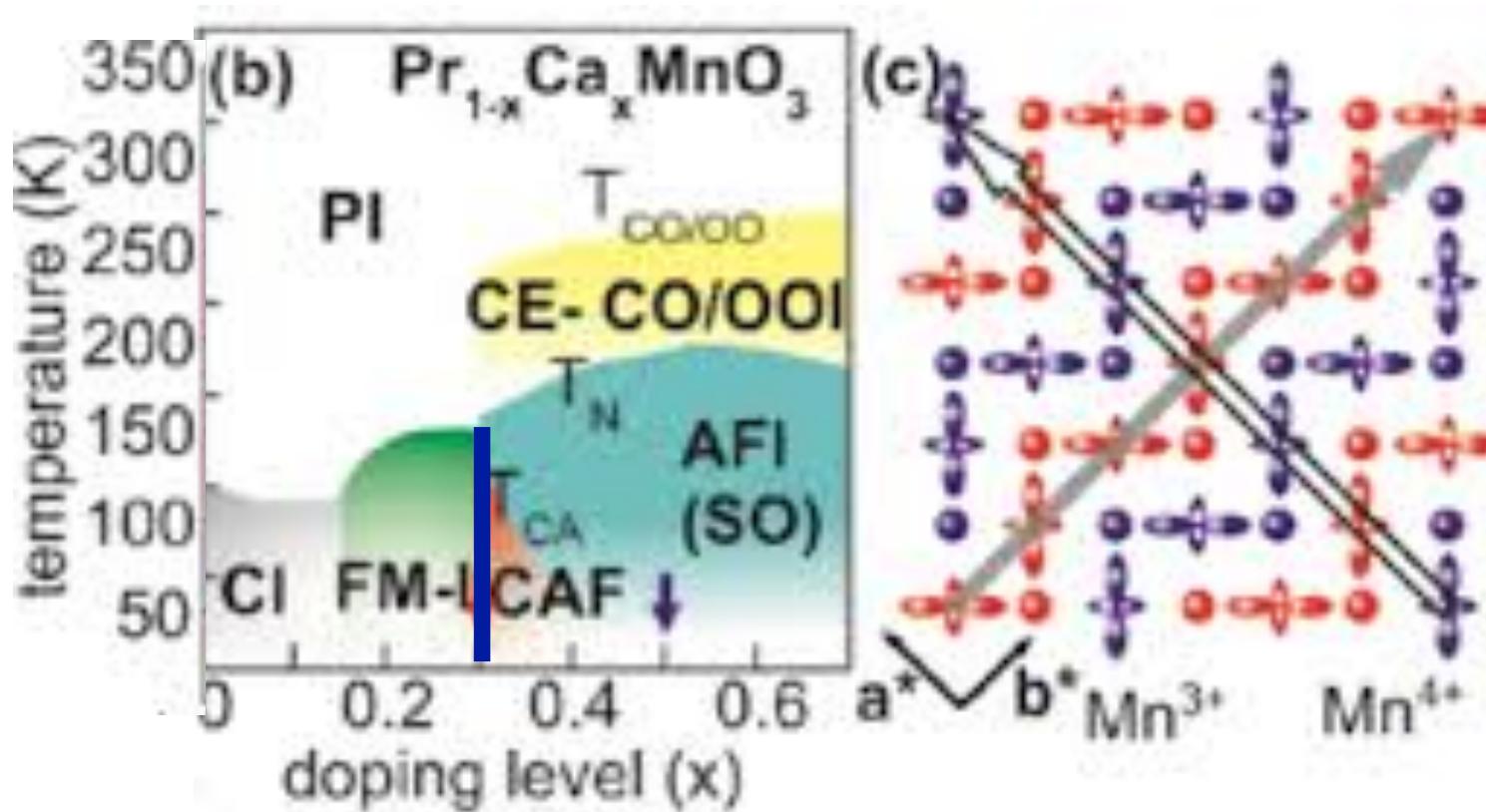
Can one control specific bond angles with light ?



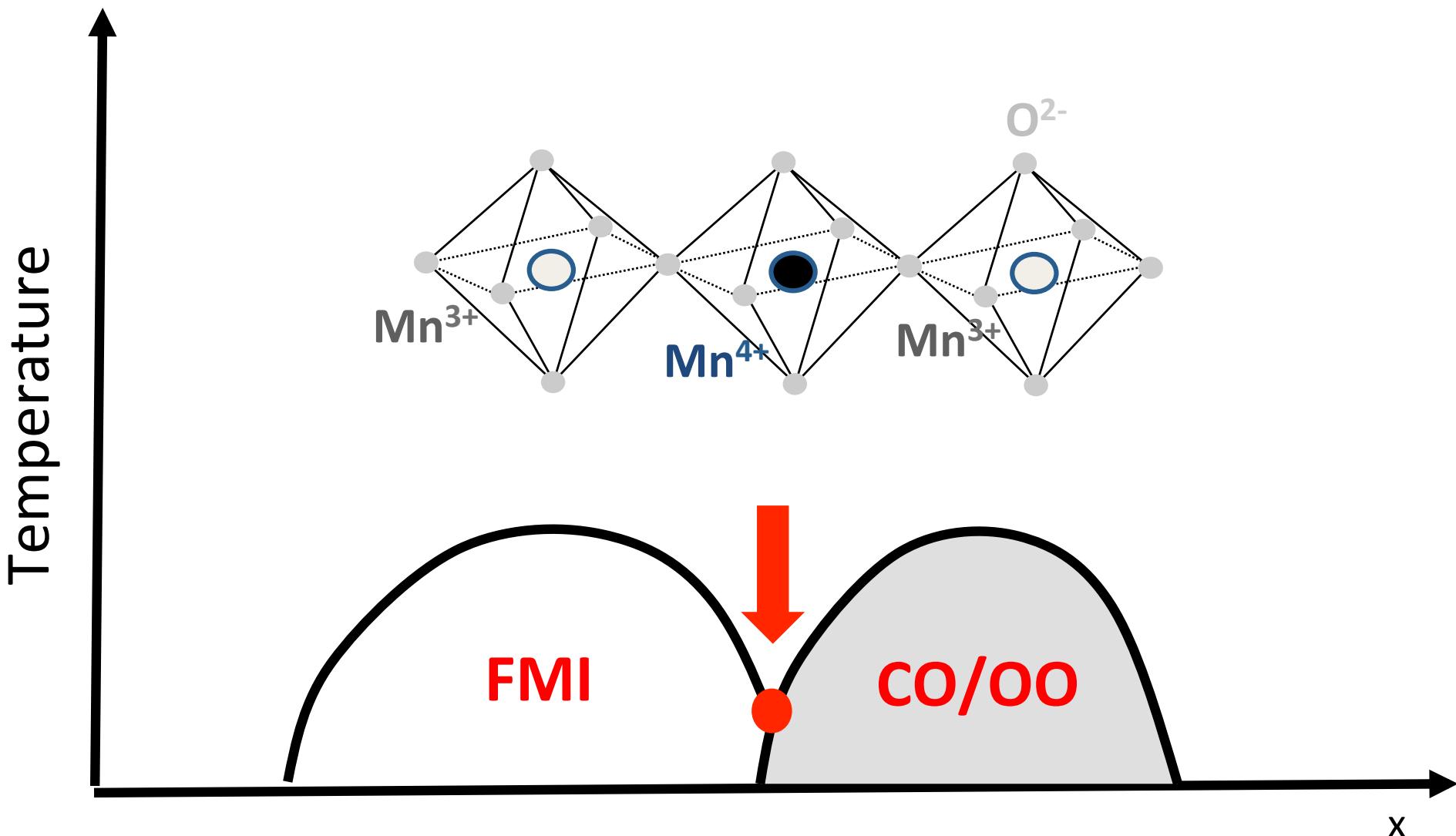
Demonstration away from phase boundary



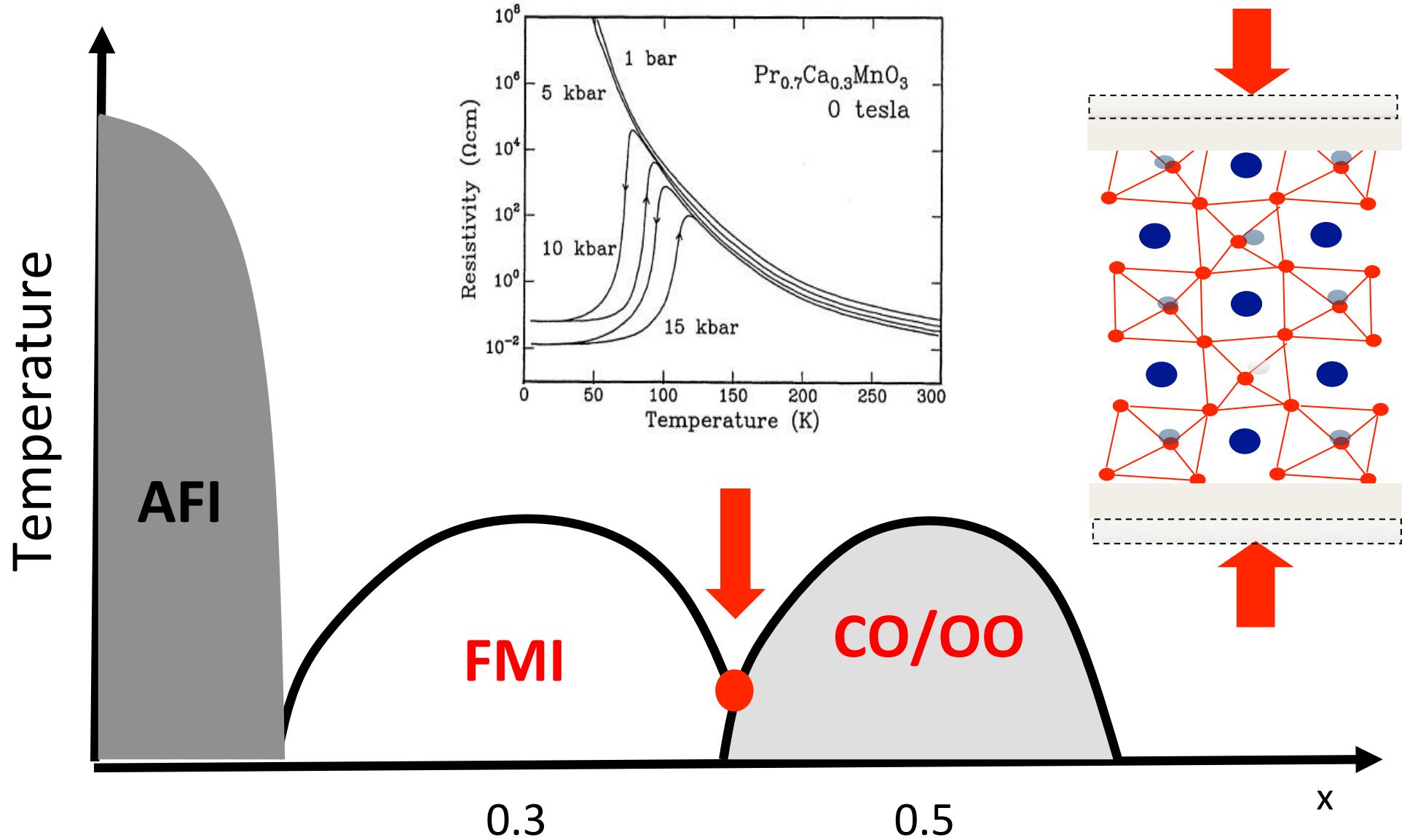
Close to a Phase Boundary: $\text{Pr}_{0.3}\text{Ca}_{0.3}\text{MnO}_3$



$\text{Pr}_{0.3}\text{Ca}_{0.3}\text{MnO}_3$: competition between two insulators



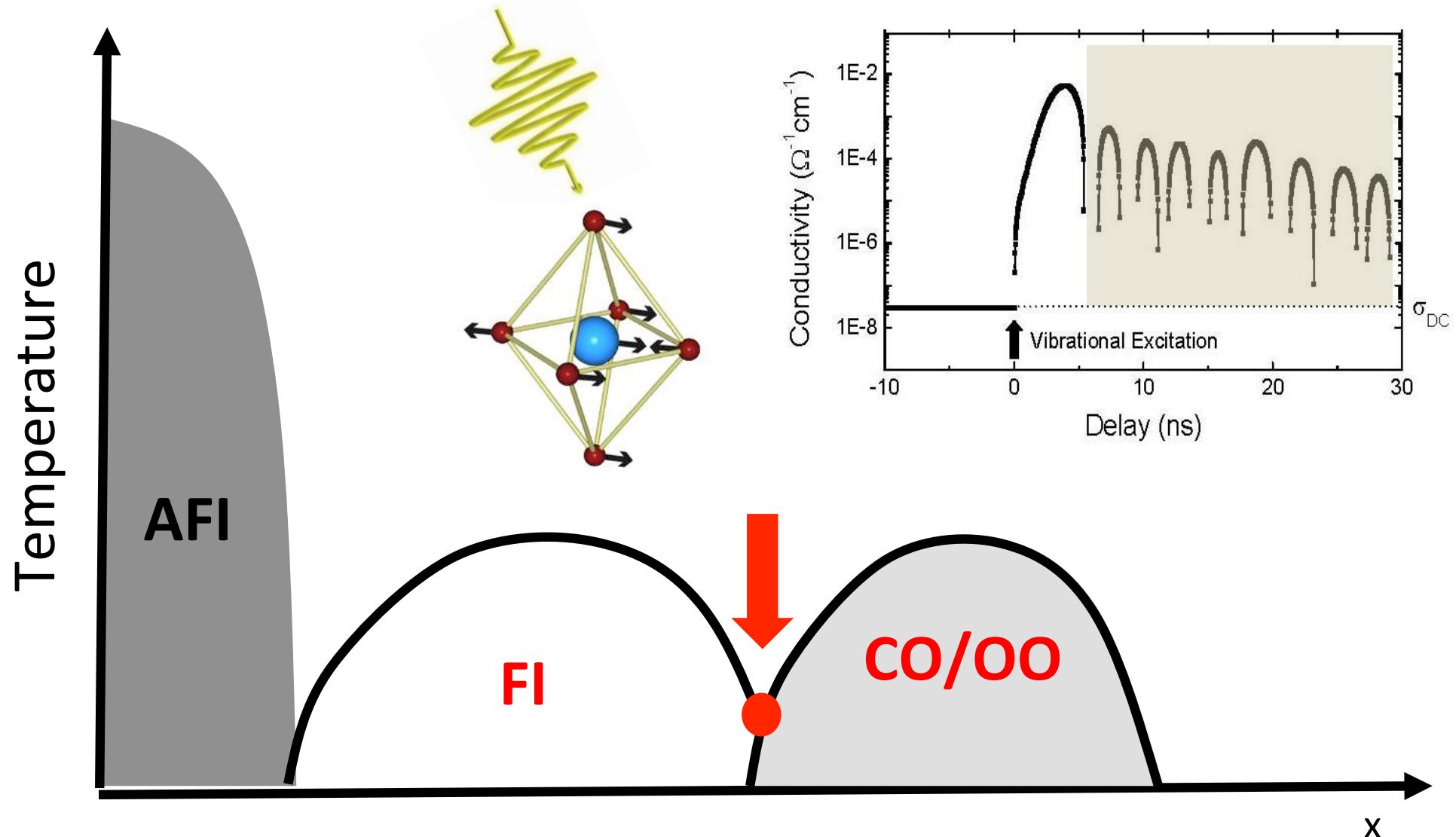
$\text{Pr}_{0.3}\text{Ca}_{0.3}\text{MnO}_3$: a hidden metallic phase



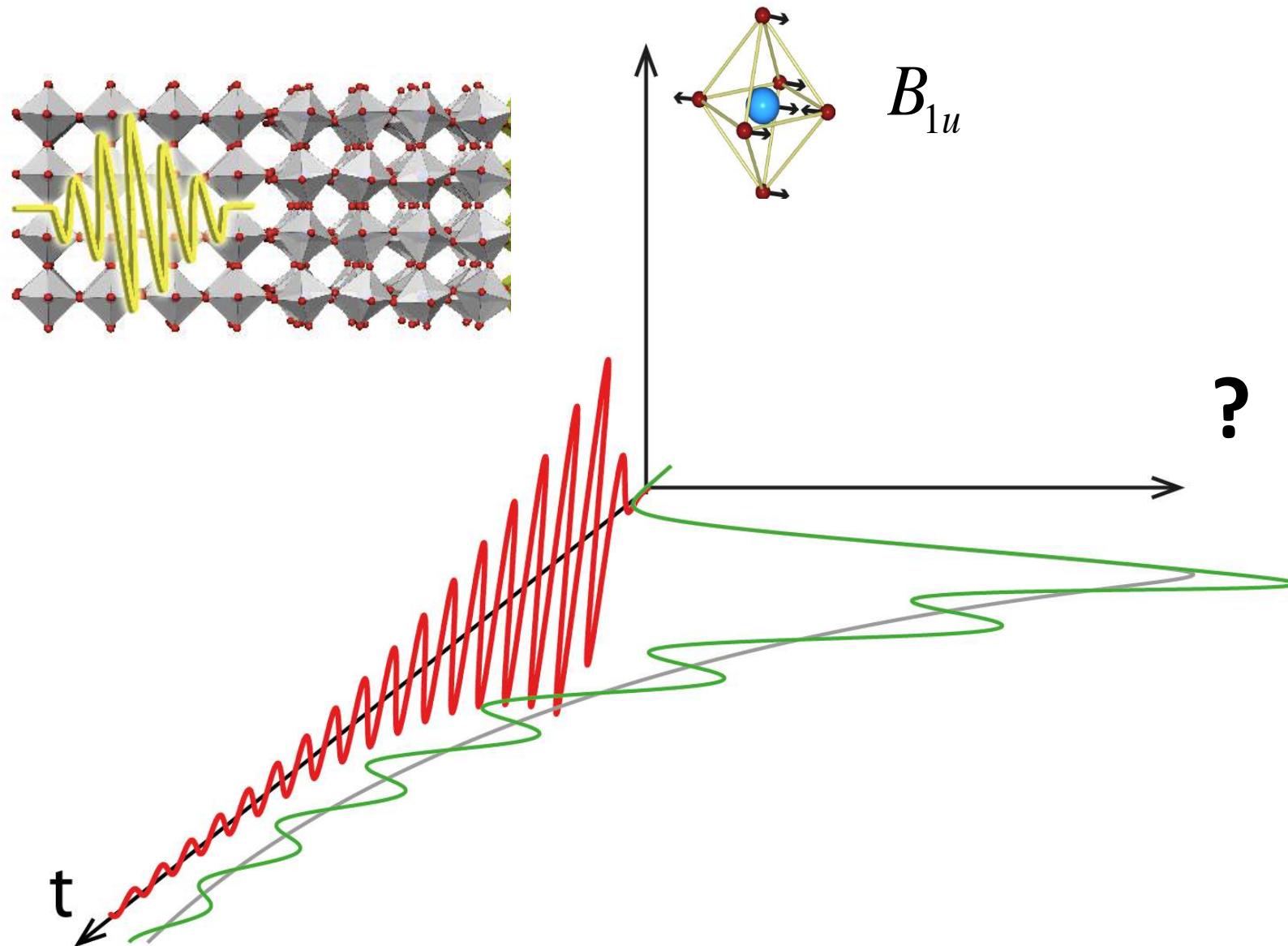
H. Hwang et al., Phys. Rev. Lett. 52, 15046 (1995)



$\text{Pr}_{0.3}\text{Ca}_{0.3}\text{MnO}_3$: excite B_{1u} mode induces hidden metal



What is the dynamical lattice distortion ?



B_{1u} drives A_{1g} mode

$$U_{\text{int}} = A Q_{ir}^2 Q_2$$

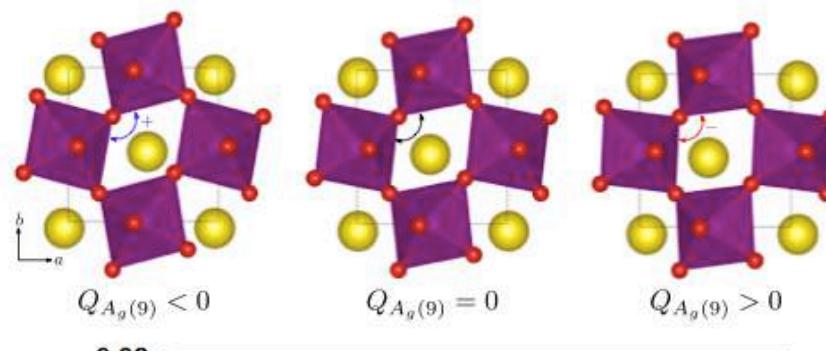
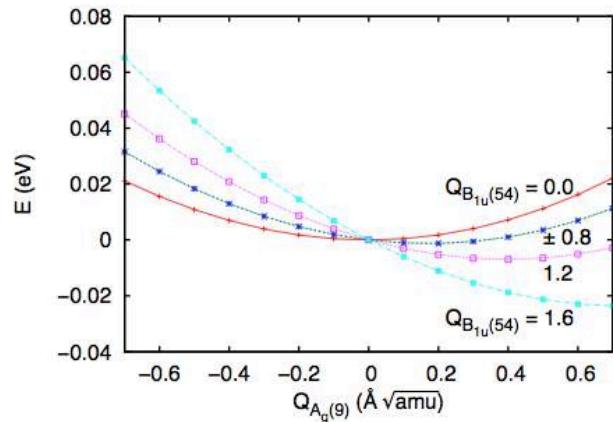


$$B_{1u}^2 A_{1g}$$

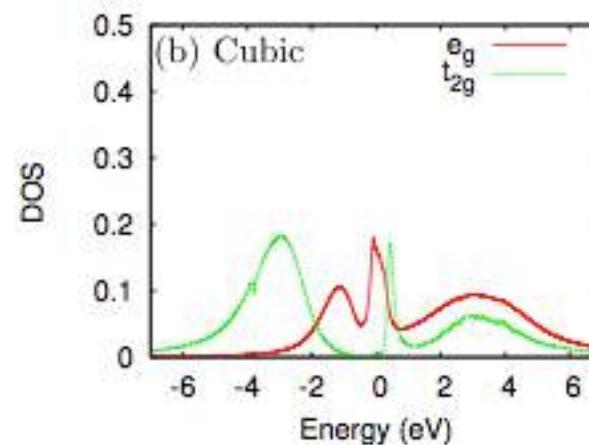
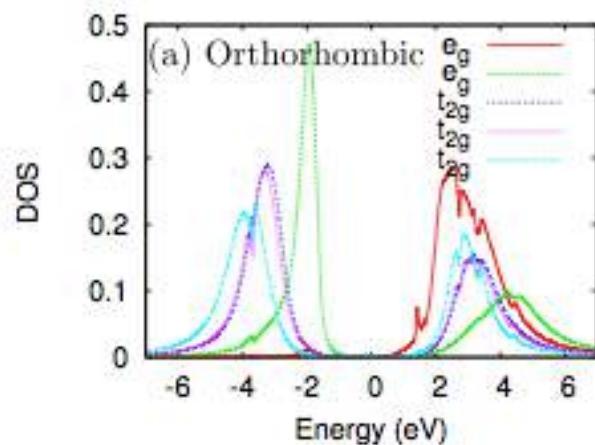


B_{1u} stretch drives A_{1g} rotations

Frozen Phonon

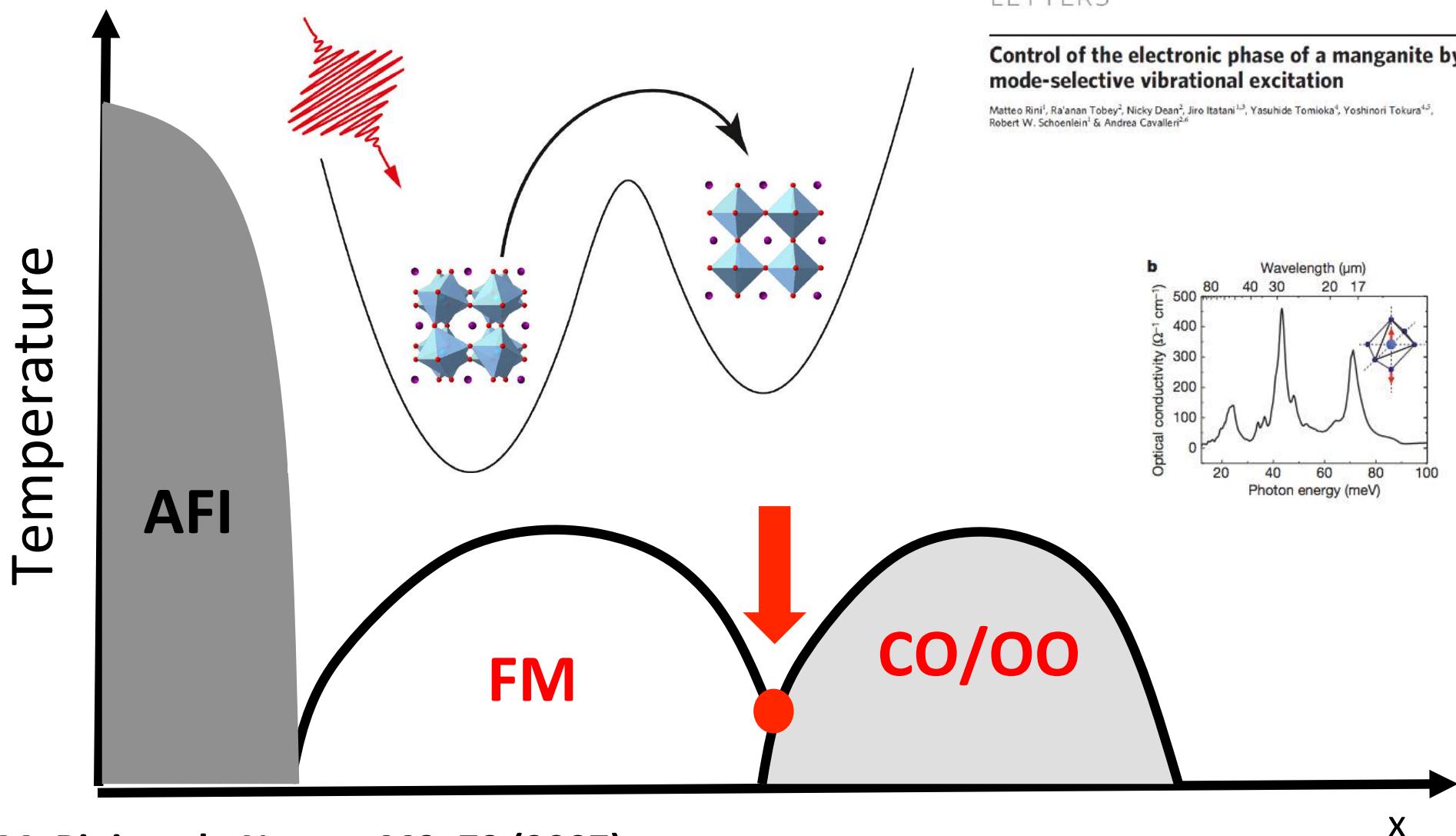


Electronic Structure in the distorted state -> metallic



A. Subedi, A. Cavalleri, A. Georges *Phys. Rev B* 89, 330301 (2014)

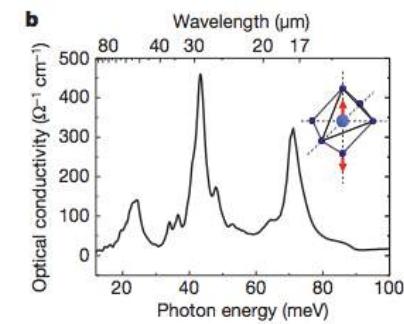
$\text{Pr}_{0.3}\text{Ca}_{0.3}\text{MnO}_3$: control of bond tilt...after all



LETTERS

Control of the electronic phase of a manganite by mode-selective vibrational excitation

Matteo Rini¹, Raanan Tobej², Nicky Dean², Jiro Itatani^{1,3}, Yasuhide Tomioka⁴, Yoshinori Tokura^{4,5}, Robert W. Schoenlein¹ & Andrea Cavalleri^{2*}

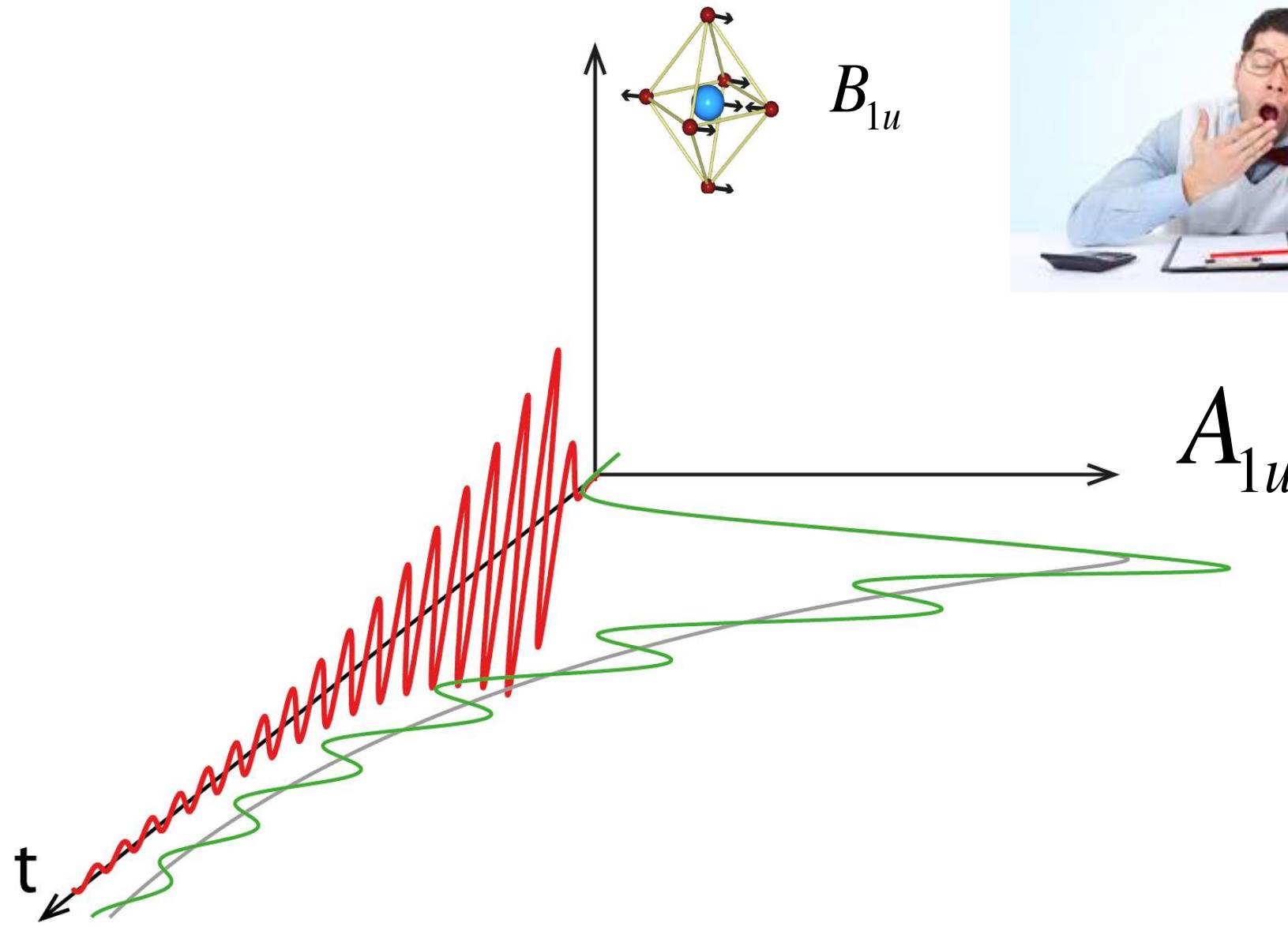


M. Rini et al., Nature 449, 72 (2007)

A. Subedi, A. Cavalleri, A. Georges Phys. Rev B 89, 330301 (2014)



Centrosymmetric – only A modes



What else can I do ?

Manipulate inversion symmetry

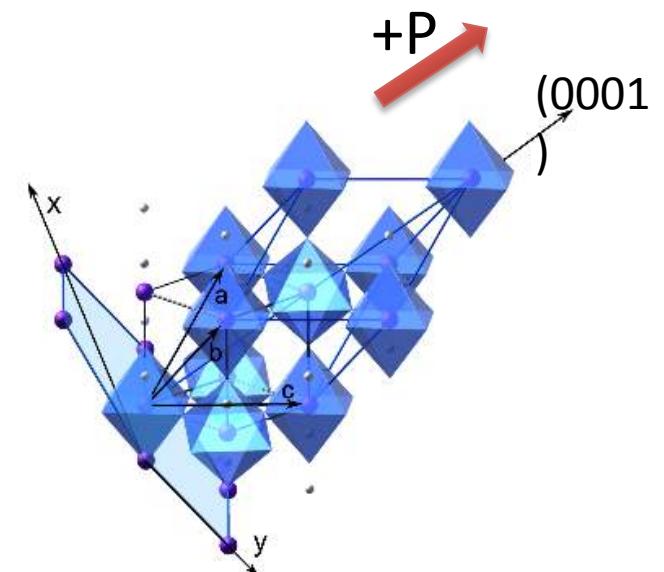
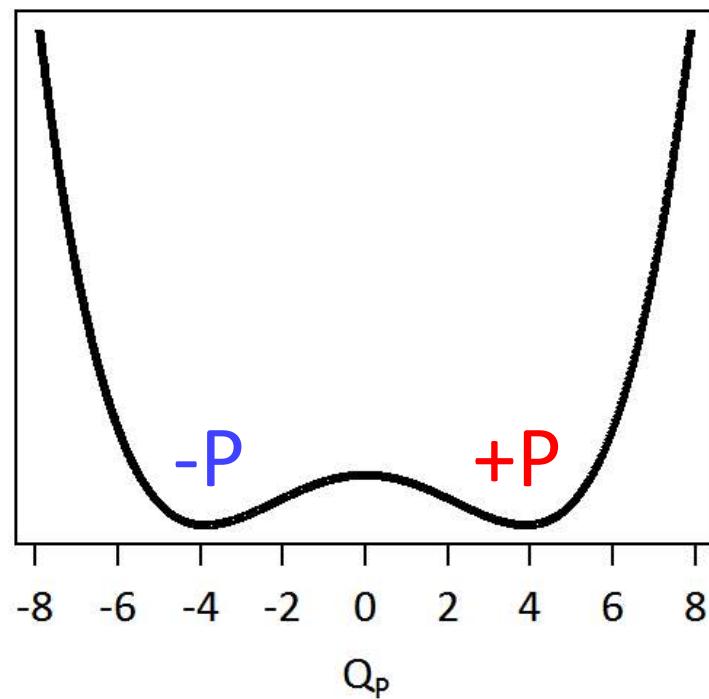
Manipulate time reversal symmetry



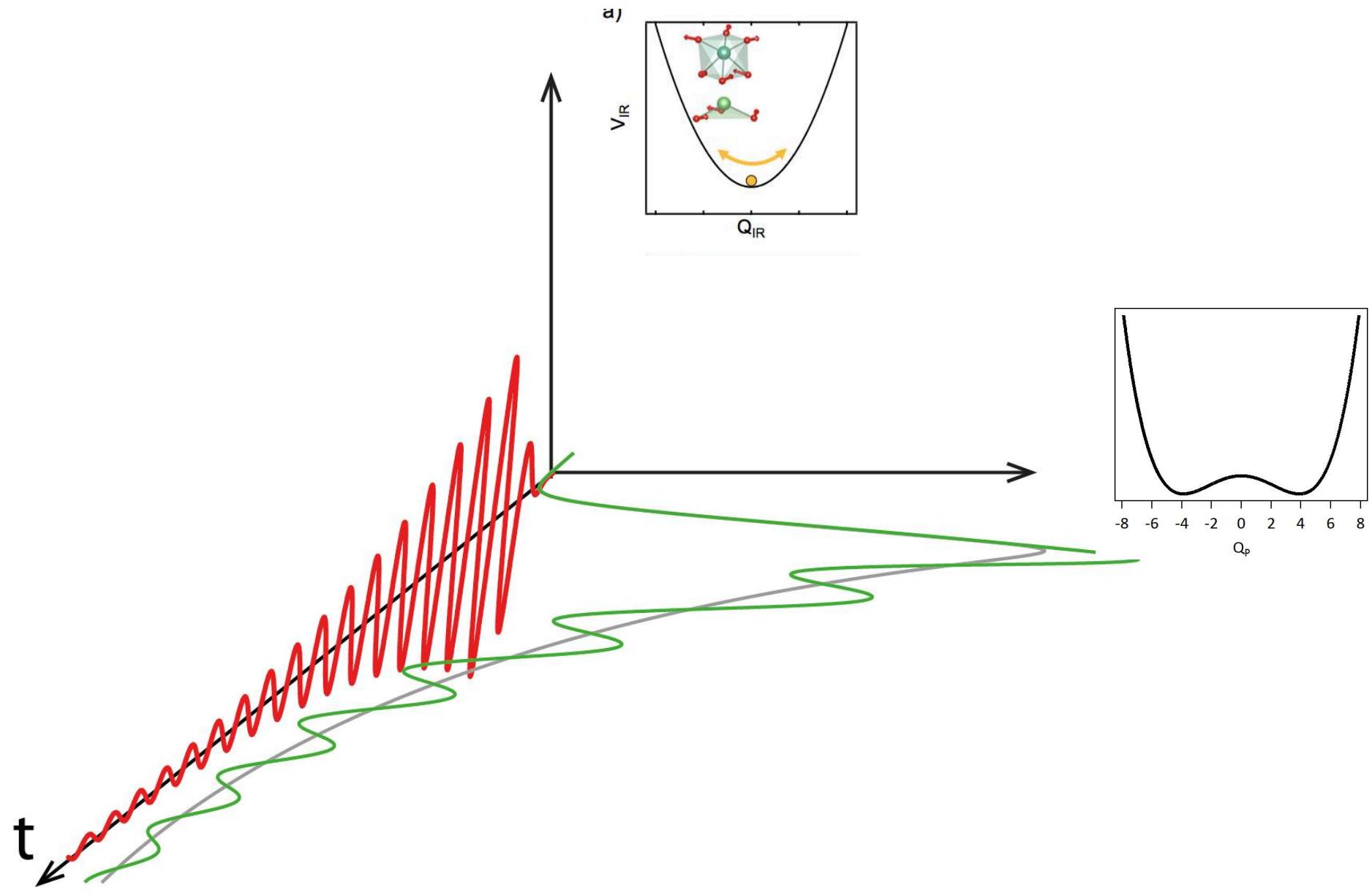
Ferroelectric: no centre of inversion

$$V_P = \frac{1}{2}\omega_P^2 Q_P^2 + aQ_P^3 + bQ_P^4$$

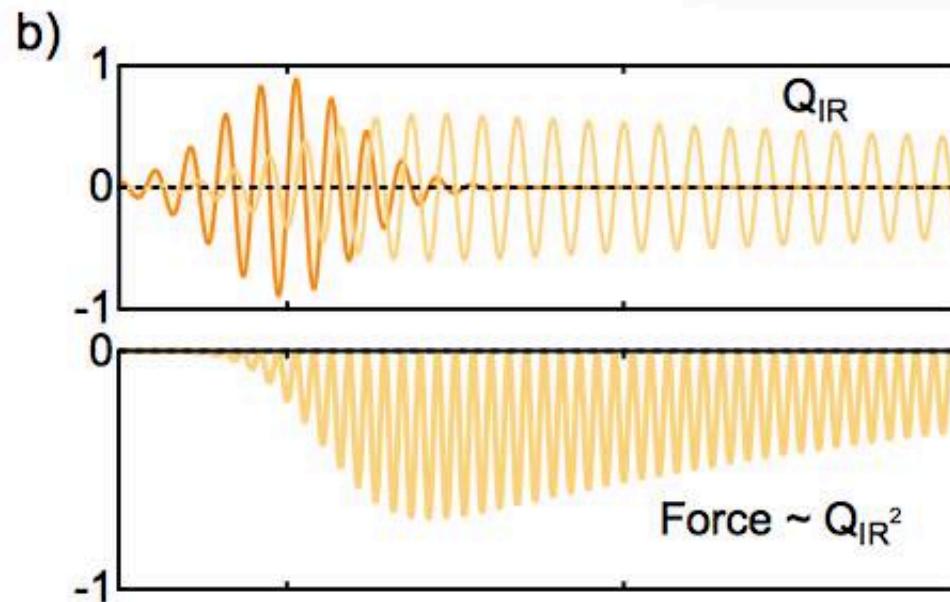
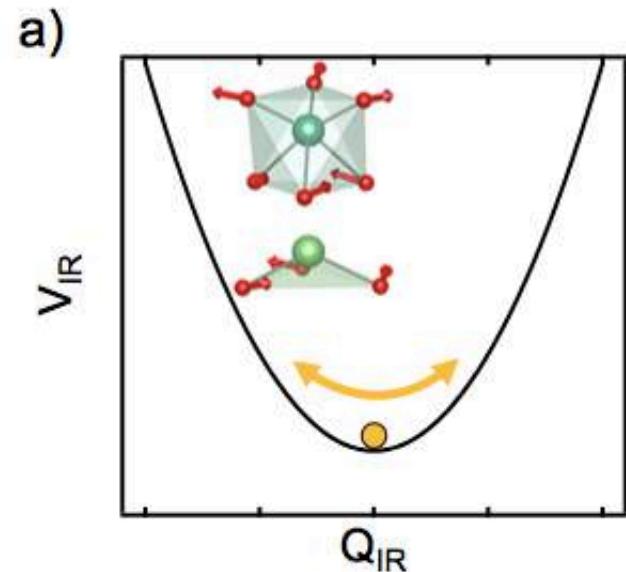
LiNbO₃



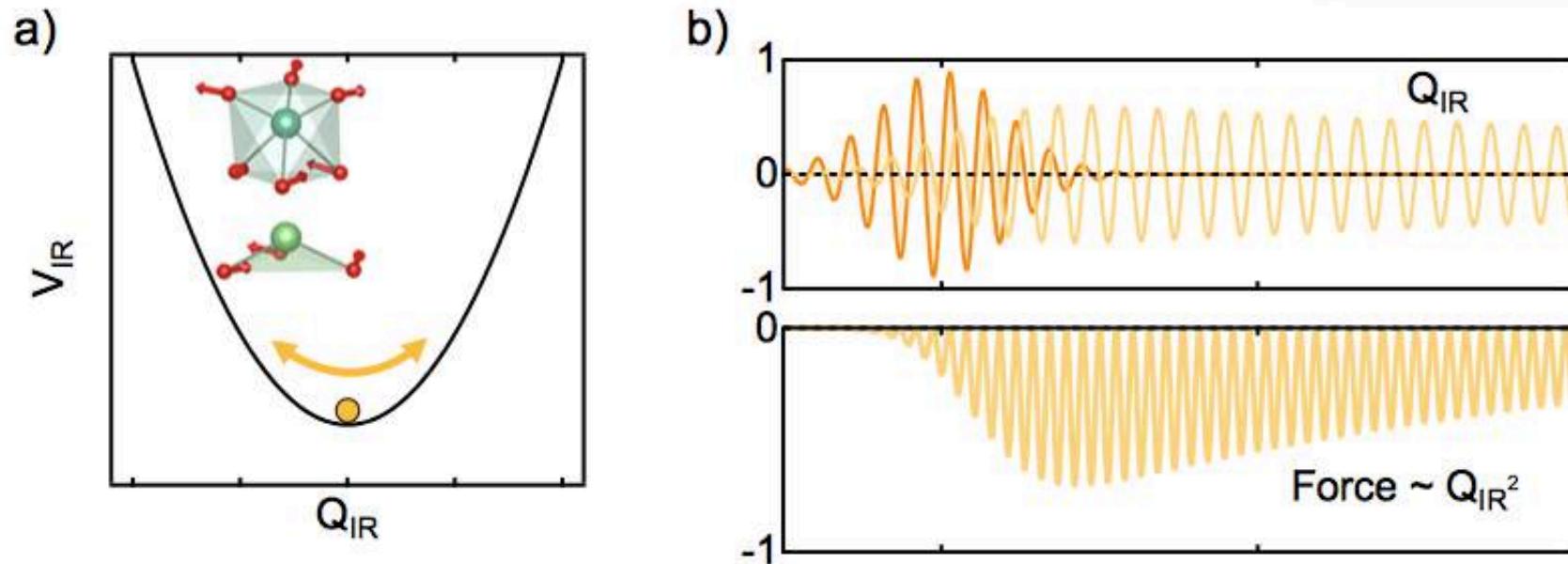
Not centrosymmetric – coupling odd modes



Pump an auxilliary mode



Force on ferroelectric mode

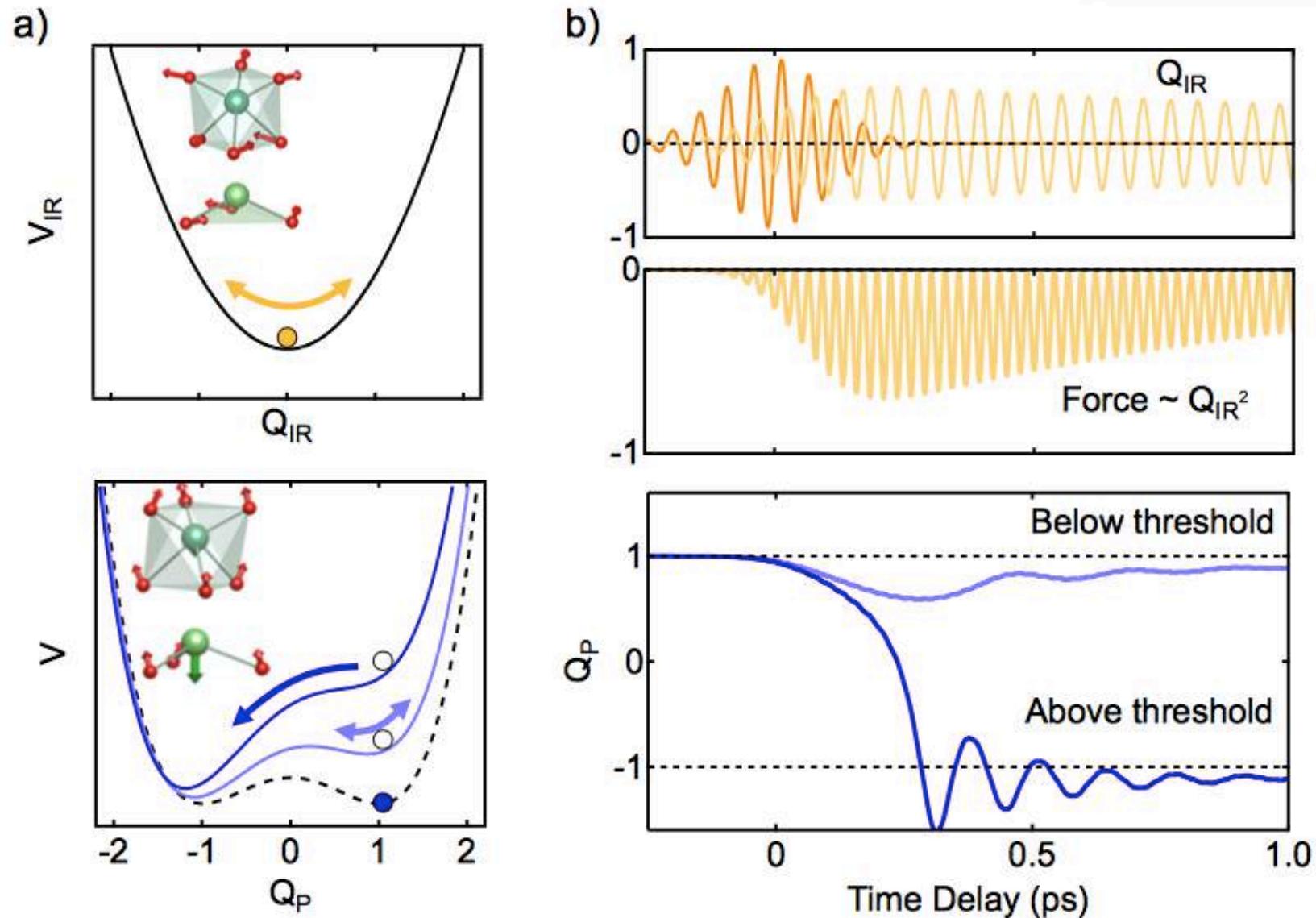


$$\ddot{Q}_{IR} + \gamma_{IR} \dot{Q}_{IR} + \omega_{IR}^2 Q_{IR} = 2aQ_p Q_{IR} + f(t),$$

$$\ddot{Q}_p + \gamma_p \dot{Q}_p - \omega_p^2 Q_p + c_p Q_p^3 = aQ_{IR}^2,$$

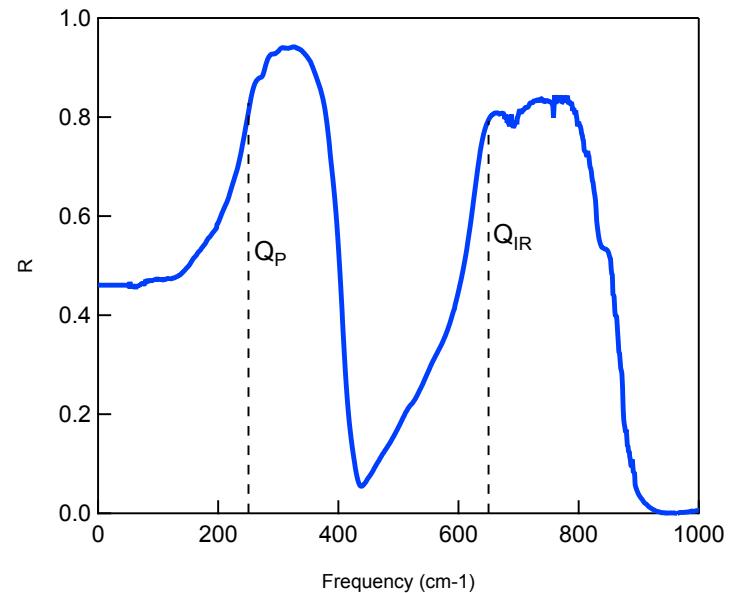
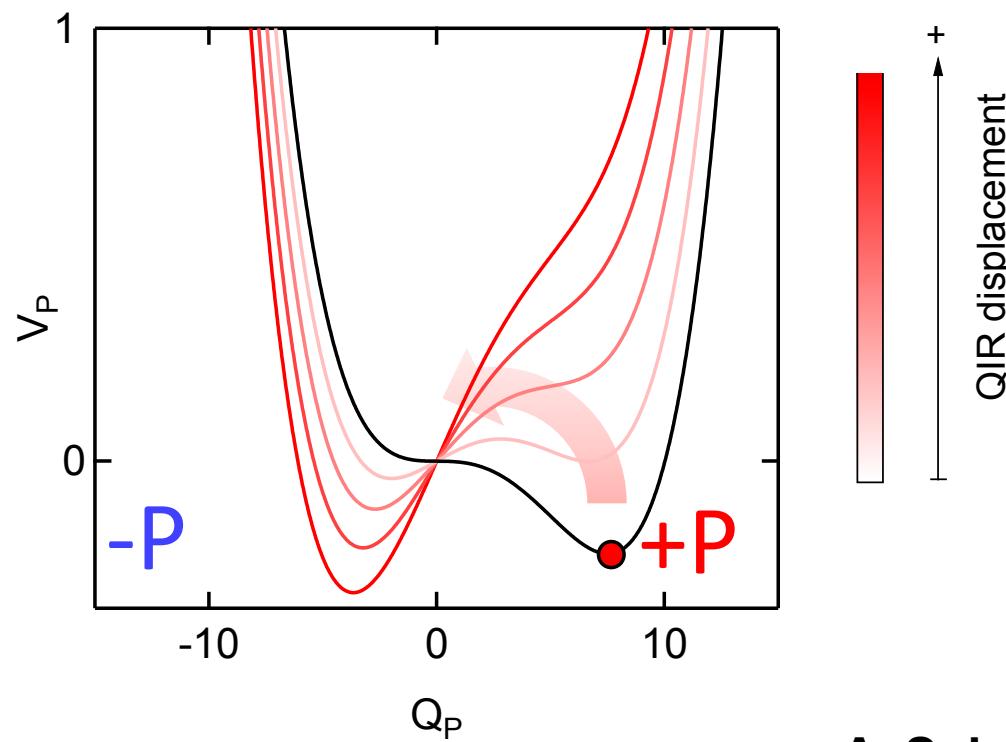


Force on ferroelectric mode



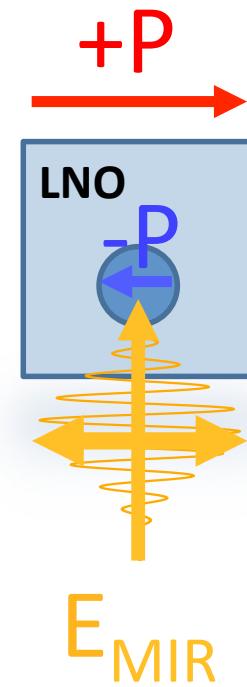
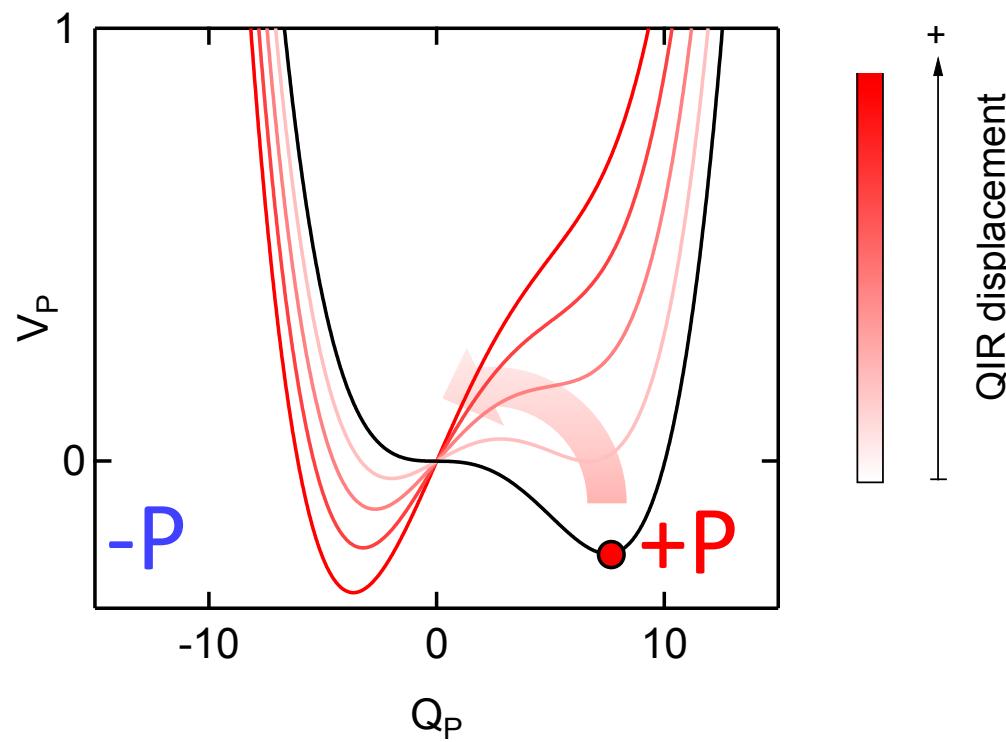
Coupling to Ferroelectric polarization

$$V_P = \frac{1}{2}\omega_P^2 Q_P^2 + aQ_P^3 + bQ_P^4 + AQ_{ir}^2 Q_P$$

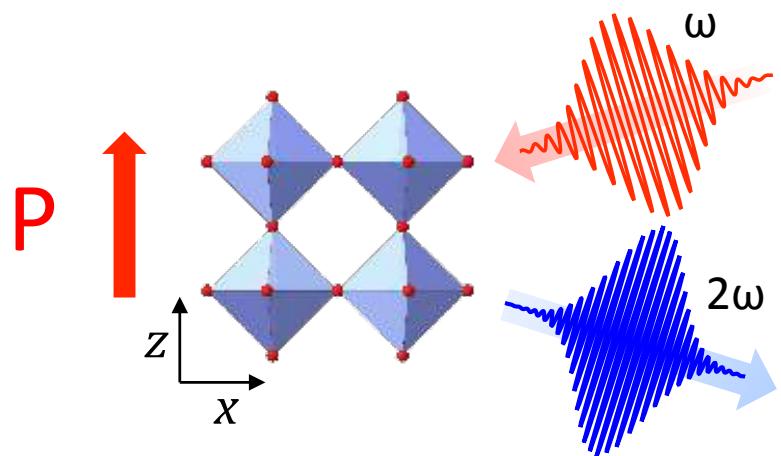


Coupling to Ferroelectric polarization

$$V_P = \frac{1}{2}\omega_P^2 Q_P^2 + aQ_P^3 + bQ_P^4 - A Q_{ir}^2 Q_P$$



Probing Ferroelectric polarization



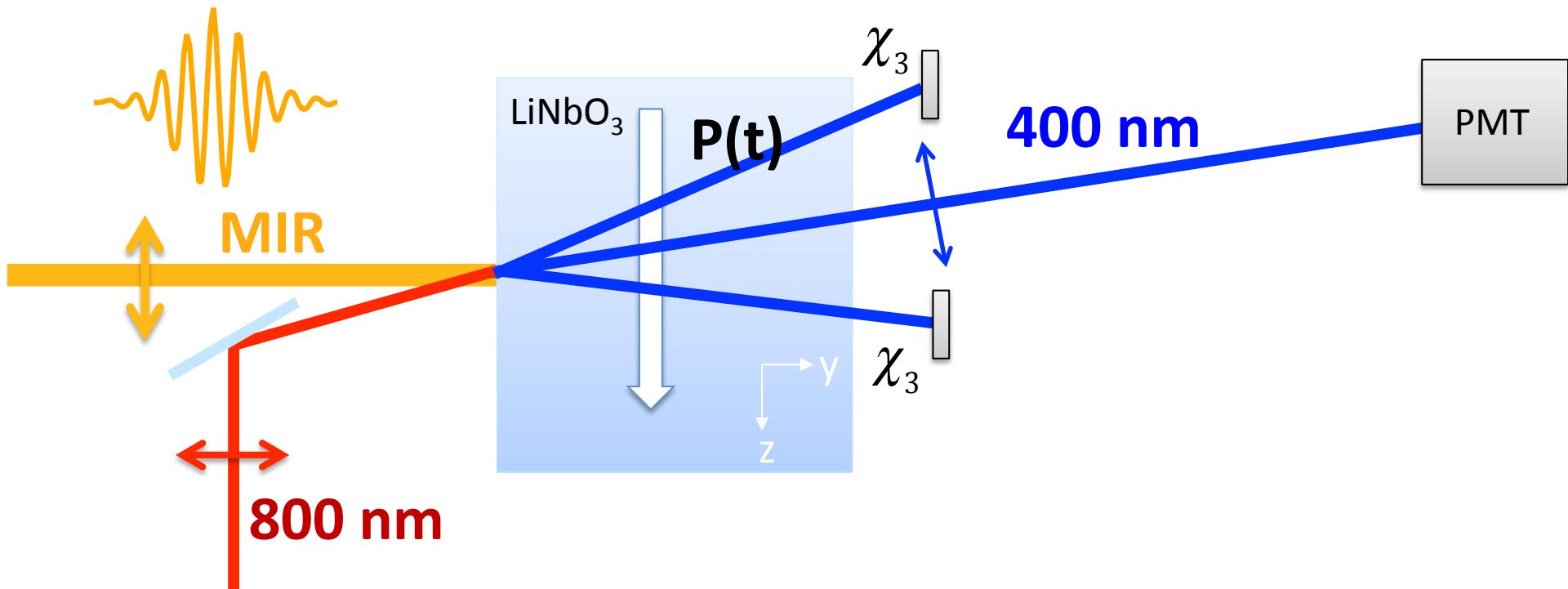
SHG only possible in non-centrosymmetric materials

$$P^{(2)} = \epsilon_0 \cdot \chi^{(2)} \cdot E_{(800\text{nm})}^2$$

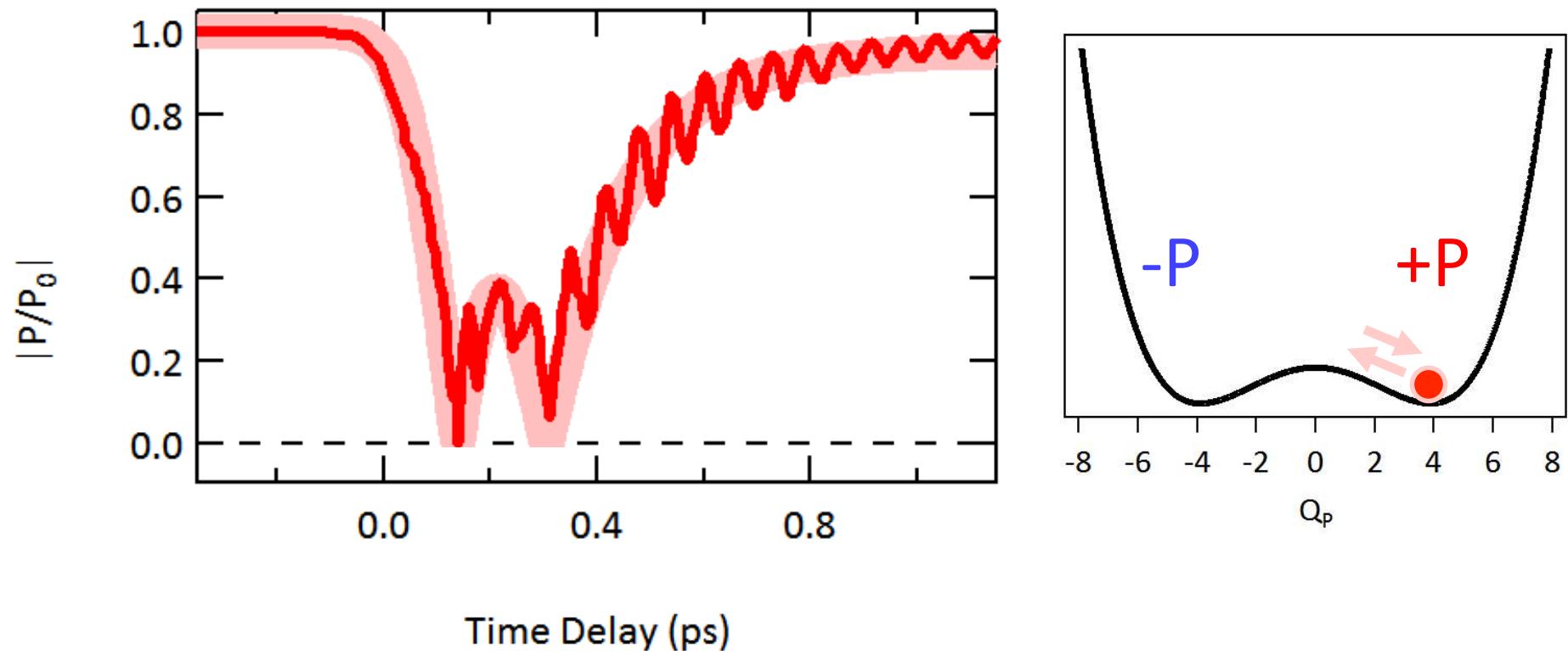


Probing Ferroelectric polarization

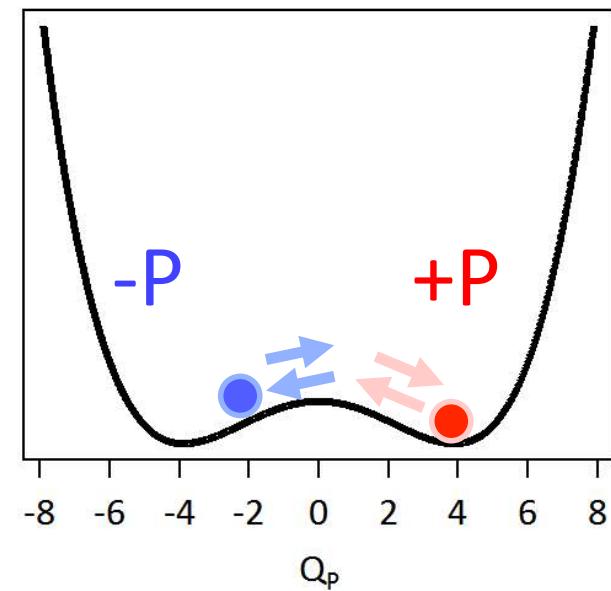
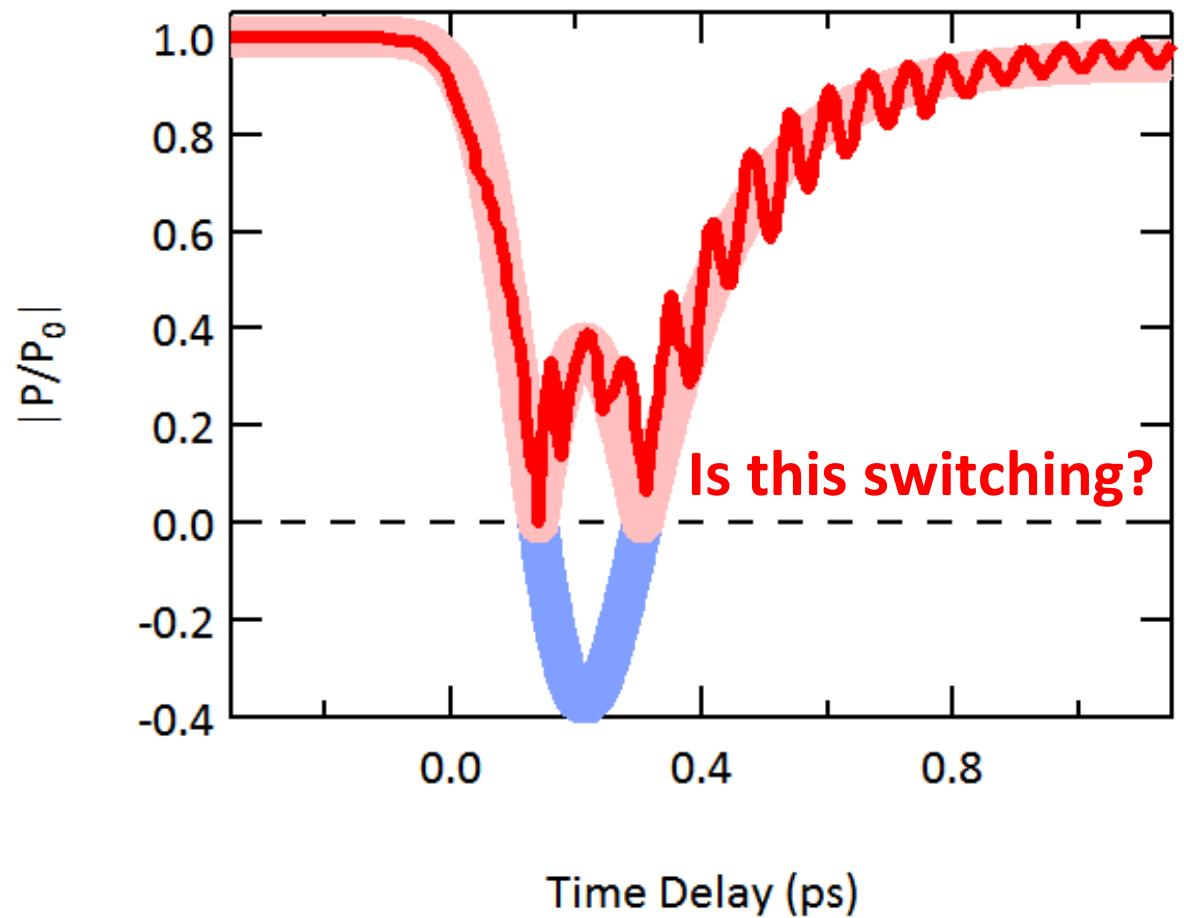
Measure time-resolved second harmonic intensity



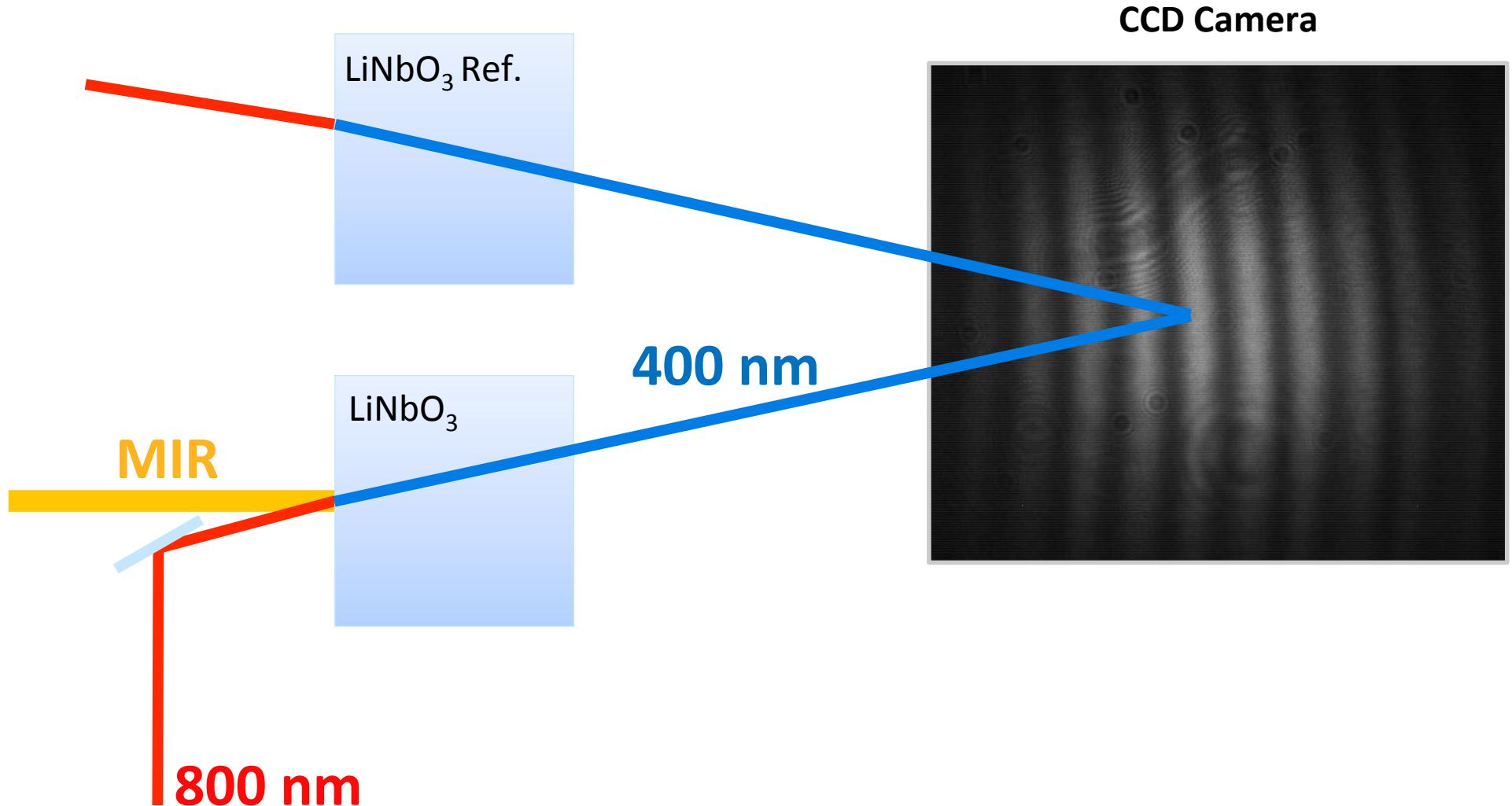
drops to zero...bounces....goes back to zero



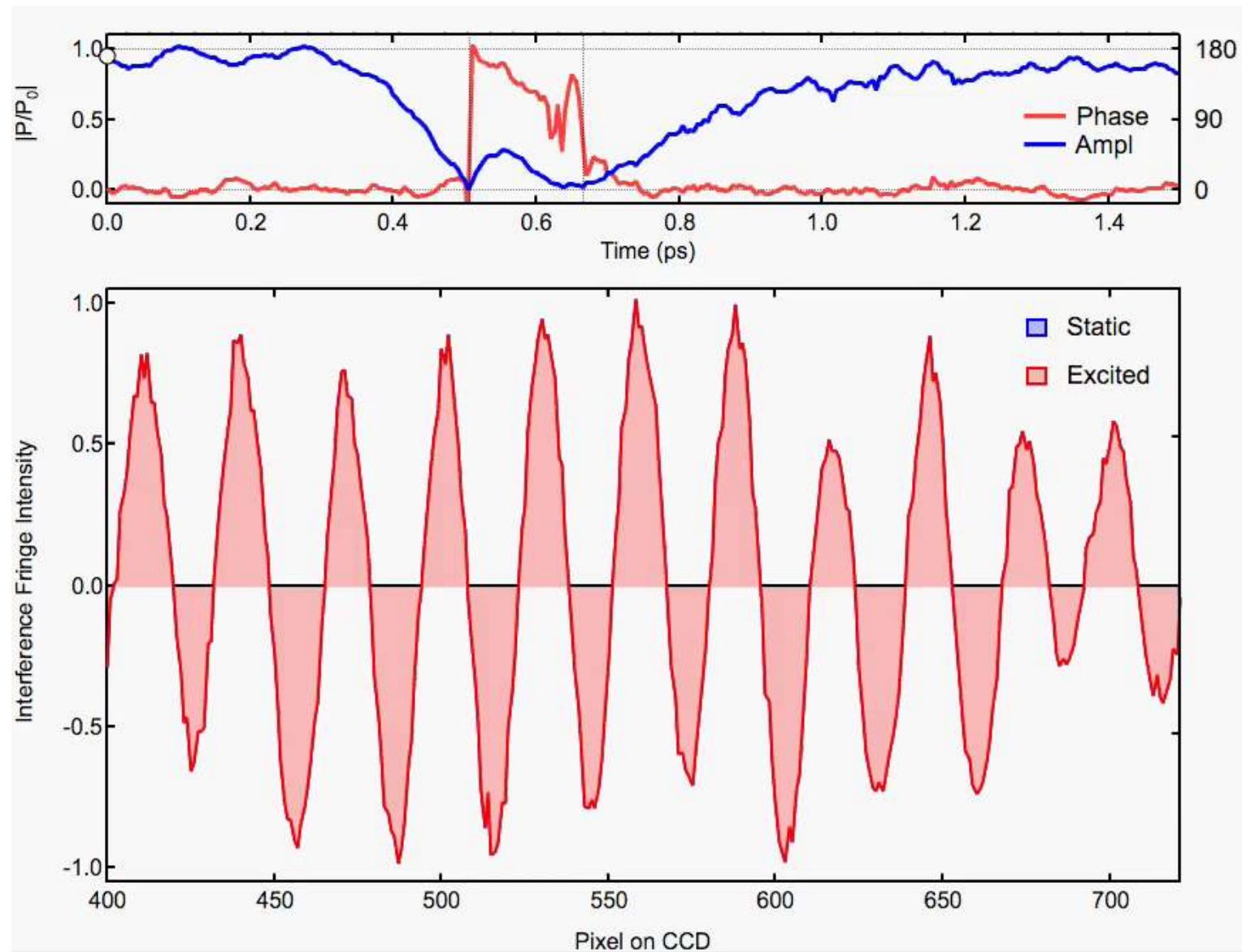
Is this switching?



Phasing Ferroelectric Polarization

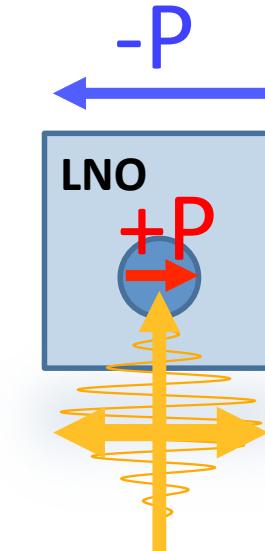
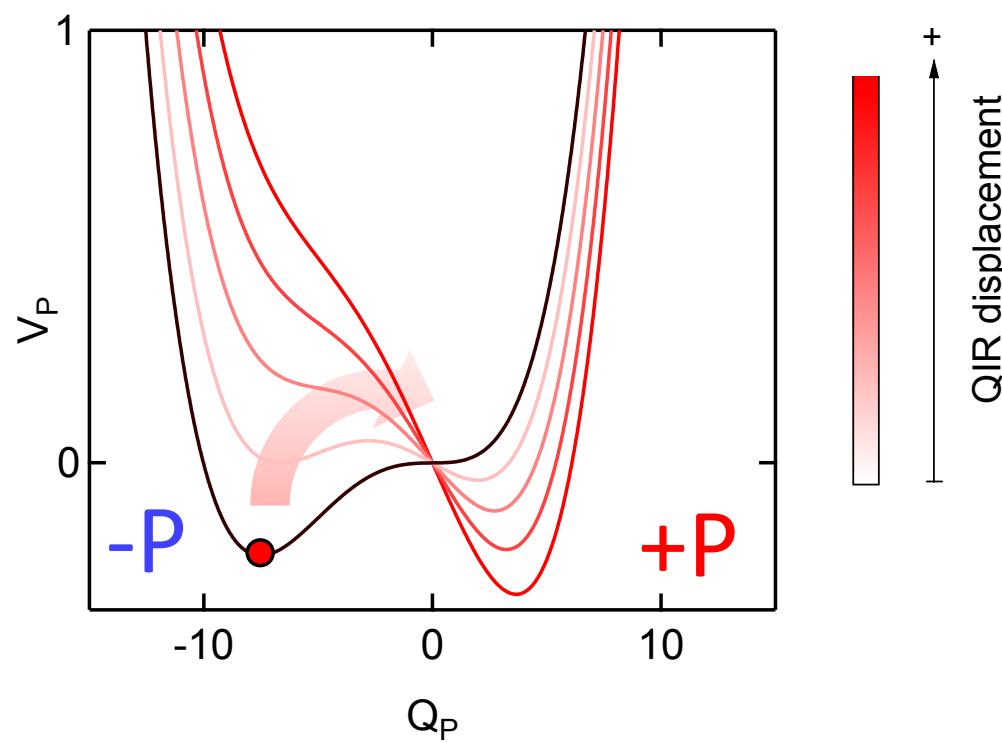


Phasing Ferroelectric Polarization



Ferroelectric Switching in LNO

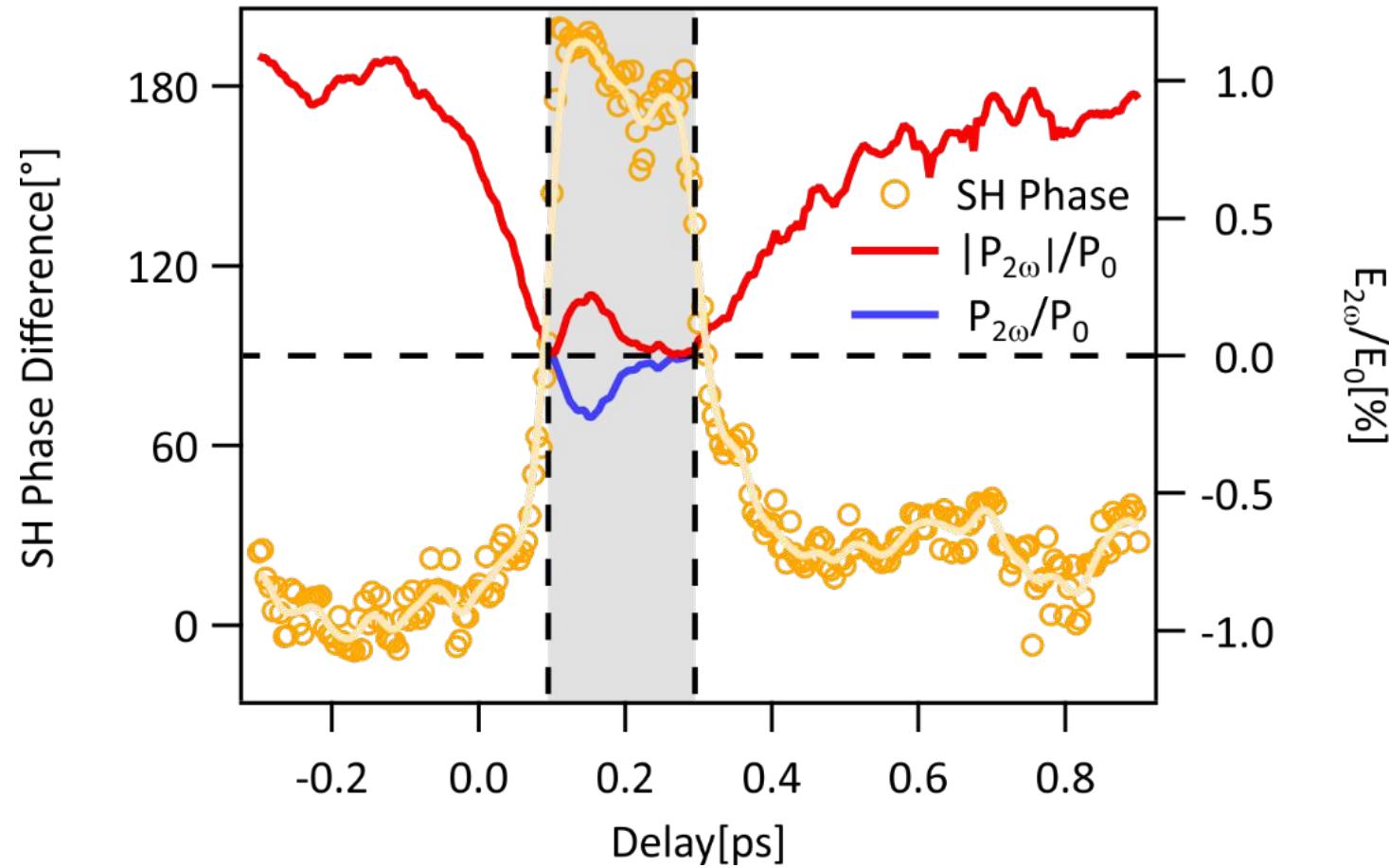
$$V_P = \frac{1}{2}\omega_P^2 Q_P^2 + aQ_P^3 + bQ_P^4 - A Q_{ir}^2 Q_P$$



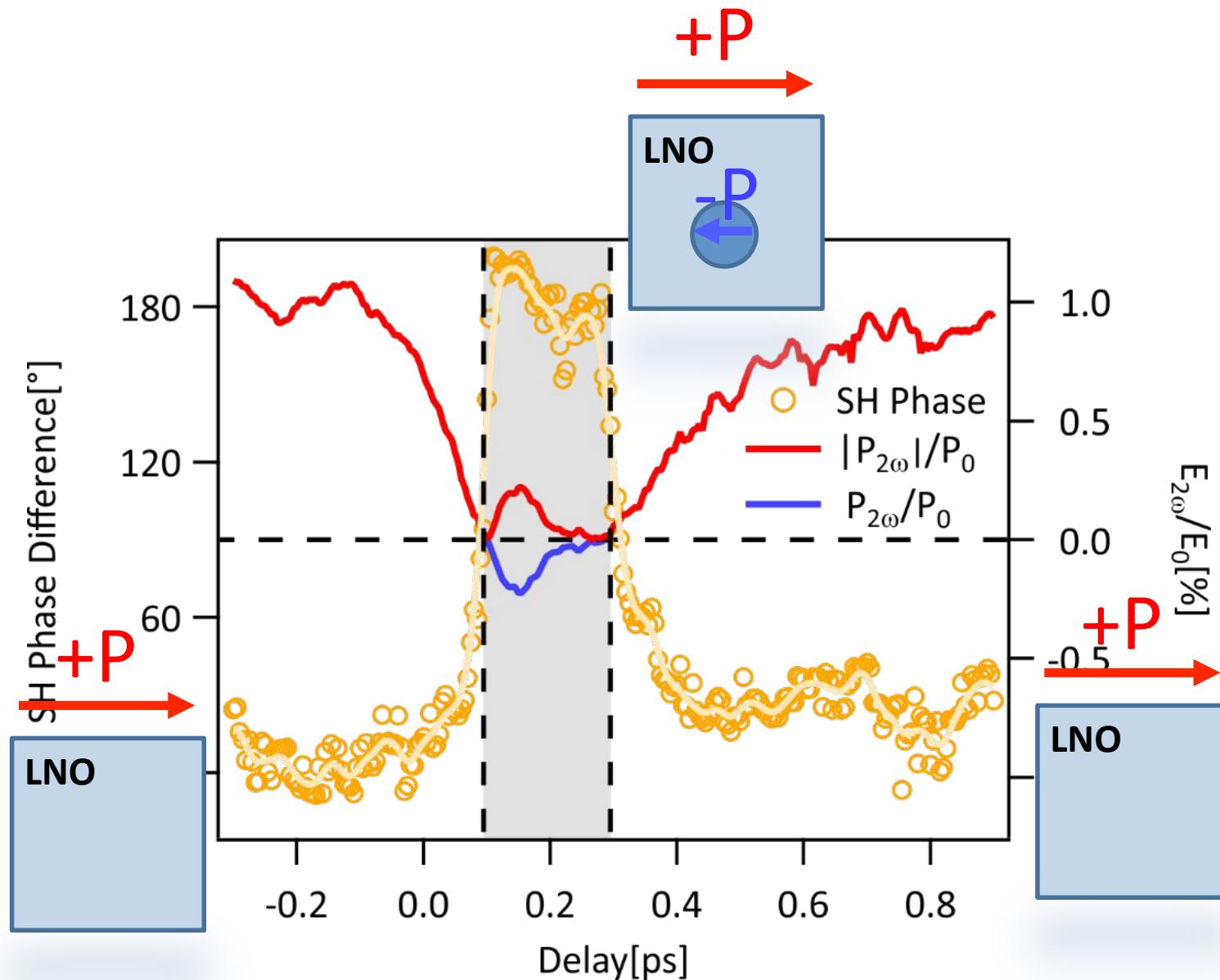
“Direction” given by initial state



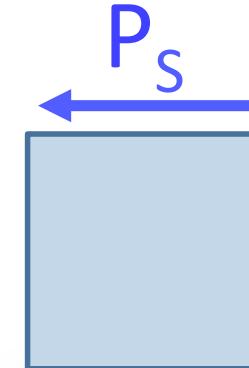
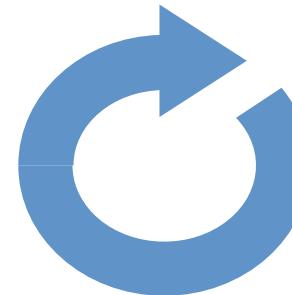
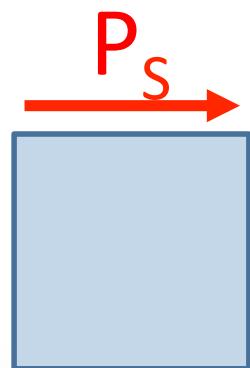
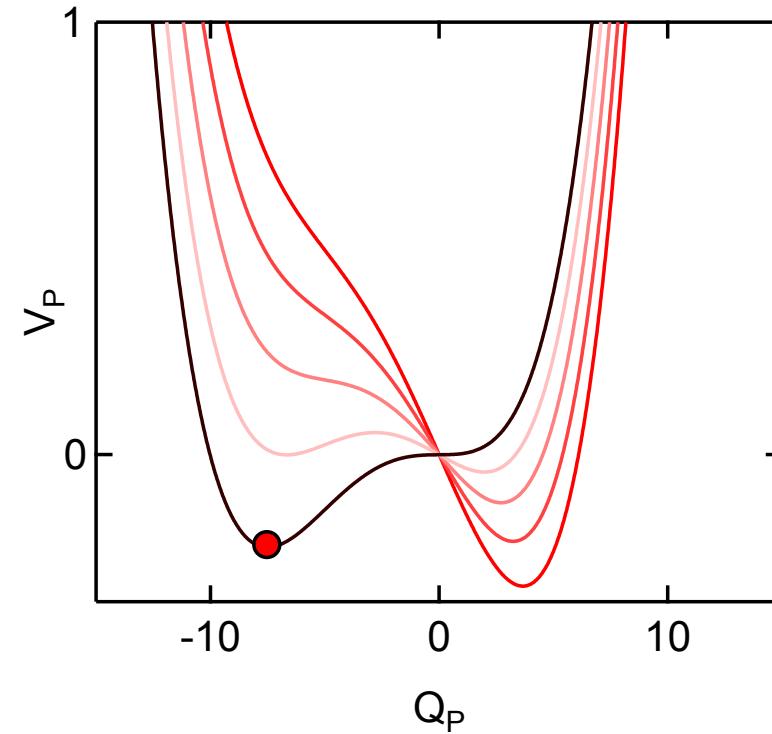
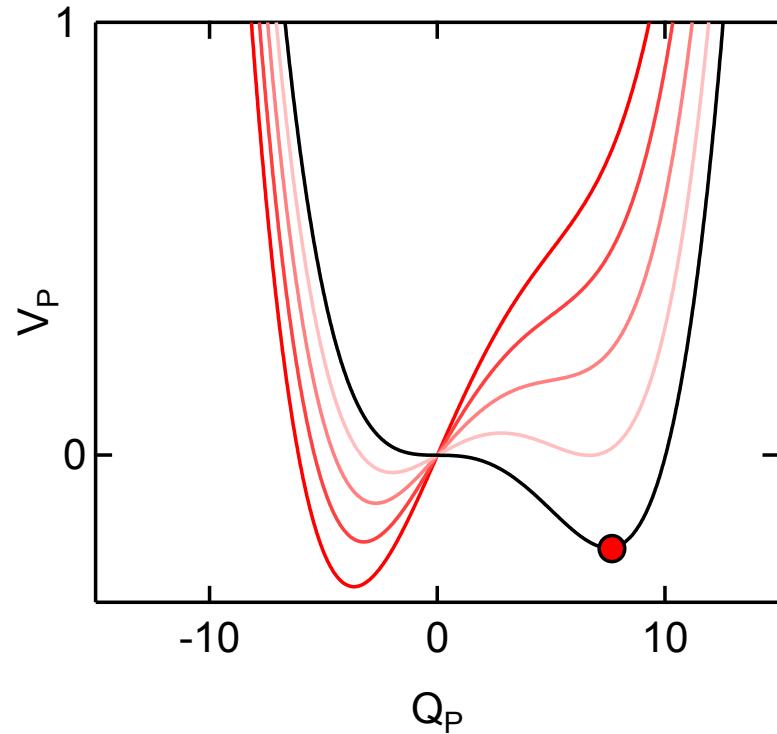
Phasing Polarization Switching



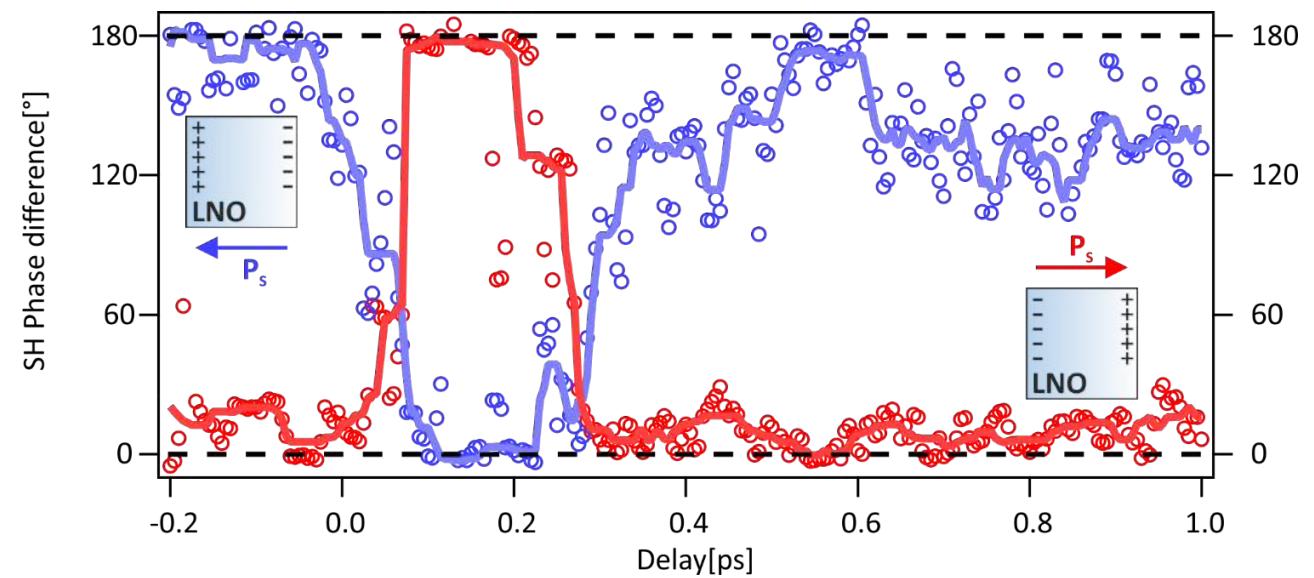
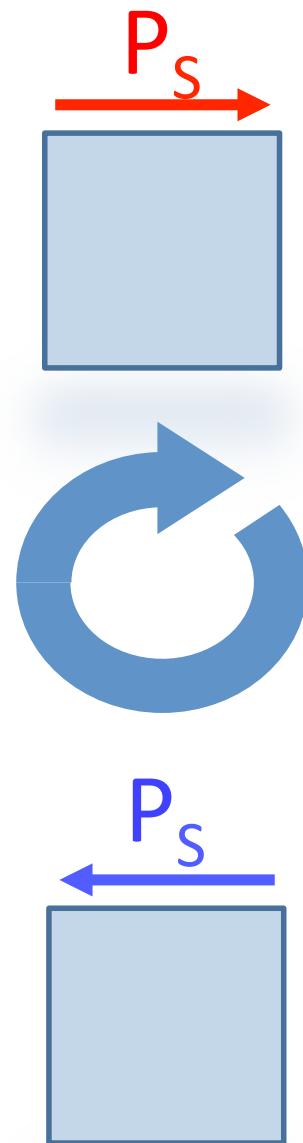
Phasing Ferroelectric Polarization



Starting from different direction



Bi-directional switching



Up to now I discussed only lowest order

In analogy with nonlinear optics

$Q_{IR}^2 \cdot Q_2$ (lattice control)



$Q_{IR}^2 \cdot Q_2^2$ (phonon squeezing)

Q_{IR}^4 (parametric phonon amplification)

$Q_{IR1} \cdot Q_{IR2} e^{i\Theta} \cdot s$ (controlling time reversal invariance)

$Q_{IR}^2 \cdot U$ (controlling correlations)



What else?

In analogy with nonlinear optics

$Q_{IR}^2 \cdot Q_2$ (lattice control)

$Q_{IR}^2 \cdot Q_2^2$ (phonon squeezing)

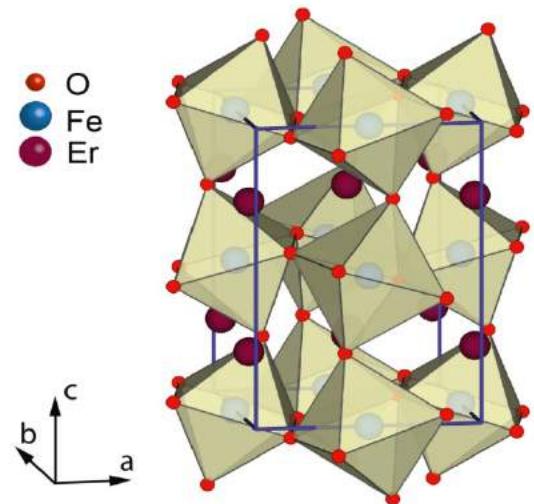
Q_{IR}^4 (parametric phonon amplification)

$Q_{IR1} \cdot Q_{IR2} e^{i\Theta} \cdot s$ (controlling time reversal invariance) ←

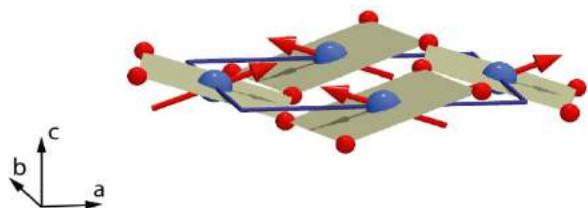
$Q_{IR}^2 \cdot U$ (controlling correlations)



Can we break time reversal symmetry ?



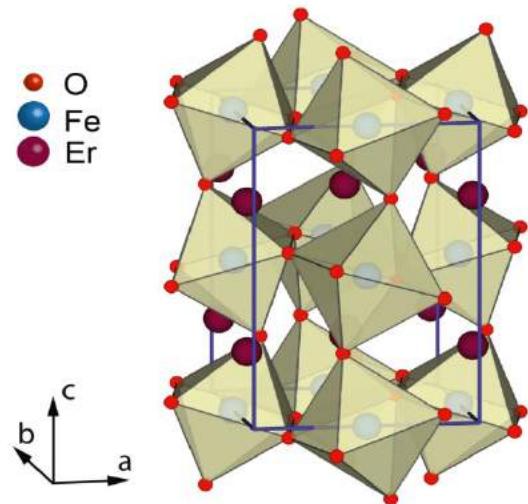
ErFeO_3



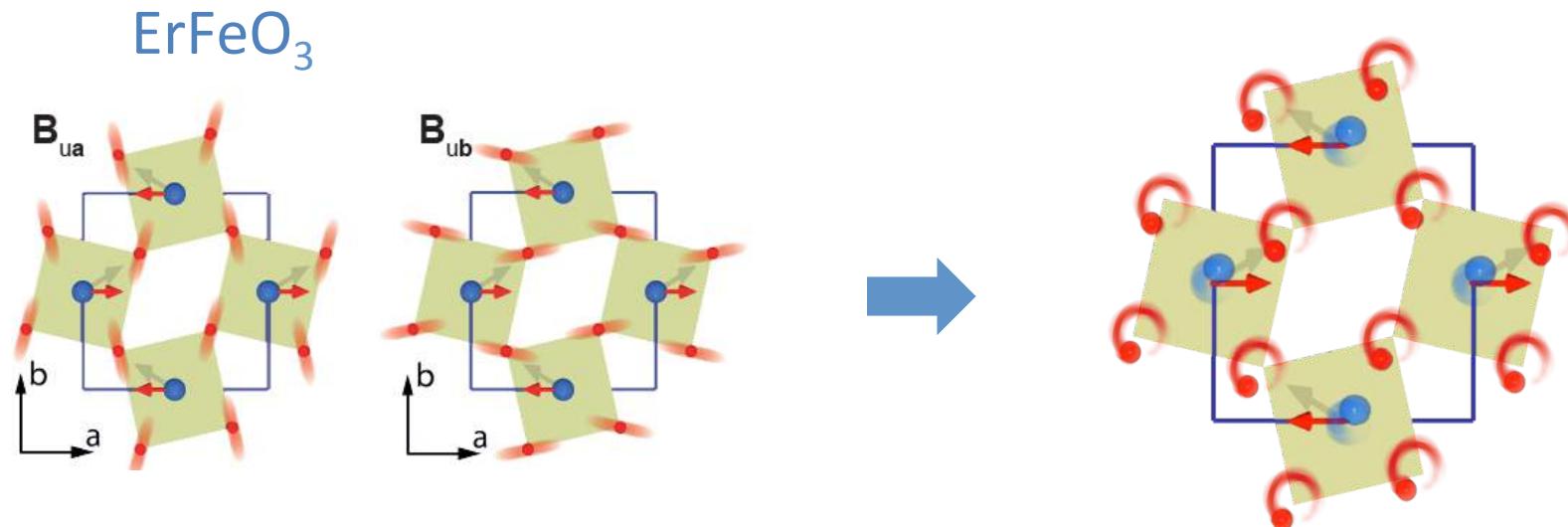
Orthorombically distorted perovskite
Antiferromagnetic insulator



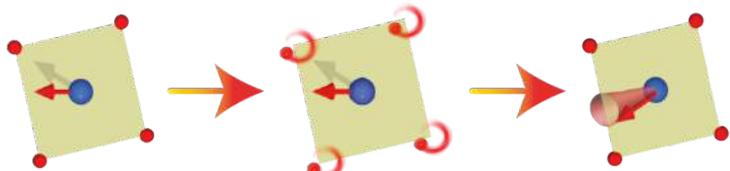
Exciting more than one mode



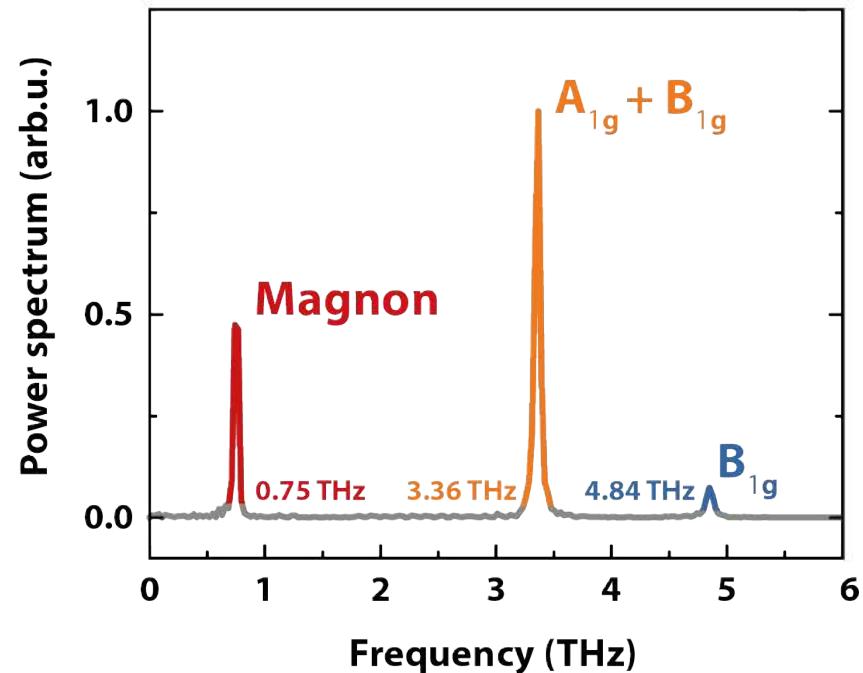
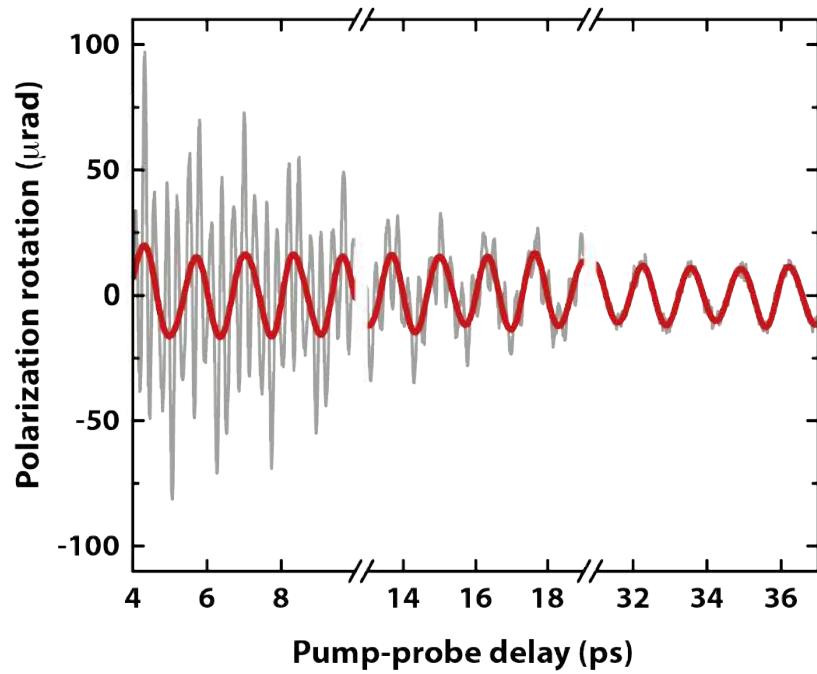
Simultaneous excitation of **two lattice modes** with controlled **relative phase**.



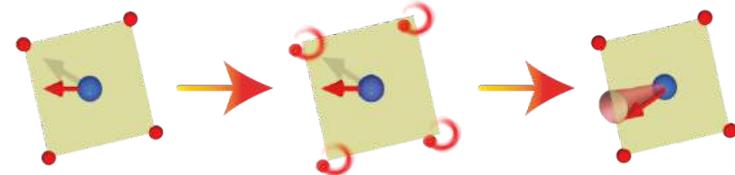
Break time reversal symmetry



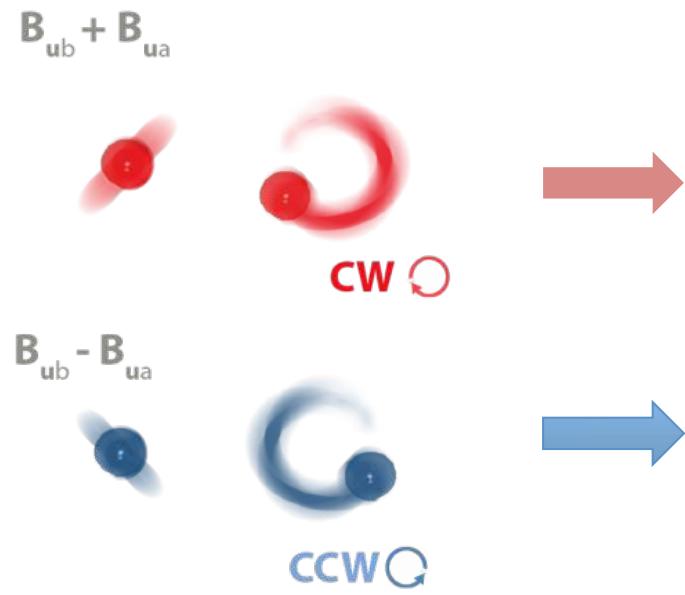
$$H_{eff} = i\alpha Q_{IR1}Q_{IR2}^* M$$



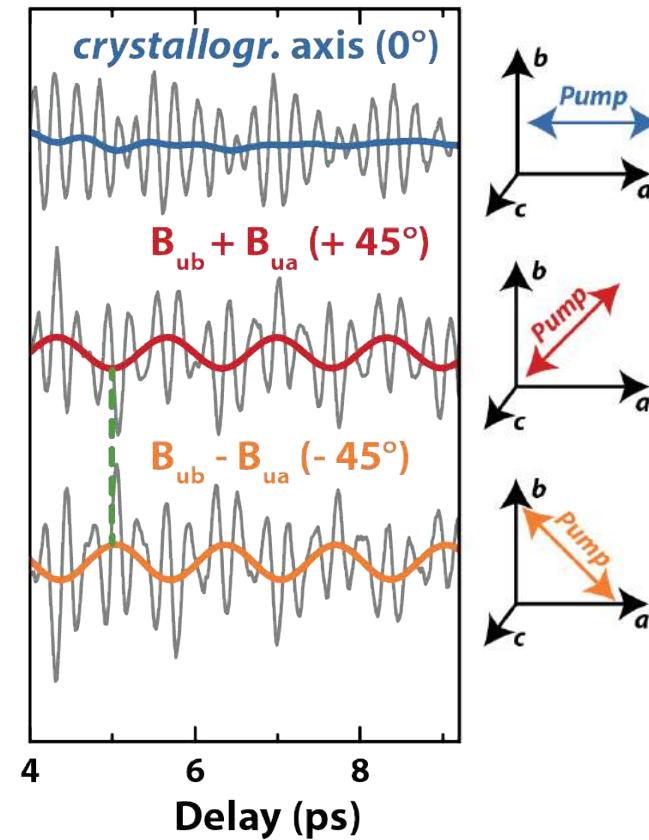
Coherent control of magnetic mode



$$H_{eff} = i\alpha Q_{IR1}Q_{IR2}^* M$$

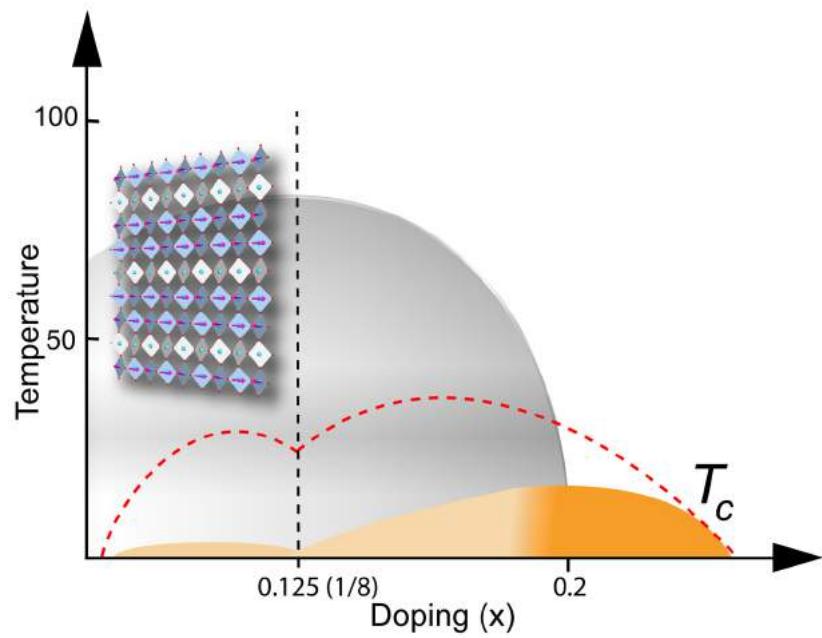


Polarization dependence

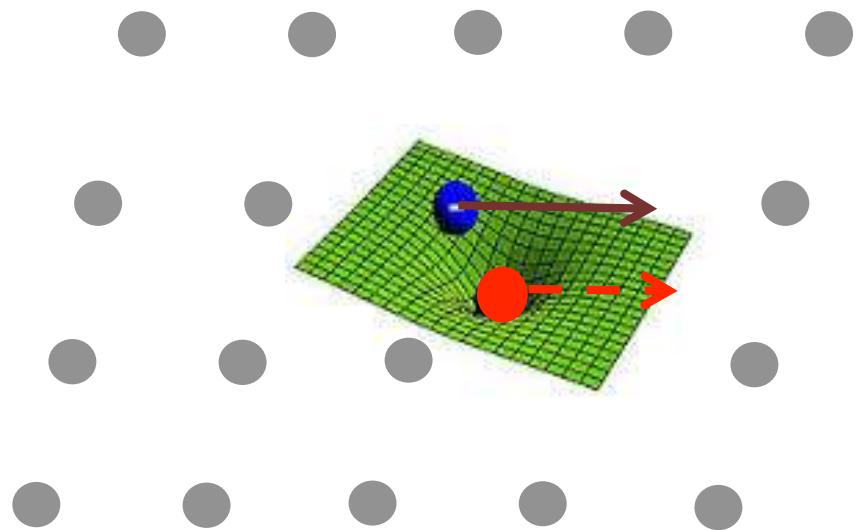


Controlling superconductivity

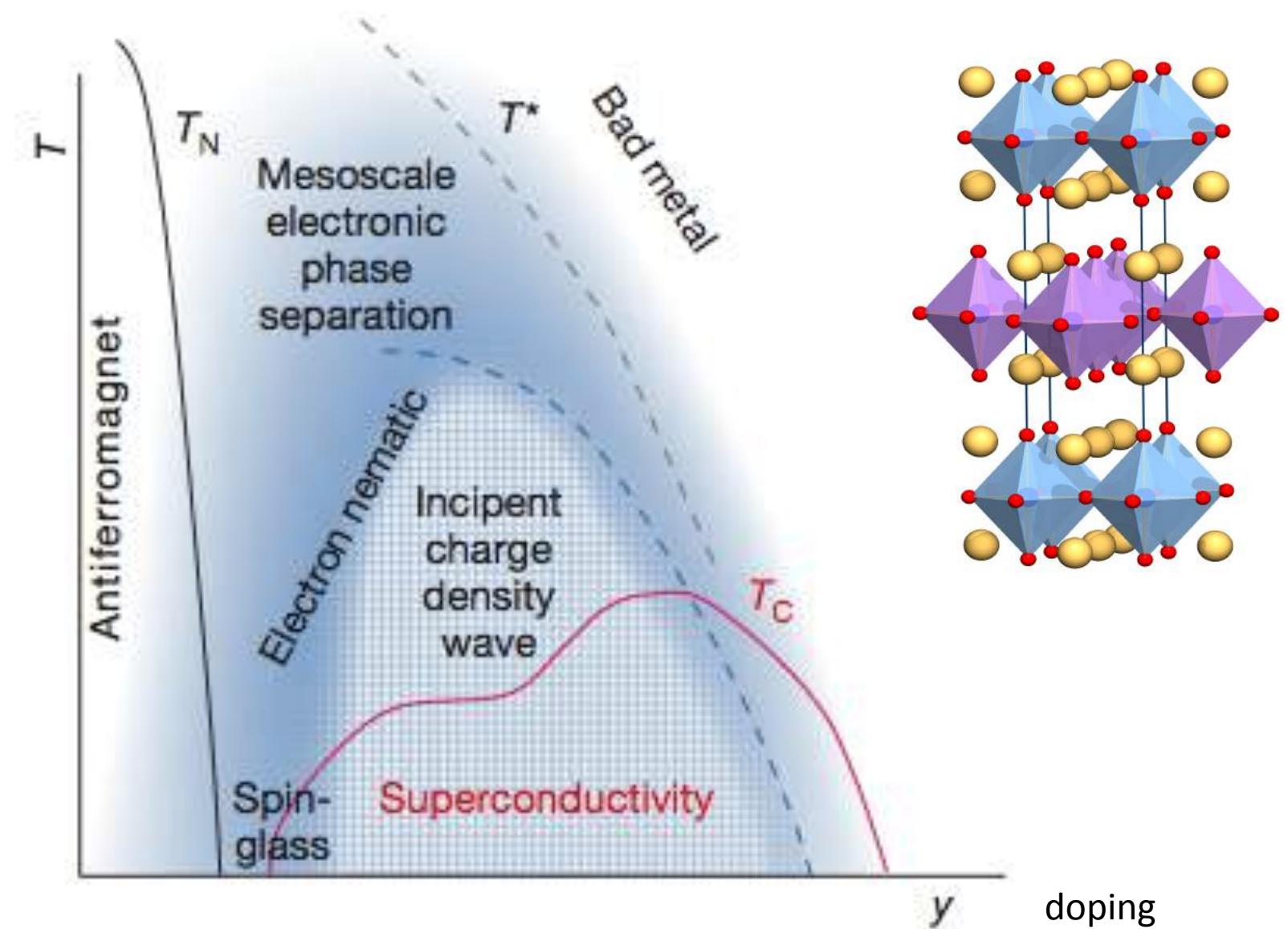
Lattice distortions may quench SC



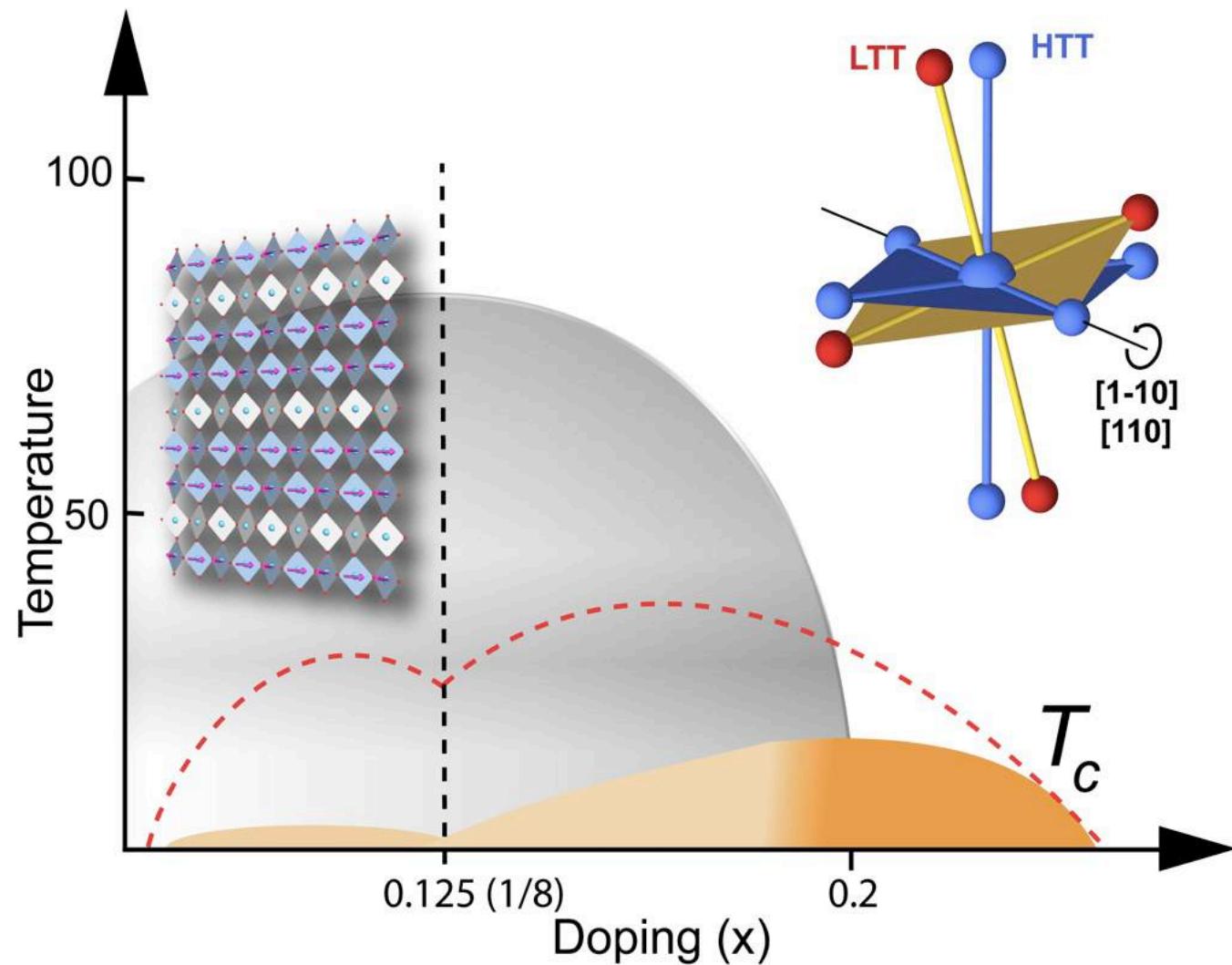
Lattice distortions may promote SC



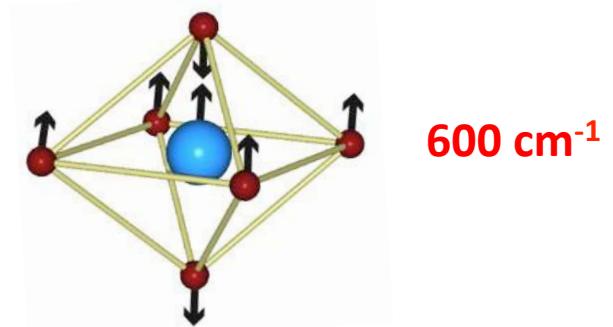
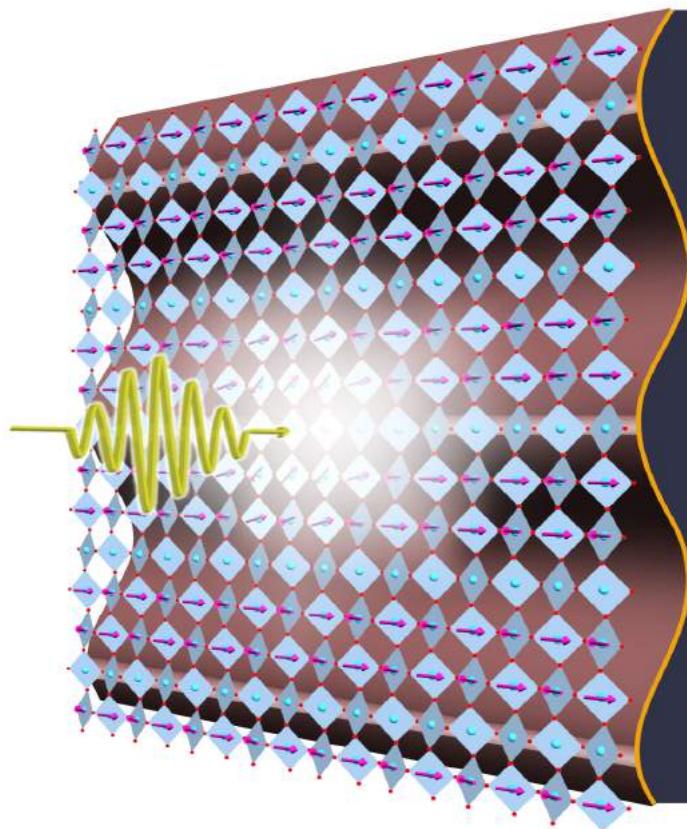
Cuprate superconductors: competing orders and hidden phases



Eu:LSCO_{1/8} stripe charge order



Excitation of in plane Cu-O stretch



16 μm wavelength
 μJ pulses
MV/cm fields



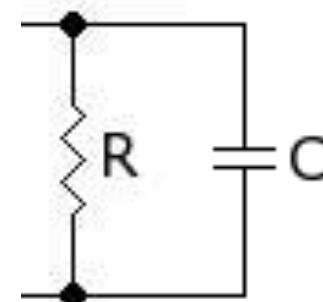
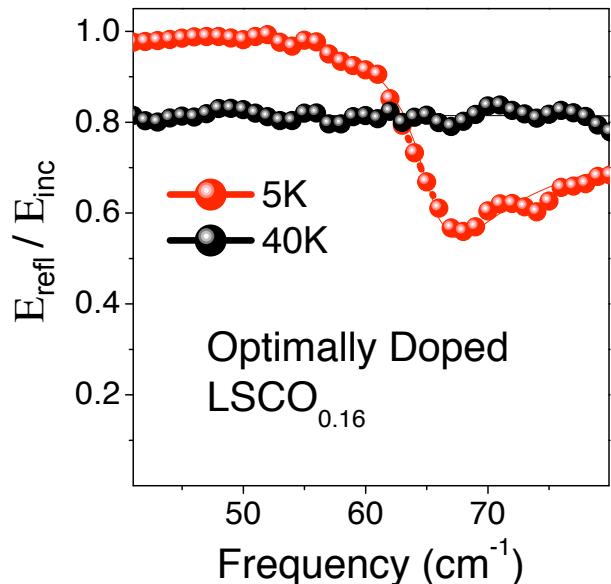
With Hide Takagi
MPI Stuttgart

Probing the transient state

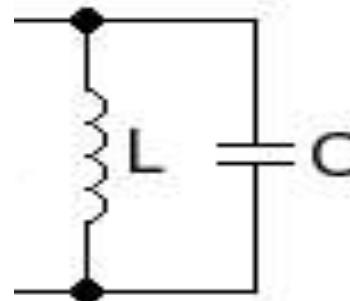
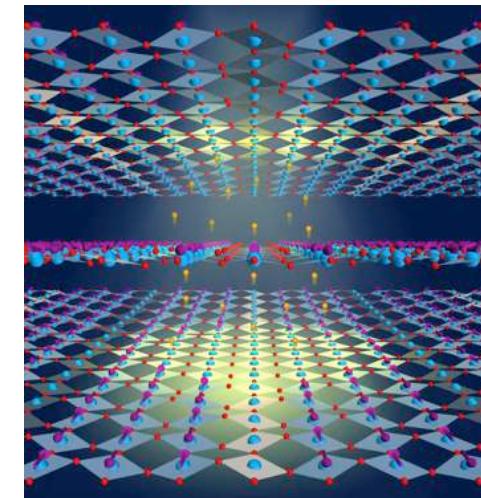
How do I recognize a transient superconductor ?



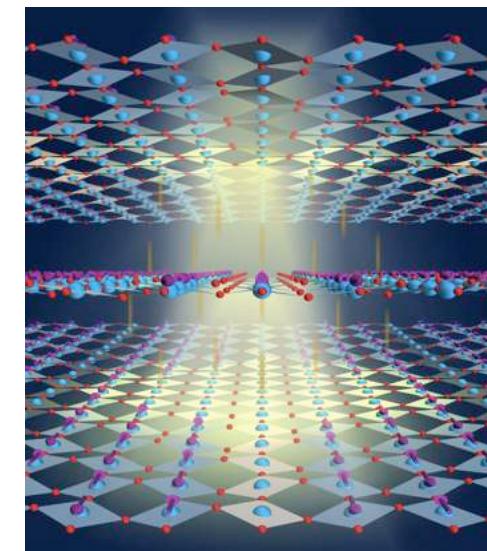
Josephson Plasmon



$T > T_c$



$T < T_c$

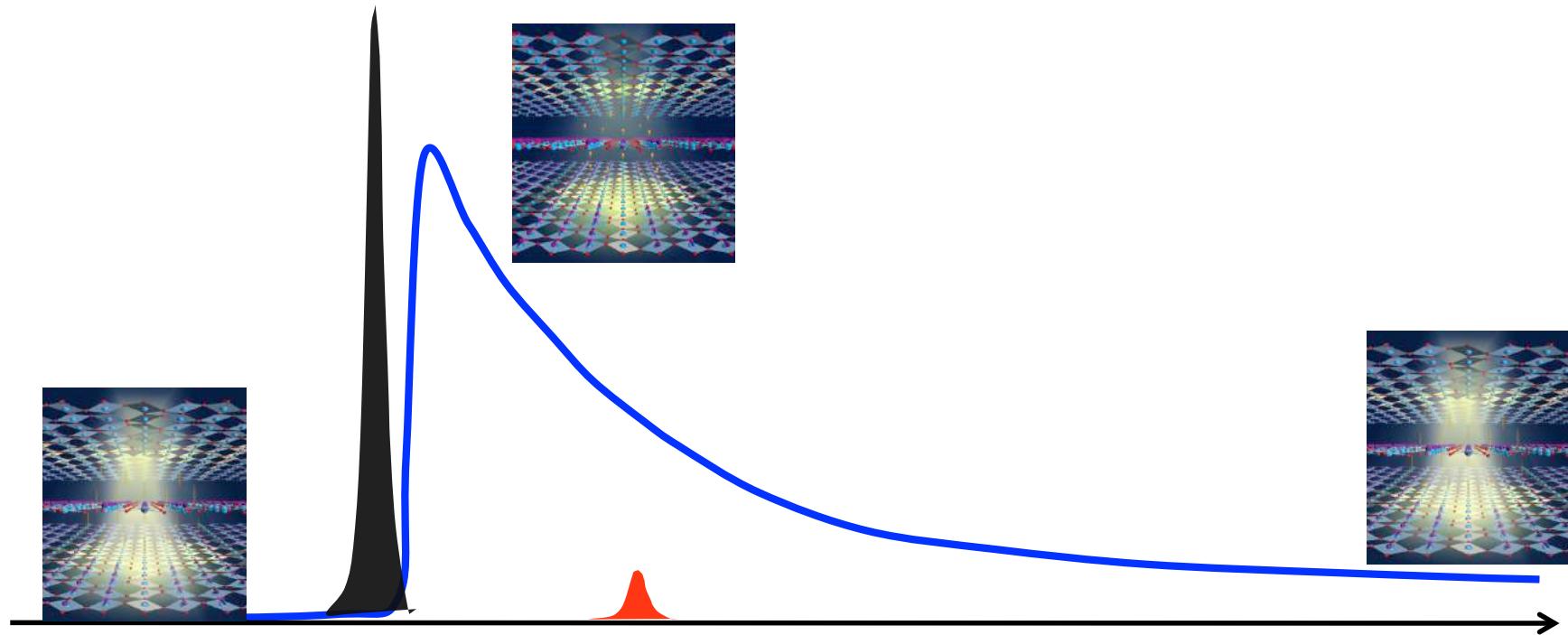


Kresin and Morawitz PRB (1988)

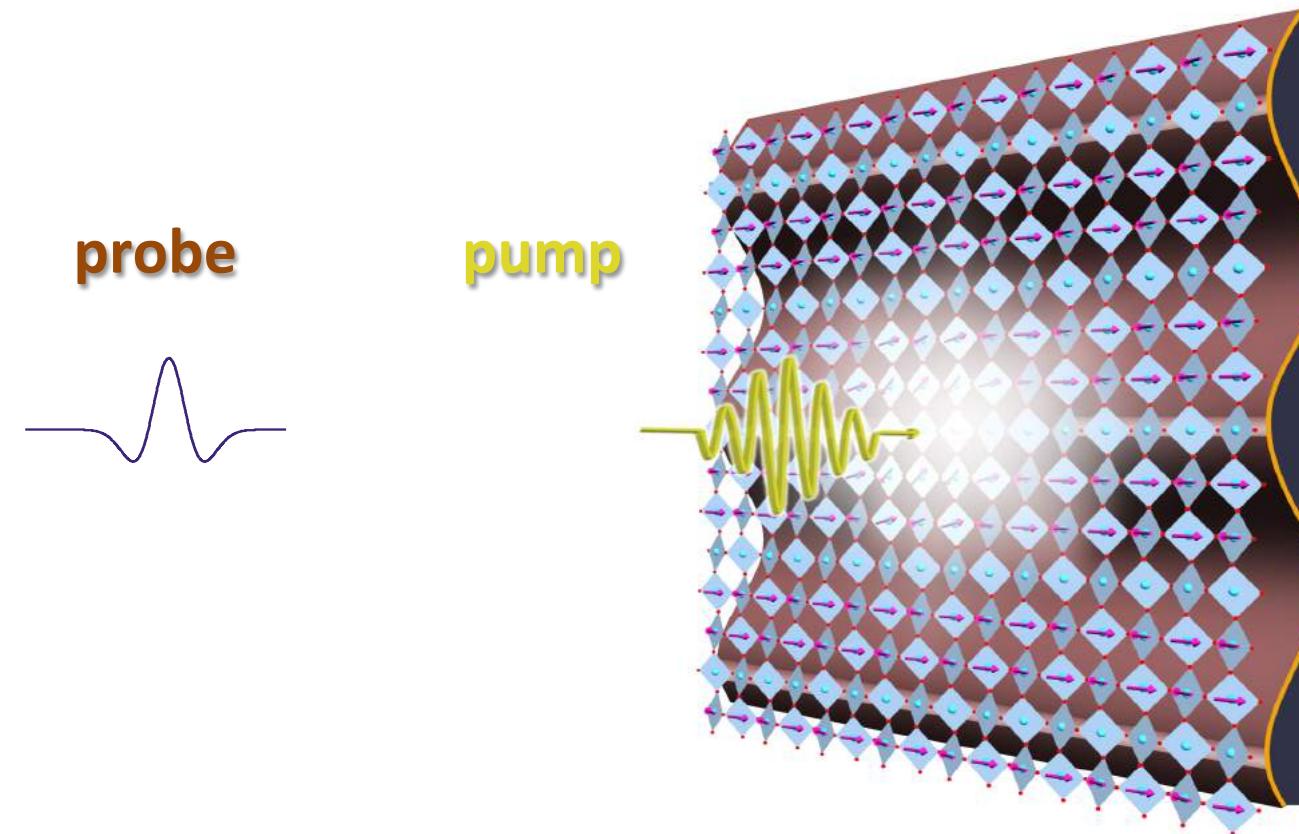
van der Marel and A. A. Tsvetkov Czech. J. Phys. (1996)



Probing the transient state



Mid-IR pump / THz Probe Spectroscopy



A light Induced Josephson plasma edge

Equilibrium LSCO
Superconducting (eq.)

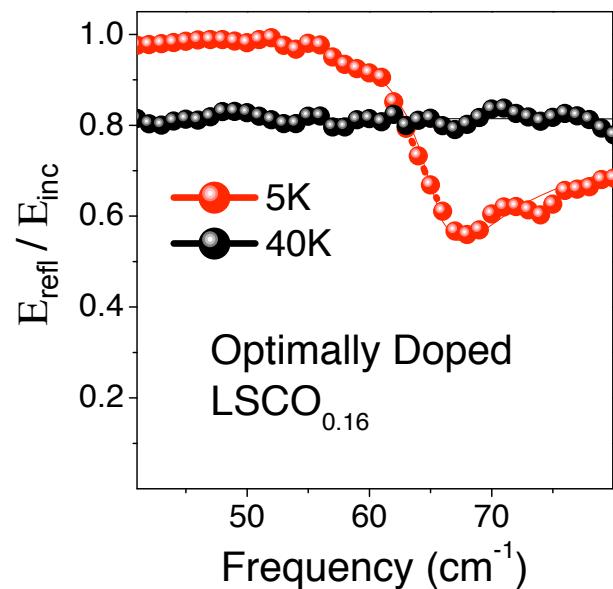
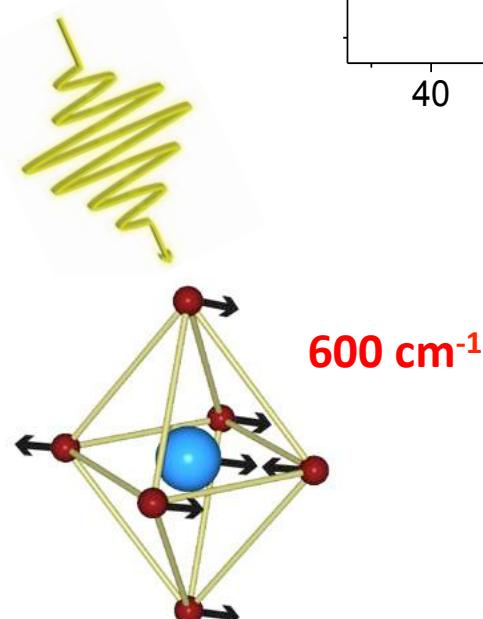
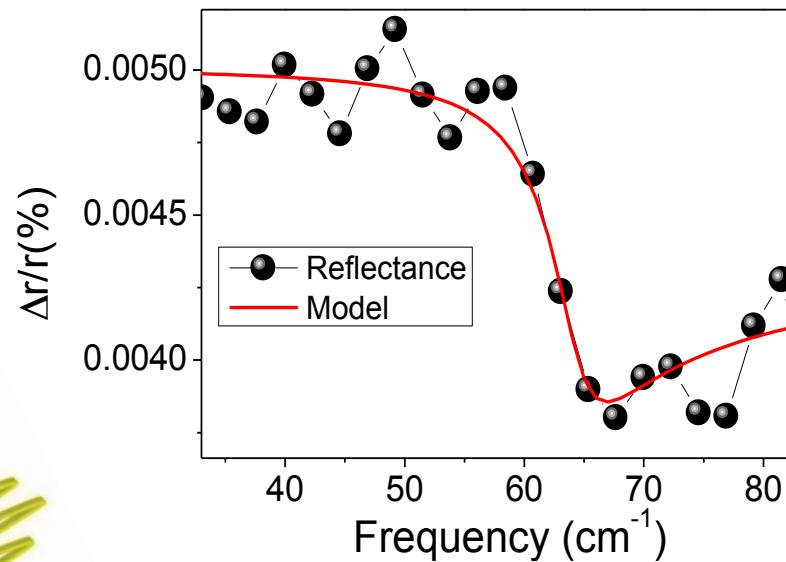
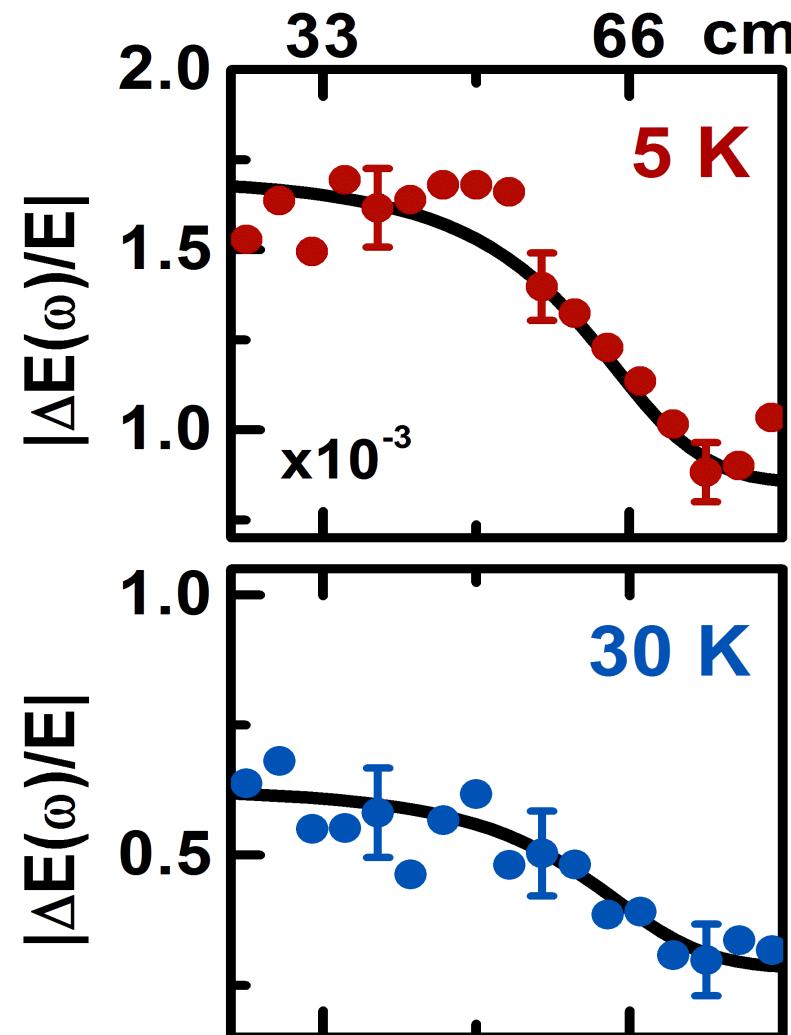
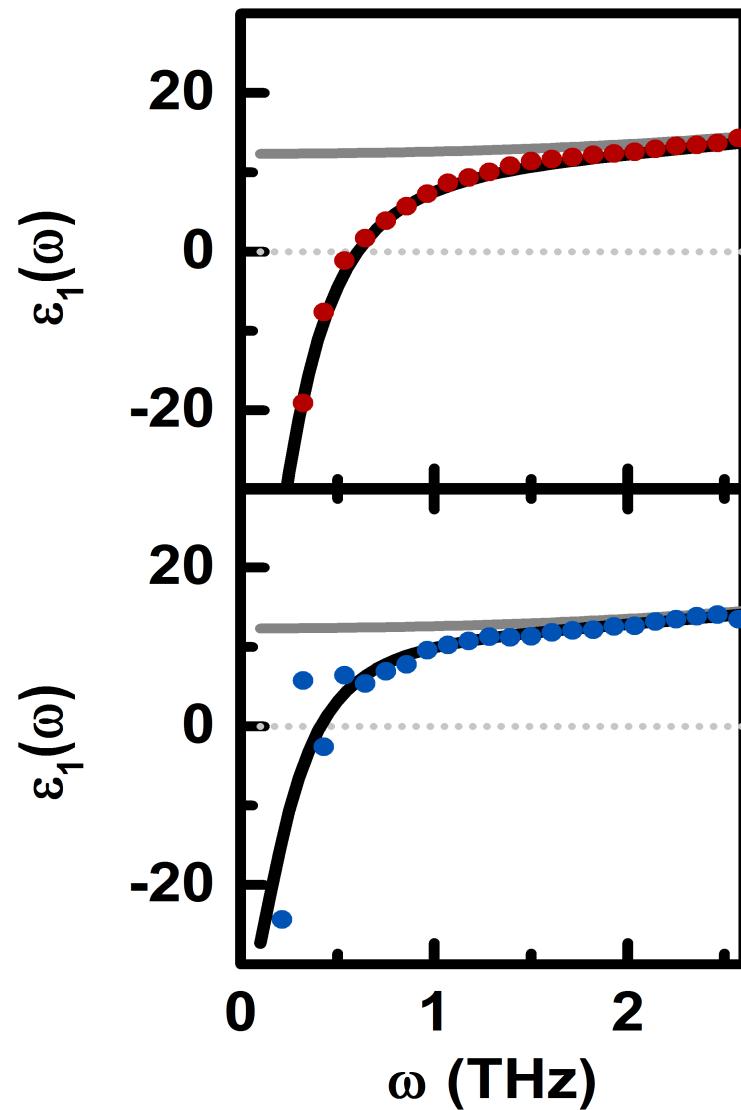


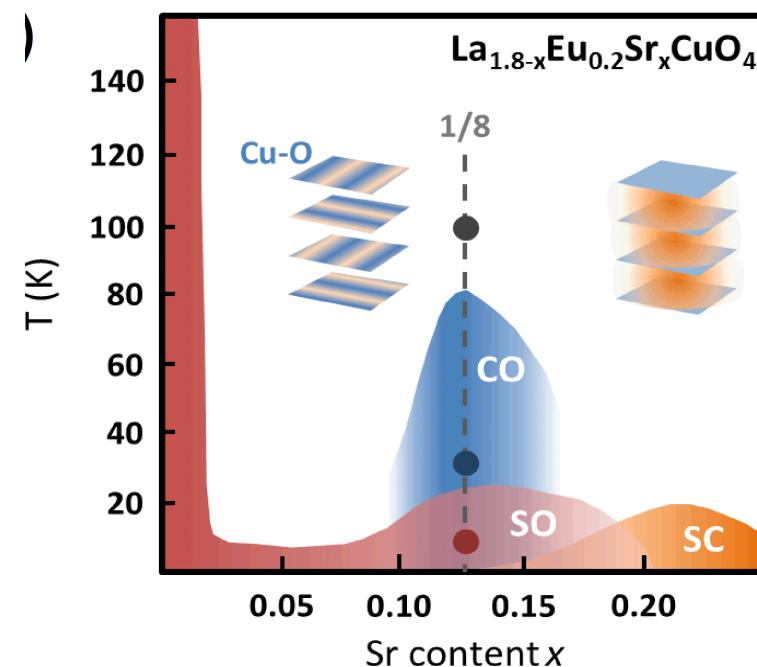
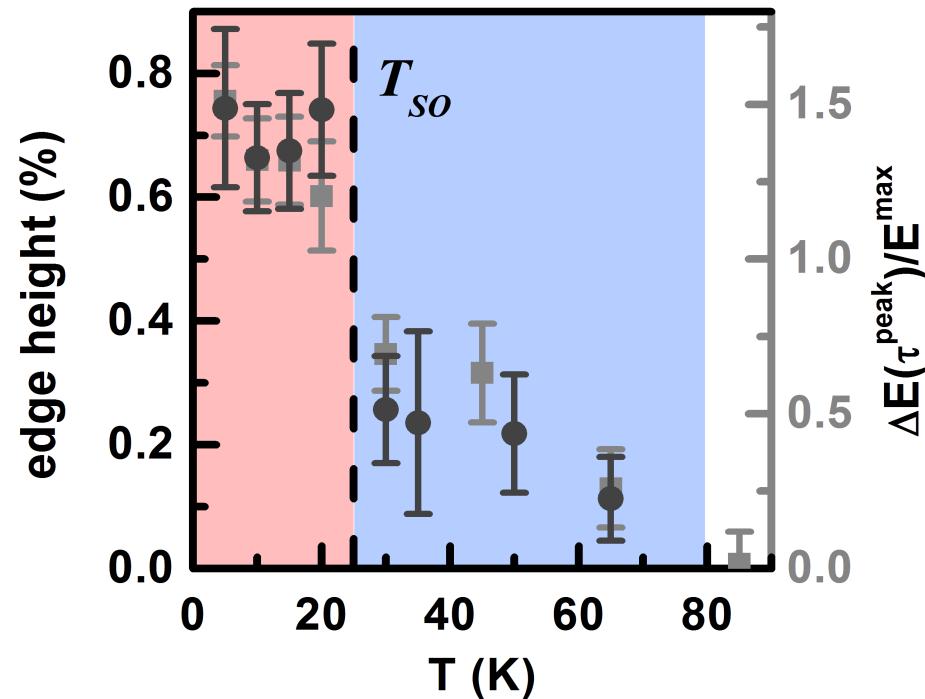
Photo-induced LESCO
Superconducting (non eq.)



Plasma mode where $\varepsilon_1(\omega)$ crosses zero



Plasma mode up to $T_{\text{CO}} = 80$ K



Probing the transient state

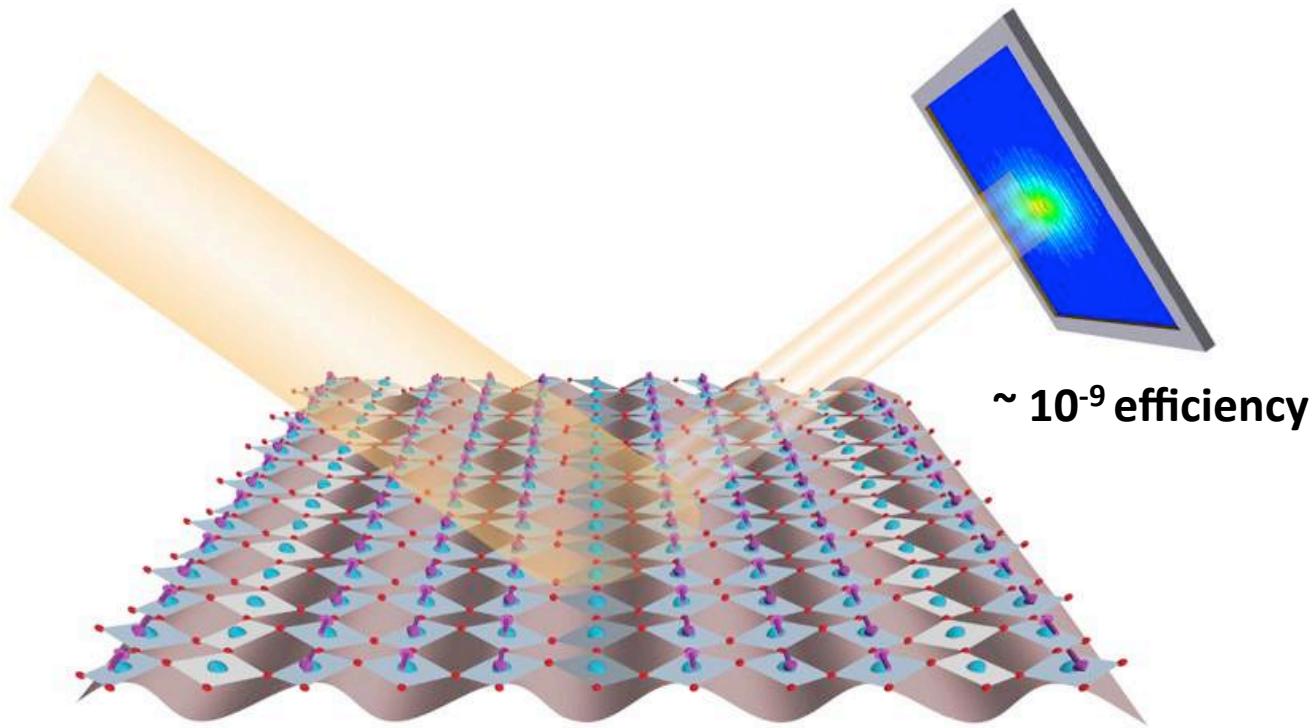
Am I melting charge stripes with light ?



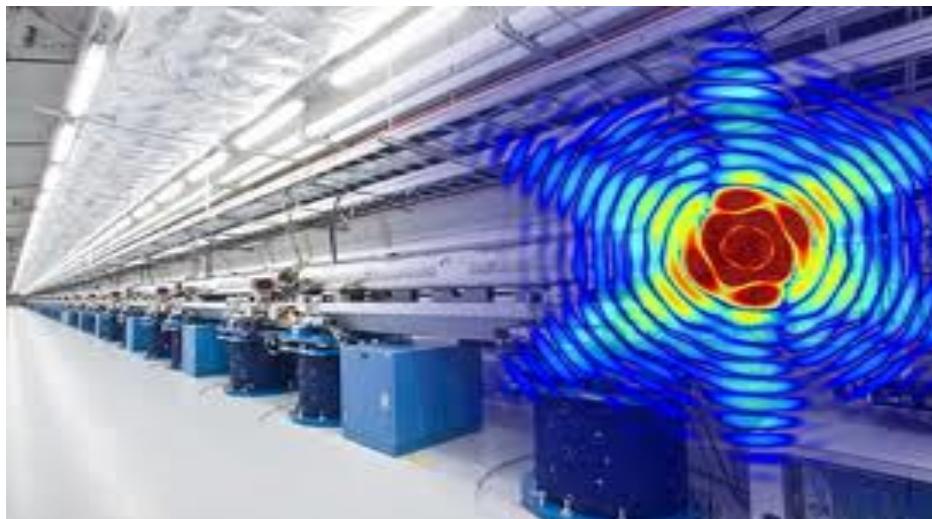
Charge stripes are seen by soft x-ray scattering

O Kedge

(0.25, 0, 0.65)

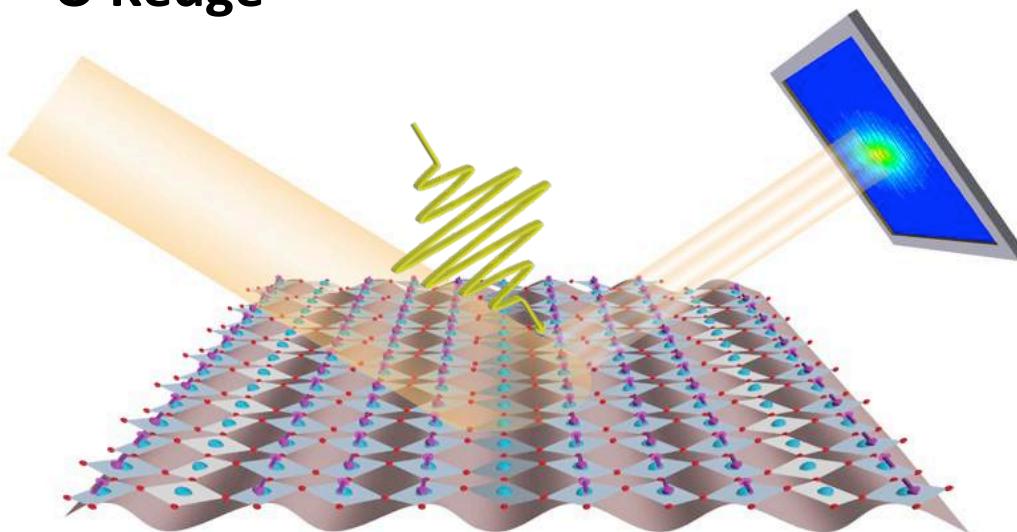


Ultrafast soft X-ray diffraction

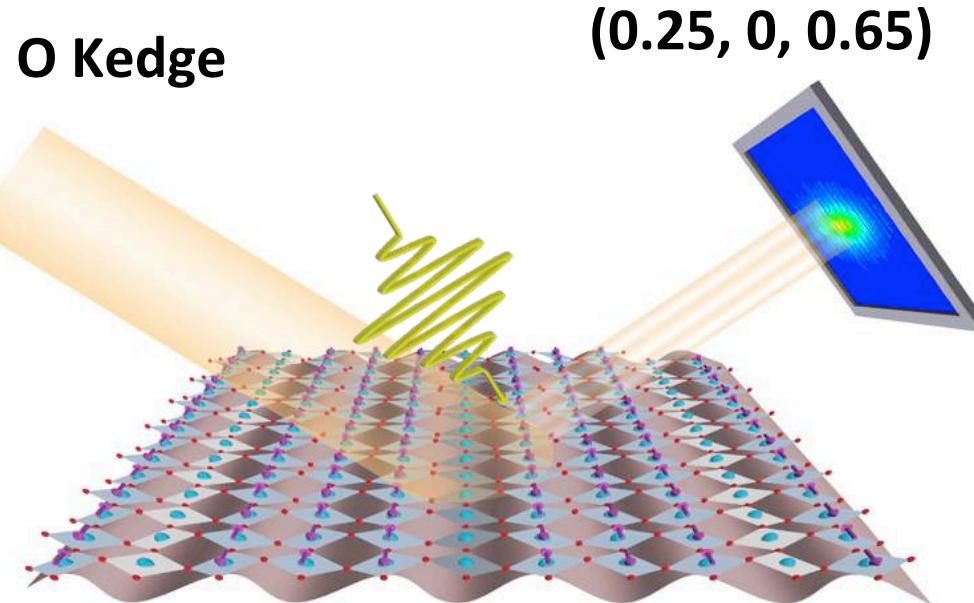
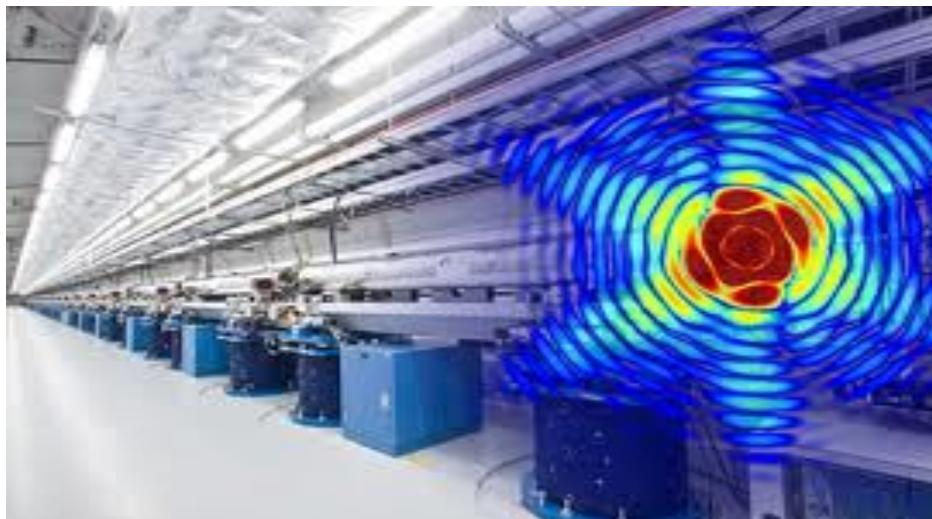


O Kedge

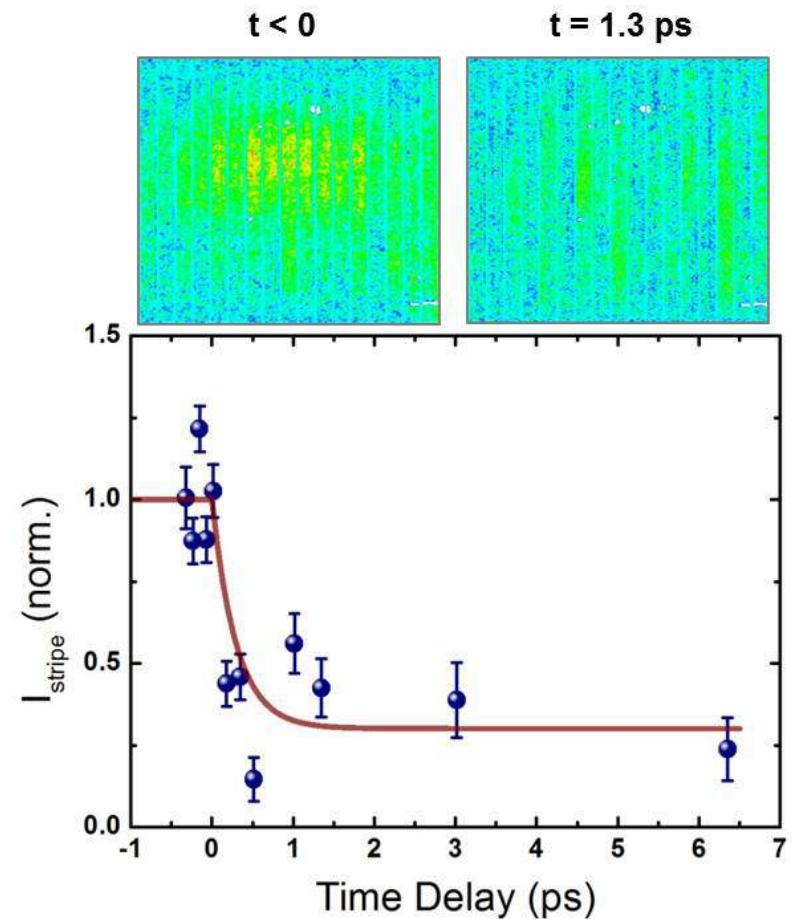
(0.25, 0, 0.65)



Ultrafast soft X-ray diffraction



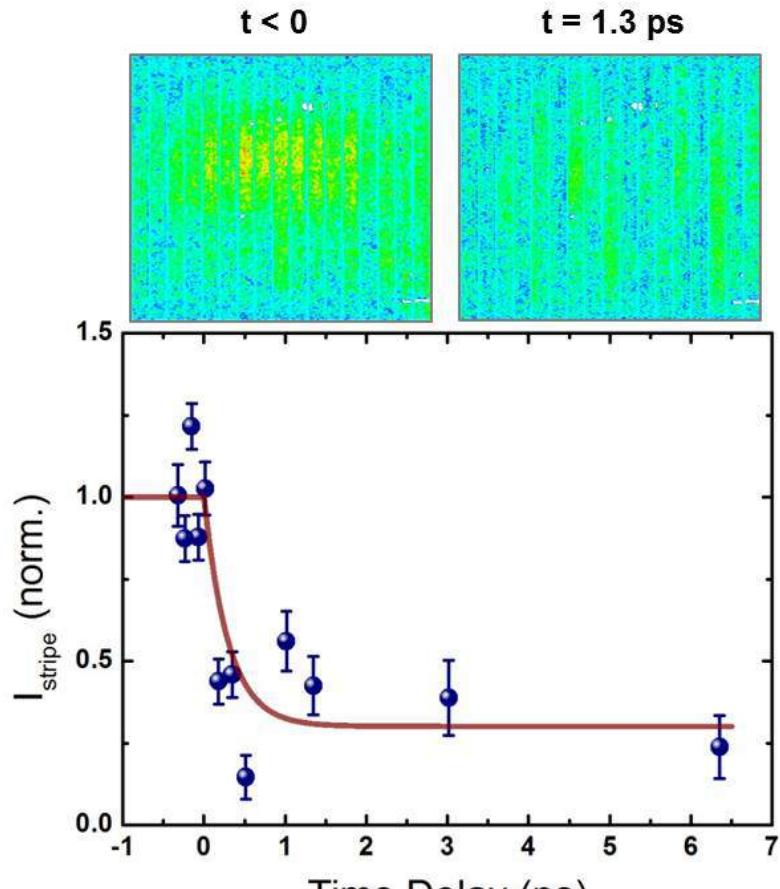
M. Foerst et al., Phys Rev Lett 112, 157002 (2014)



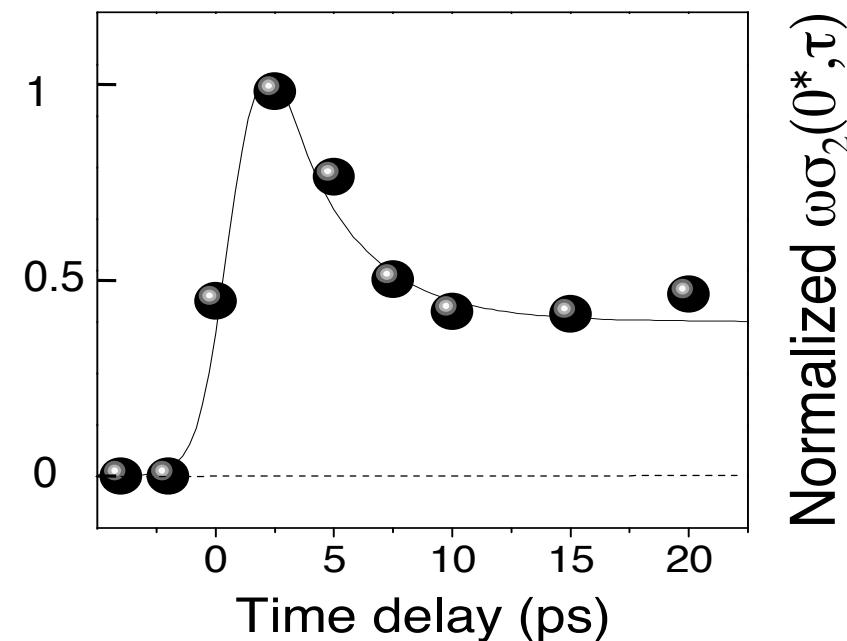
With John Hill, BNL

Charge stripe melting - superconductivity

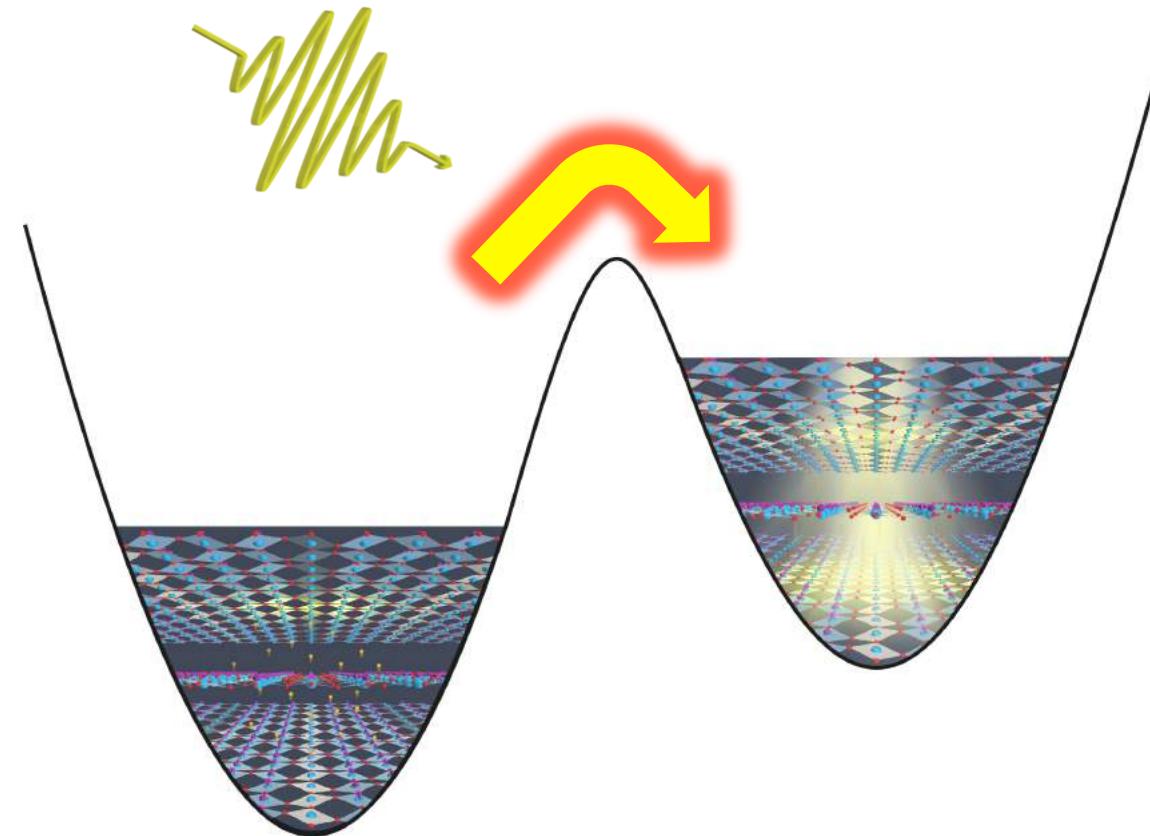
- Charge Stripes melt concomitantly with the formation of the SC



superconductivity



.....switching into a hidden phase

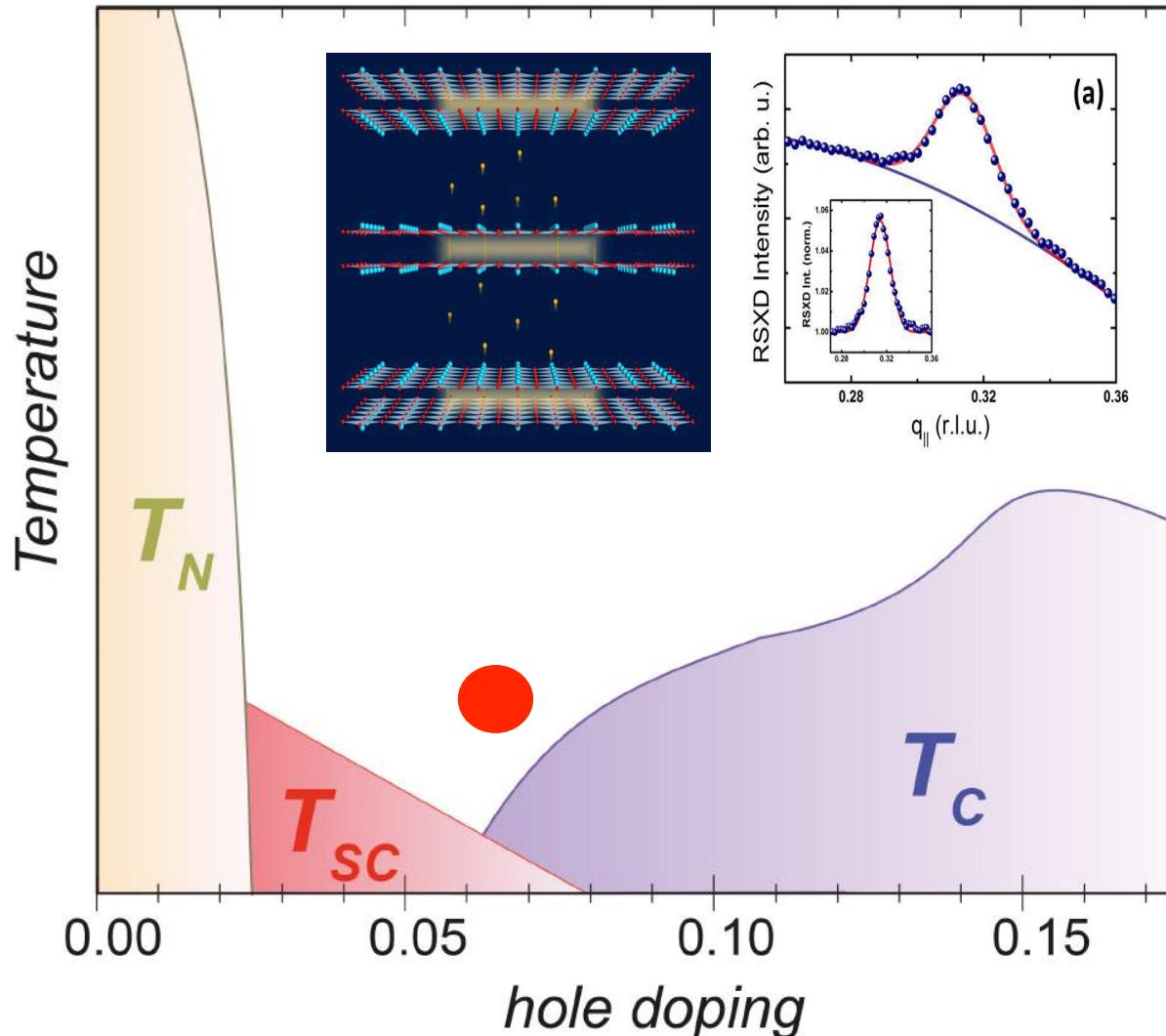


Other systems

Can I do this in other cuprates ?



YBCO: Coherence above T_c and a CDW

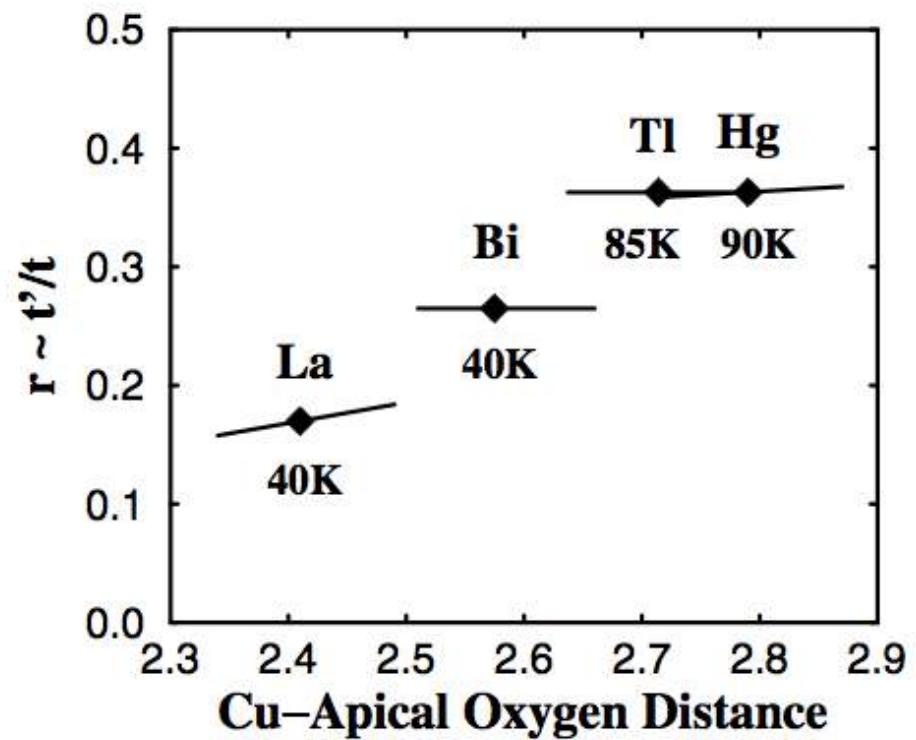
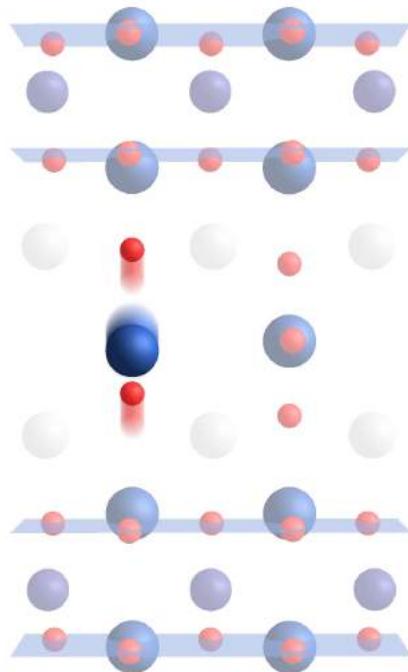


G. Ghiringhelli et al., *Science* 337, 821 (2012)

A. Dubroka et al., *Phys. Rev. Lett.* 107, 047006 (2011)

With B. Keimer
MPI Stuttgart

Apical oxygen correlates with T_c at equilibrium

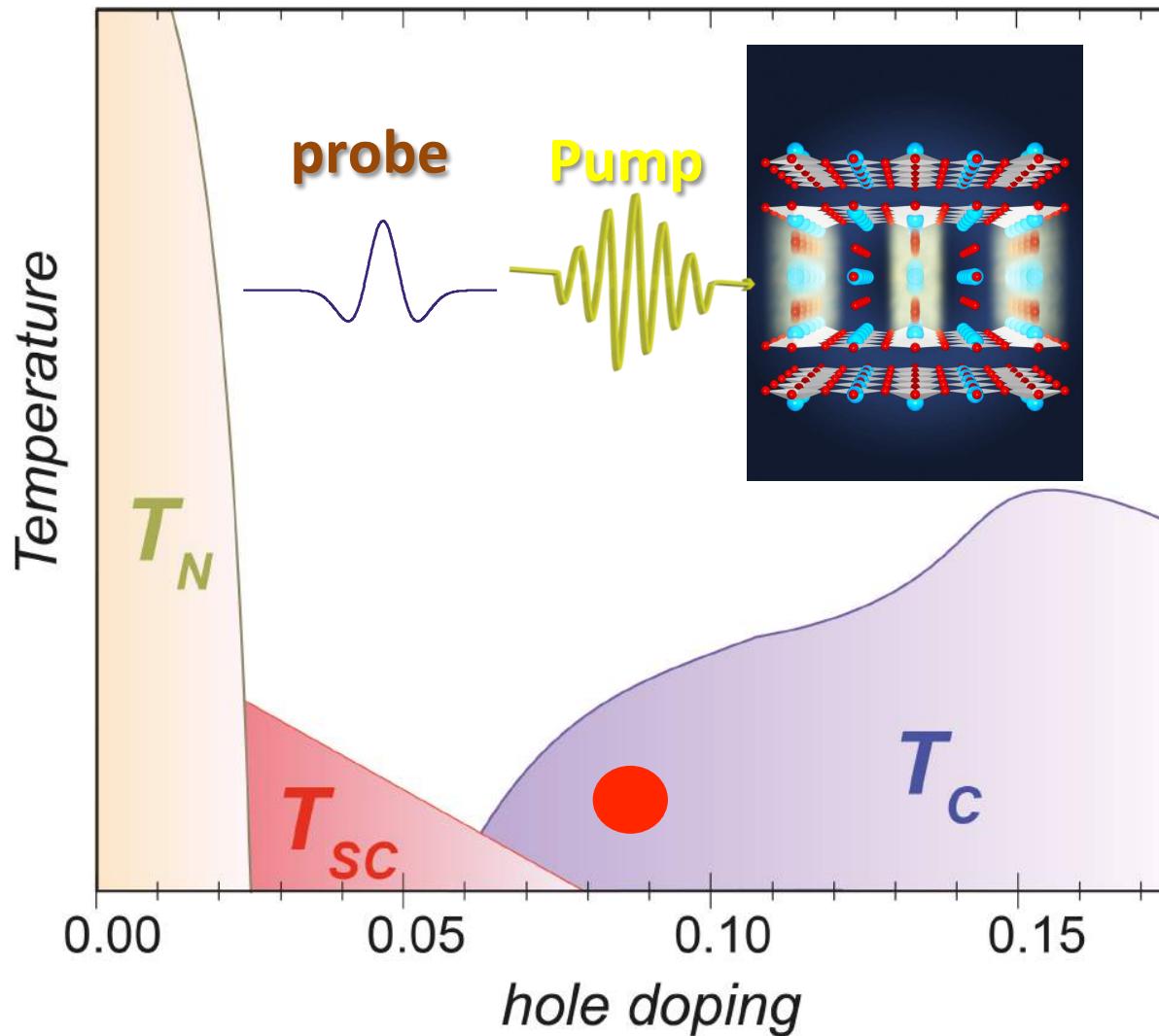


E. Pavarini et al., *PRL* 87, 047003 (2001)

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



Pump apical oxygen probe c-axis plasma

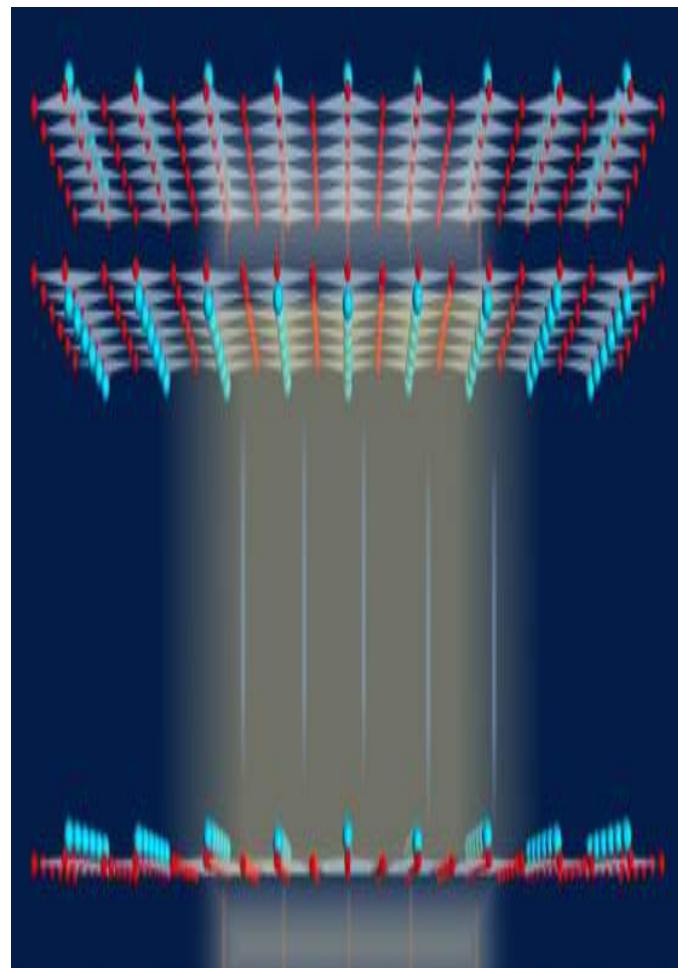
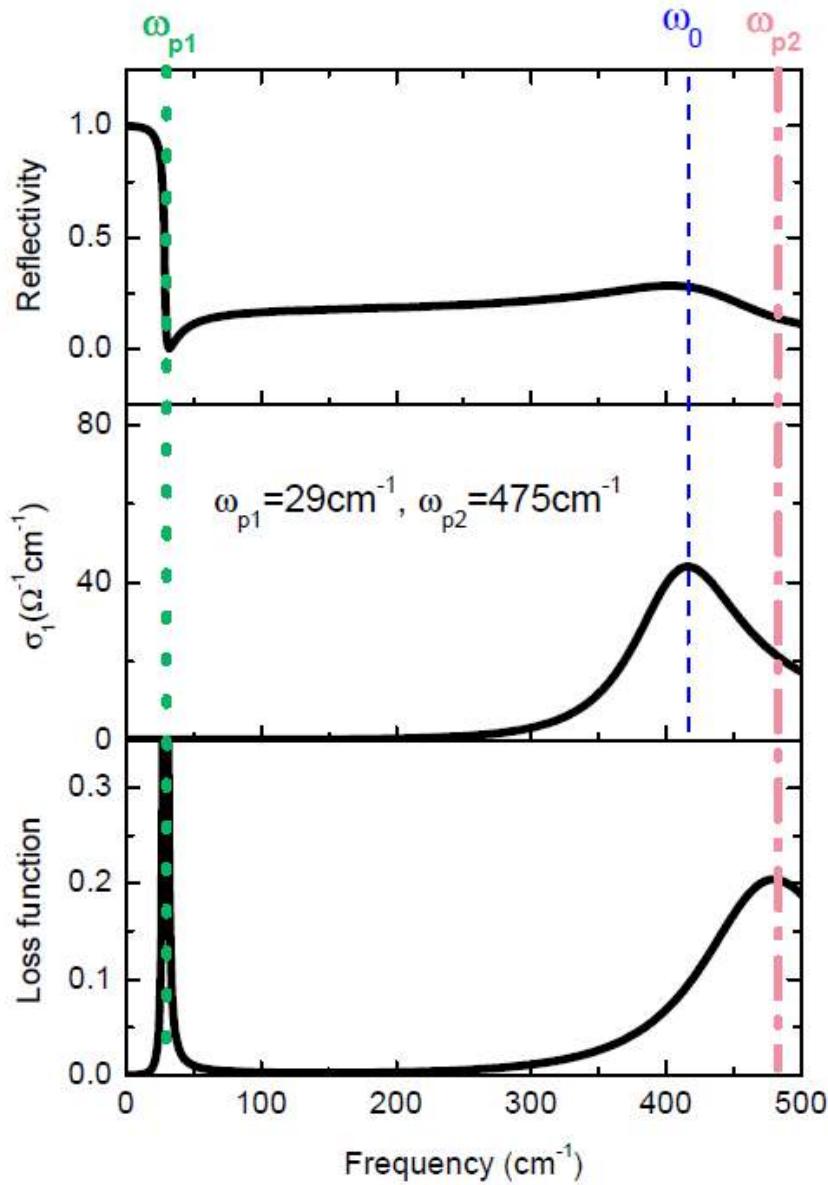


W. Hu et al. *Nature Materials* 13, 705 (2014)

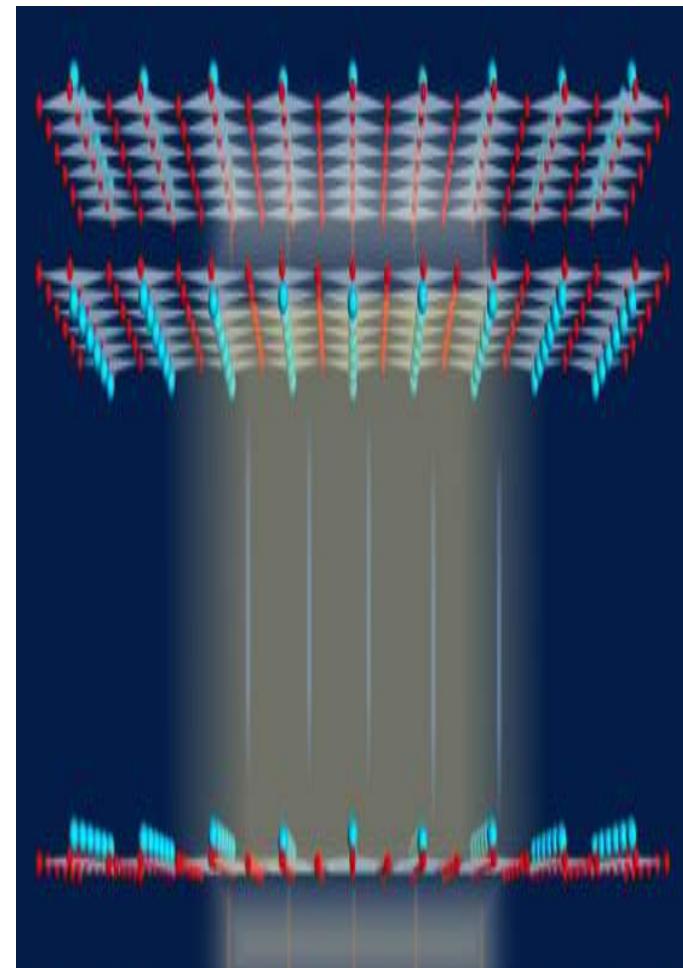
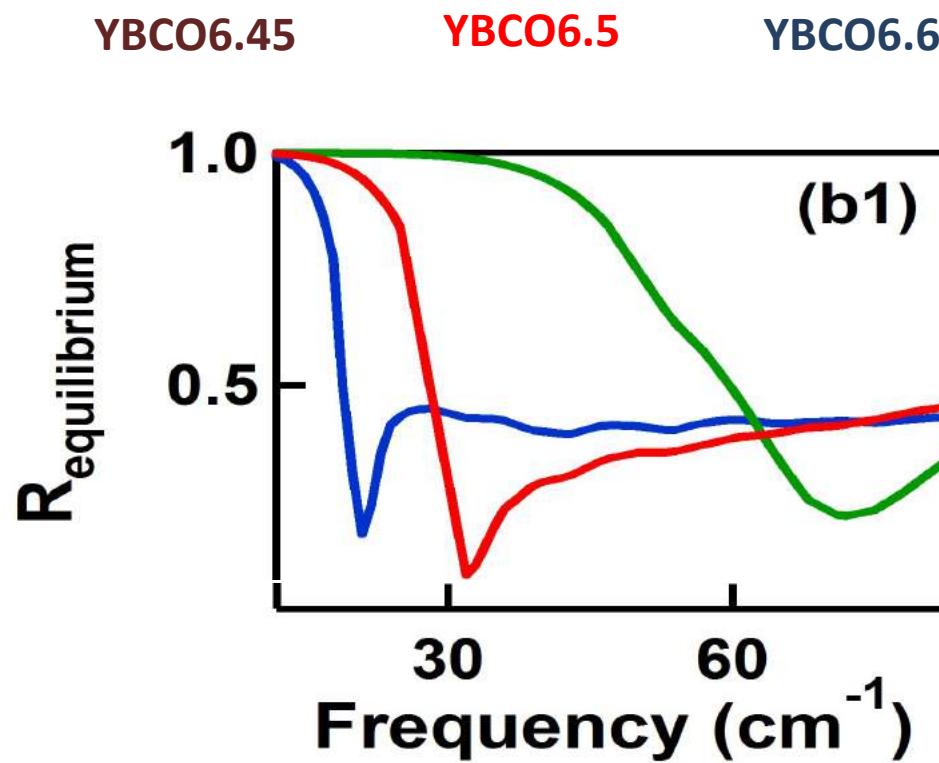
S. Kaiser, D. Nicoletti, C. Hunt et al., *Phys. Rev. B* 89, 184516 (2014)



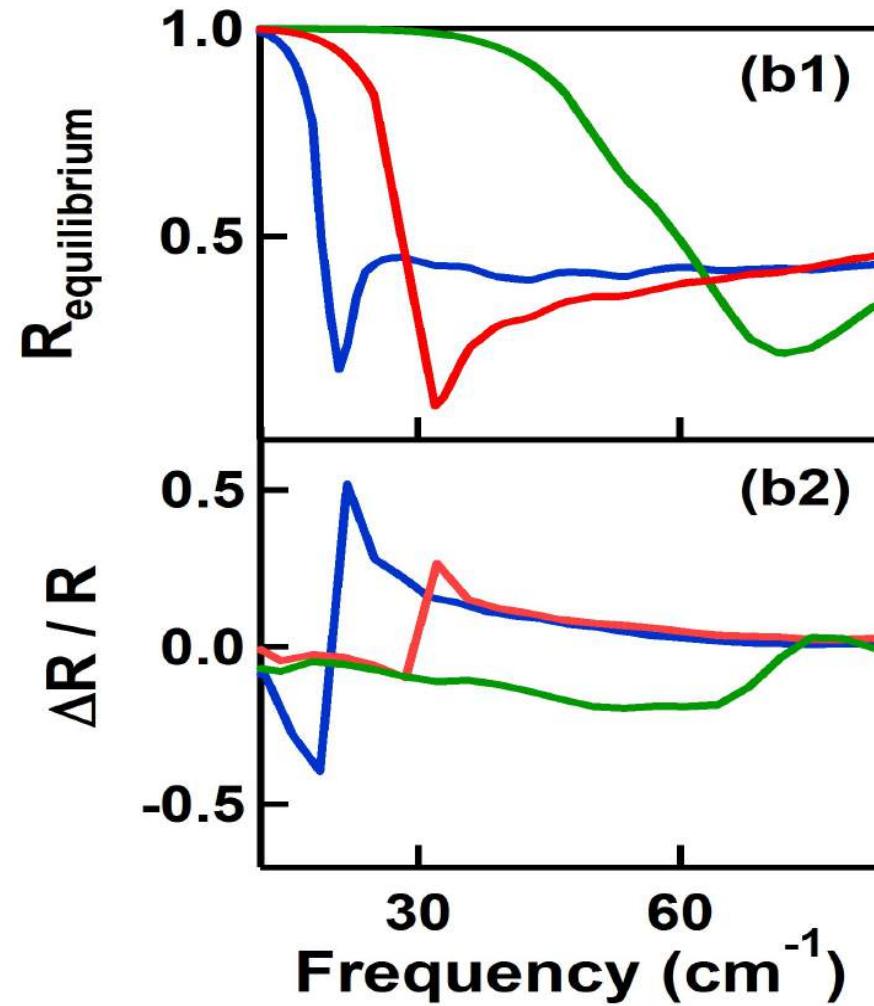
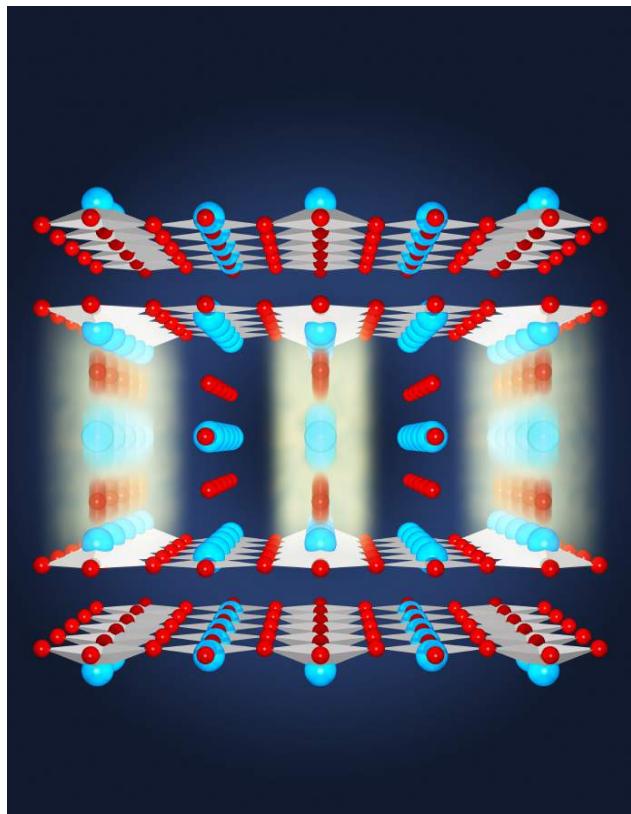
Below Tc: two plasma plasma edges



Low frequency inter-bilayer plasma edge



Below T_c: Light-induced blue shift of the edge

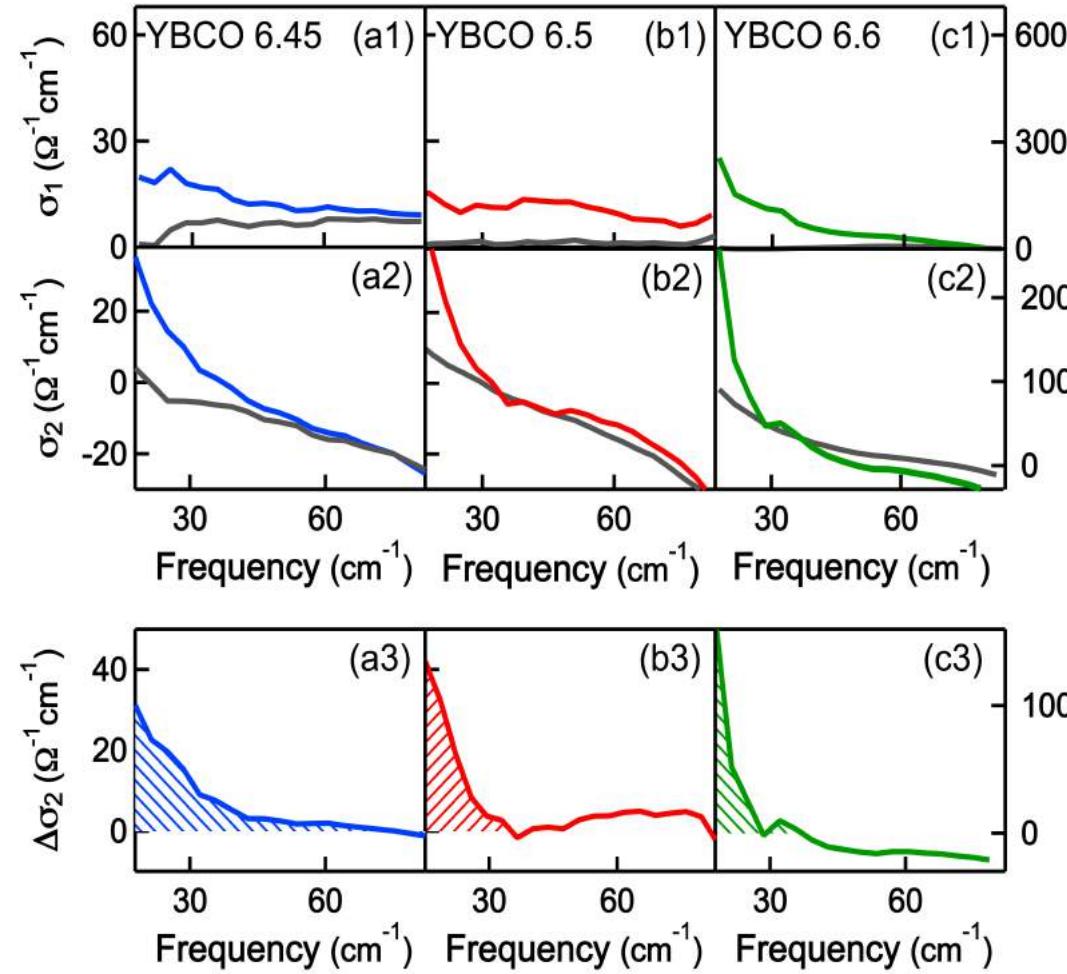


W. Hu et al. *Nature Materials* 13, 705 (2014)

S. Kaiser, D. Nicoletti, C. Hunt et al., *Phys. Rev. B* 89, 184516 (2014)



Below T_c : Enhancement of “superconductivity”

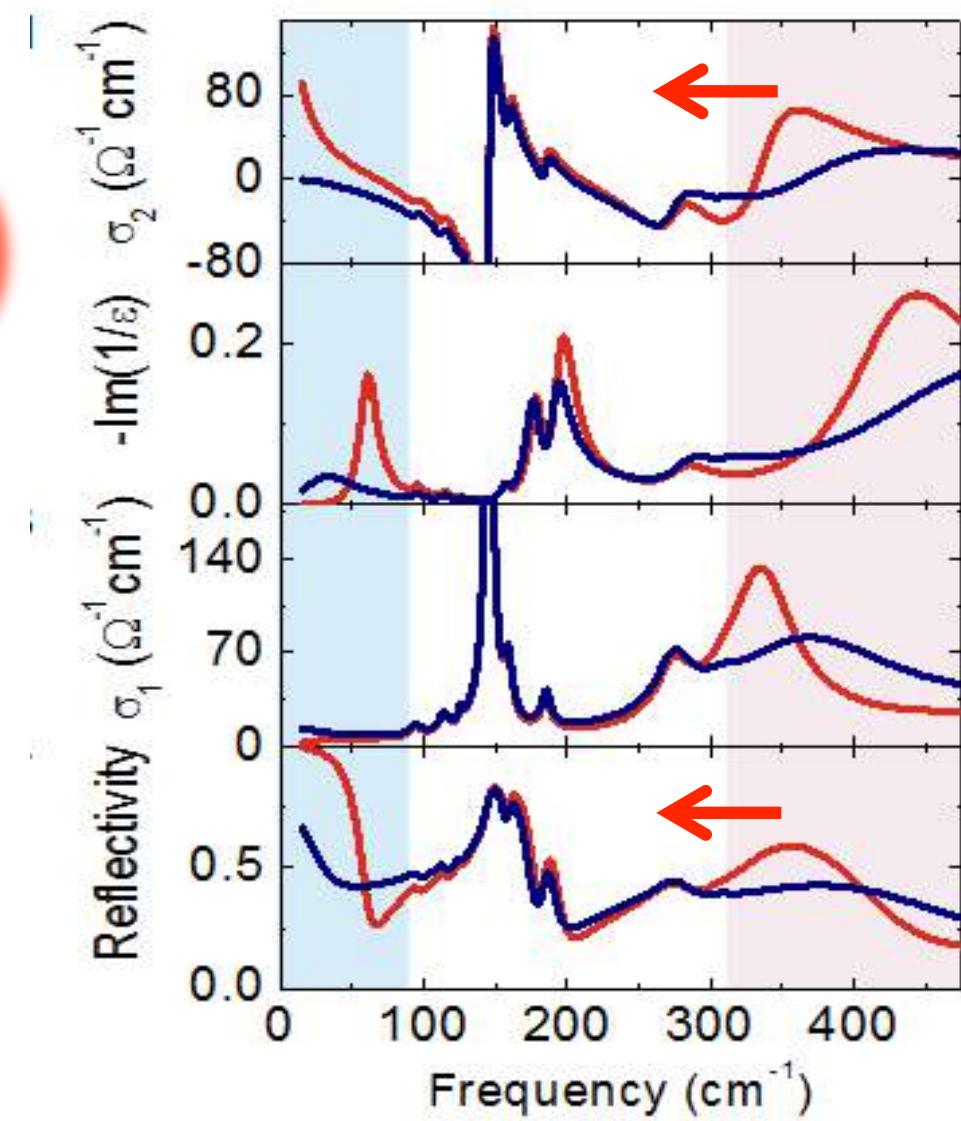
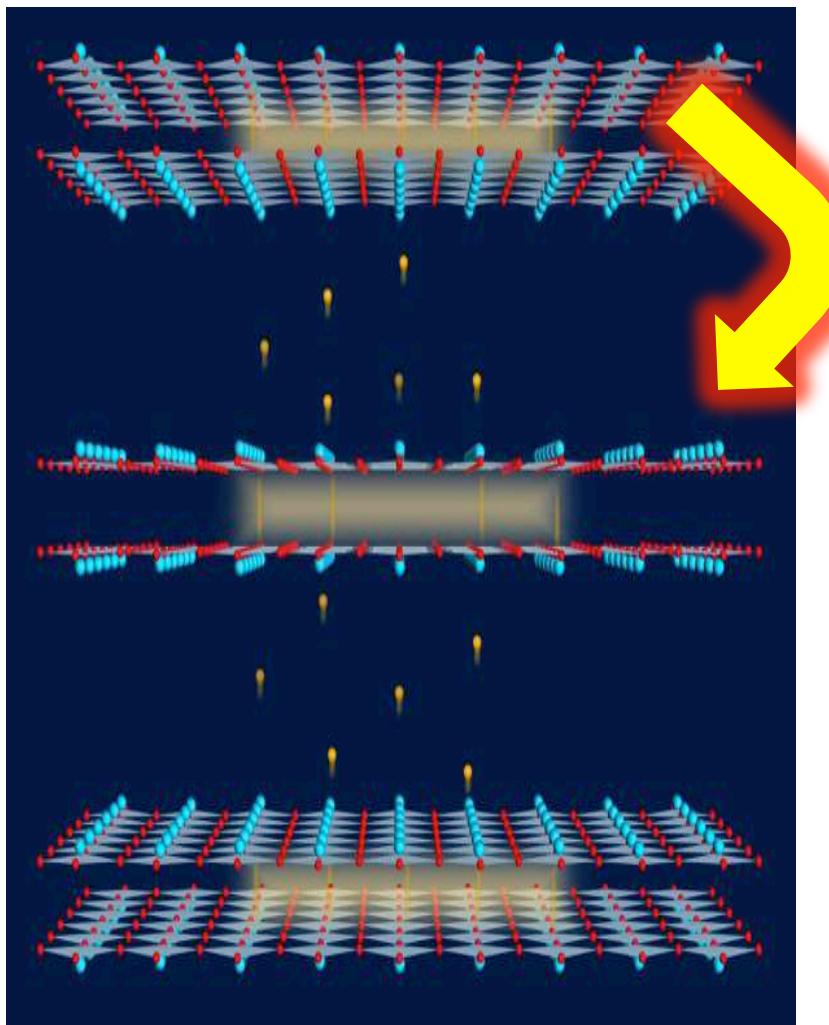


W. Hu. S. Kaiser, D. Nicoletti, C.S. Hunt et al. *Nature Materials* 13, 705 (2014)

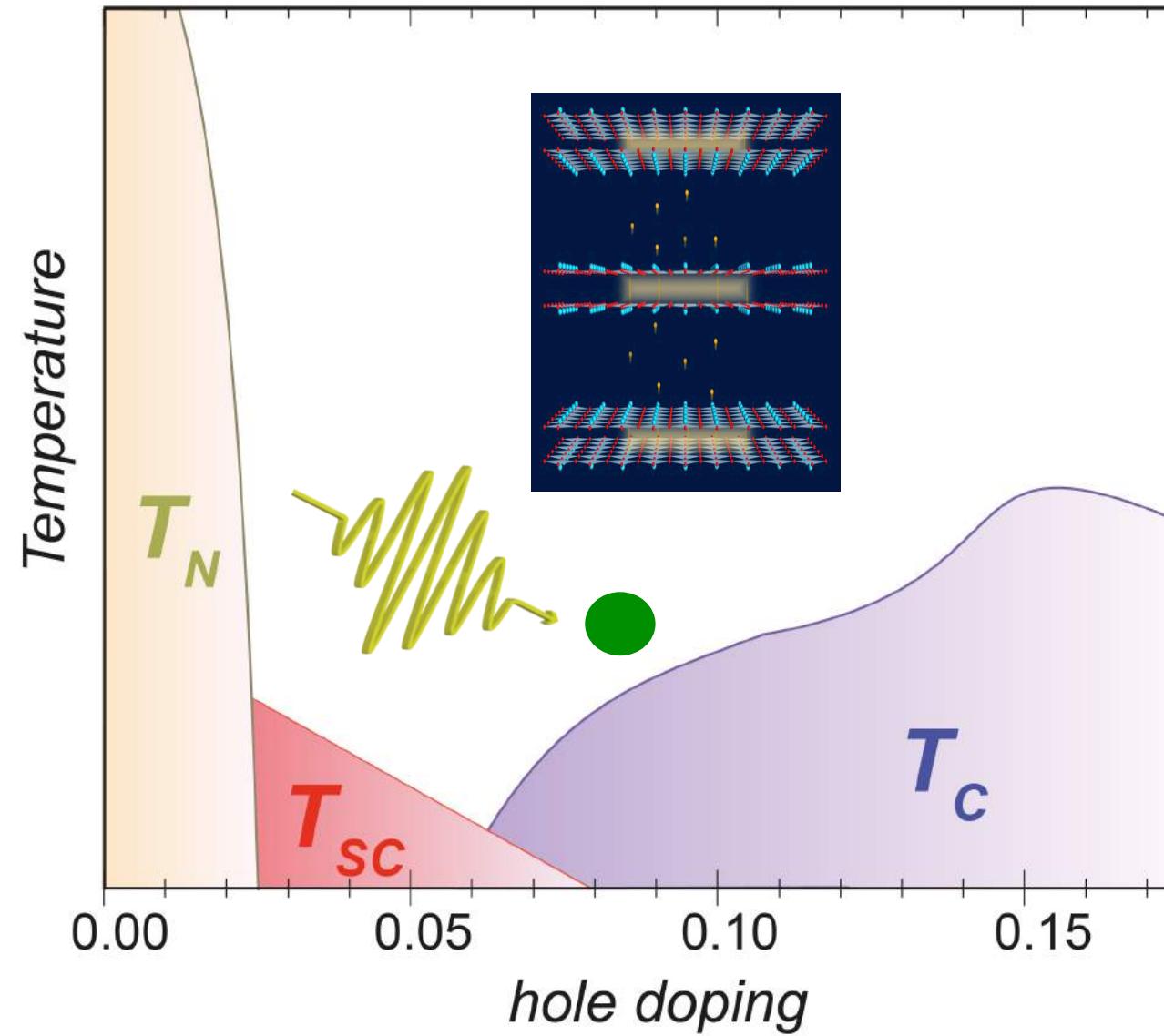
S. Kaiser, D. Nicoletti, C. Hunt et al., *Phys. Rev. B* 89, 184516 (2014)



Spectral weight from high frequency

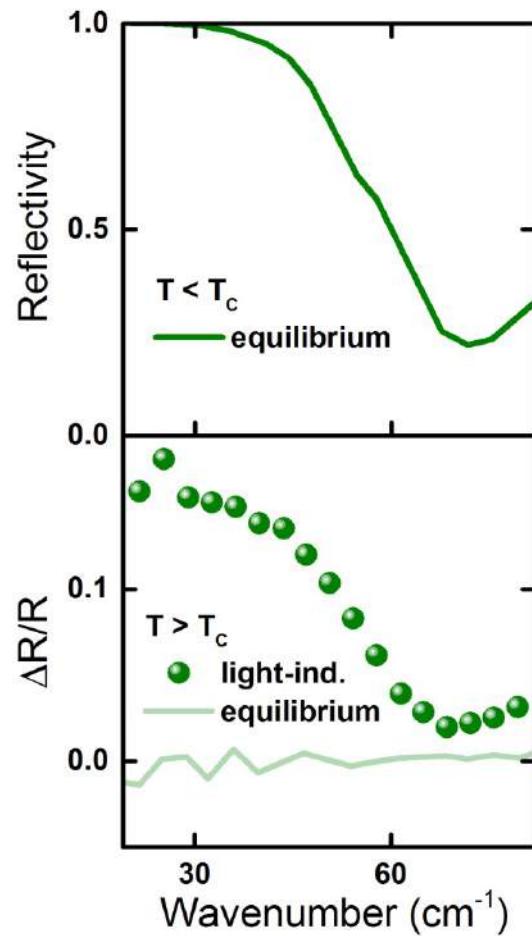


Above T_c

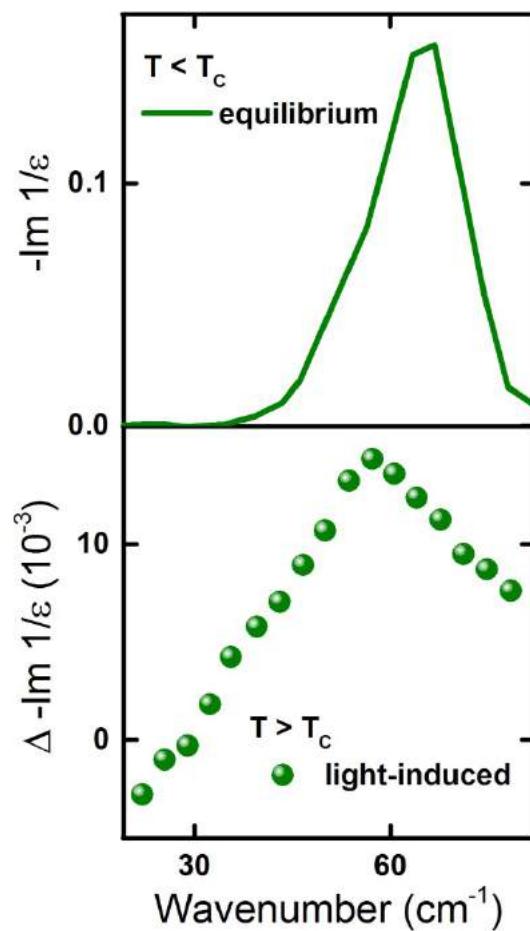


Light induced Plasma Mode – 2 X Tc

Plasma edge



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



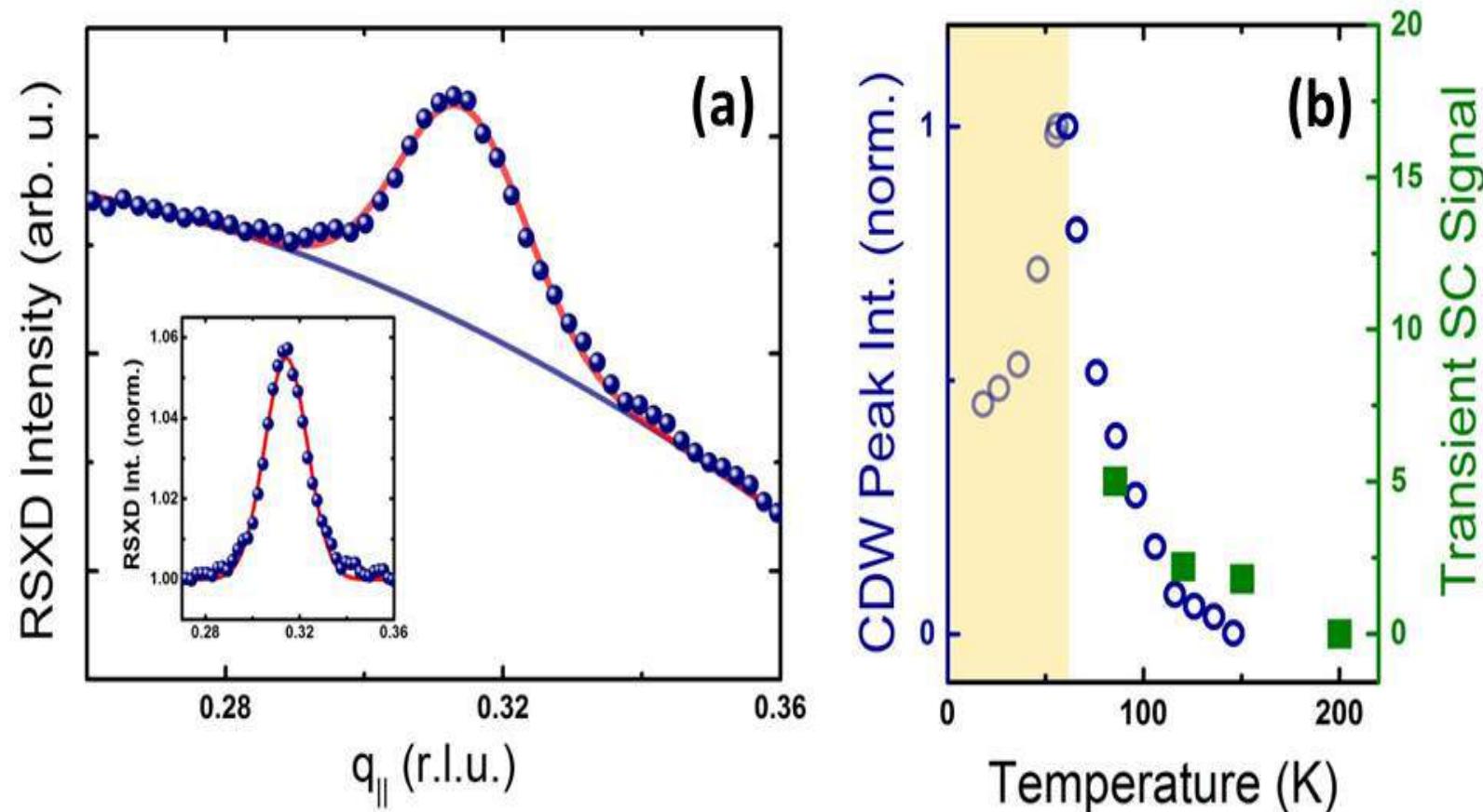
YBCO_{6.6} – 100 K

Equilibrium $T < T_c$

Light induced $T > T_c$



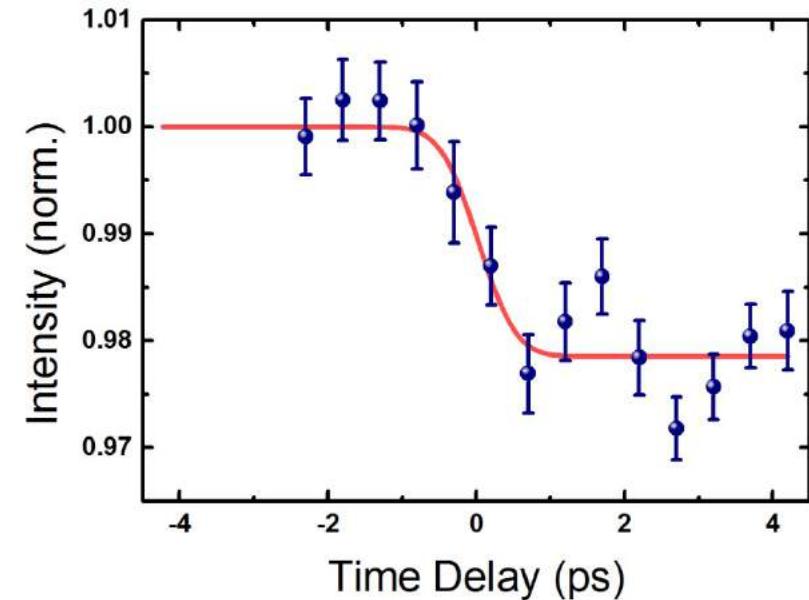
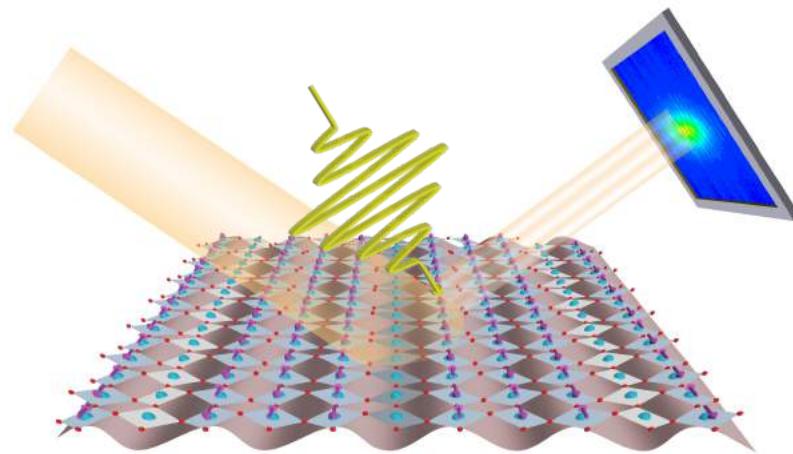
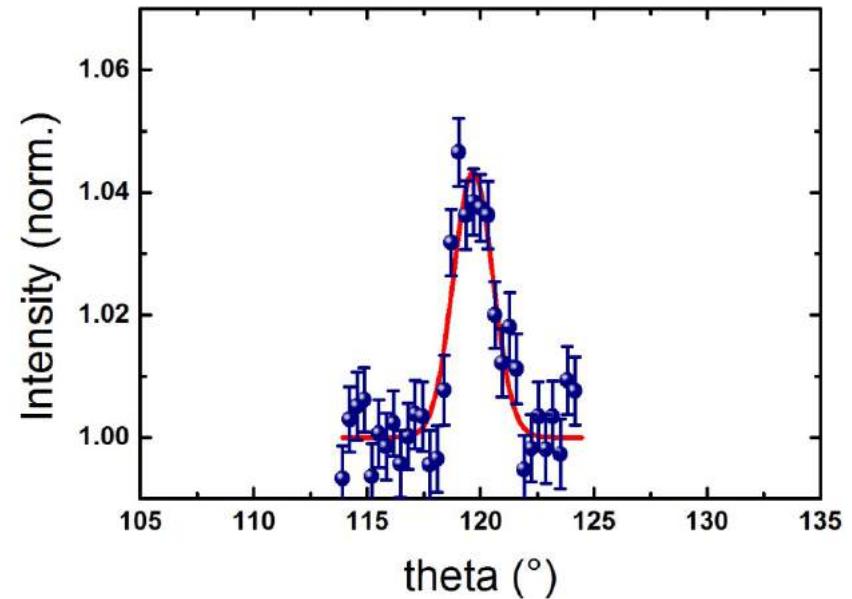
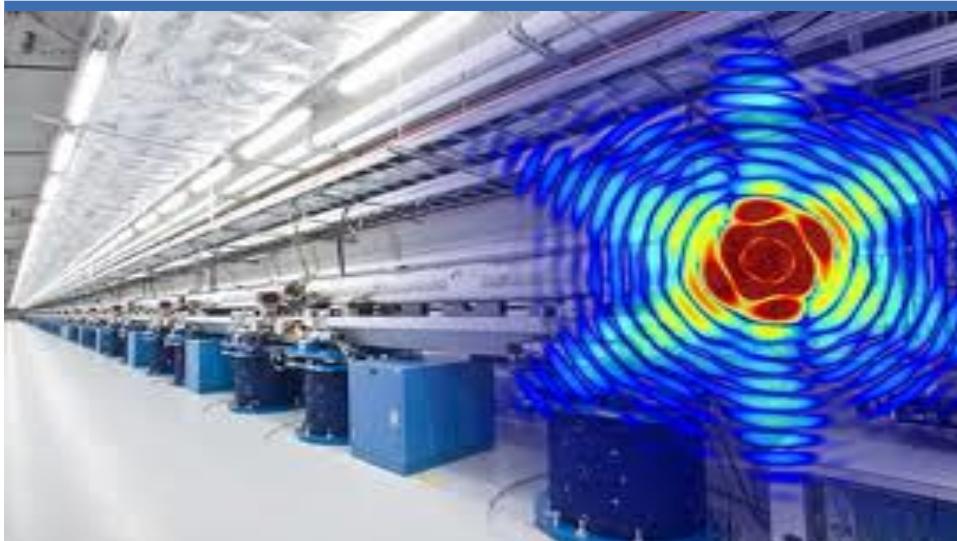
Light induced edge – follows charge order



G. Ghiringhelli et al., *Science* 337, 821 (2012)

S. Kaiser, D. Nicoletti, C. Hunt et al., *Phys. Rev. B* 89, 184516 (2014)

$\text{YBCO}_{6.6}$: Light induced CDW melting



M. Foerst et al., *Phys. Rev. B* (2014)

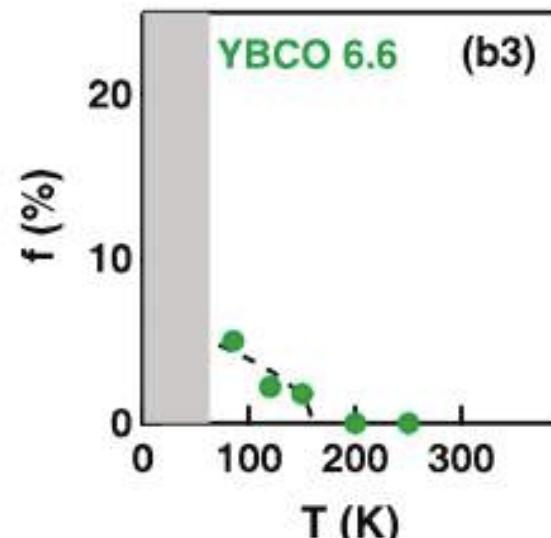
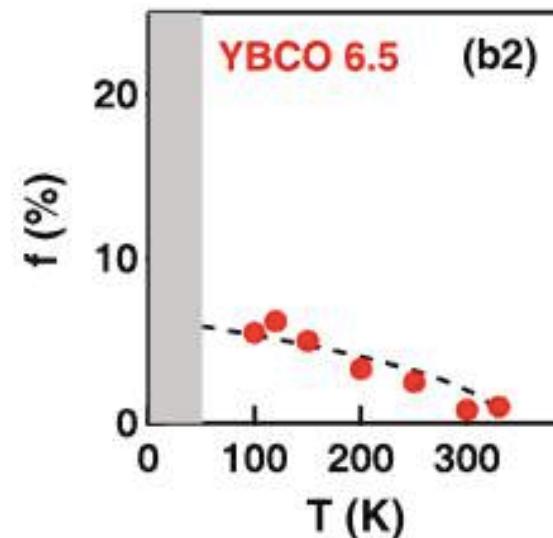
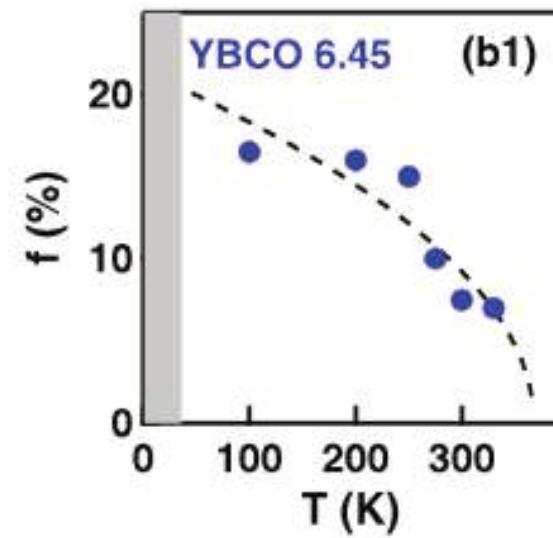


Underdoped YBCO

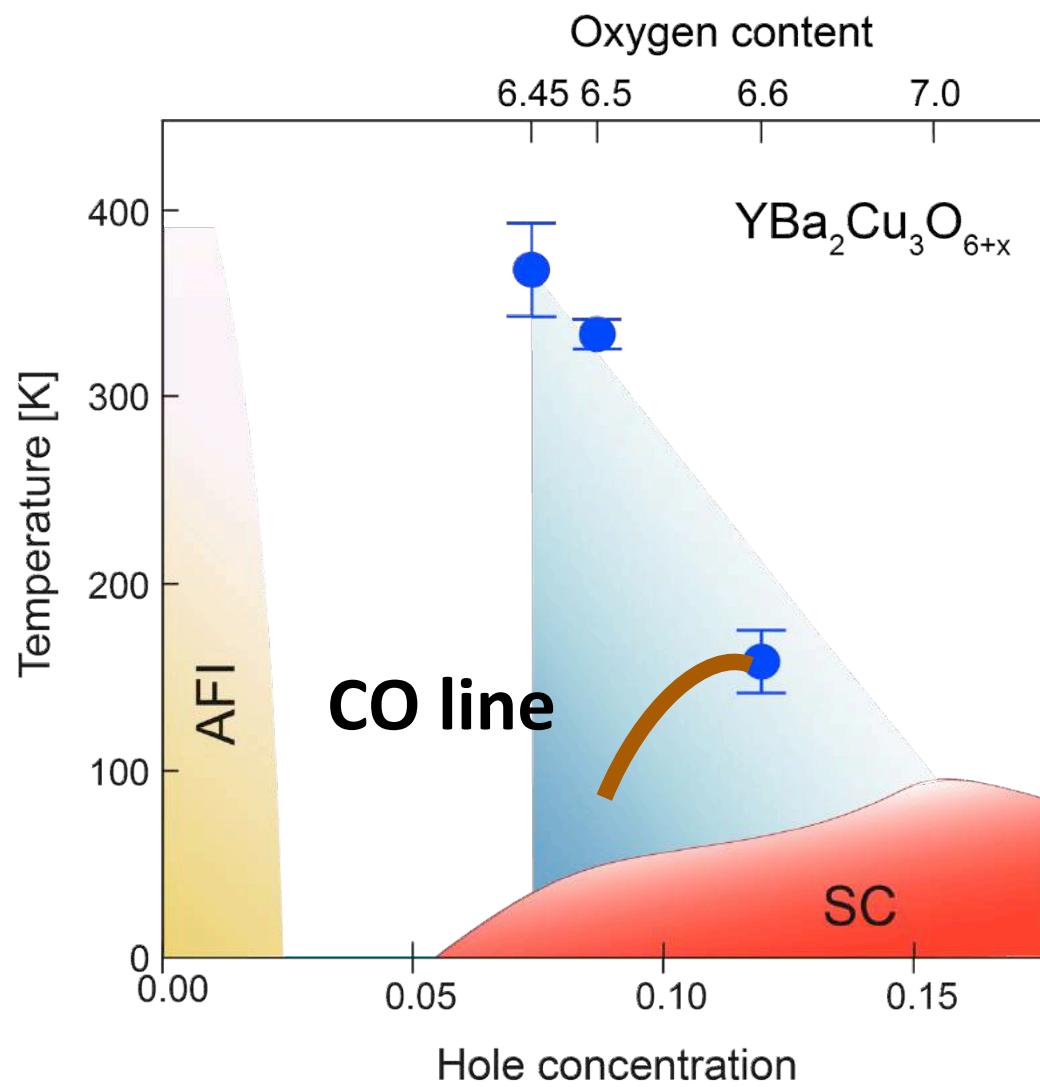
So far everything like in $\text{LESCO}_{1/8}$



Other dopings - Surprise..... Follows T^*



Follows T^*

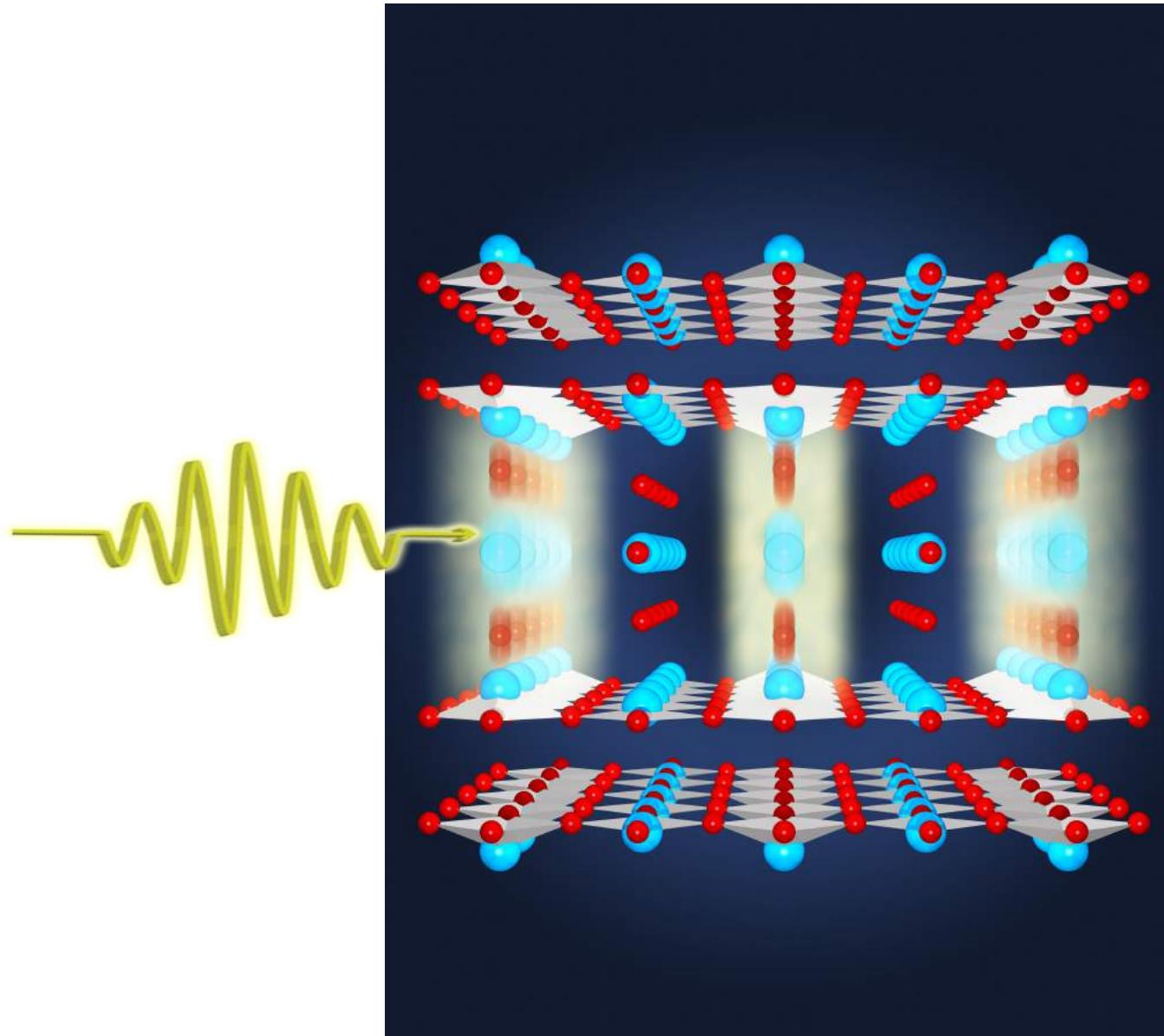


W. Hu. S. Kaiser, D. Nicoletti, C.S. Hunt et al. *Nature Materials* (2014)

S. Kaiser, D. Nicoletti, C. Hunt et al., *Phys. Rev. B* 89, 184516 (2014)



Dynamical modulation: what is going on ?

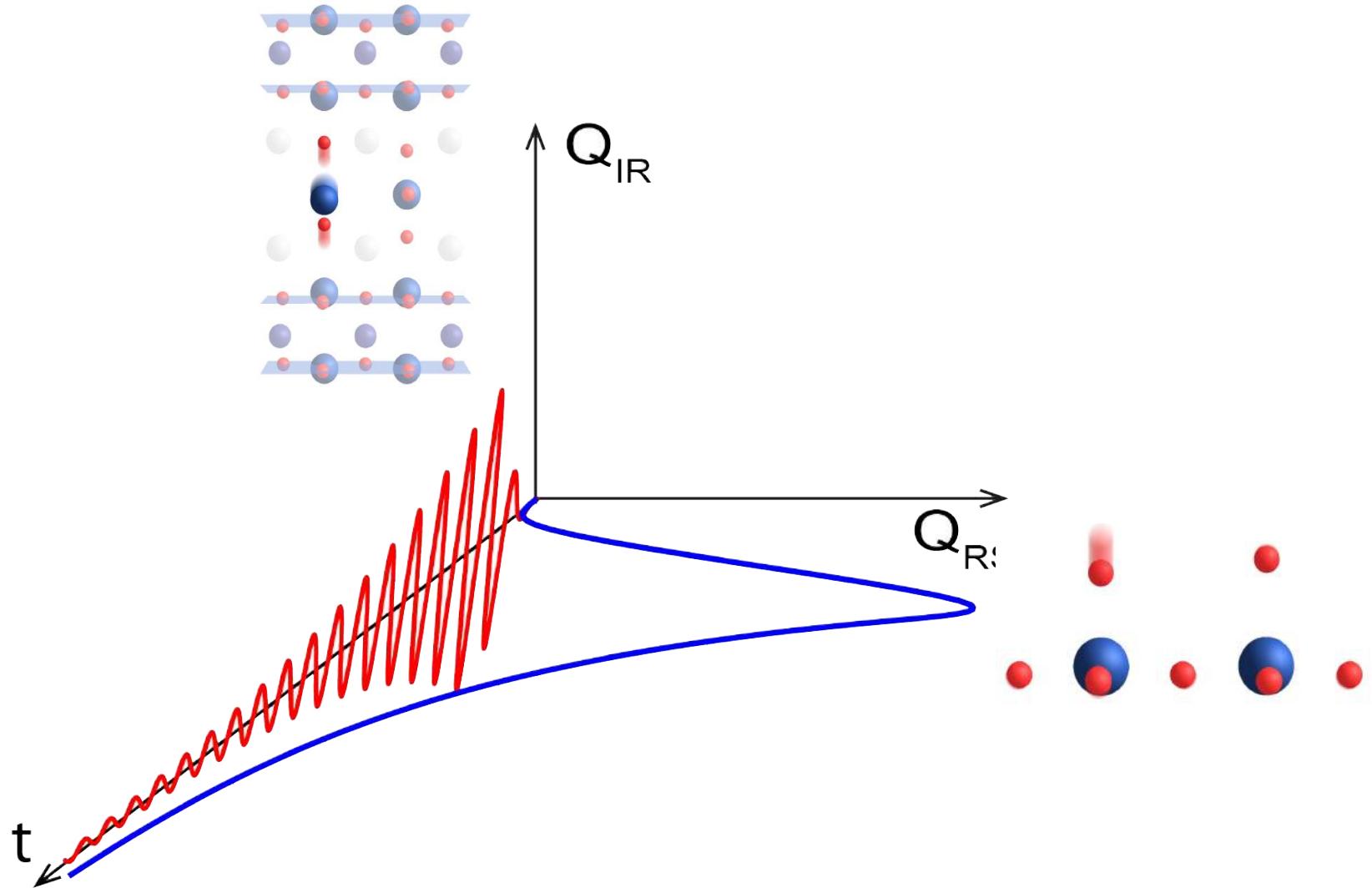


Underdoped YBCO

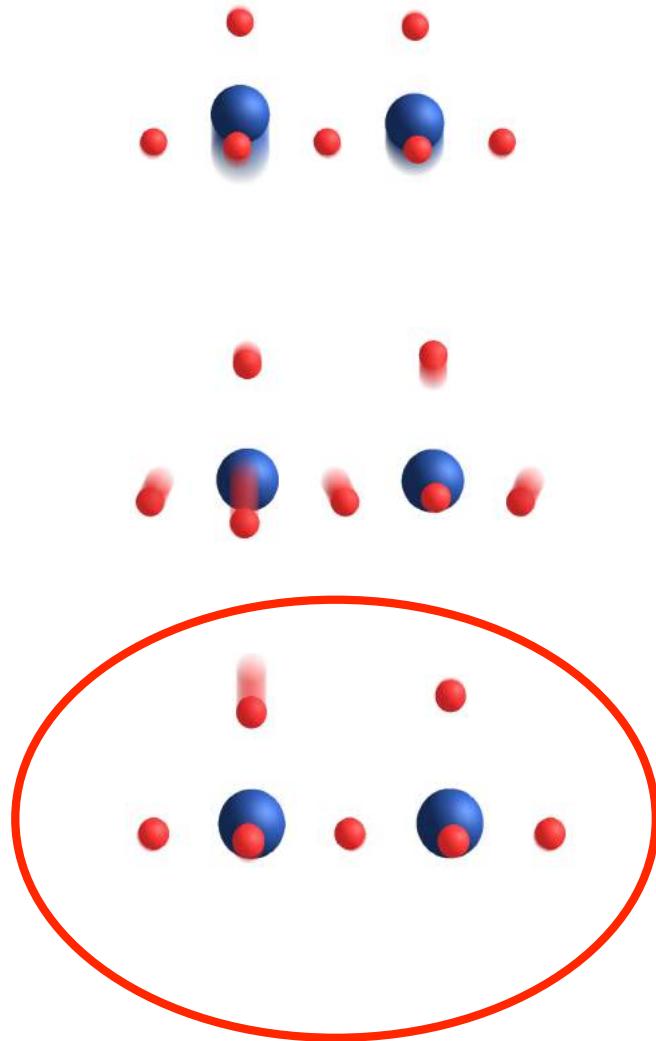
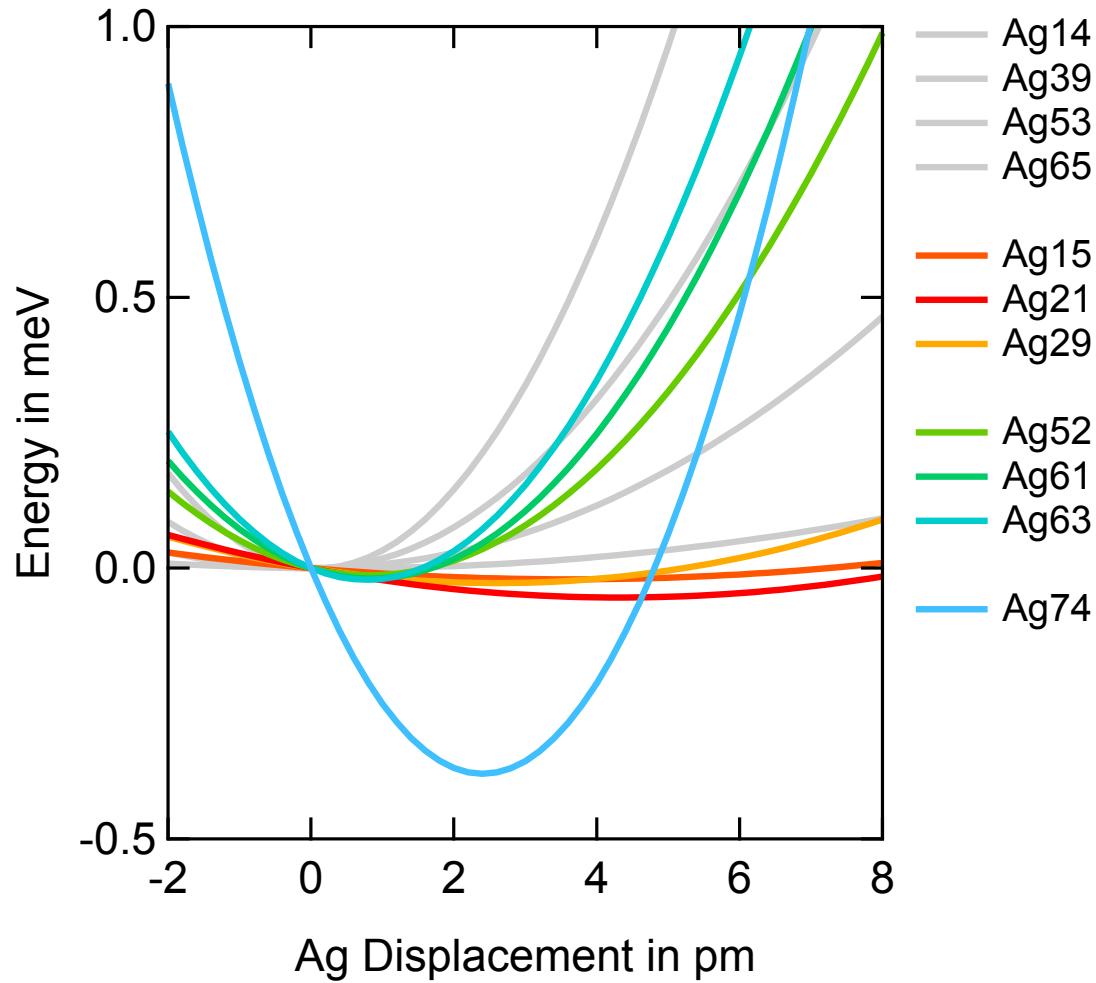
What is the lattice doing ?



Excite B_{1u} and displace along A_g



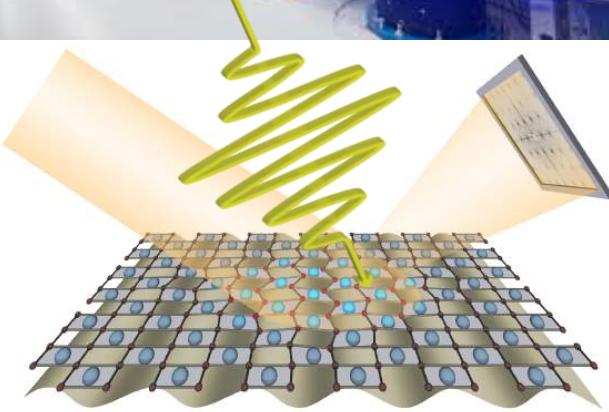
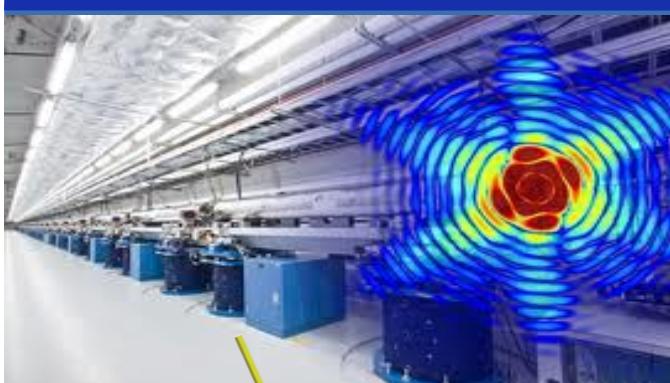
Doped YBCO: 11 A_g Raman modes



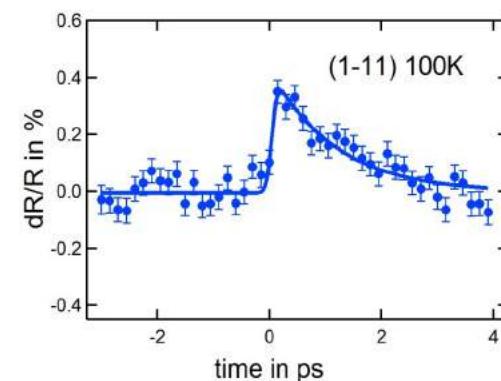
Only three Ag modes are coupled strongly with B1u



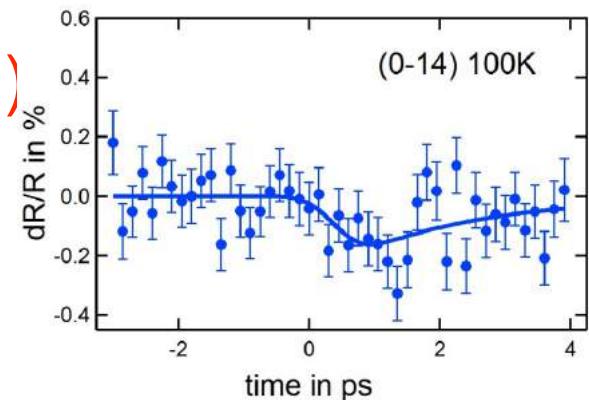
Femtosecond X-ray Scattering



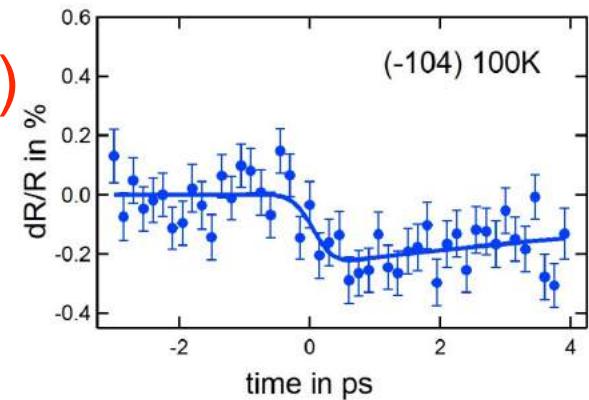
(-1,1,1)



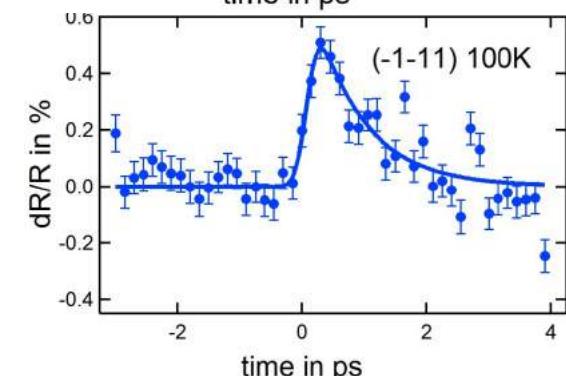
(0,-1,4)



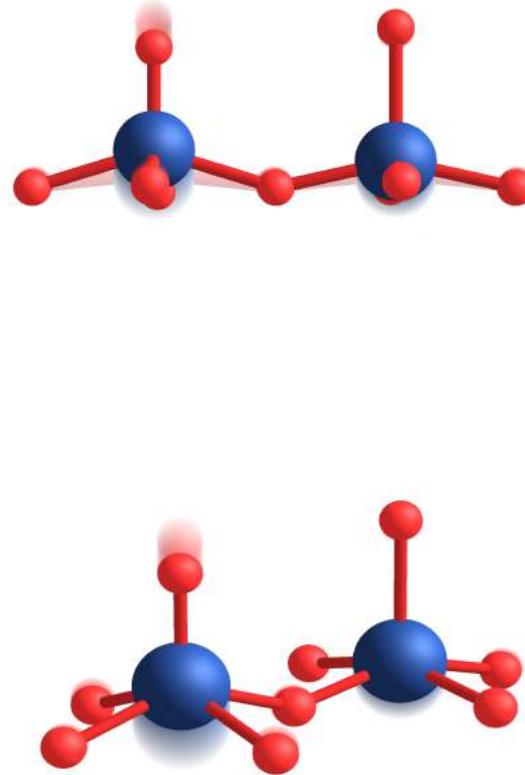
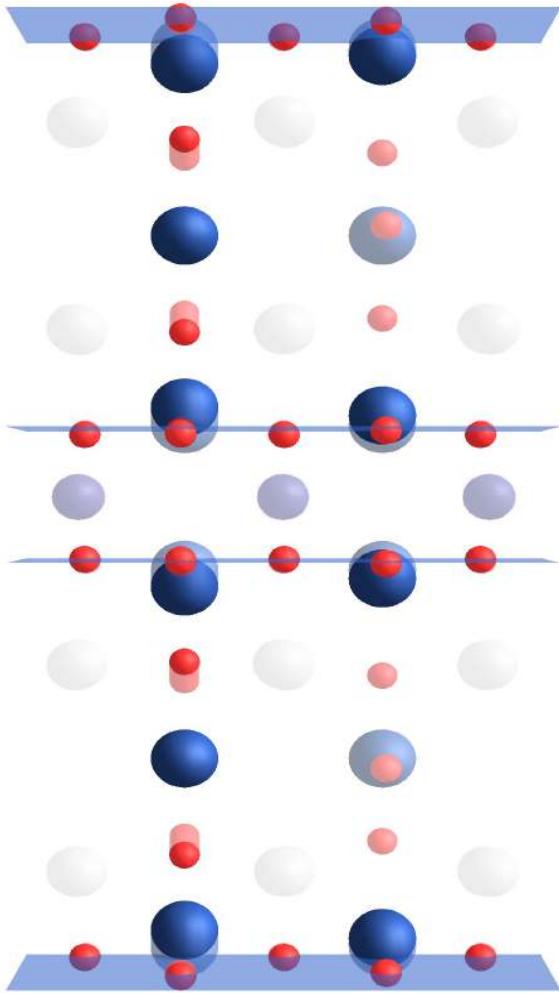
(-1,0,4)



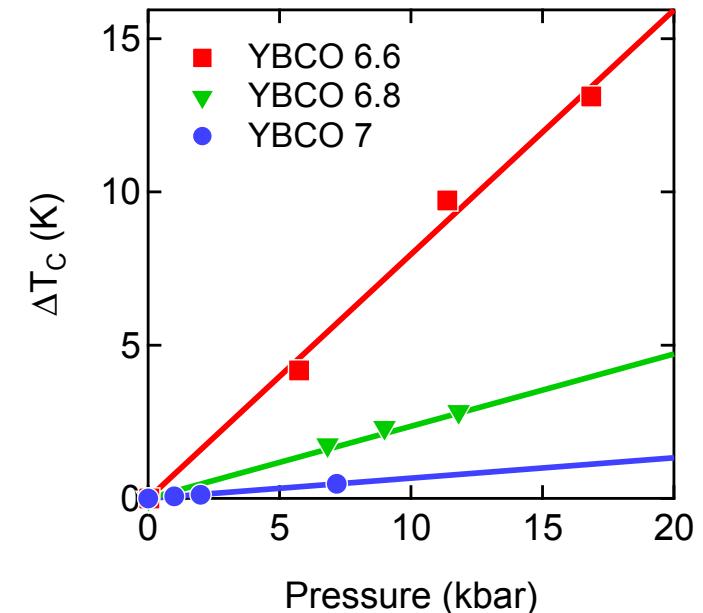
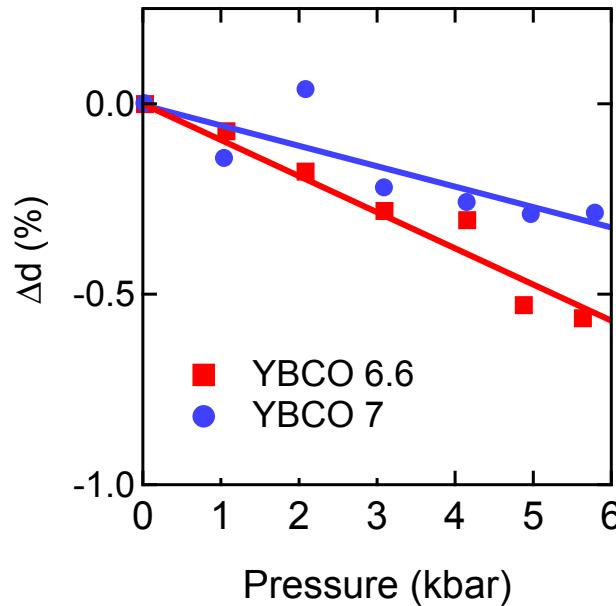
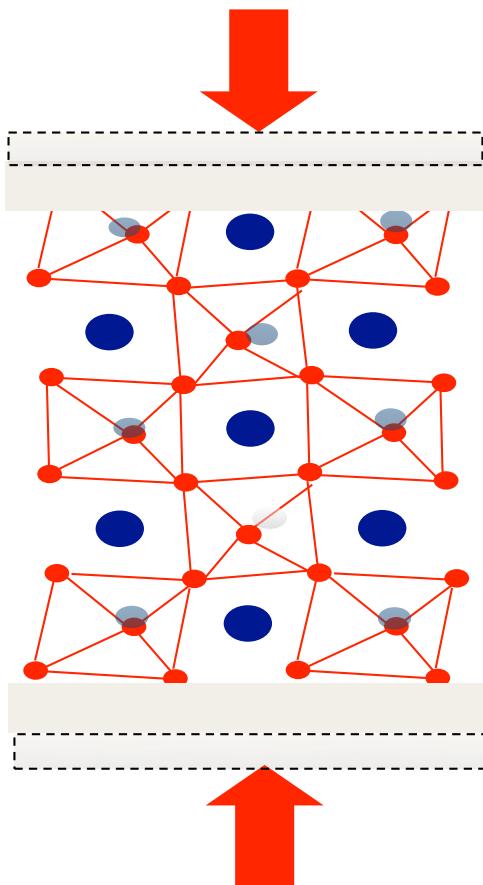
(1,-1,1)



A new, transient crystal structure



Same distortions observed under pressure



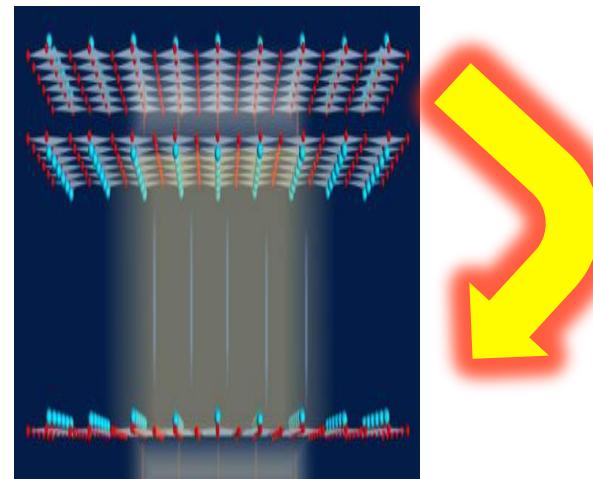
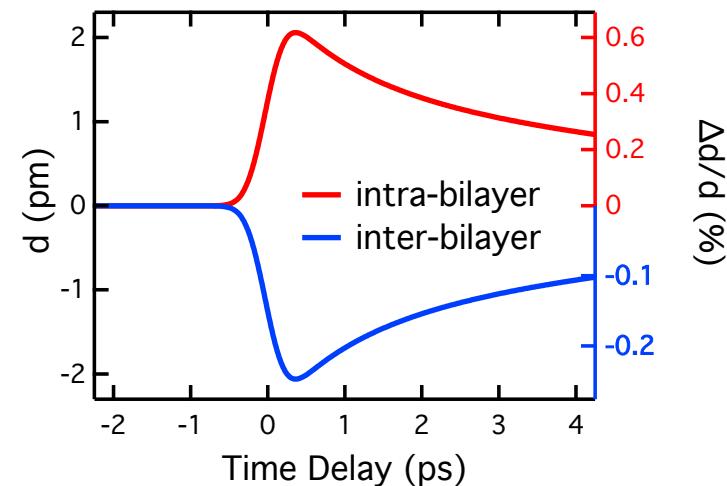
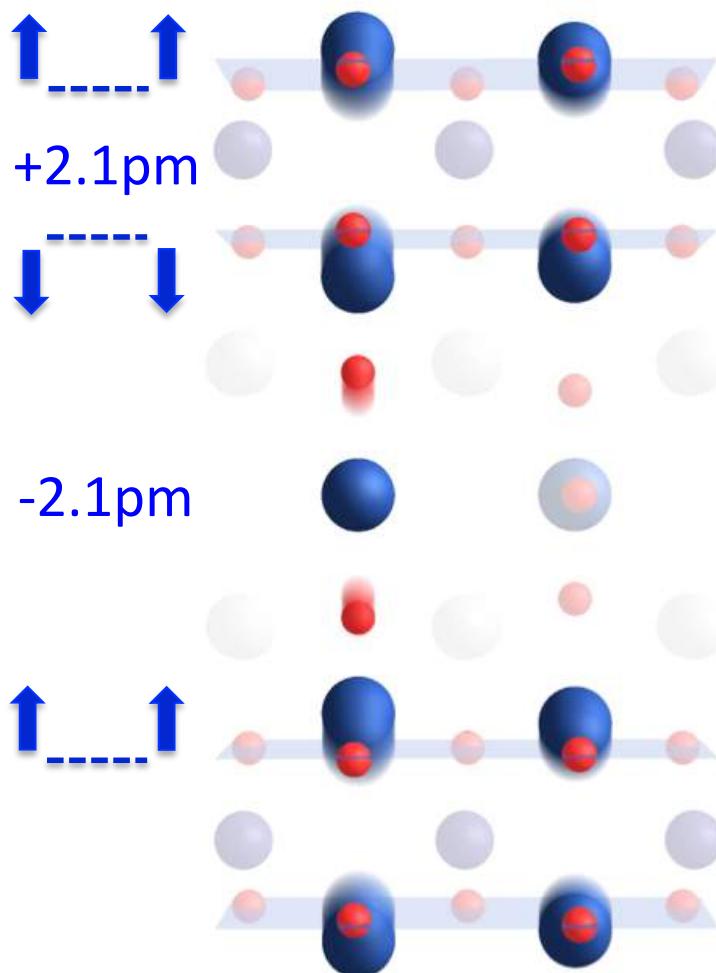
Pressure
 $d \sim 0.5 \%$
 $\Delta T_c \sim 5 - 15 \text{ K}$

- J. G. Huber et al. *Phys. Rev. B* 41, 8757 (1990)
L. E. Schirber et al. *Phys. Rev. B* 35, 8709 (1987)
B. Bucher et al. *Journal of Less-Common Metals* 164, 165, 20 (1990)
J. Jorgensen et al. *Physica C* 171, 93 (1990)

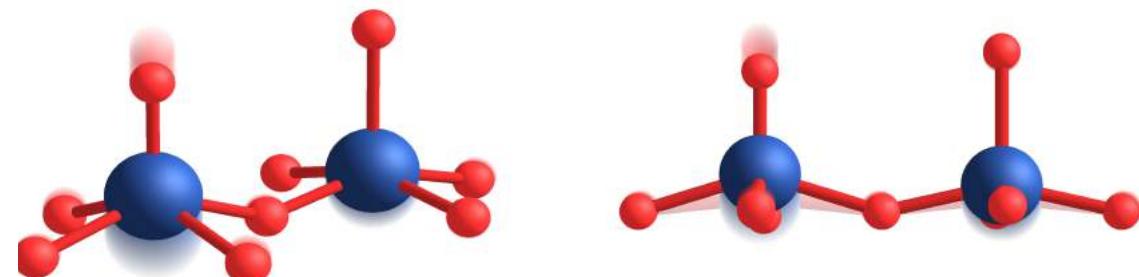
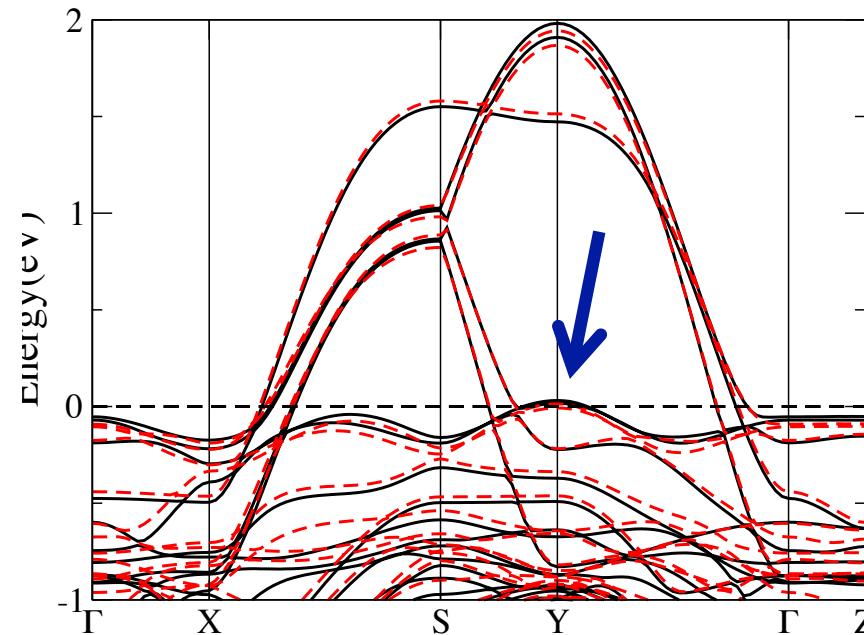
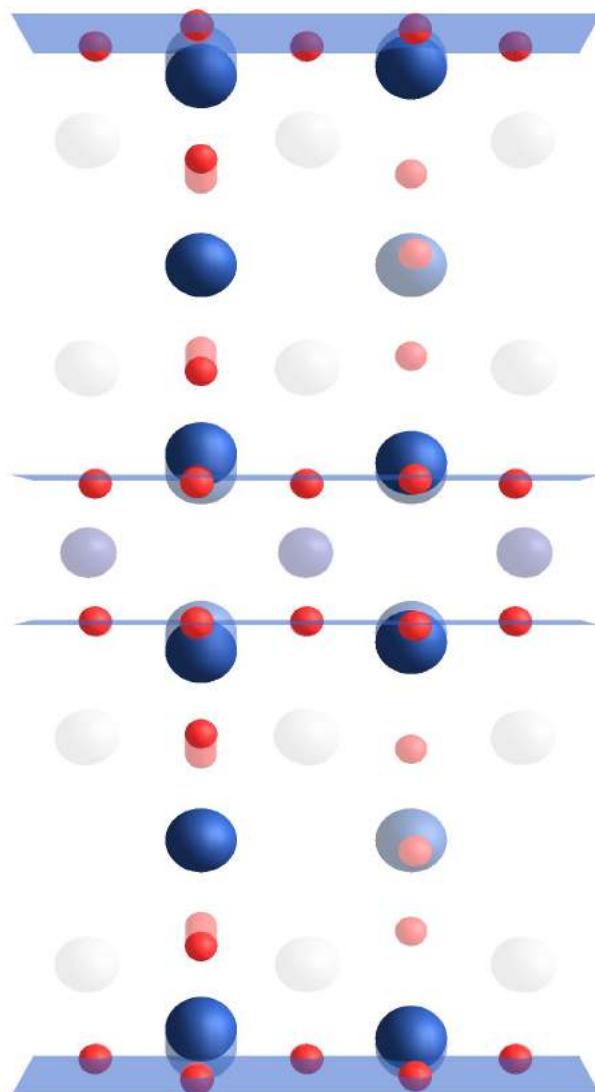
Phononics
 $d \sim 3 \%$



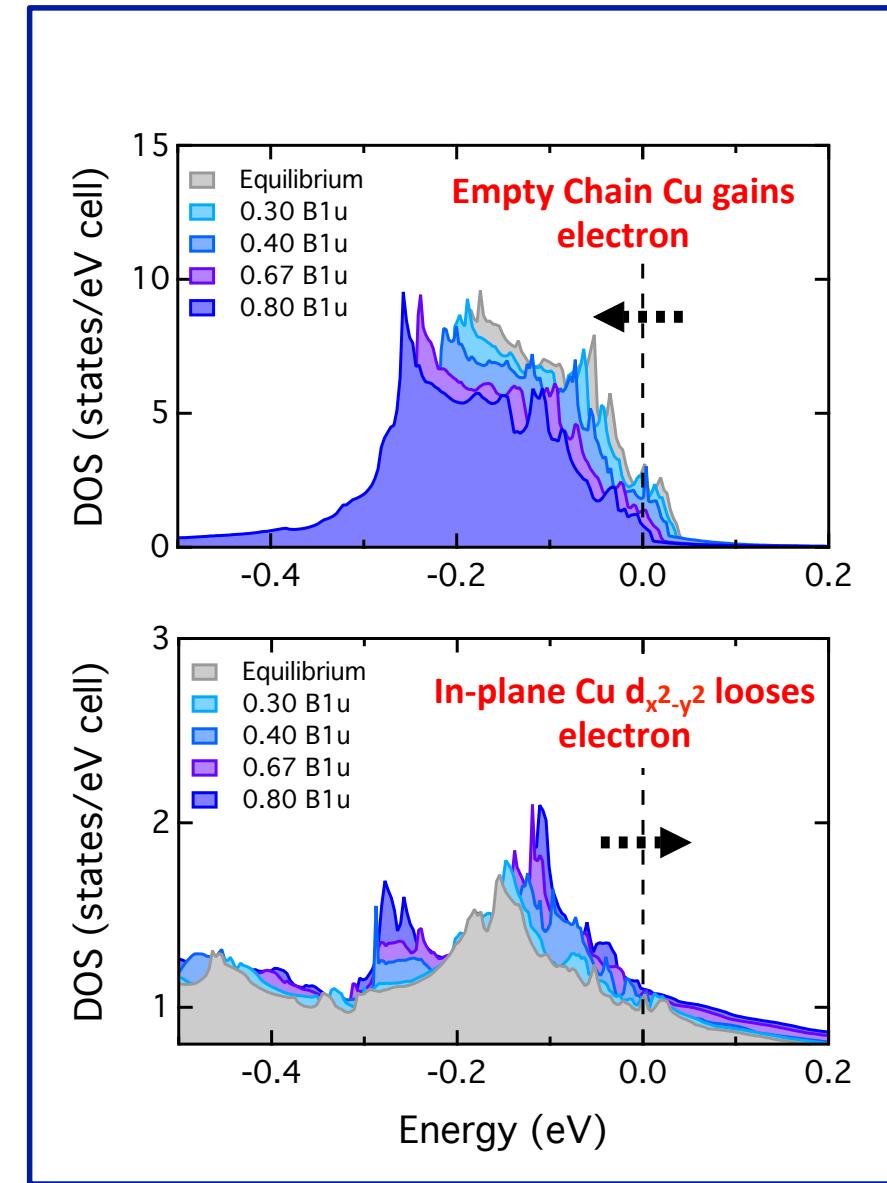
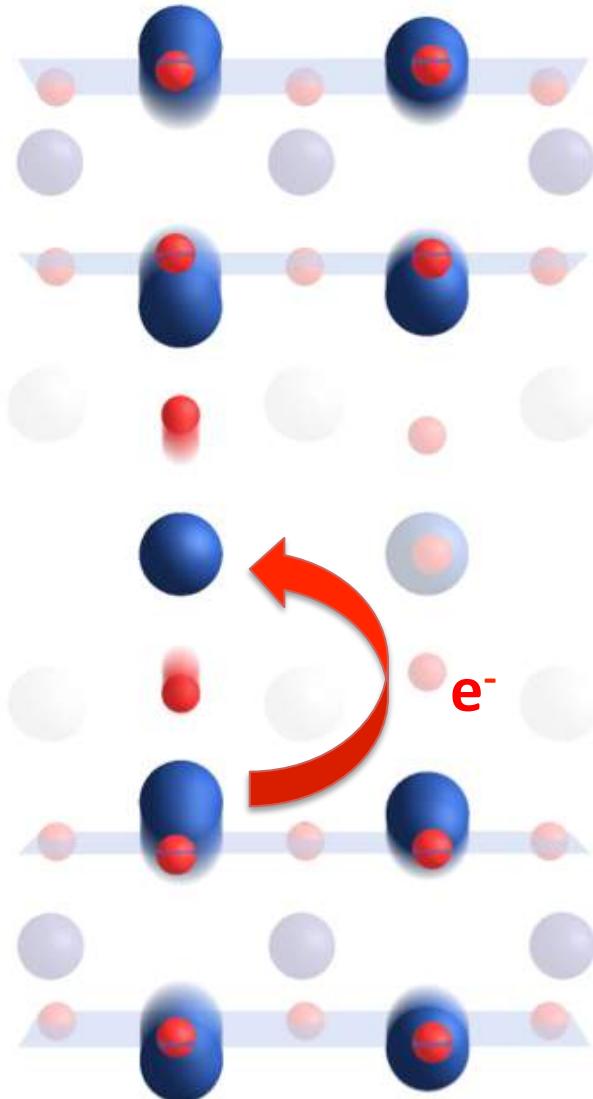
1) Staggered motion of the planes



2) Empty chain band moves down in energy

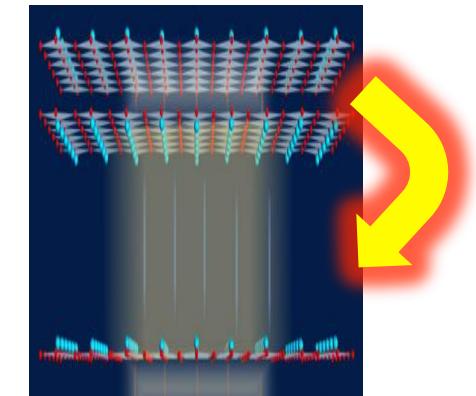


3) Charge transfer from the planes to the chains

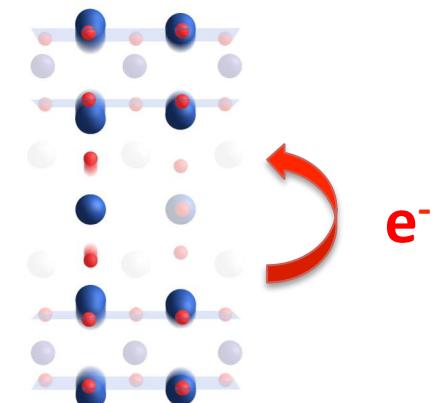


Summary: three good things

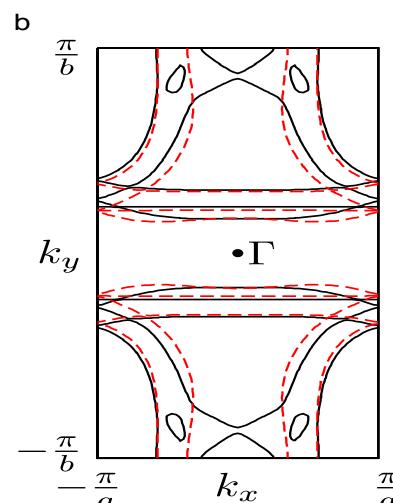
1) Staggered motion of the layers



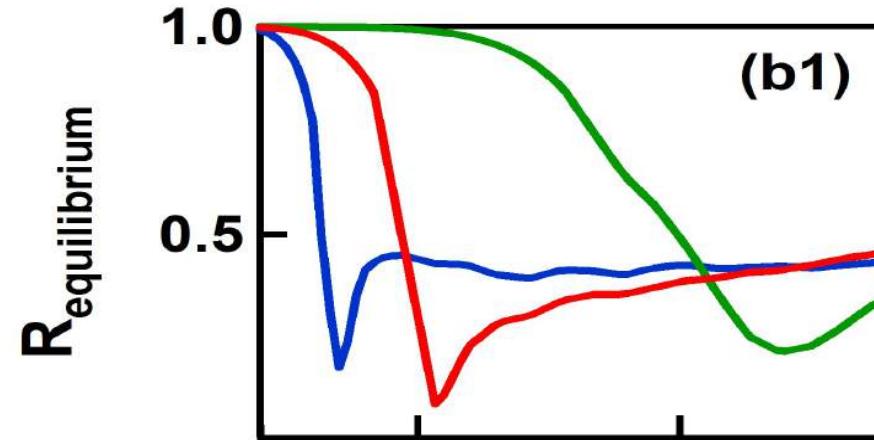
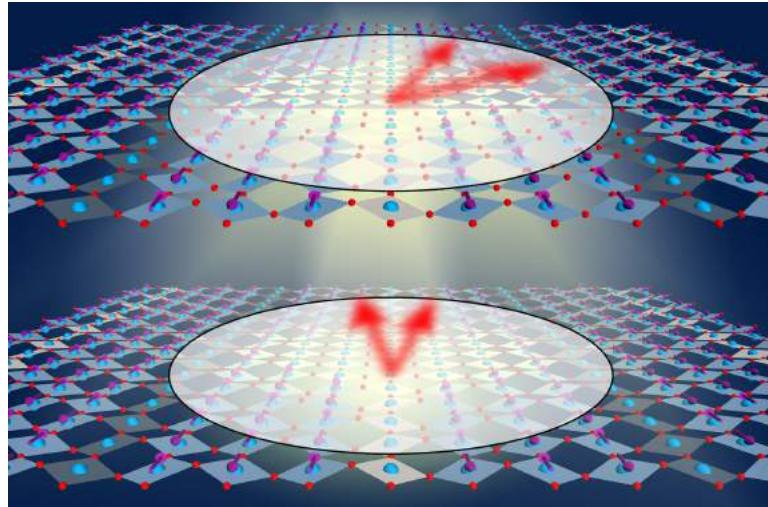
2) Charge transfer from to chains



3) dx^2-y^2 Fermi surface



Let's think about YBCO again: below T_c

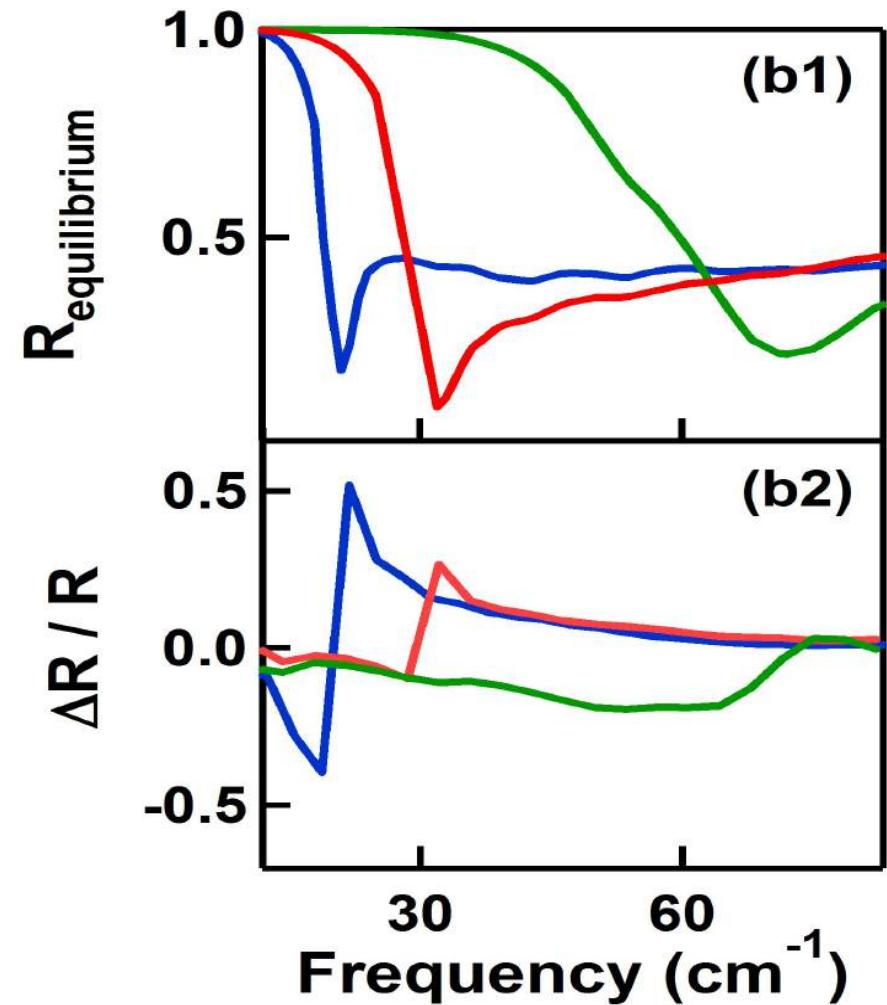
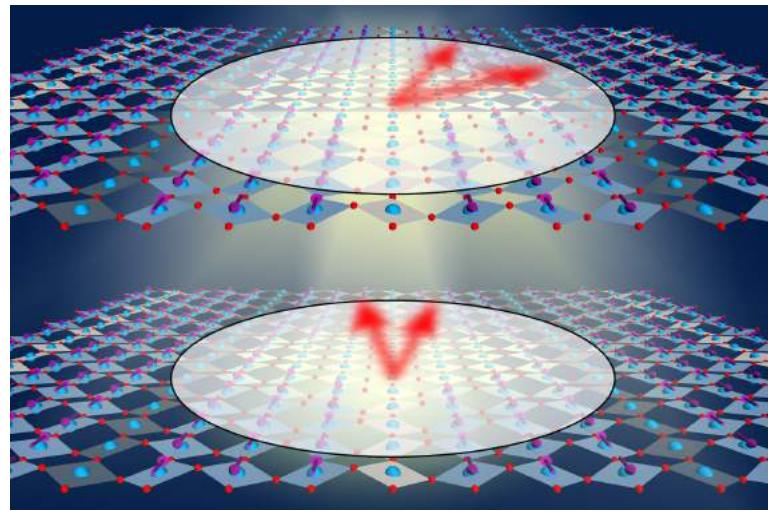


W. Hu et al. *Nature Materials* 13, 705 (2014)

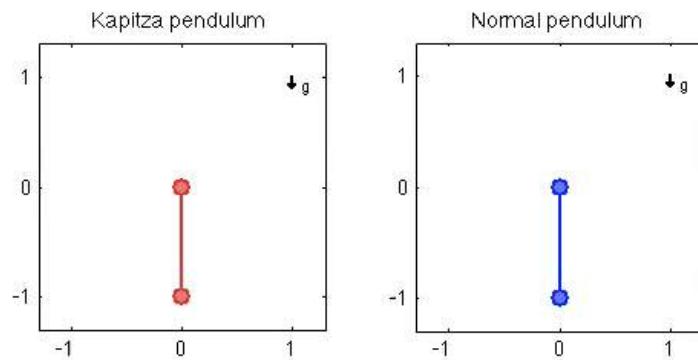
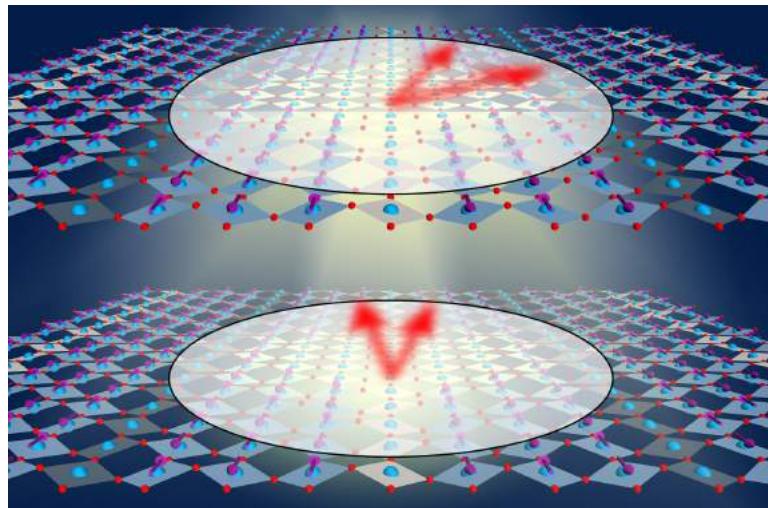
S. Kaiser, et al., *Phys. Rev. B* 89, 184516 (2014)



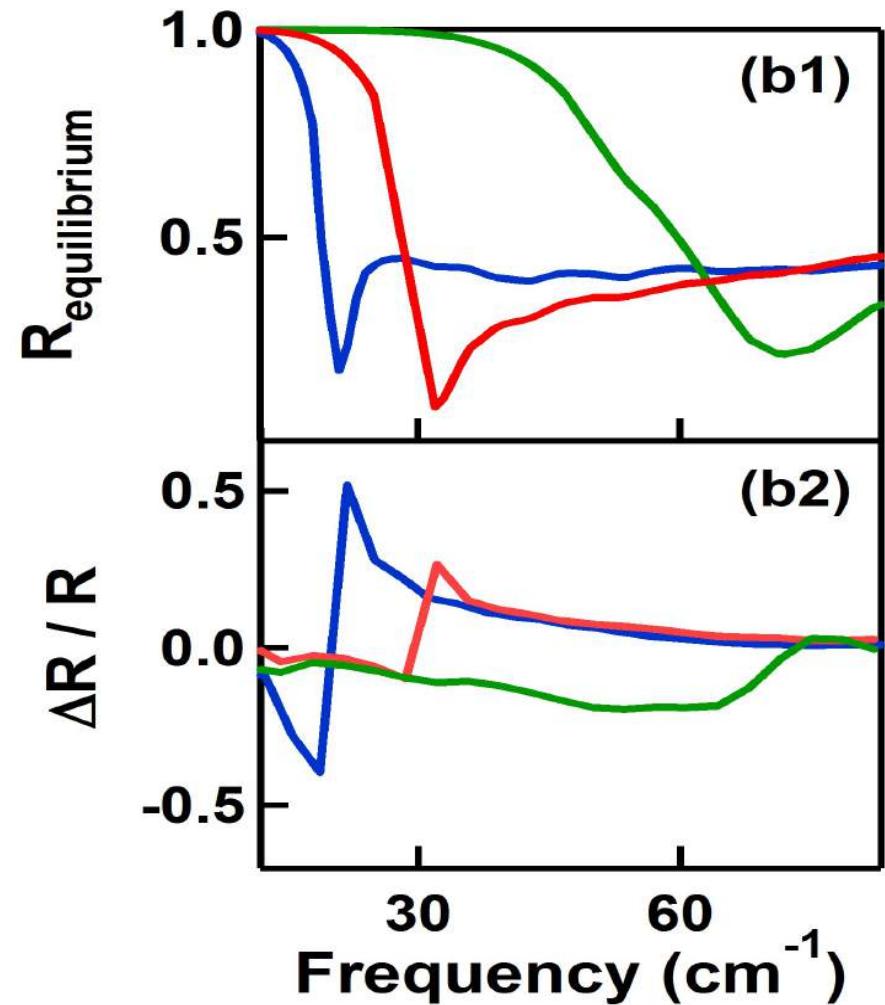
Let's think about YBCO again: below T_c



Let's think about YBCO again: below T_c

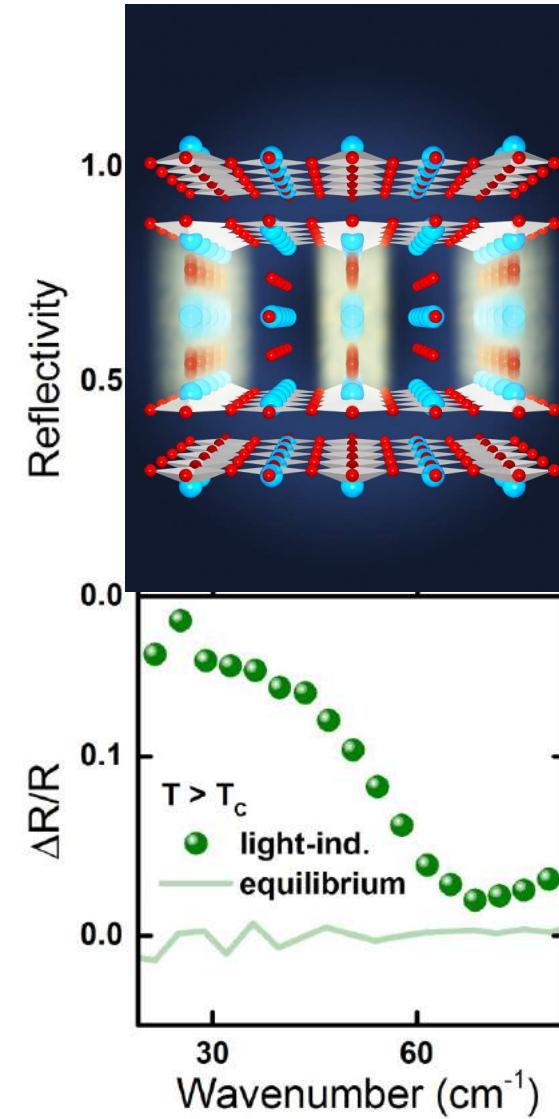
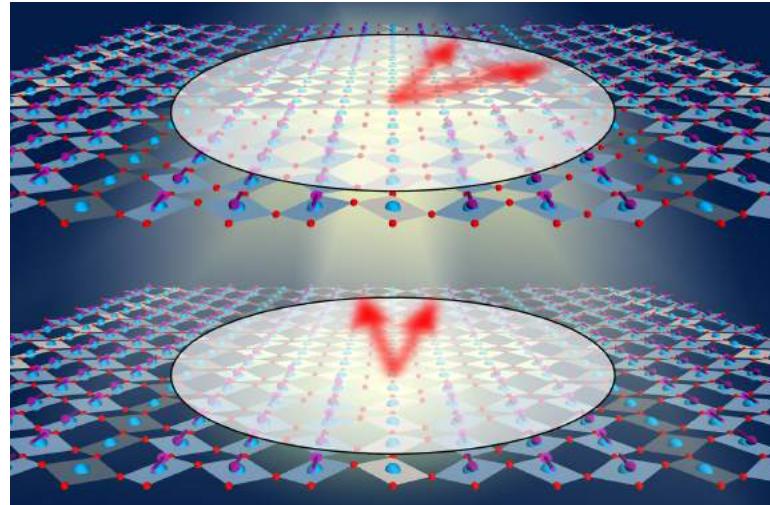


P.L. Kapitza, "Dynamic stability of a pendulum with an oscillating point of suspension,"
Zh. Eksp. Teor. Fiz. 21, 588 (1951)

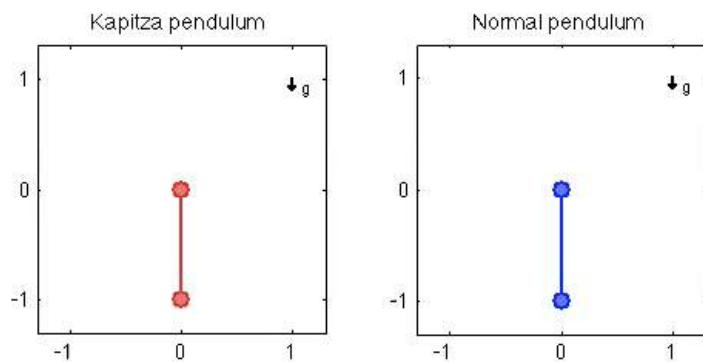
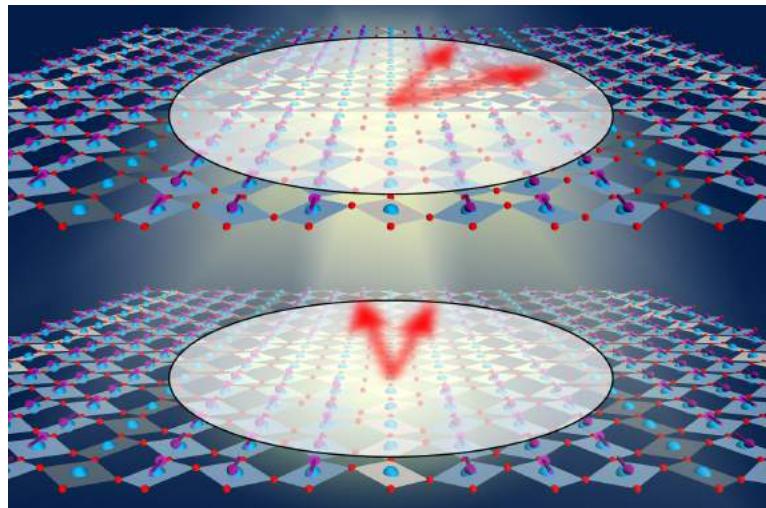


S. Kaiser, et al., *Phys. Rev. B* 89, 184516 (2014)

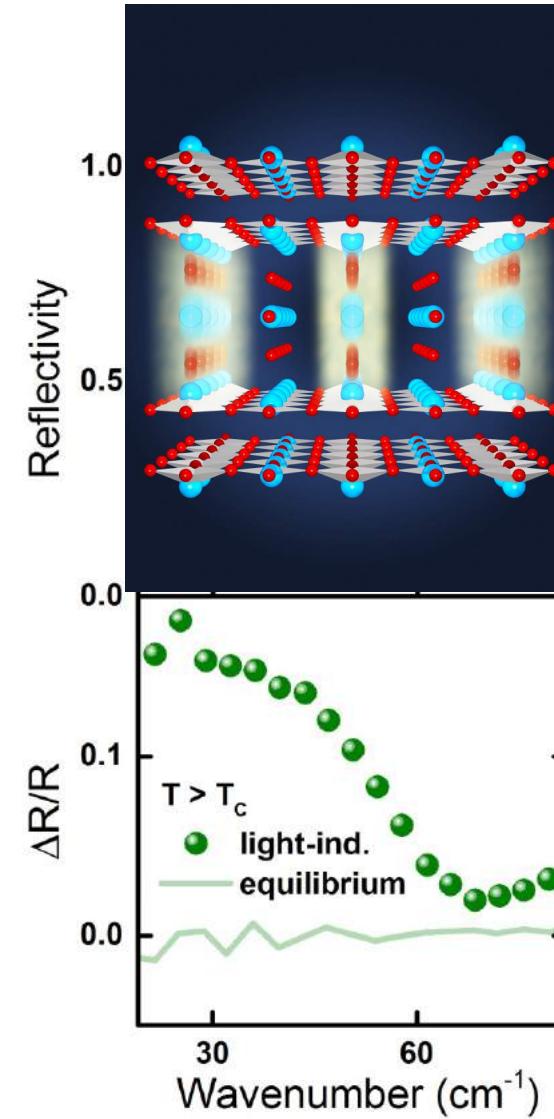
Let's think about YBCO again: above T_c



Let's think about YBCO again: above T_c



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

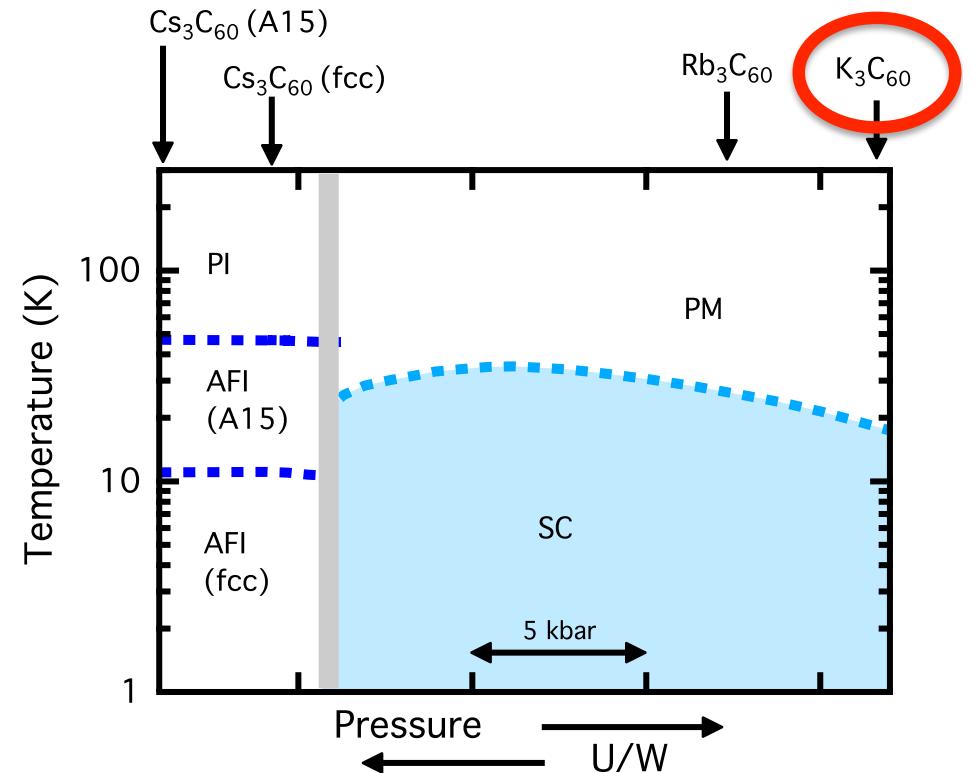
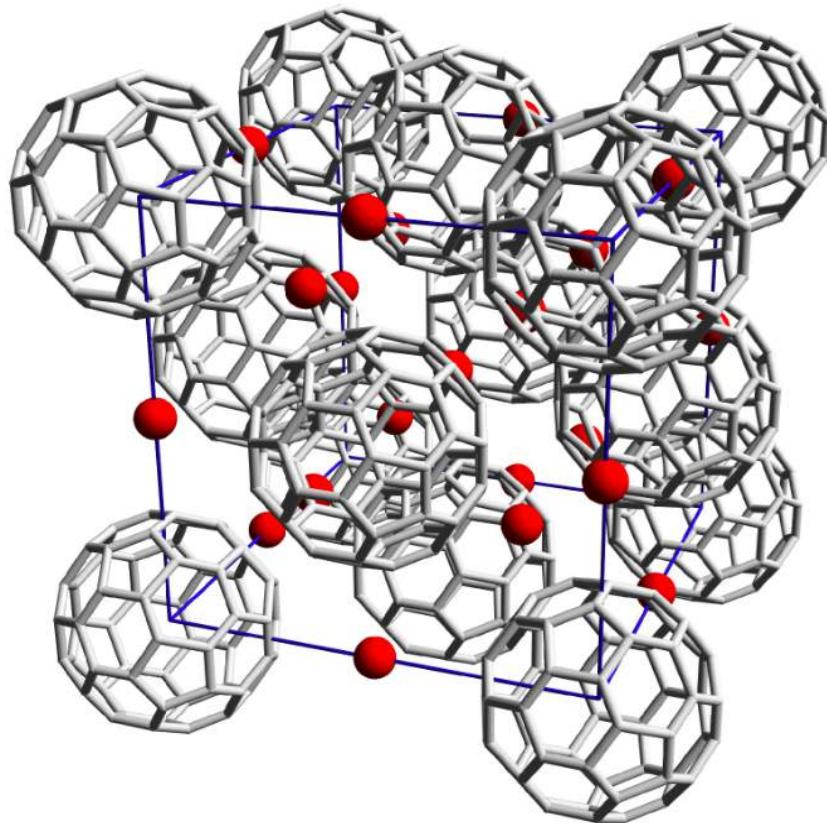


Light enhanced superconductivity

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



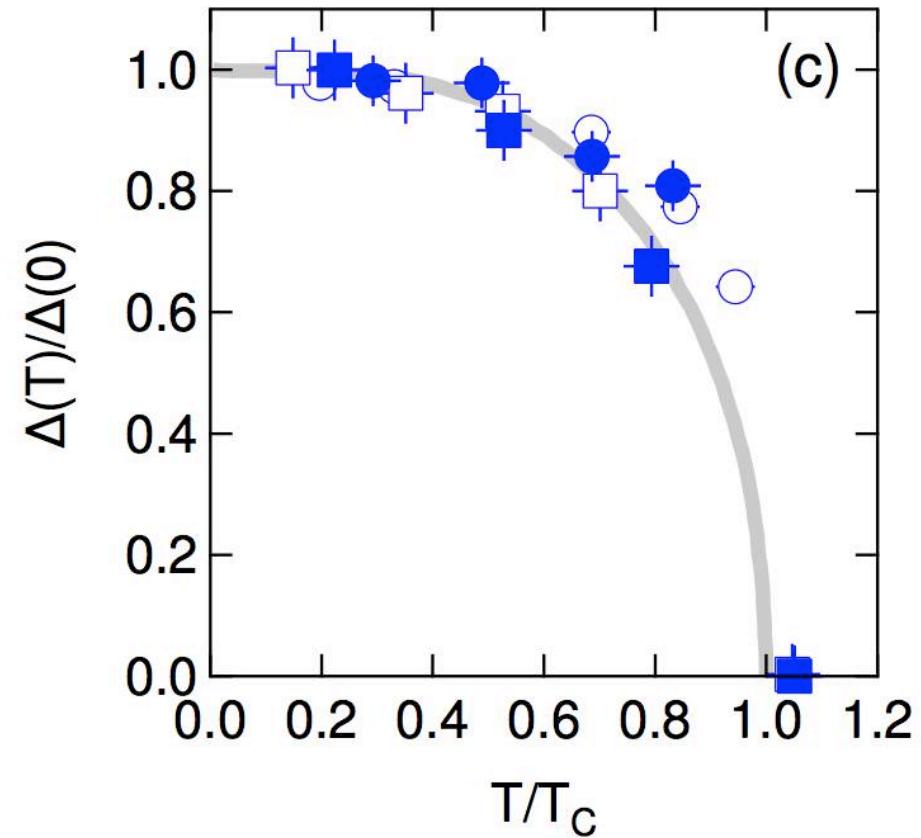
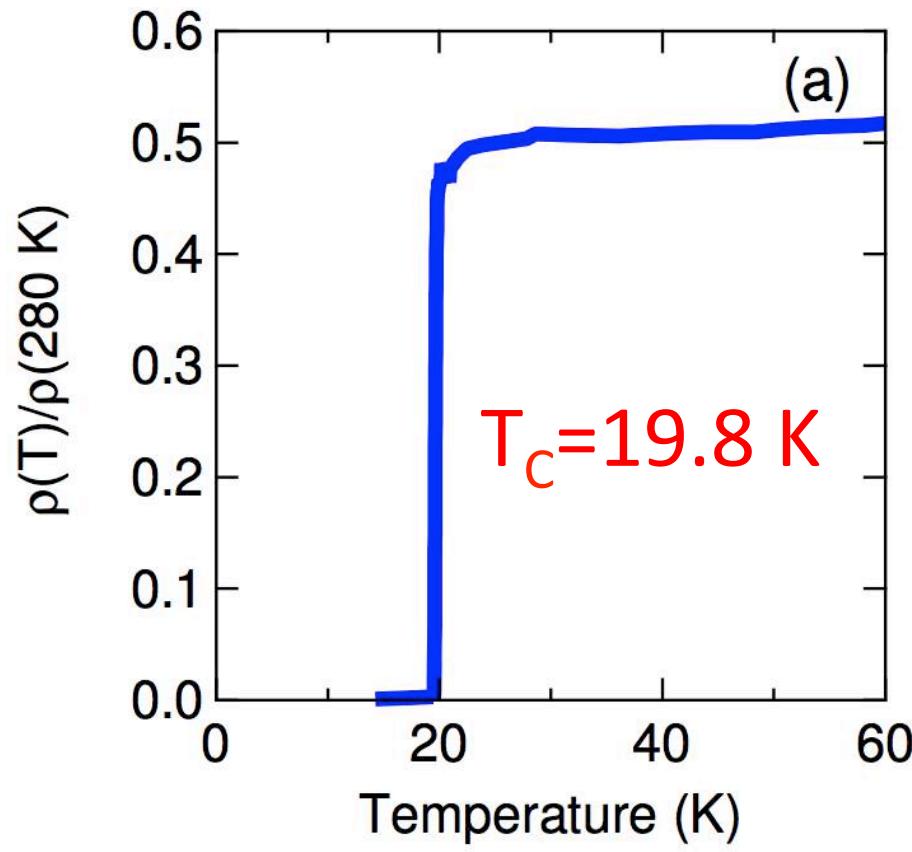
K_3C_{60} : a 20 K superconductor



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



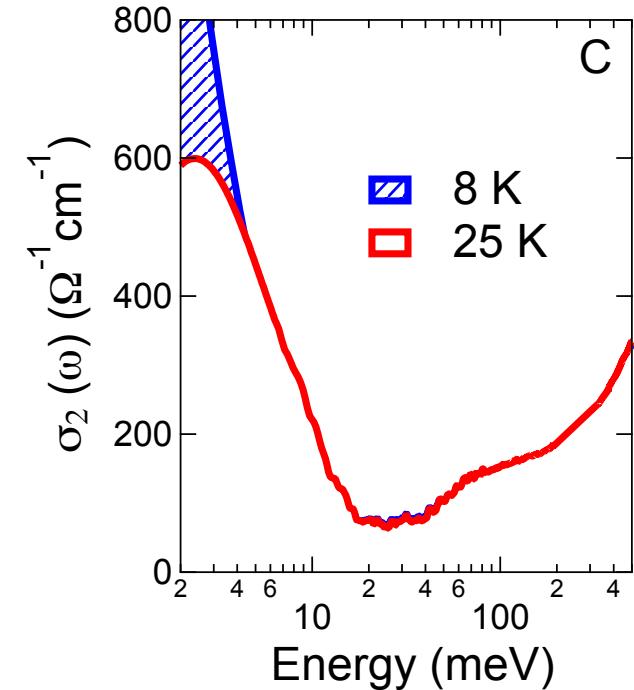
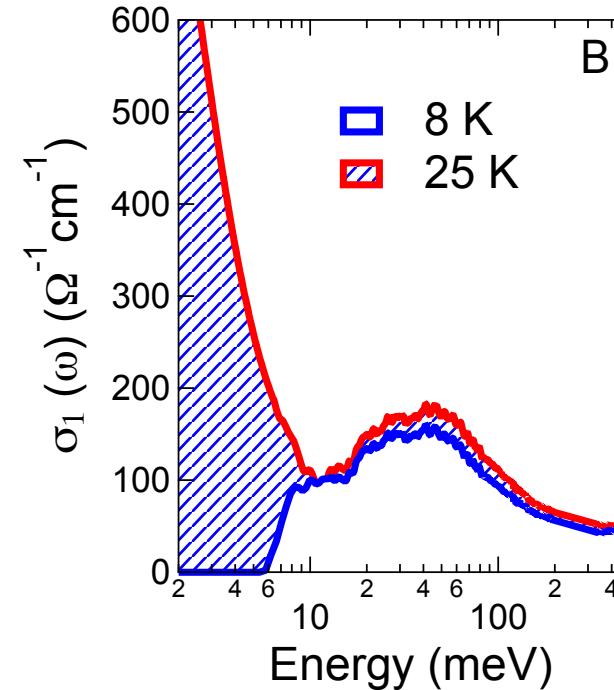
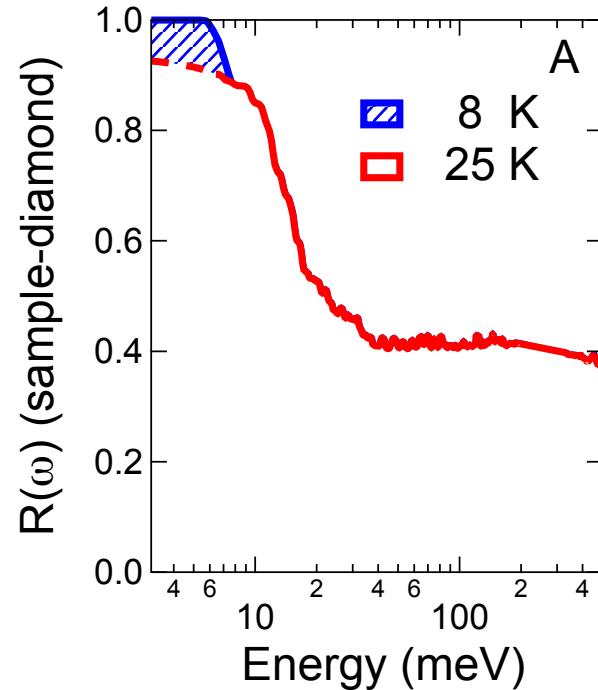
Equilibrium Superconductivity in K_3C_{60}



From literature data, MM PhD thesis



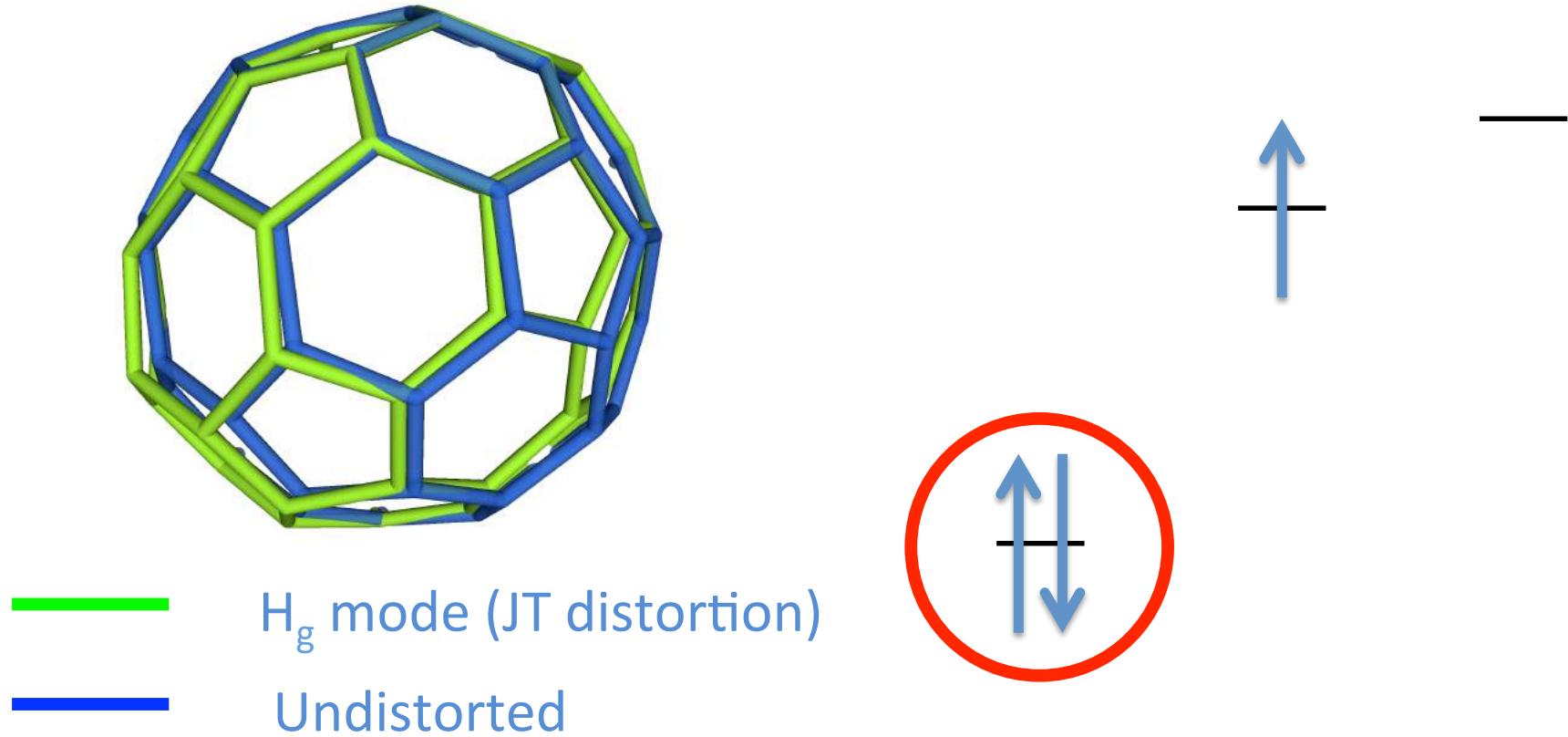
Equilibrium Superconducting Transition



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



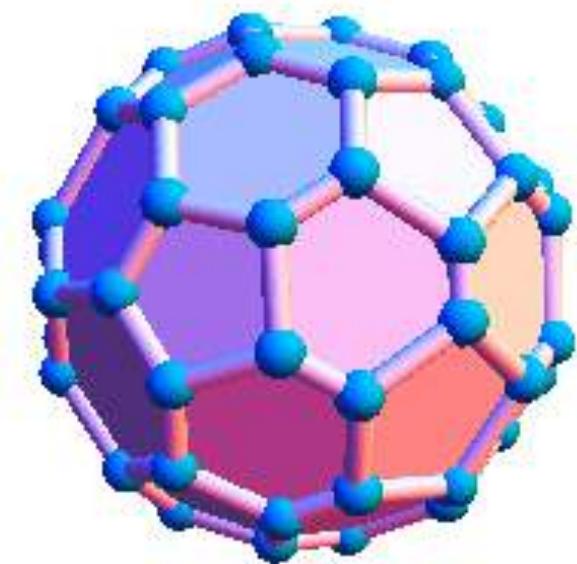
Pairing Interaction in K_3C_{60}



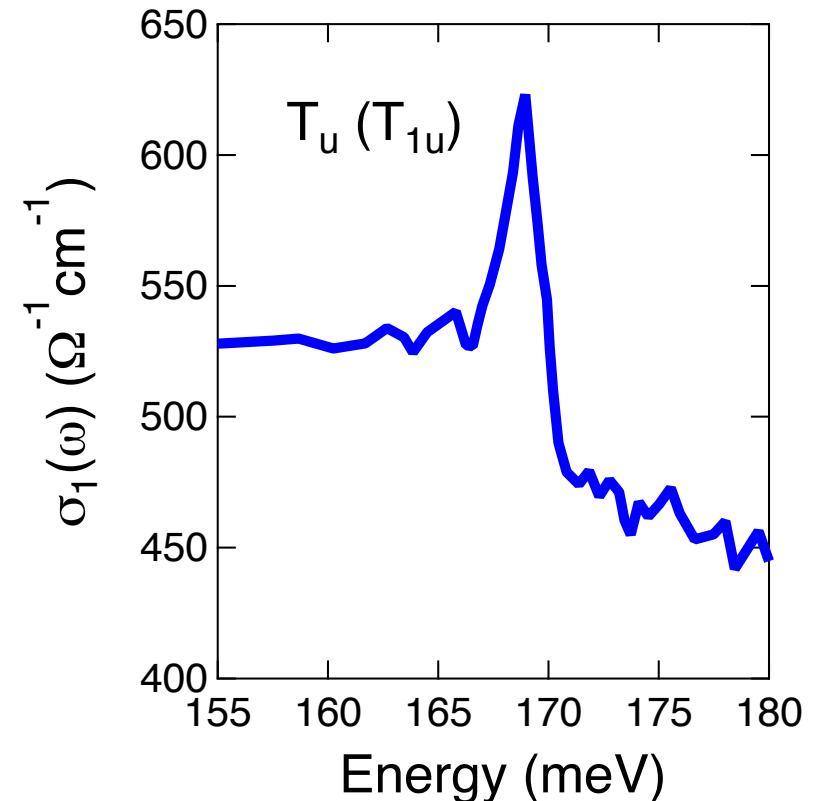
“On ball” vibrations plus correlations favor local pairing

Vibrational pump

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



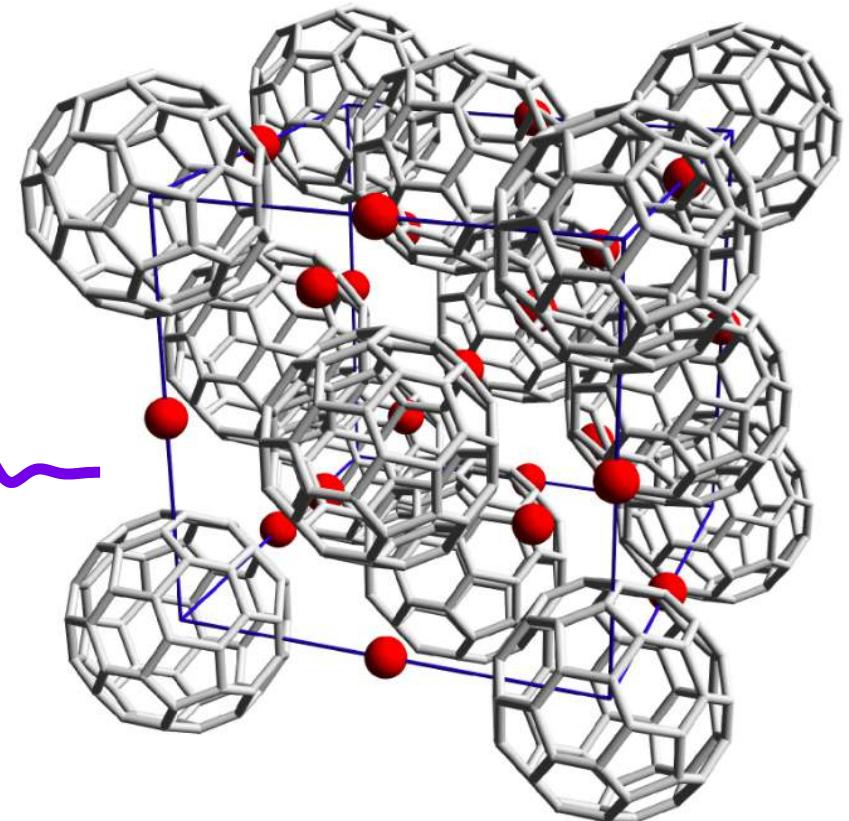
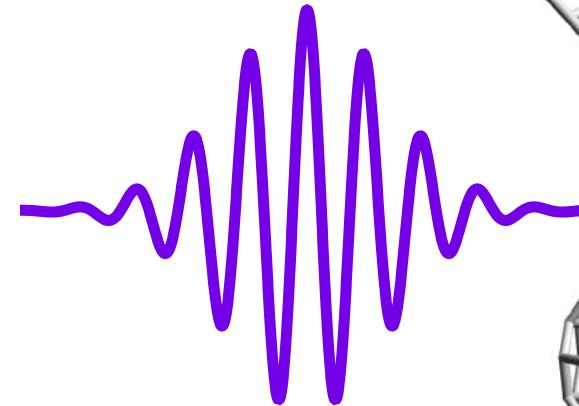
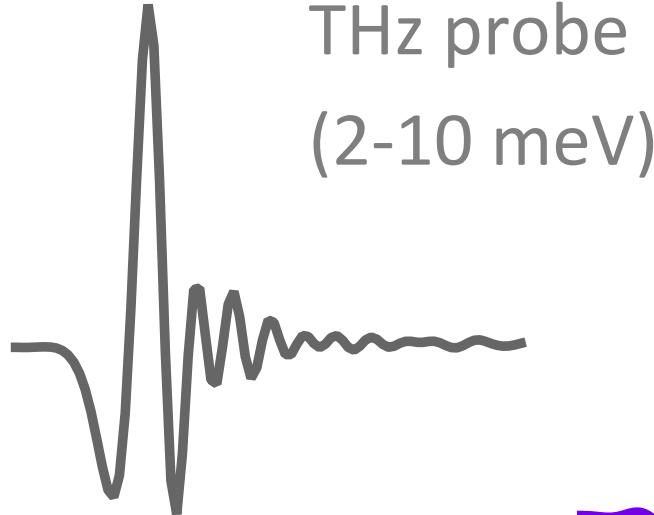
MIR pump 170 meV (7.3 μm)



Iwasa et al. PRB 51, 3678 (1995)



Vibrational pump THz probe in K_3C_{60}

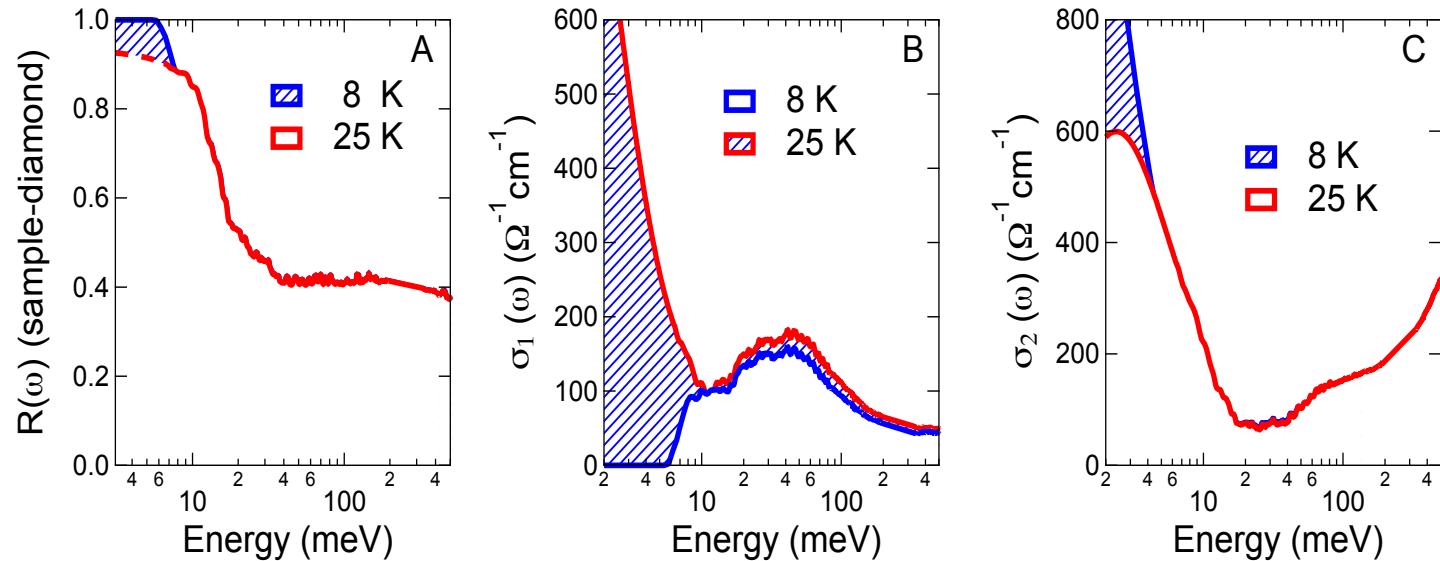


MIR pump 170 meV (7.3 μ m)

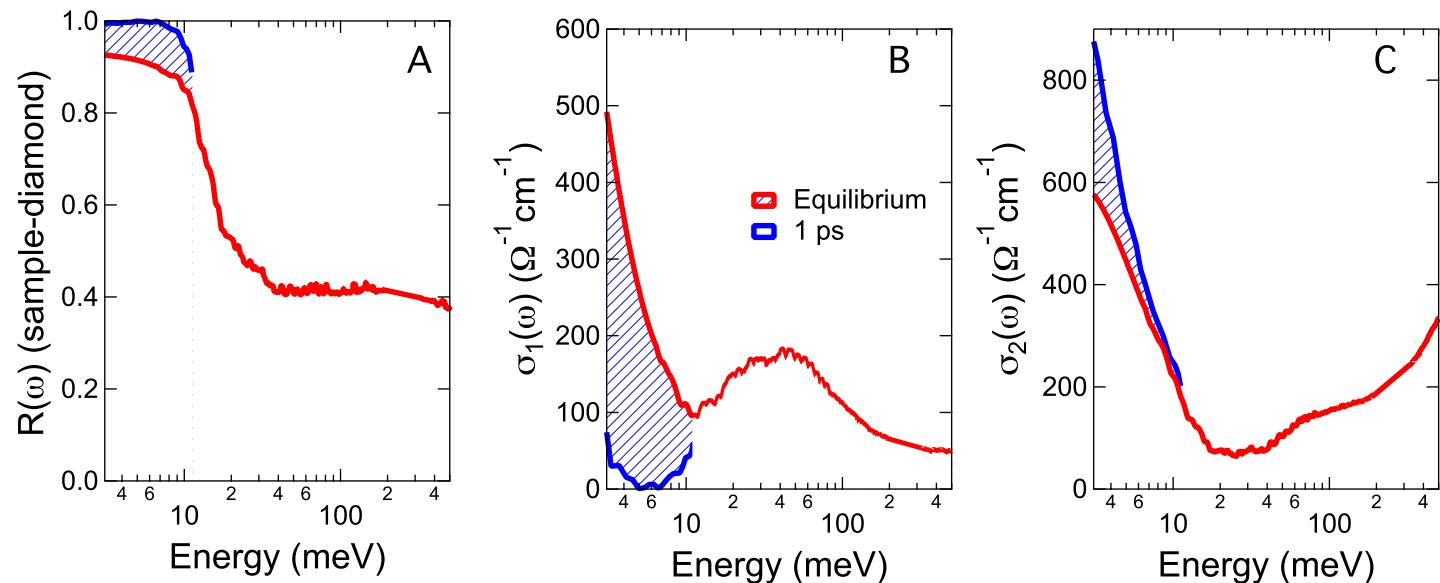


Striking similarity with the low temperature SC

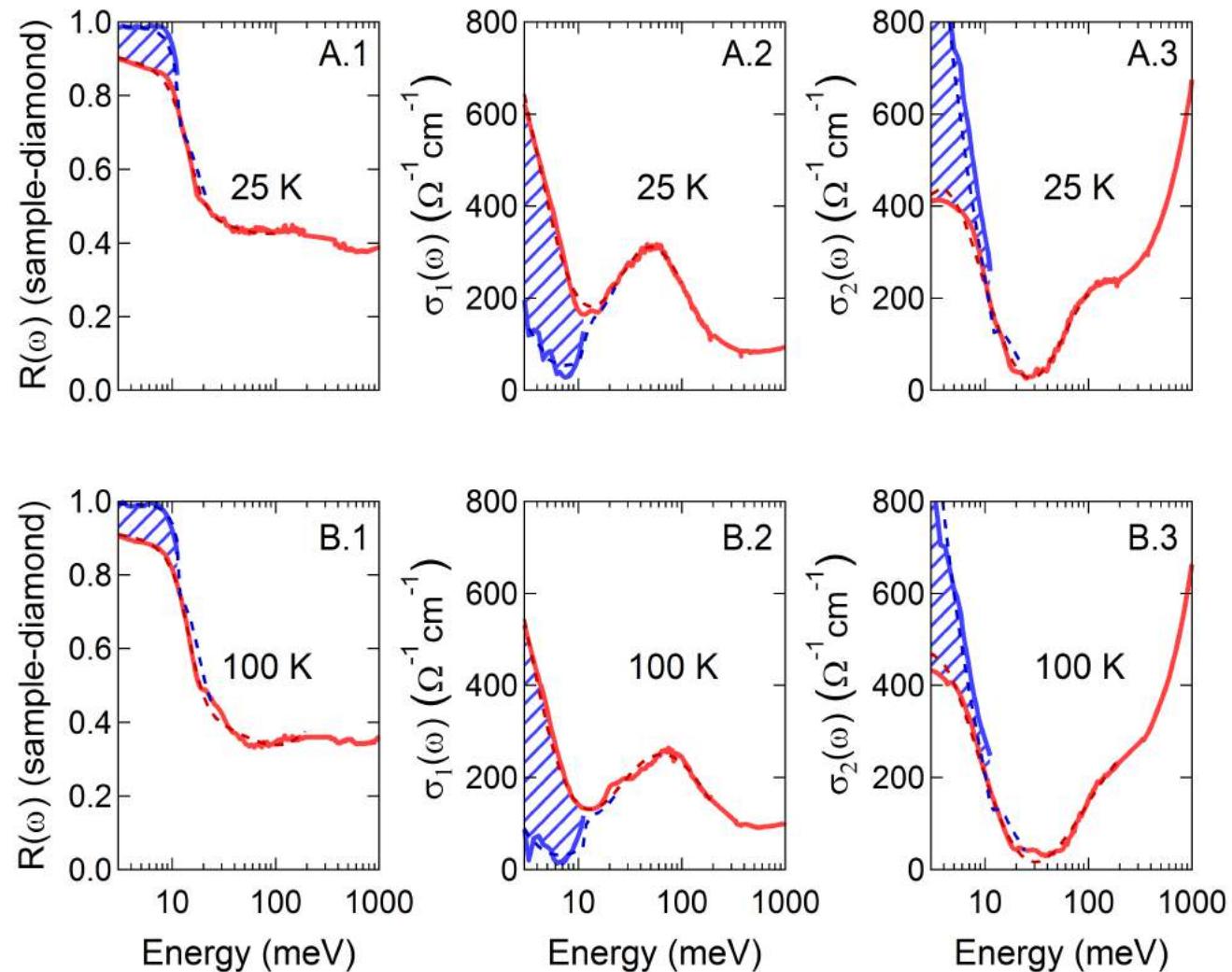
Cooling



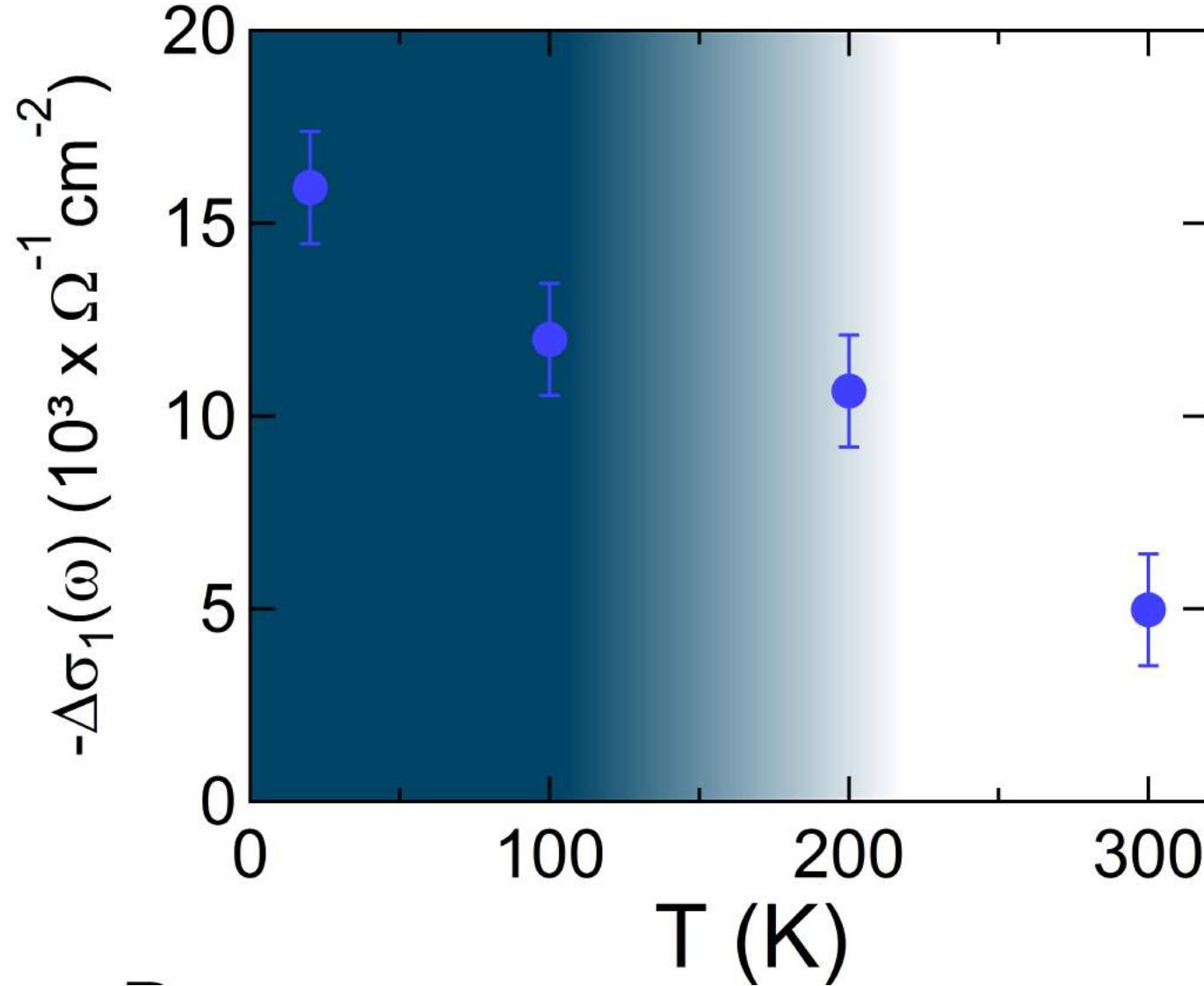
Light-induced
 $T=25\text{ K}$



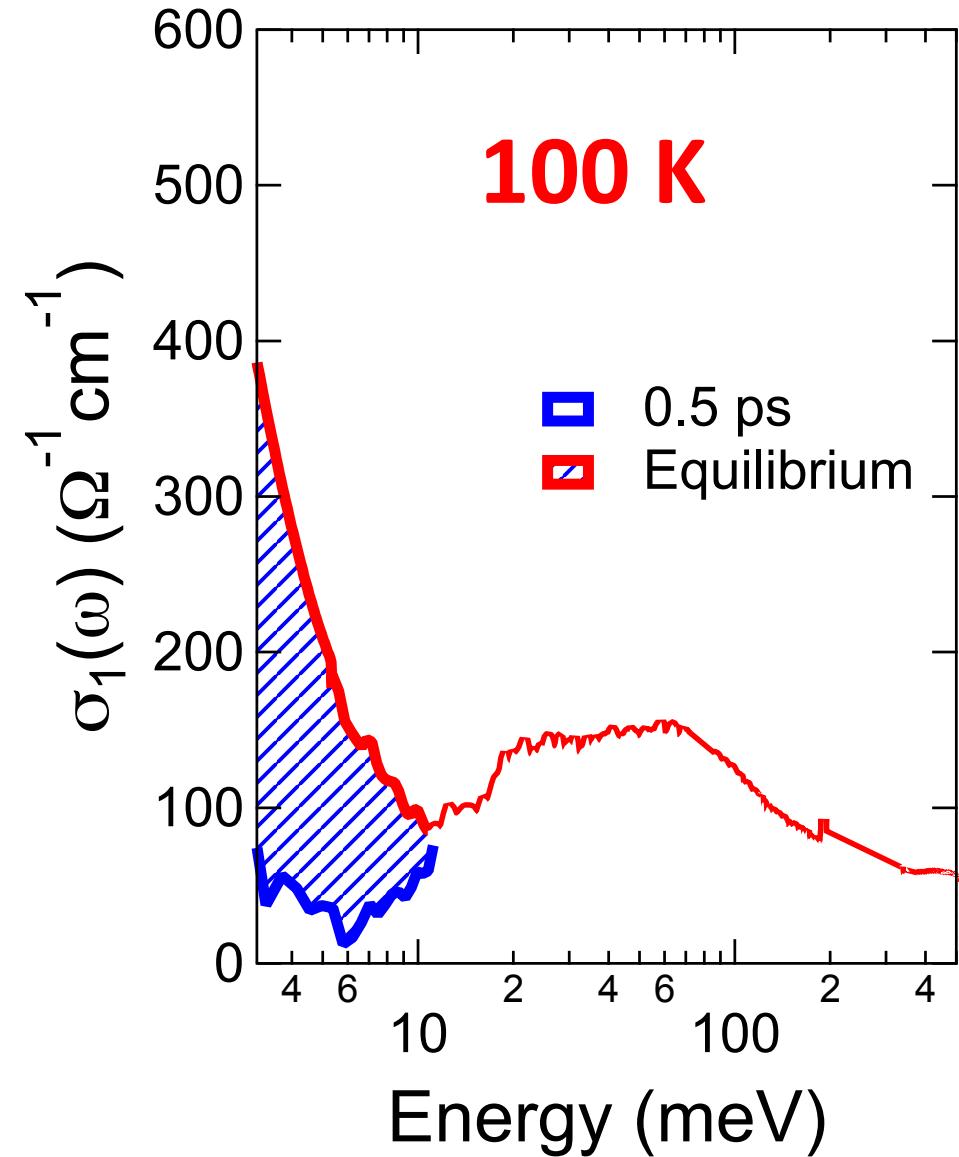
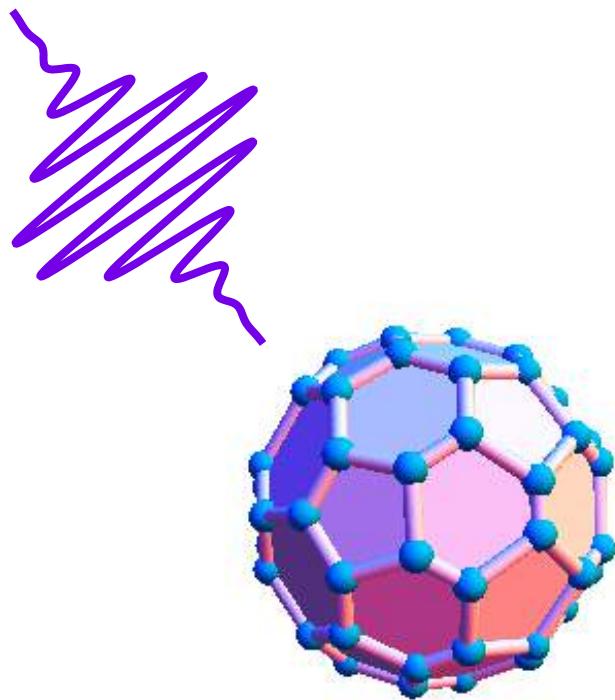
Temperature dependence



Crossover at \sim 10 times T_c



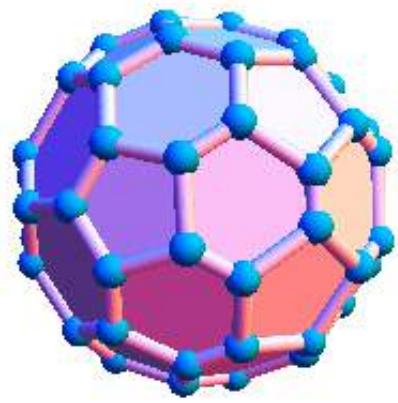
K_3C_{60} : Stimulated superconductivity ?



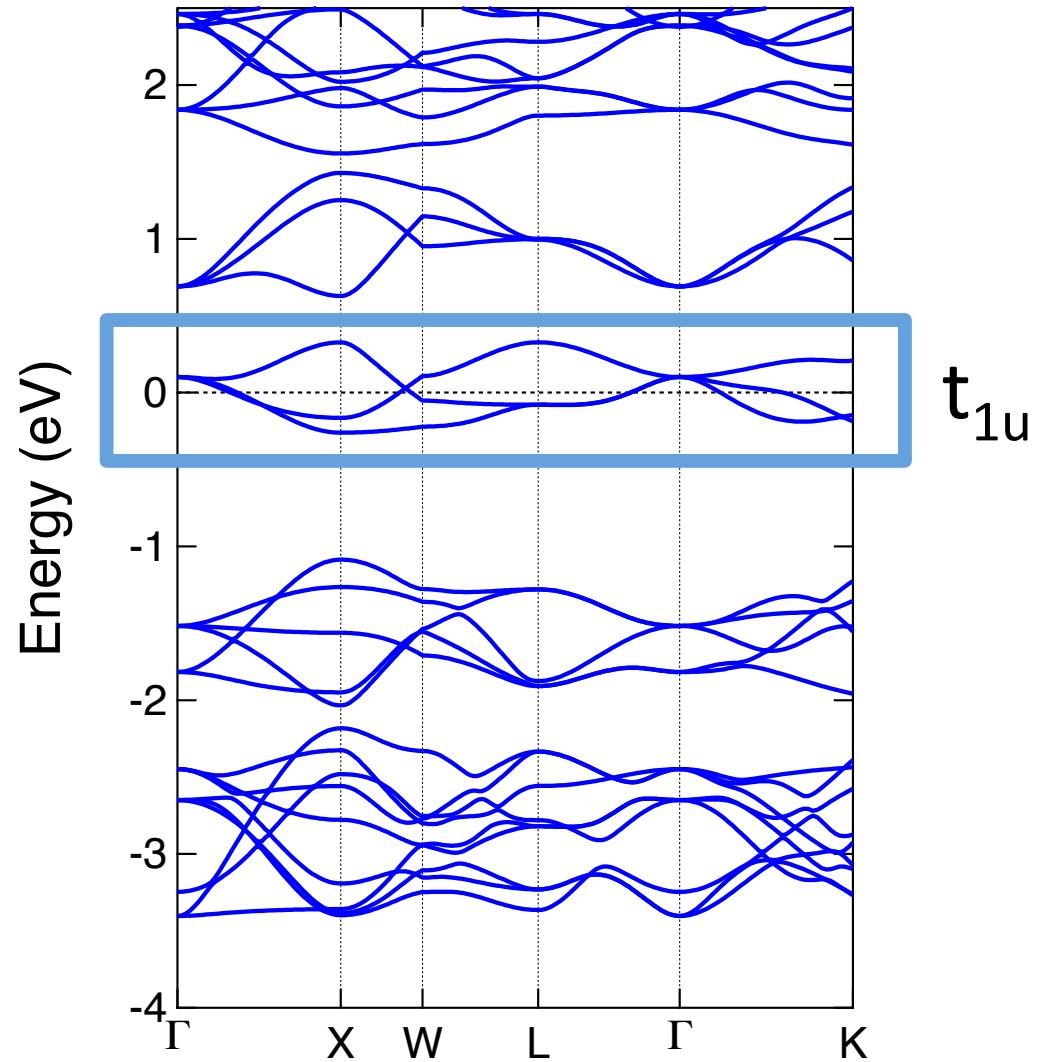
What is going on?



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

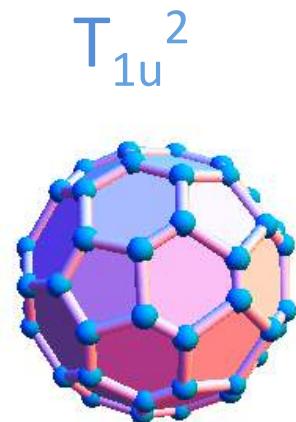


$T_{1u}(4)$
 1370 cm^{-1}

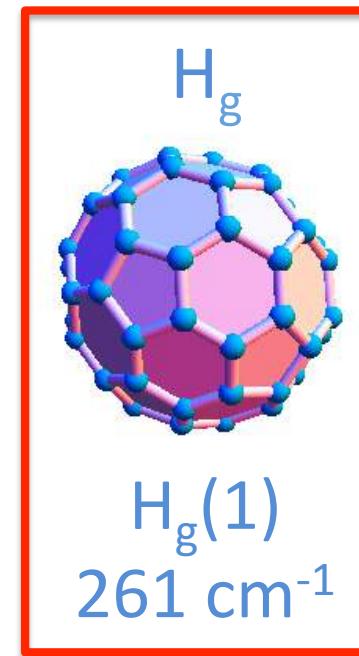


Nonlinear coupling to Jahn Teller Phonon?

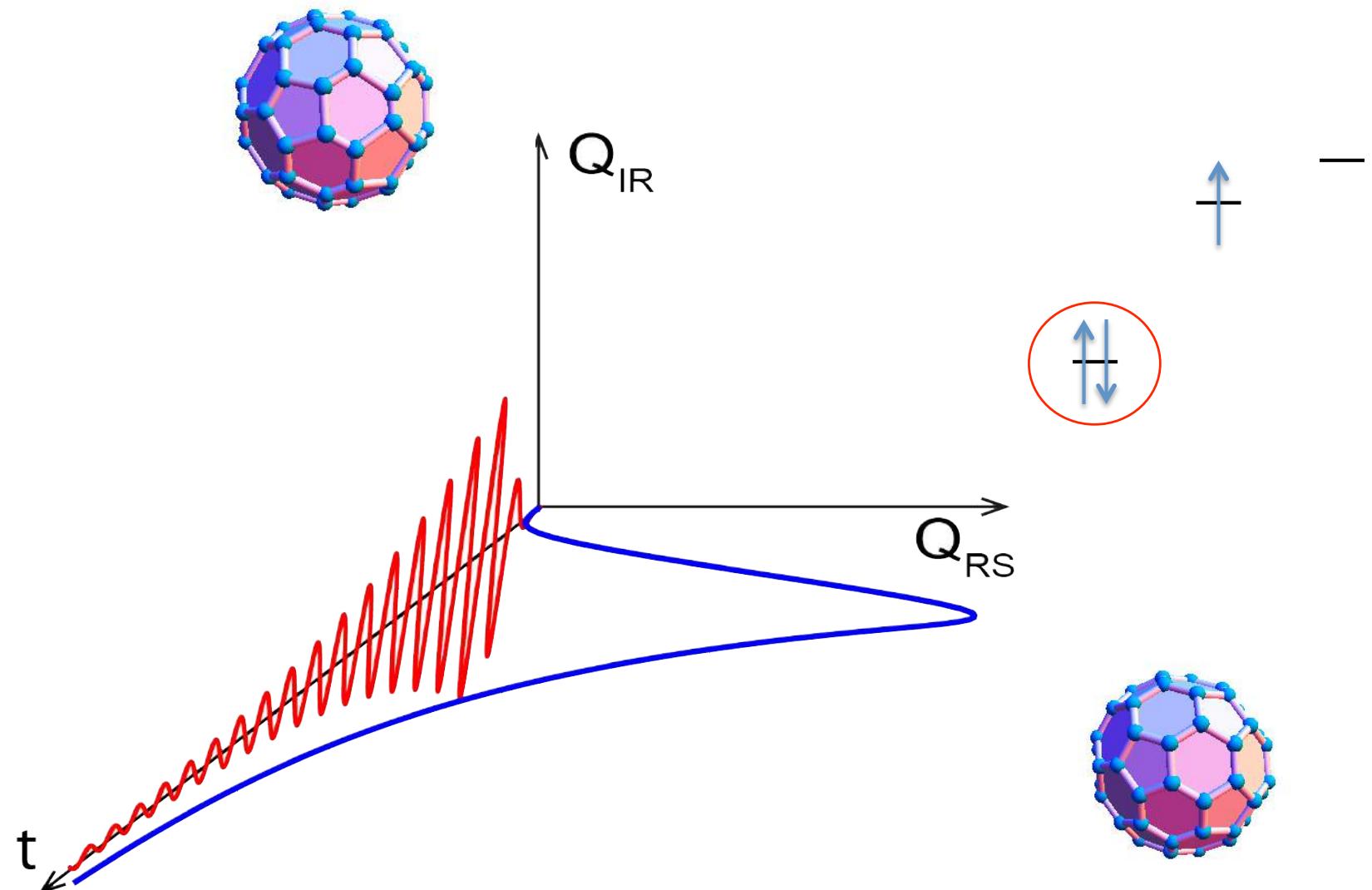
$$Q^2_{T1u} Q_{Hg}$$



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



Enhancement of pairing distortions ?



Or....new types of coupling

In analogy with nonlinear optics

$Q_{IR}^2 \cdot Q_2$ (lattice control)

$Q_{IR}^2 \cdot Q_2^2$ (phonon squeezing)

Q_{IR}^4 (parametric phonon amplification)

$Q_{IR1} \cdot Q_{IR2} e^{i\Theta} \cdot s$ (controlling time reversal invariance)

$Q_{IR}^2 \cdot U$ (controlling correlations)



Modulation of U in organics

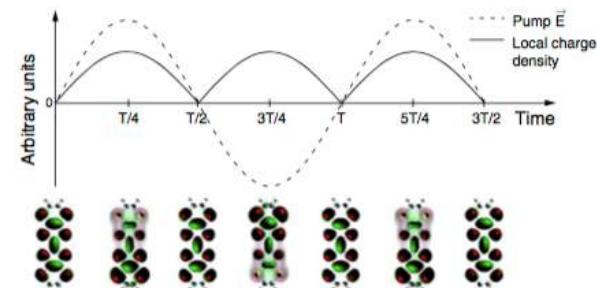
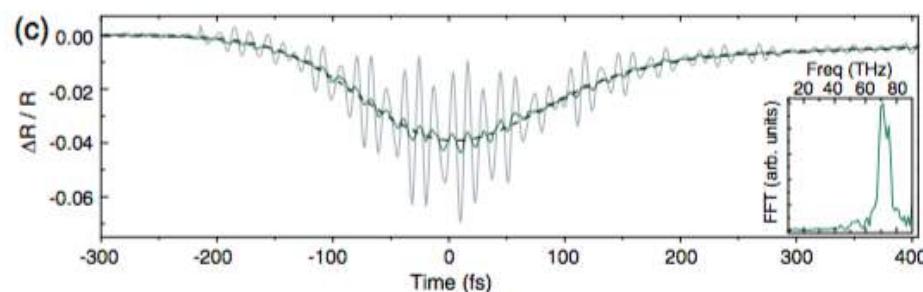
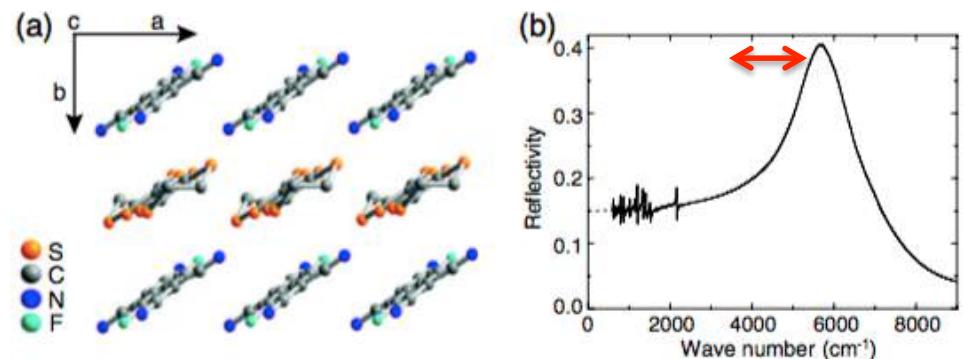
PRL 115, 187401 (2015)

PHYSICAL REVIEW LETTERS

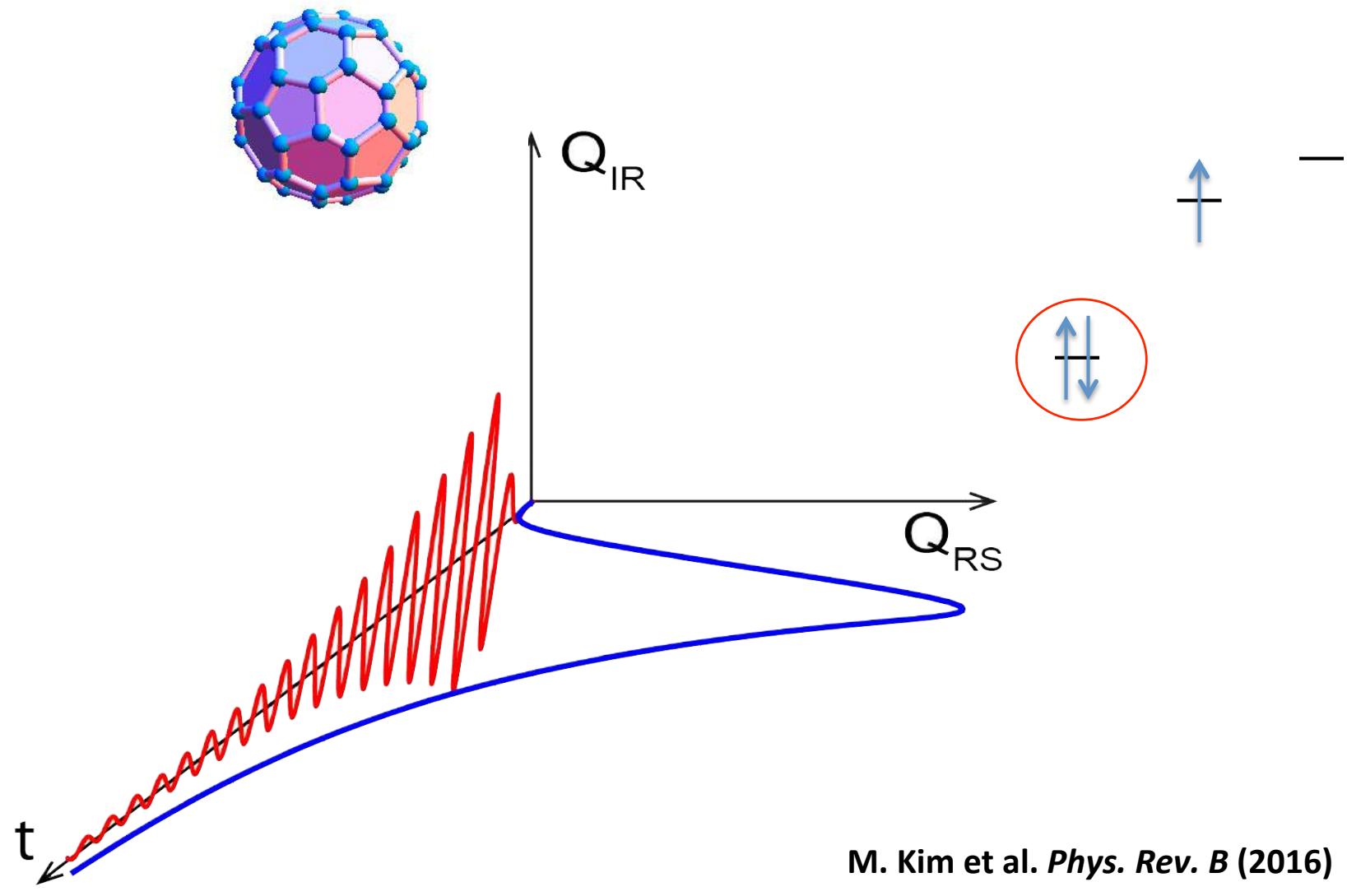
week ending
30 OCTOBER 2015

THz-Frequency Modulation of the Hubbard U in an Organic Mott Insulator

R. Singla,^{1,*} G. Cotugno,^{1,2} S. Kaiser,^{1,7,8,†} M. Först,¹ M. Mitrano,¹ H. Y. Liu,¹ A. Cartella,¹ C. Manzoni,^{1,4} H. Okamoto,⁵ T. Hasegawa,⁶ S. R. Clark,^{2,9} D. Jaksch,^{2,3} and A. Cavalleri^{1,2,‡}



Nonlinear coupling to correlation energy ?



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

G. Mazza, A. Georges (2017)



Nonlinear phononic control

