Chaire de Physique de la Matière Condensée

Matériaux et dispositifs à fortes corrélations électroniques

II.3 Contrôle sélectif par la lumière
et « Phononique non-linéaire »

Antoine Georges

Cycle 2014-2015
18 mai 2015 – II.3

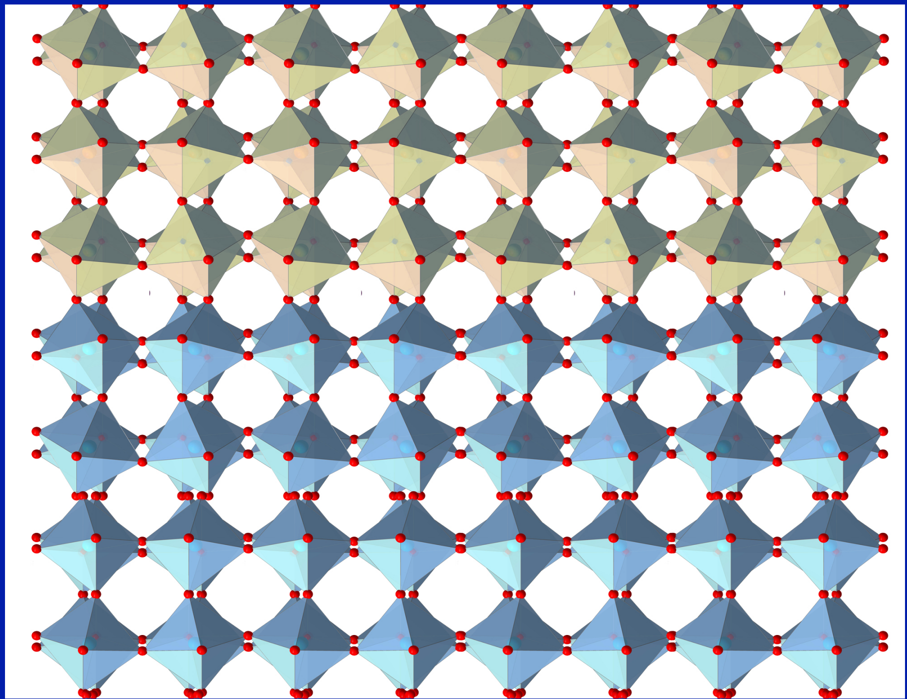
Can we teach correlated quantum materials to do what we want them to: SELECTIVE CONTROL of structure (and electronic structure) ?

"Frontiers in Quantum Materials Control"
ERC-Synergy project QMAC
A.Cavalleri, A.G., D.Jaksch, J.M. Triscone

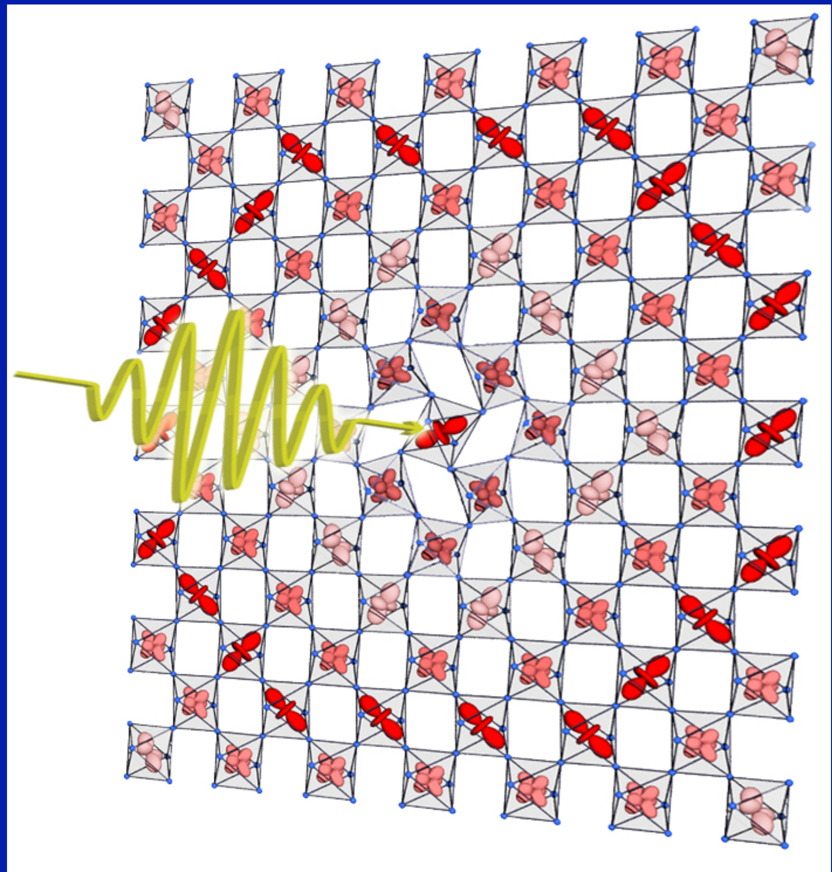
European Research Council

http://www.mpsd.mpg.de/48916/Q-MAC-start

Two routes to structural control



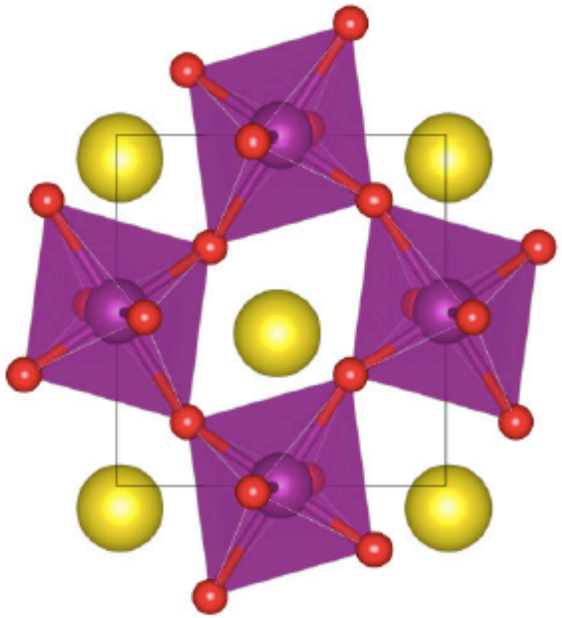
Selective control with LIGHT



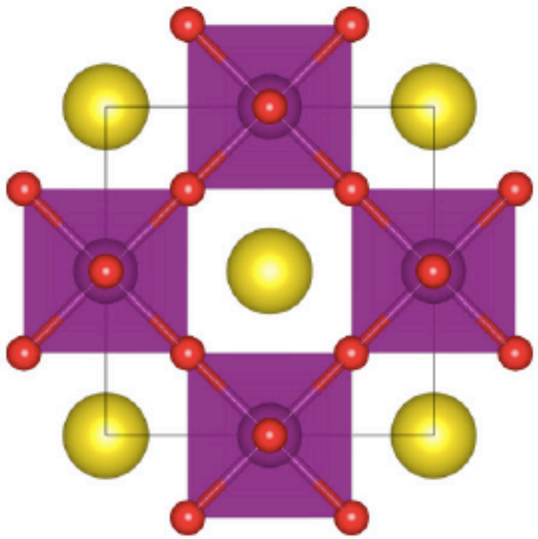
Artificial Materials:
Strained films and
Heterostructures; “Oxytronics”

Many talks at this conference

Structure determines Function



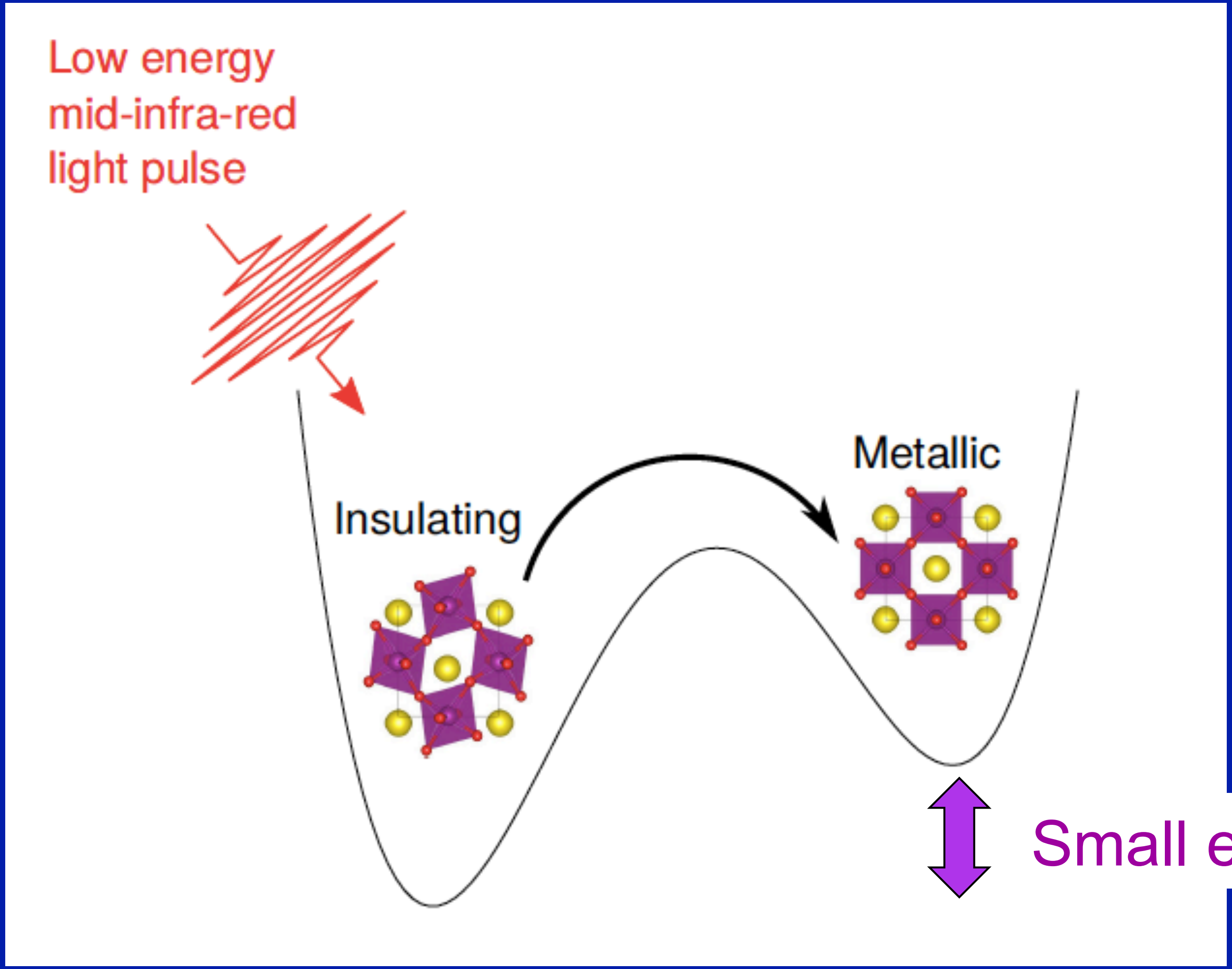
Large rotation/distortion
Insulating



No rotation/distortion
Metallic
Undistorted phase not synthesized

Aim: control key electronic energy scales such as:
1) Bandwidth 2) energy splitting between orbitals
3) Superexchange, etc…

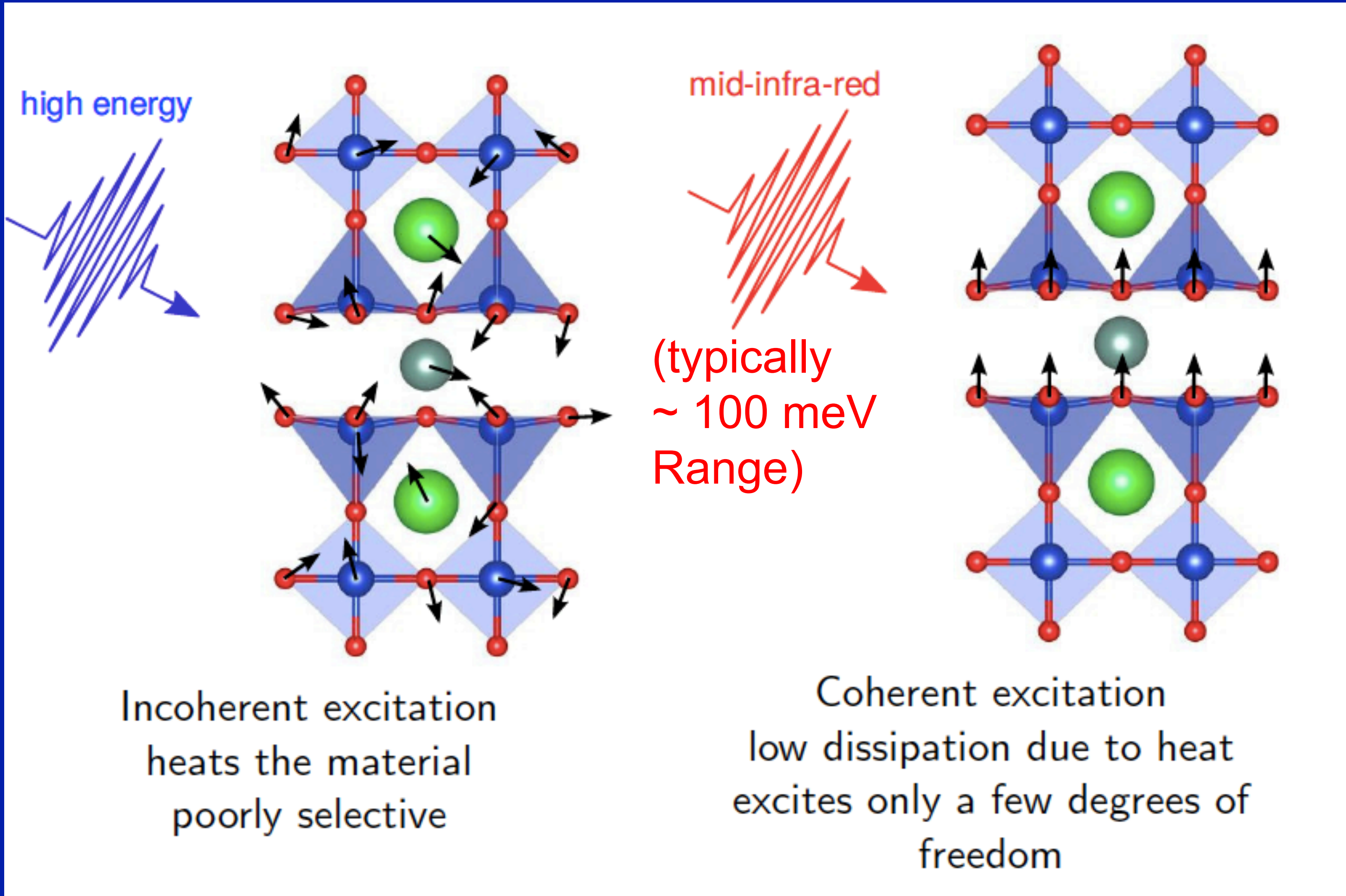
Selective control with resonant light



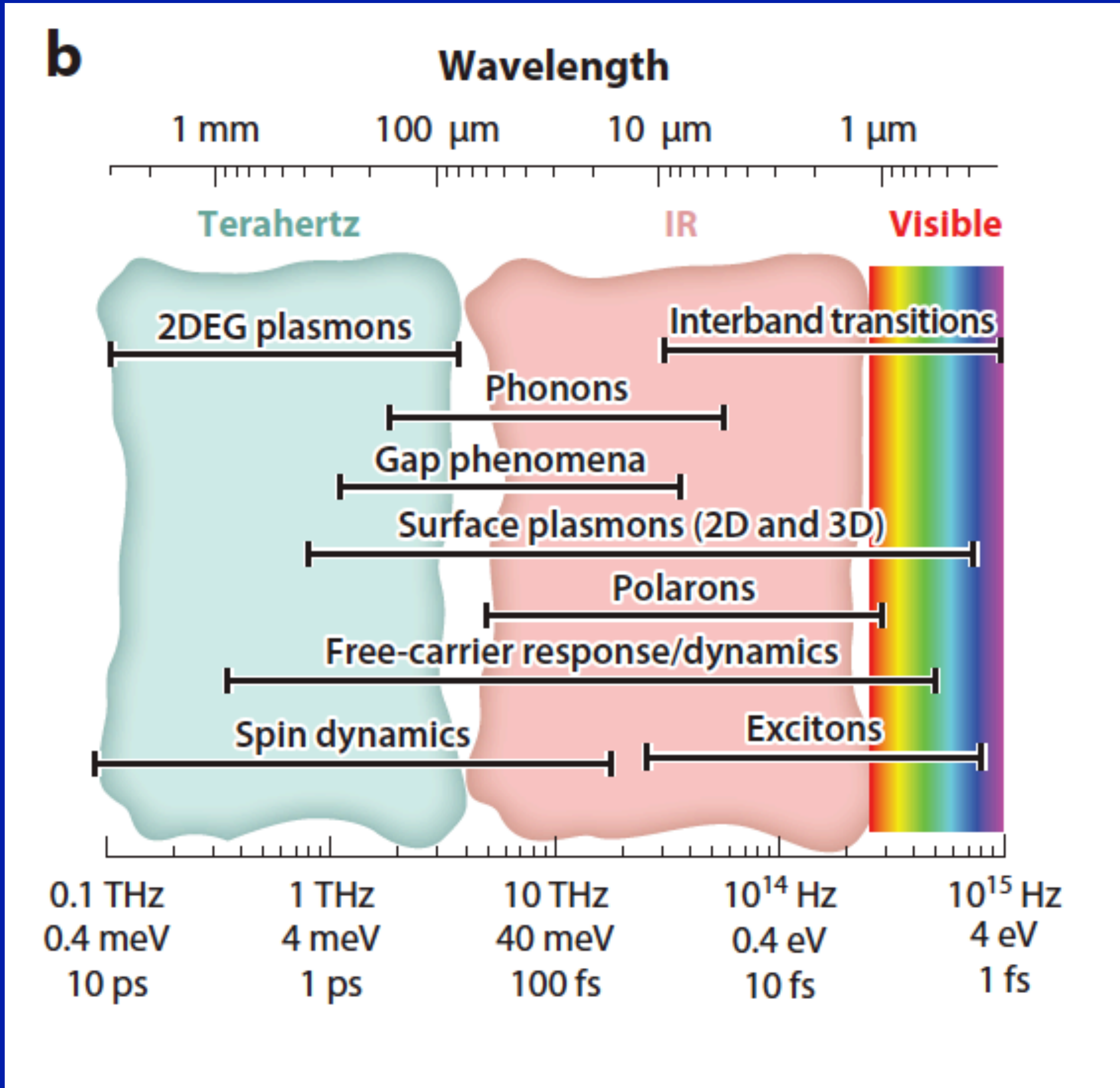
Small energy difference ?

Create transient structures which cannot be synthesized at equilibrium !

Incoherent vs. Coherent Control



Pioneering experiments by Andrea Cavalleri et al.



From Zhang and Averitt Ann Rev Mat Res 2014

Pioneering experiment: Metallization of a Manganite by selective excitation of mid-IR structural mode

Exciting an IR-active phonon
(up and down shaking of octahedra 71 meV ~ 17 THz)
in an insulating manganite *induces an Insulator-to-Metal Transition*

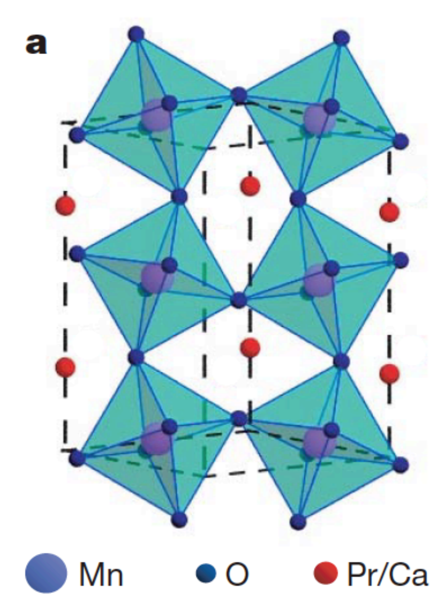
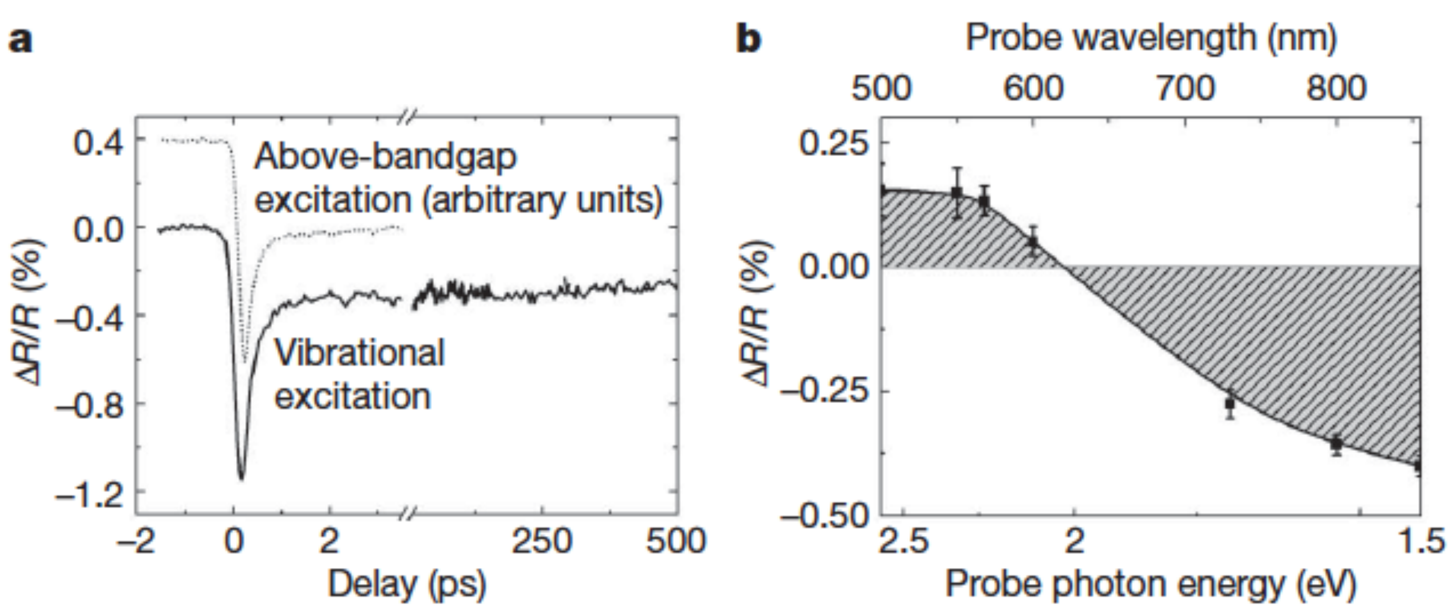


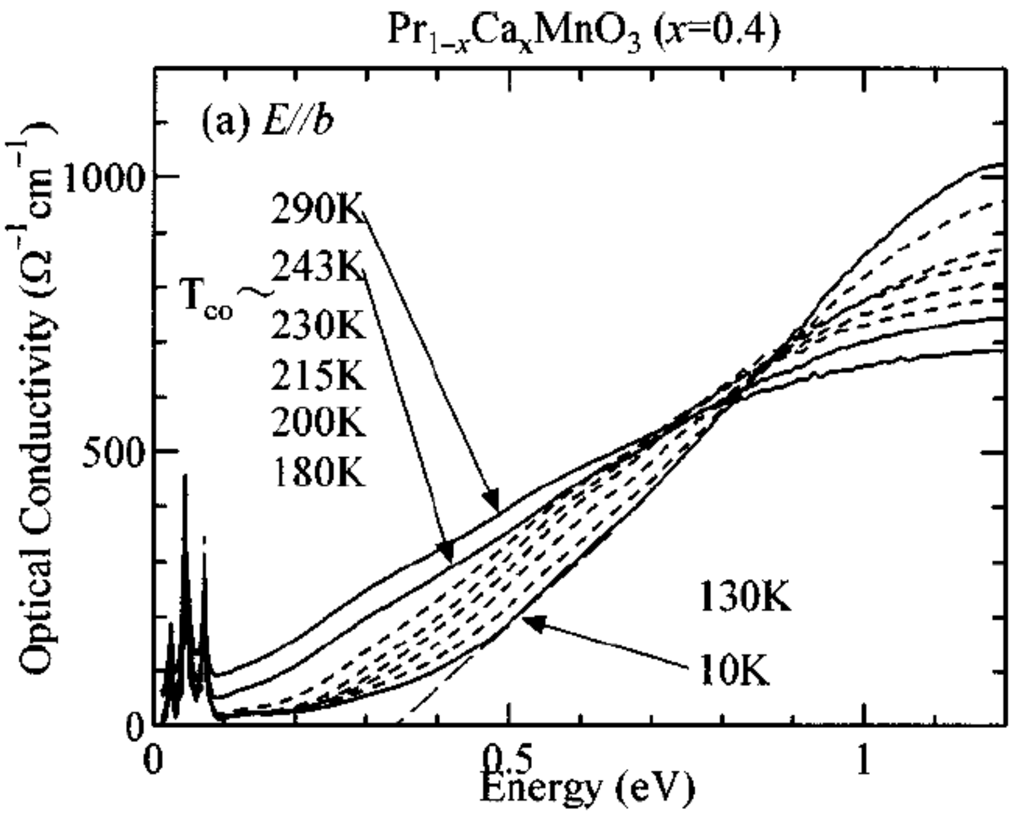
Figure 1 | $Pr_{0.7}Ca_{0.3}MnO_3$



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

Change of reflectivity at 800nm

From other experiments (B-field): signature of metallization



Metallization also seen Directly from Time-resolved dc-conductivity Measurement !

Optical conductivity <u>increases</u> above 0.8 eV as the <u>insulating state is formed</u> and the gap develops

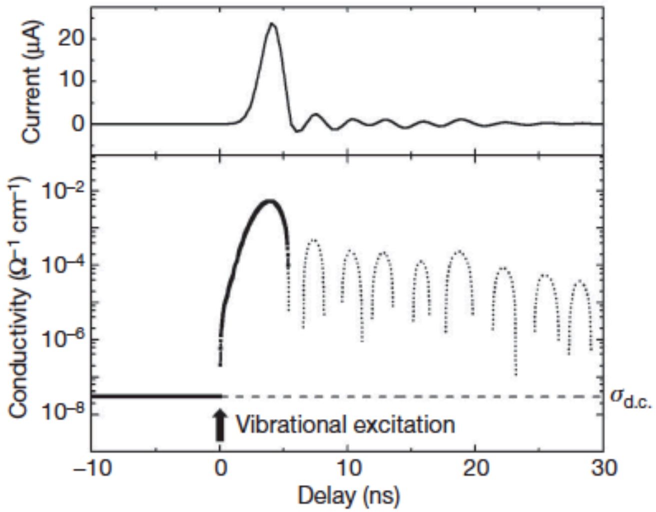
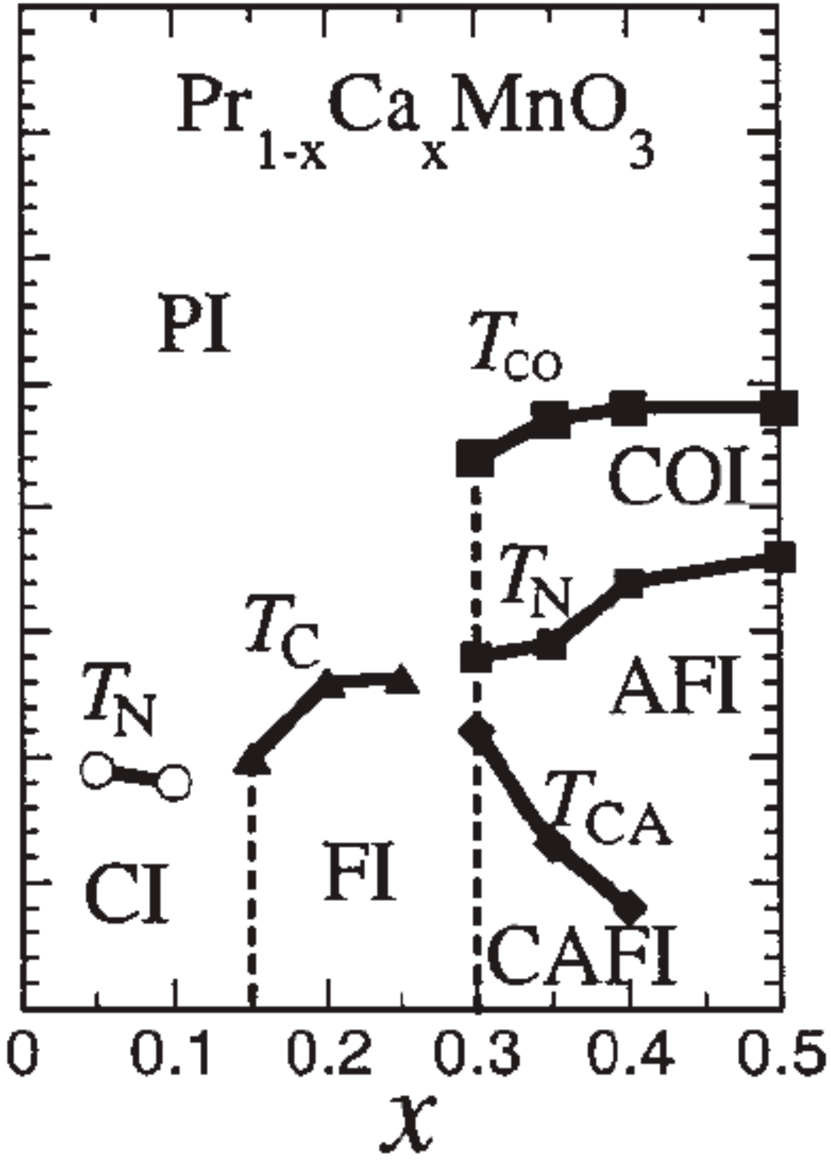
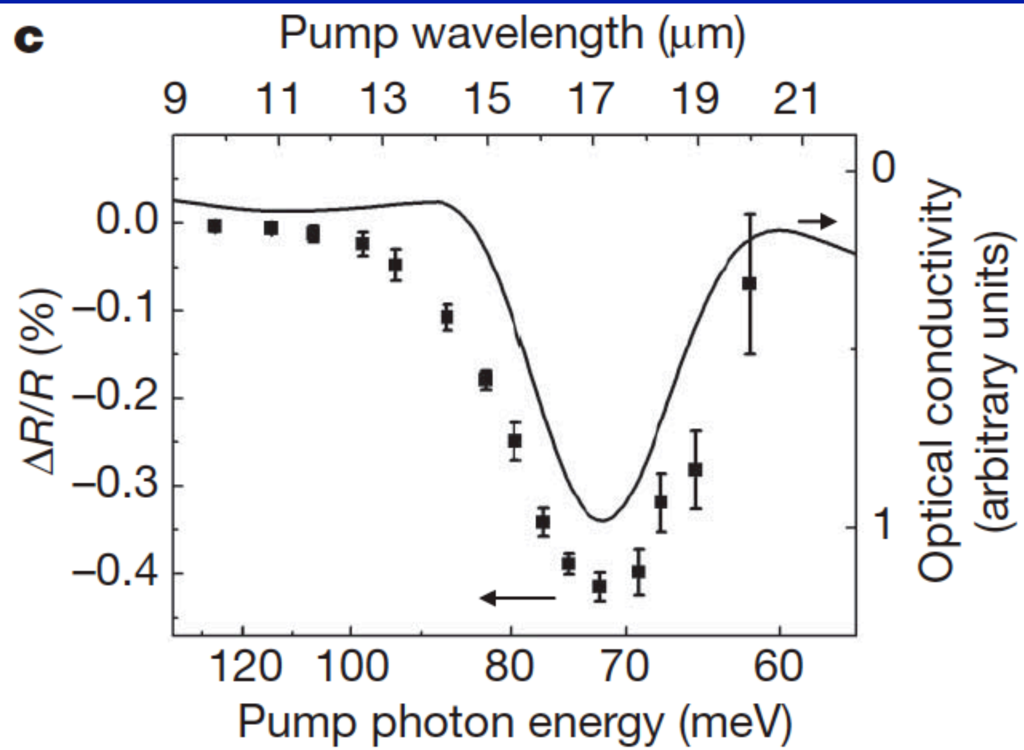


Figure 3 | Time-dependent transport measurement. Vibrational excitation of the Mn–O stretching mode results in a ~10^3 increase in the sample current (upper panel) and a corresponding ~10^5 increase in the sample conductivity (lower panel). The metastable metallic phase is formed and relaxes within the experimental time resolution of 4 ns. The current oscillations following the main pulse are due to electronic ringing and cannot be converted accurately into sample conductivity, so the derived conductivity oscillations are shown as a dotted line. The dashed line shows the d.c. conductivity of the insulating phase of $Pr_{0.7}Ca_{0.3}MnO_3$ at 30 K.



Not heating !
PCMO is an insulator for all values of x

Resonant phenomena
(with ~70 meV mode)



PUZZLE:

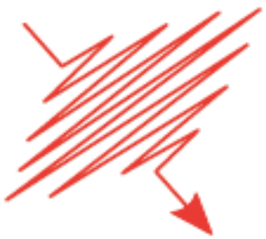
- Light couples directly only to dipolar-active modes

- Distortions most relevant to electronic structure correspond to rotations, tilts, or JT modes

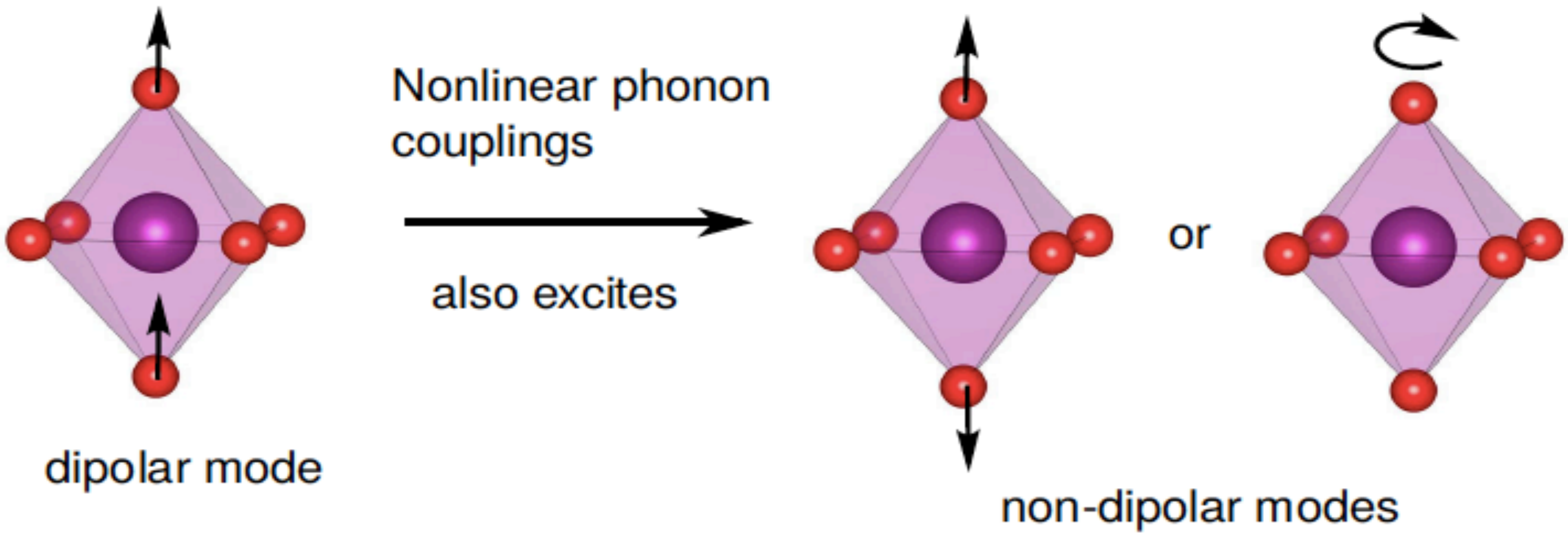
- Those modes do not carry a dipolar moment !

"Non-Linear Phononics"

Key qualitative idea: Först et al. Nature Phys 7, 854 (2011)
Microsopic theory: Subedi, Cavalleri and AG, PRB 89 22031R (2014)



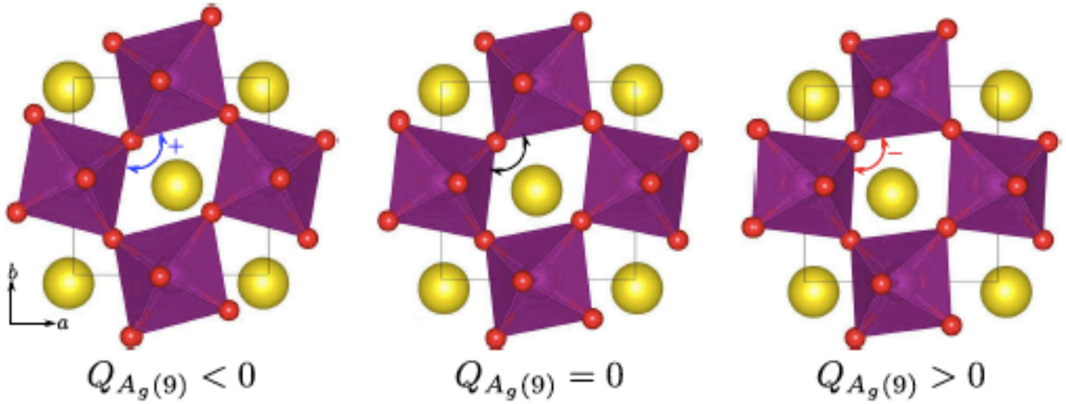
cf `Ionic Raman Scattering'

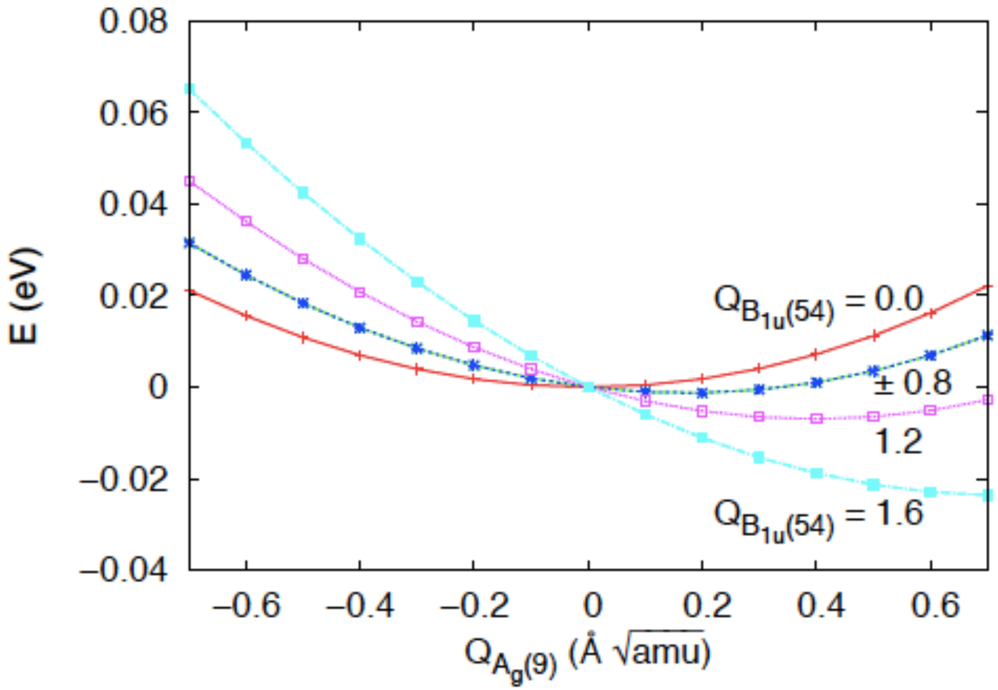


PHYSICAL REVIEW B **89**, 220301(R) (2014)

Theory of nonlinear phononics for coherent light control of solids

Alaska Subedi,[1] Andrea Cavalleri,[2,3] and Antoine Georges[1,4,5]





Energy surface for $PrMnO_3$ as a function of Raman mode for different amplitudes of pumped IR mode

TABLE I: Calculated zone center phonon frequencies and the symmetries of selected modes of orthorhombic $PrMnO_3$. The mode number is given in the parenthesis.

Calc. freq. (cm^{-1})		Symmetry
97.43		$A_g(4)$
154.80	Octahedral rotations ~ 19 meV	$A_g(9)$
231.10		$A_g(21)$
267.58		$A_g(23)$
351.07		$A_g(36)$
479.49		$A_g(47)$
552.09		$A_g(51)$
622.12	c-axis `shaking' of octahedra ~ 77 meV	$B_{1u}(54)$
633.38		$B_{1u}(56)$
639.95		$B_{2u}(58)$
660.54		$B_{3u}(60)$

NL coupling to a symmetry-preserving Raman mode: cubic

$$V(Q_{\mathrm{R}}, Q_{\mathrm{IR}}) = \frac{1}{2}\Omega_{\mathrm{R}}^2 Q_{\mathrm{R}}^2 + \frac{1}{2}\Omega_{\mathrm{IR}}^2 Q_{\mathrm{IR}}^2 + \frac{1}{3}a_3 Q_{\mathrm{R}}^3 + \frac{1}{4}b_4 Q_{\mathrm{IR}}^4 - \frac{1}{2}g\, Q_{\mathrm{R}} Q_{\mathrm{IR}}^2 \qquad (1)$$

$$\begin{aligned} \ddot{Q}_{\mathrm{IR}} + \Omega_{\mathrm{IR}}^2 Q_{\mathrm{IR}} &= g\, Q_{\mathrm{R}} Q_{\mathrm{IR}} - b_4 Q_{\mathrm{IR}}^3 + F(t) \\ \ddot{Q}_{\mathrm{R}} + \Omega_{\mathrm{R}}^2 Q_{\mathrm{R}} &= \frac{1}{2} g\, Q_{\mathrm{IR}}^2 - a_3 Q_{\mathrm{R}}^2 \, . \end{aligned}$$

Note: $Q_{IR}^2(t) \propto \cos^2 \Omega_{IR} t$ has a finite mean-value !

Effective potential seen by Raman mode, time-averaged over IR mode:

$$V_{\text{eff}}(Q_{\text{R}}) = \frac{1}{2}\Omega_{\text{R}}^2 Q_{\text{R}}^2 + \frac{1}{3}a_3 Q_{\text{R}}^3 - \frac{1}{4} g Q_{\text{IR,max}}^2 Q_{\text{R}} \quad (11)$$

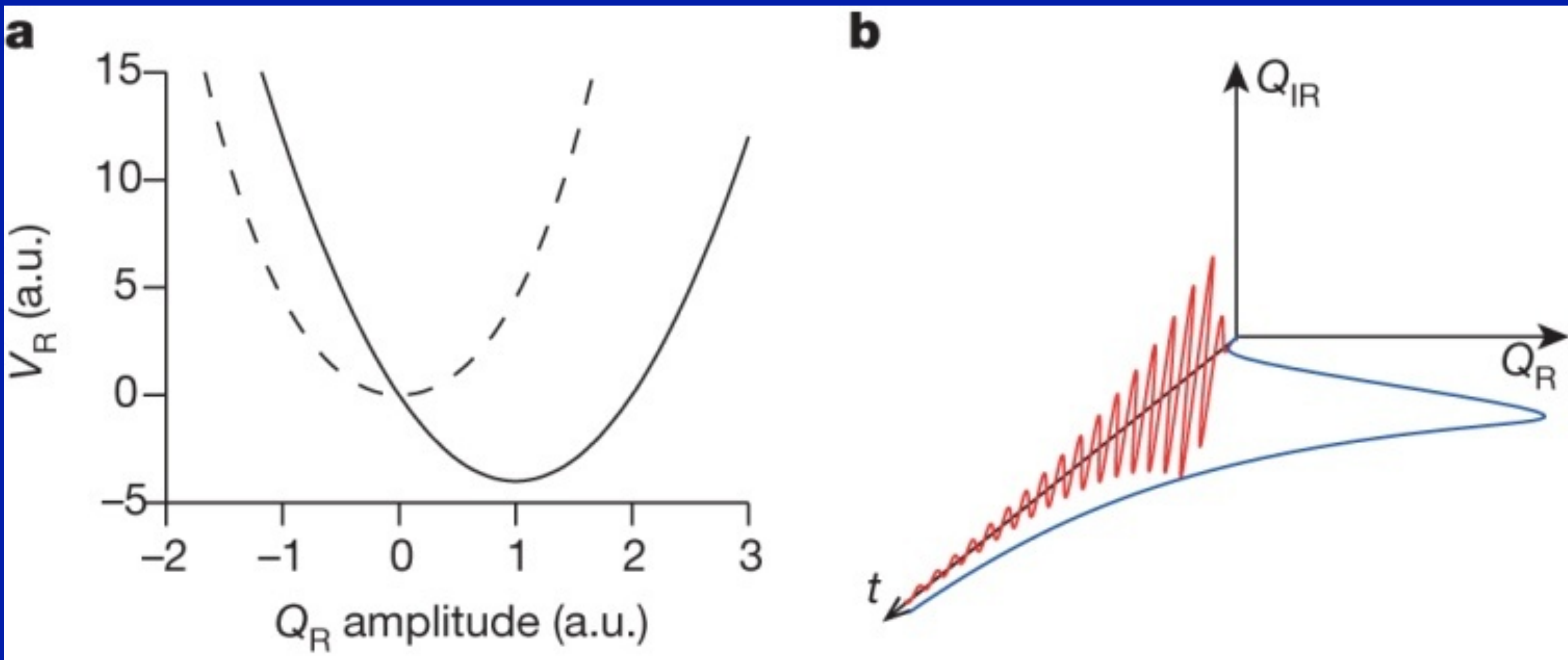
The displaced position δQ_{R} corresponds to the minimum of this potential given by $a_3 \delta Q_{\text{R}}^2 + \Omega_{\text{R}}^2 \delta Q_{\text{R}} - g Q_{\text{IR,max}}^2/4 = 0$, and thus reads

$$\delta Q_{\text{R}} = \frac{\Omega_{\text{R}}^2}{2a_3}\left[\sqrt{1 + \frac{a_3 g Q_{\text{IR,max}}^2}{\Omega_{\text{R}}^4}} - 1\right] \quad (12)$$

$$\simeq \frac{g}{4\Omega_{\text{R}}^2} Q_{\text{IR,max}}^2 - \frac{1}{16} a_3 \frac{g^2 Q_{\text{IR,max}}^4}{\Omega_{\text{R}}^6} + \cdots$$

The mechanism:

Upon pumping (fast IR mode),
the effective potential seen by the slow (Raman-active) mode has a shifted minimum, corresponding to a change of the structure



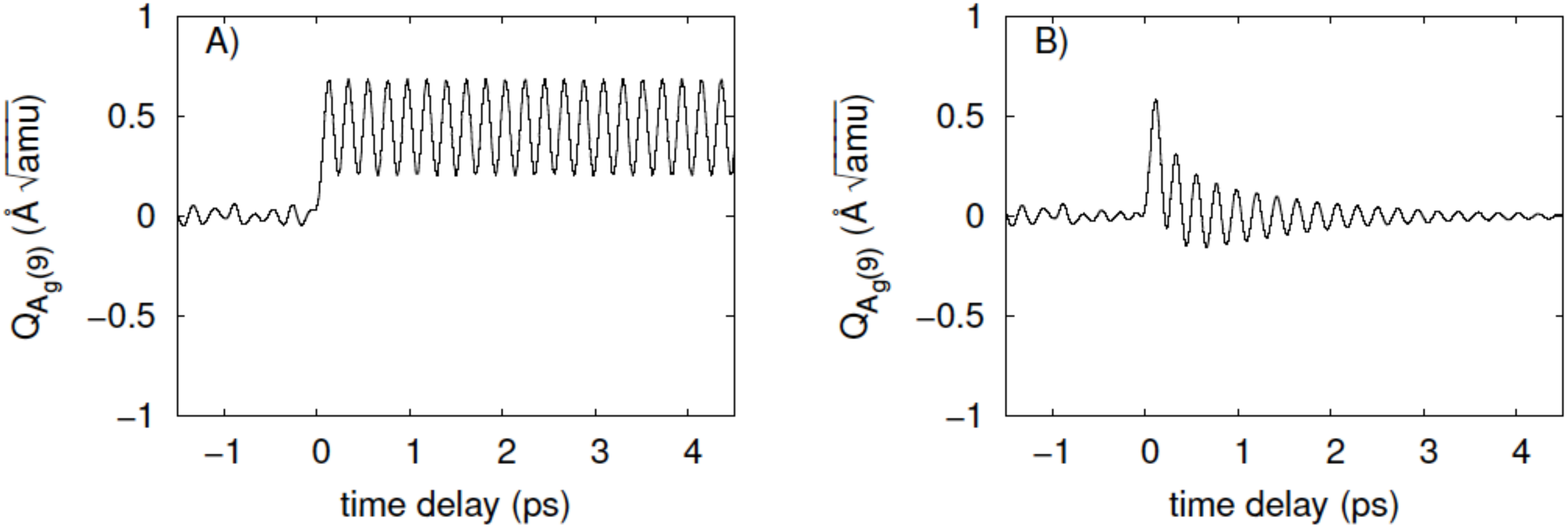
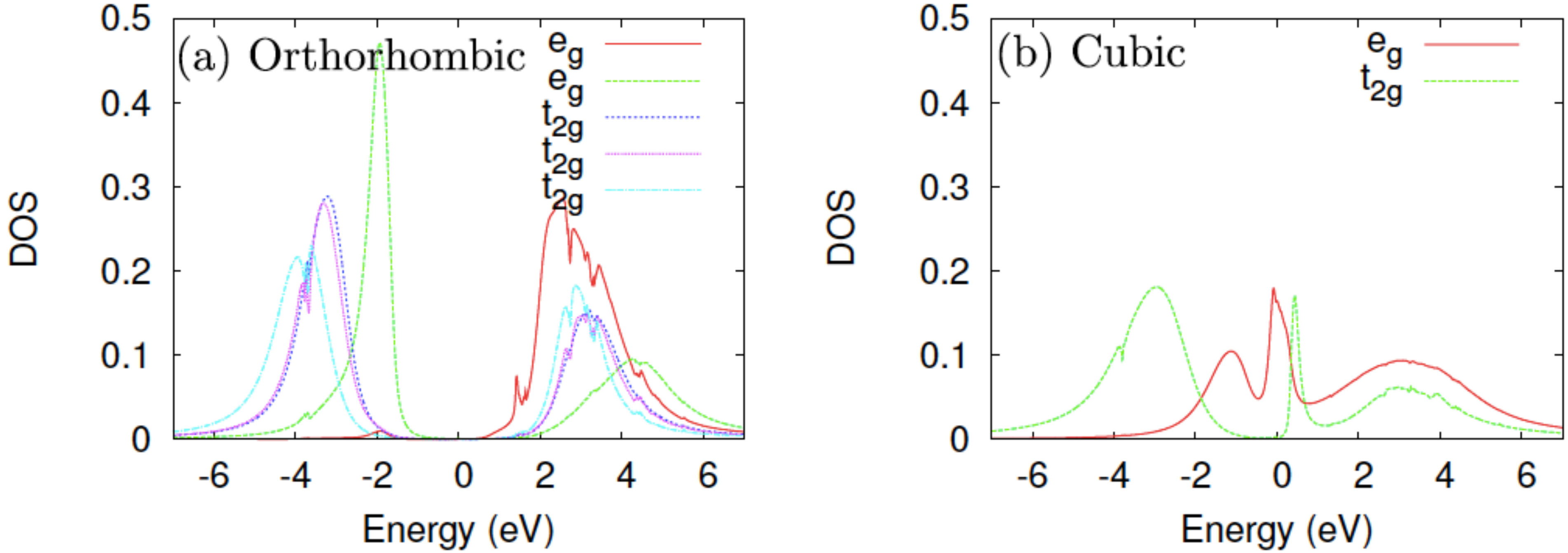


FIG. 5: Dynamics of the Raman $A_g(9)$ mode for PMO (cubic coupling). Left panel: dynamics without damping. Right panel: dynamics with damping values of 5% for both $B_{1u}(54)$ and $A_g(9)$ modes.

Displacement of the Raman phonon away from equilibrium position

Reducing orthorombic distortion increases bandwidth and leads to metallic phase
(can be quantified by a DMFT+DFT calculation:)



- Explains experimental observation of Rini et al.
- Provides a quantitative framework to predict how the structure changes upon resonant excitation of the IR mode

Time-resolved X-ray diffraction

(@ Free-Electron Lasers)

Direct evidence of displacement of Raman modes

see later in the talk, for YBCO

(also Foerst et al. on LSMO, submitted)

Coupling to a symmetry-breaking Raman mode:

Q^2Q^2 coupling

→ a different universality class !

Non-perturbative phenomena

…yet to be demonstrated experimentally

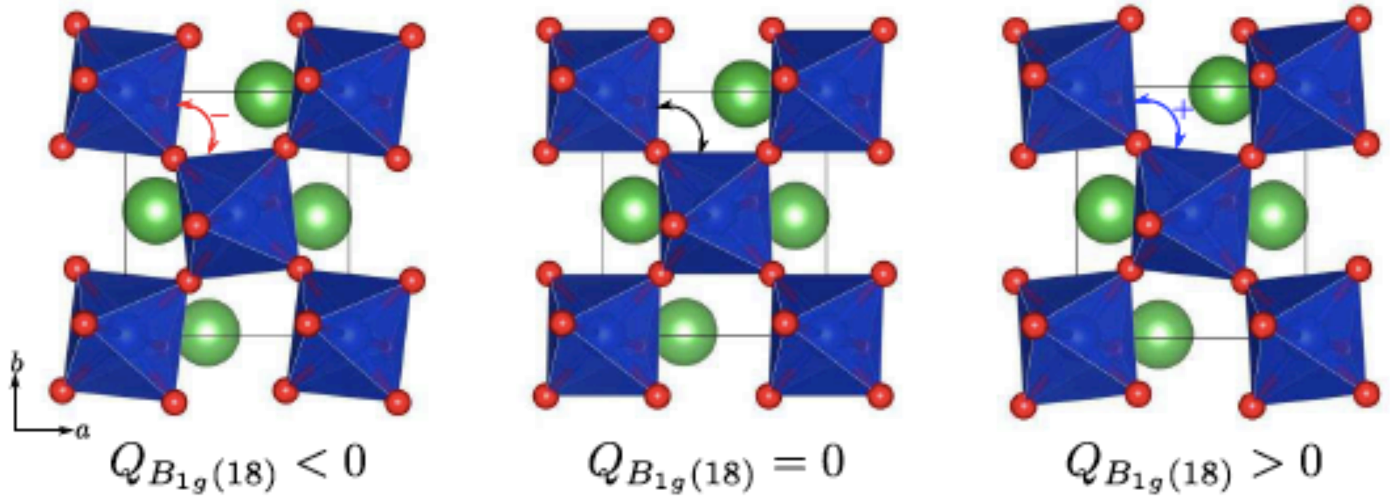
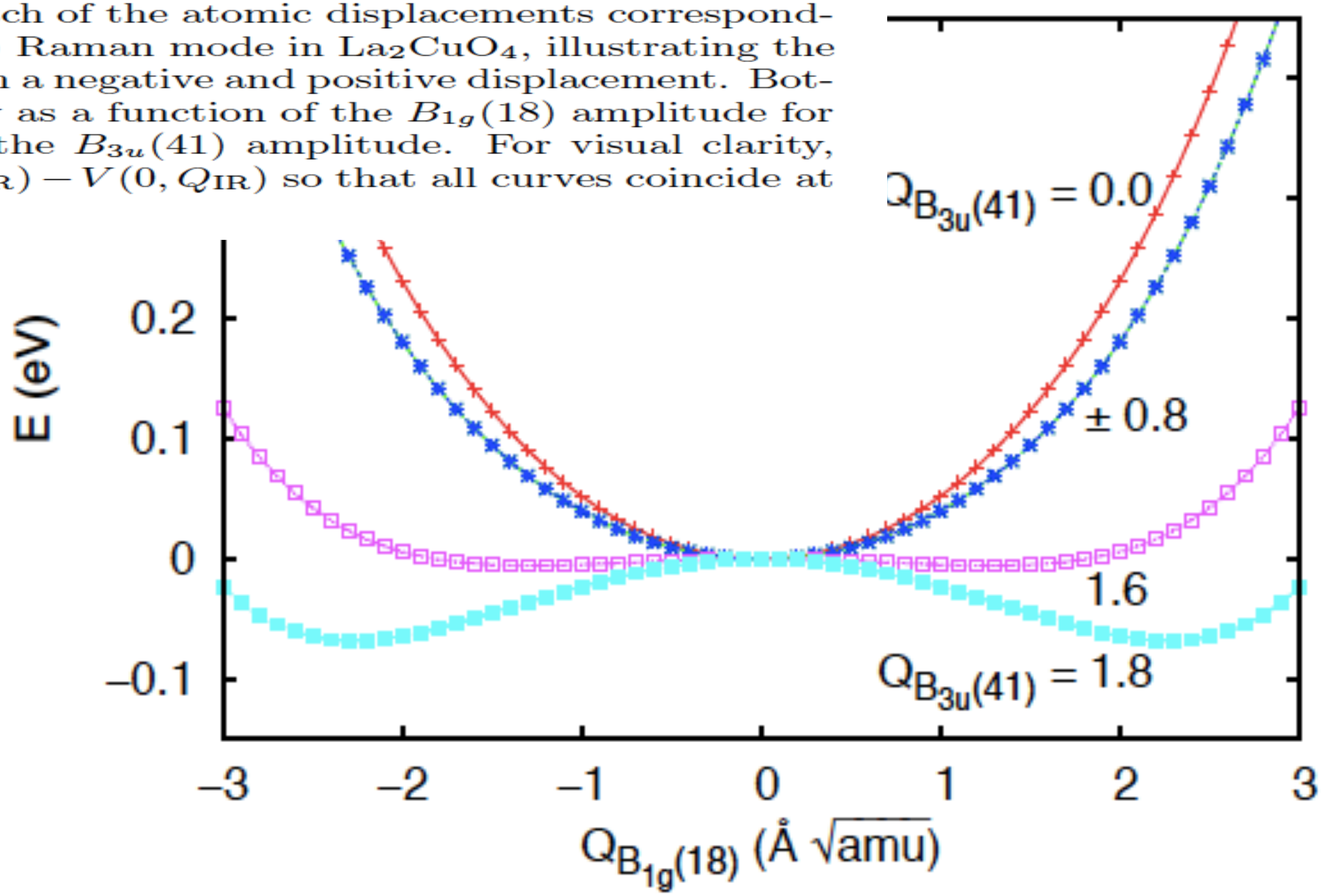


FIG. 2: Top: Sketch of the atomic displacements corresponding to the $B_{1g}(18)$ Raman mode in La_2CuO_4, illustrating the symmetry between a negative and positive displacement. Bottom: Total energy as a function of the $B_{1g}(18)$ amplitude for several values of the $B_{3u}(41)$ amplitude. For visual clarity, we plot $V(Q_R, Q_{IR}) - V(0, Q_{IR})$ so that all curves coincide at $Q_R = 0$.



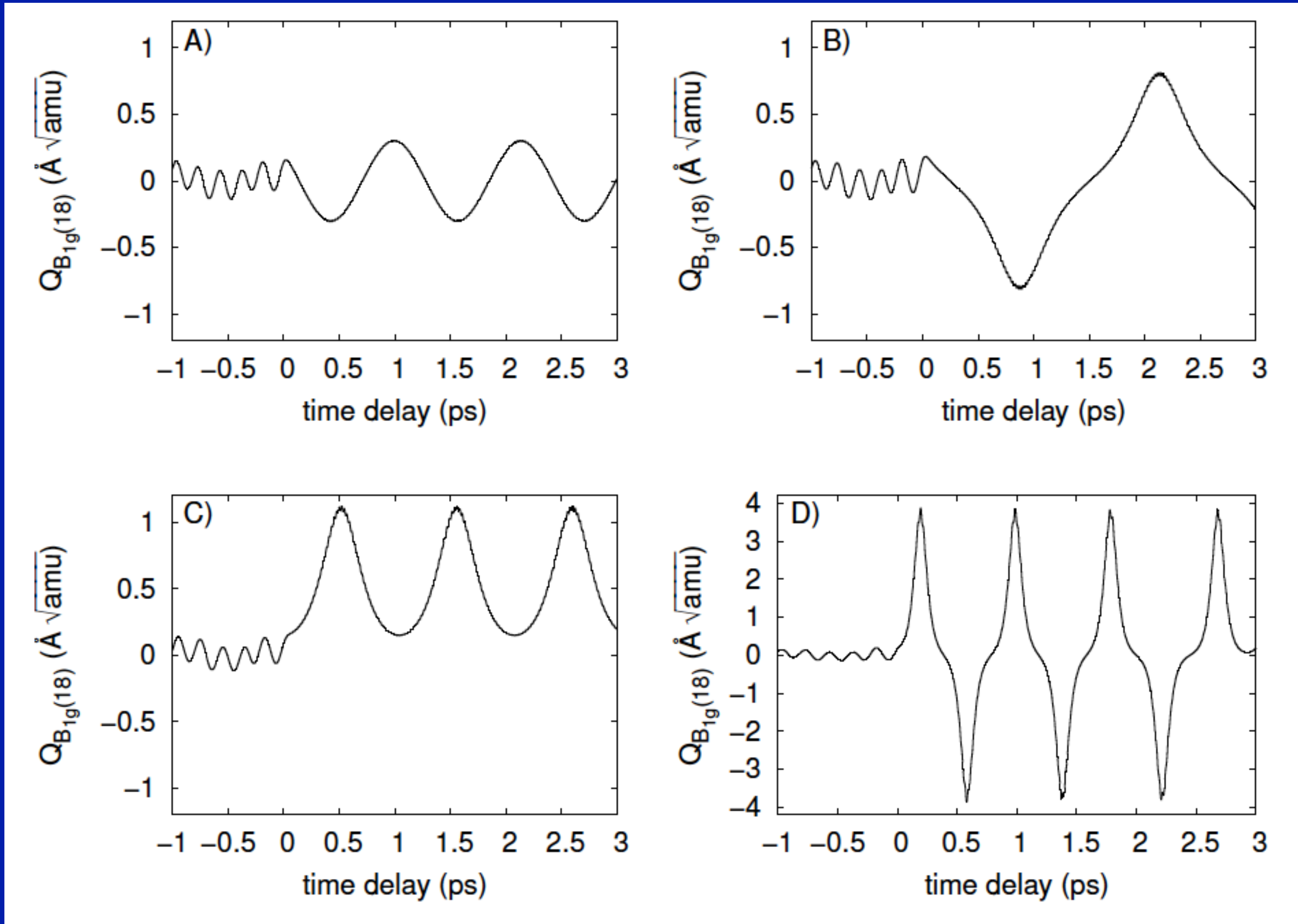
Q^2Q^2 coupling, parametric oscillators, Kapitza pendulum and all that…

$$V(Q_{\mathrm{R}}, Q_{\mathrm{IR}}) = \frac{1}{2}\Omega_{\mathrm{R}}^2 Q_{\mathrm{R}}^2 + \frac{1}{2}\Omega_{\mathrm{IR}}^2 Q_{\mathrm{IR}}^2 + \frac{1}{4}a_4 Q_{\mathrm{R}}^4 + \frac{1}{4}b_4 Q_{\mathrm{IR}}^4 - \frac{1}{2}g\, Q_{\mathrm{R}}^2 Q_{\mathrm{IR}}^2 . \qquad (2)$$

$$\ddot{Q}_{\mathrm{IR}} + \Omega_{\mathrm{IR}}^2 Q_{\mathrm{IR}} = g\, Q_{\mathrm{R}}^2 Q_{\mathrm{IR}} - b_4 Q_{\mathrm{IR}}^3 + F(t)$$

$$\ddot{Q}_{\mathrm{R}} + \Omega_{\mathrm{R}}^2 Q_{\mathrm{R}} = g\, Q_{\mathrm{R}} Q_{\mathrm{IR}}^2 - a_4 Q_{\mathrm{R}}^3$$

Very different type of coupling: ~ parametric oscillator
Frequency softening. Dynamical instability



Softening of the Raman mode, Dynamical threshold for displacement (driven parametric oscillator)

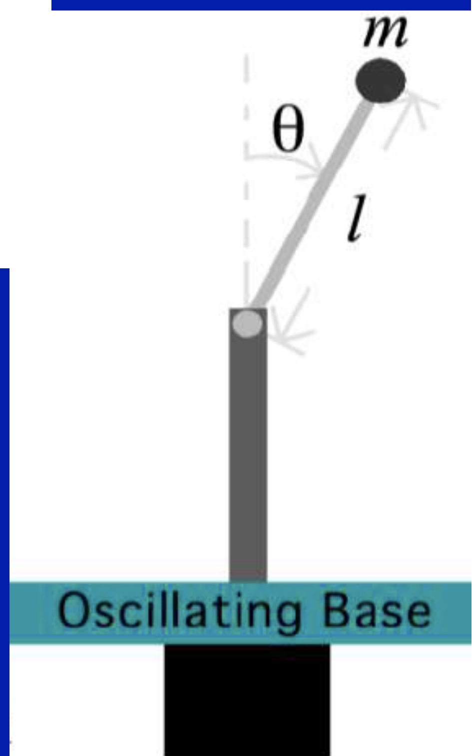
$$V_{\rm eff}(Q_{\rm R}) = \frac{1}{2}\Omega_{\rm R}^2 Q_{\rm R}^2 \left[1 - \frac{g Q_{\rm IR,max}^2}{2\Omega_{\rm R}^2}\right] + \frac{1}{4} a_4 Q_{\rm R}^4 \quad (16)$$

The motion becomes unstable when this effective potential acquires a negative curvature, so that to first approximation (ie neglecting corrections of order $\Omega_{\rm R}/\Omega_{\rm IR} \ll 1$, see below) the instability threshold is given by:

$$\frac{g Q_{\rm IR,max}^2}{2\Omega_{\rm R}^2} = 1 \Rightarrow \quad (17)$$

$$\Rightarrow F_c = \sqrt{\frac{2}{g}} \frac{\Omega_{\rm R}}{\Omega_{\rm IR}} \left[\int_{-\infty}^{+\infty} d\tau \tau^2 \Phi(\tau)\right]^{-1} = \frac{\Omega_{\rm R}}{\Omega_{\rm IR}} \frac{1}{\sqrt{\pi g}\sigma^3}$$

Dynamical stabilisation: the dynamical threshold is larger than that corresponding to the instability of the static potential
Cf. Kapitsa’s pendulum



From Light-induced MIT…
… to Light-induced Superconductivity

Light-Induced Superconductivity in a Stripe-Ordered Cuprate
D. Fausti *et al.*
Science **331**, 189 (2011);
DOI: 10.1126/science.1197294

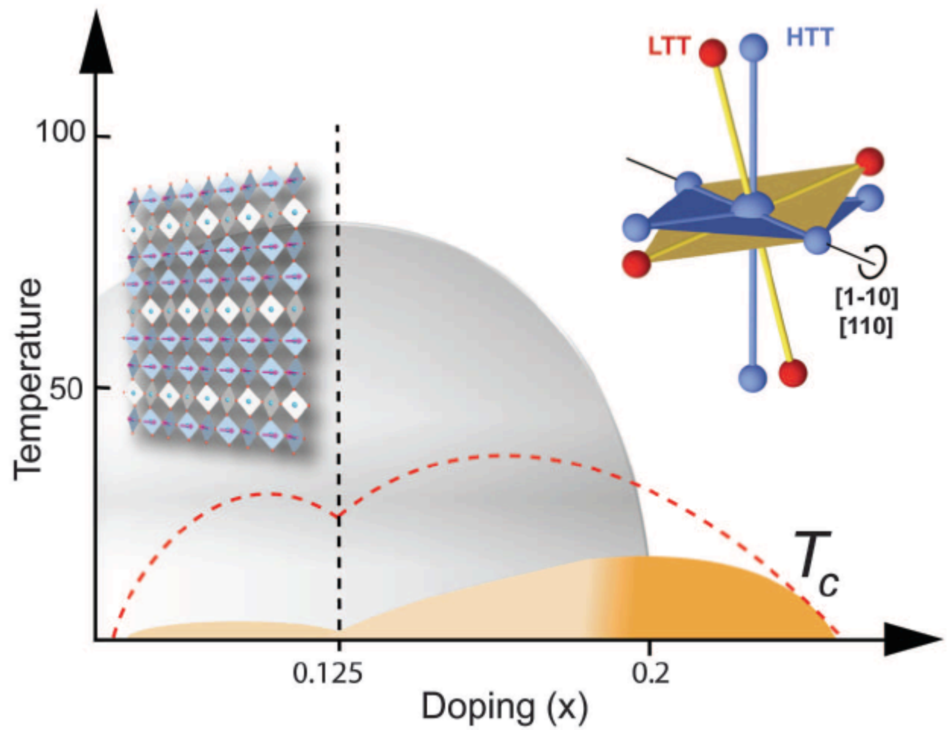
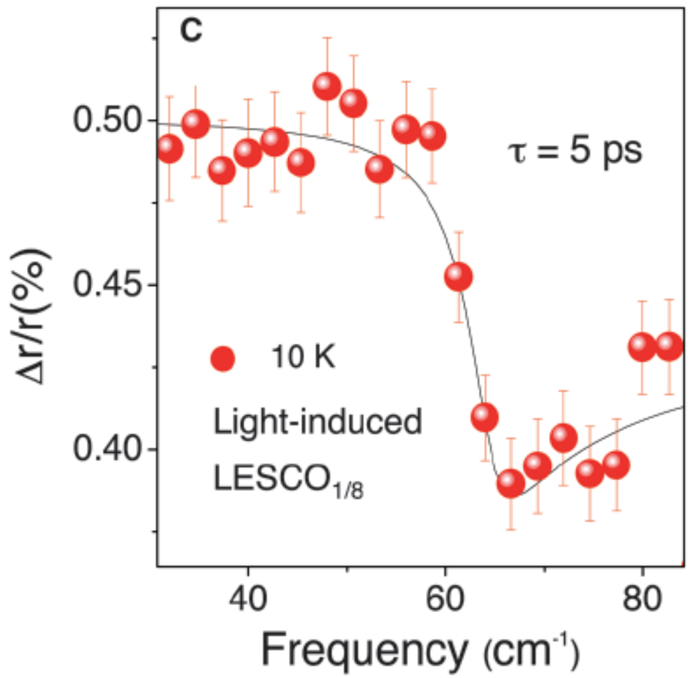


Fig. 1. Schematic phase diagram for $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$. Superconductivity (yellow area) is quenched at all doping levels (gray area) below 0.2, emerging only at very low temperatures. At 0.125 doping, a static 1D modulation of charges and spins, the stripe state, emerges in the planes. This stripe phase (left inset) is associated with a LTT distortion, in which the oxygen octahedrals in the crystal are tilted (right inset). The red dashed curve marks the boundary for superconductivity in compounds of the type $La_{2-x}Sr_xCuO_4$, in which the LTT structural modulation is less pronounced.



compound down to the lowest temperatures. (**C**) Transient *c*-axis reflectance of $LESCO_{1/8}$, normalized to the static reflectance. Measurements are taken at 10 K, after excitation with IR pulses at 16 μm wavelength. The appearance of a plasma edge at 60 cm^{-1} demonstrates that the photoinduced state is superconducting.

Report of Light-induced SC in YBCO far above T_c !

arXiv:1205.4661

PHYSICAL REVIEW B **89**, 184516 (2014)

Optically induced coherent transport far above T_c in underdoped $YBa_2Cu_3O_{6+\delta}$

S. Kaiser,[1,*] C. R. Hunt,[1,4] D. Nicoletti,[1] W. Hu,[1] I. Gierz,[1] H. Y. Liu,[1] M. Le Tacon,[2] T. Loew,[2] D. Haug,[2] B. Keimer,[2] and A. Cavalleri[1,3,†]

nature materials

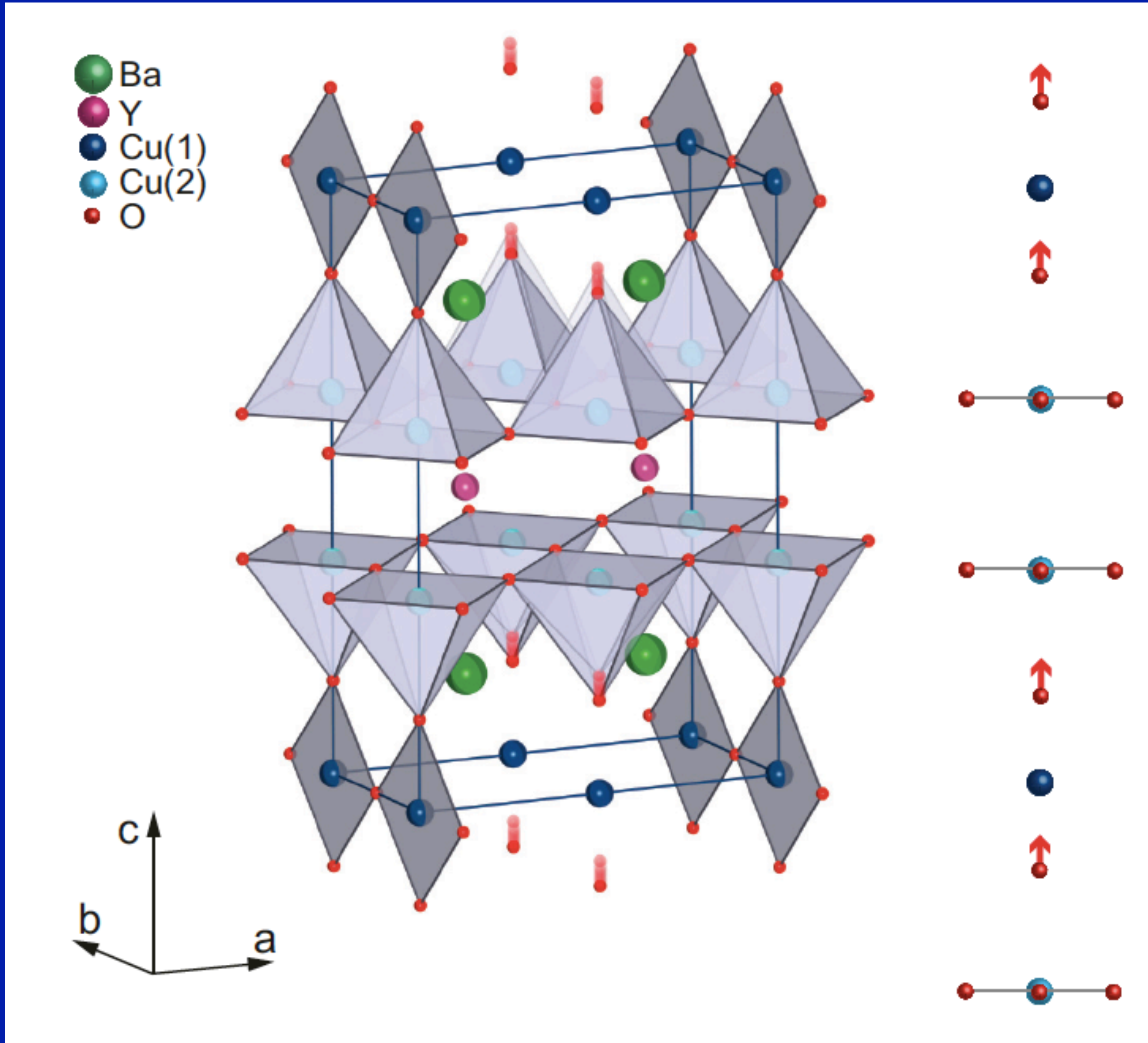
ARTICLES

PUBLISHED ONLINE: 11 MAY 2014 | DOI: 10.1038/NMAT3963

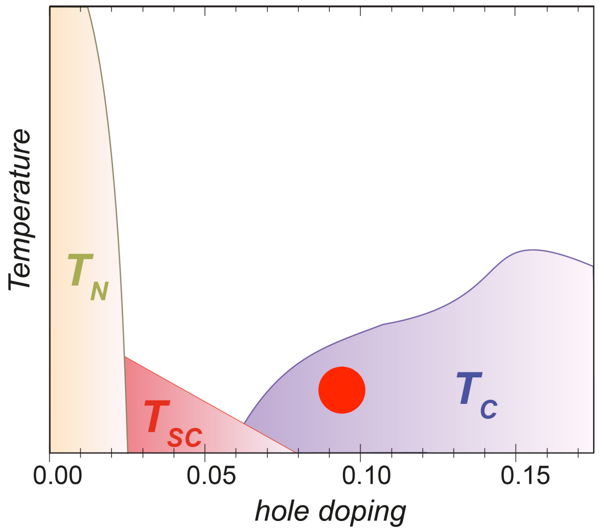
Optically enhanced coherent transport in $YBa_2Cu_3O_{6.5}$ by ultrafast redistribution of interlayer coupling

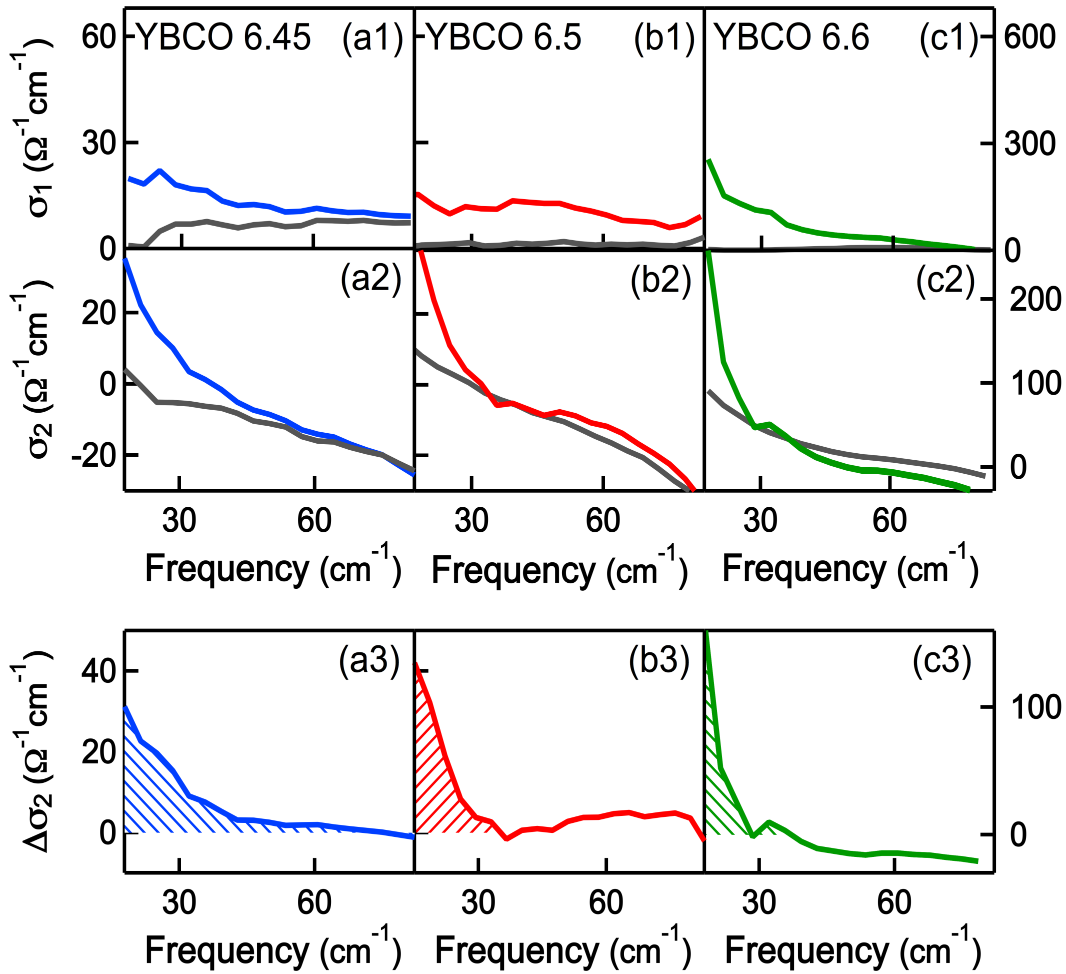
W. Hu[1†], S. Kaiser[1†], D. Nicoletti[1†], C. R. Hunt[1,2†], I. Gierz[1], M. C. Hoffmann[1], M. Le Tacon[3], T. Loew[3], B. Keimer[3] and A. Cavalleri[1,4★]

Pump 20THz B_{1u} mode: shaking apical oxygens



Below T_c: Enhancement of superconductivity





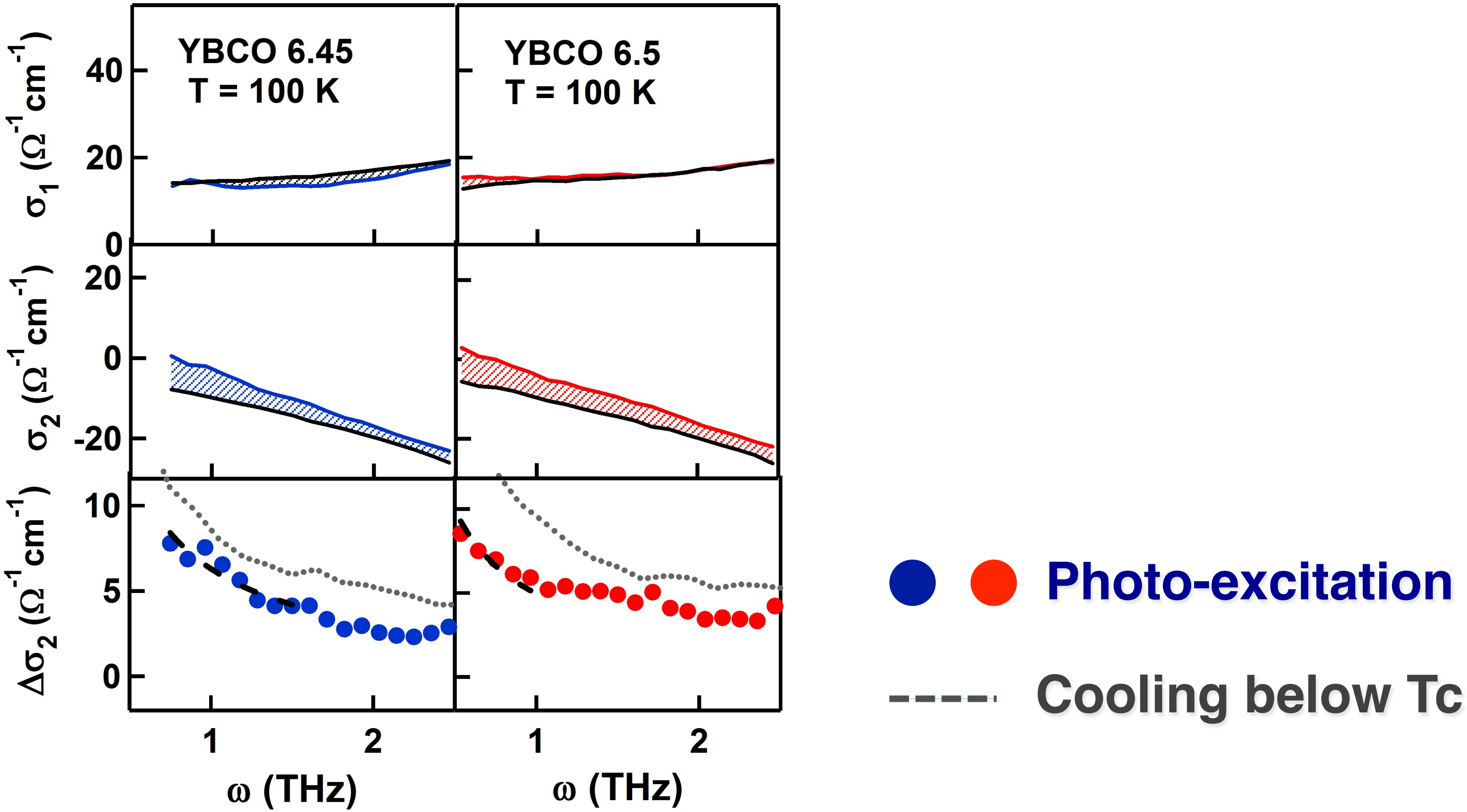
Grey lines: equilibrium
Colour: pumped state @10K
Courtesy A.Cavalleri

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)

Above T_c increase in σ_2 : same as by cooling

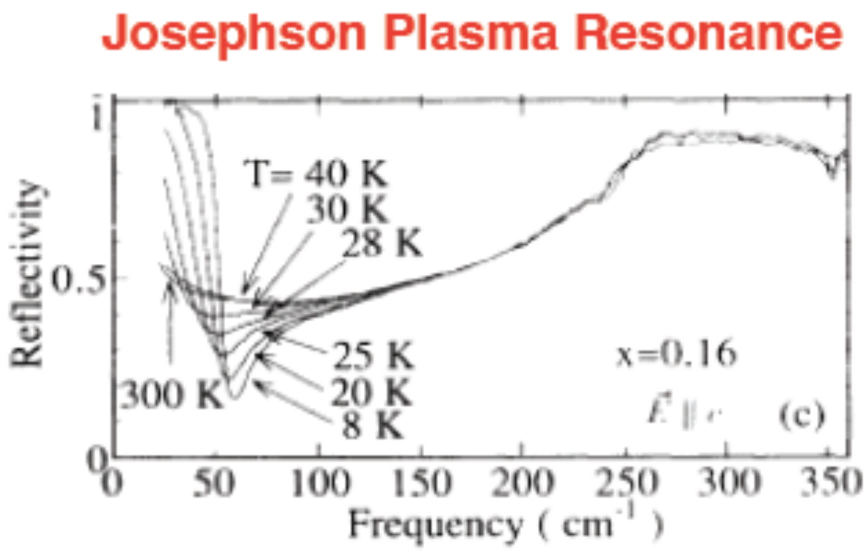
mpsd



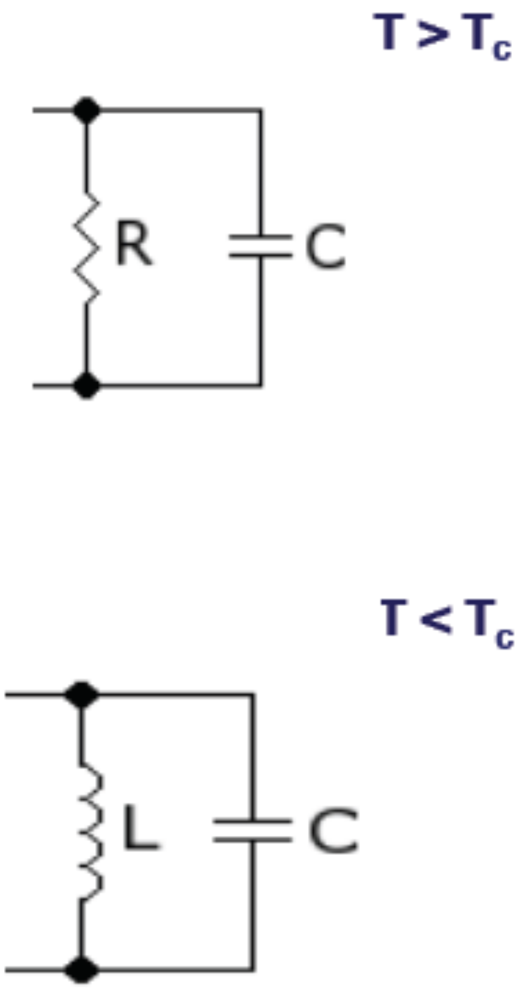
W. Hu. S. Kaiser, D. Nicoletti, C.S. Hunt et al. Nature Materials (2014)

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

Diagnostics: Josephson Plasma Resonance

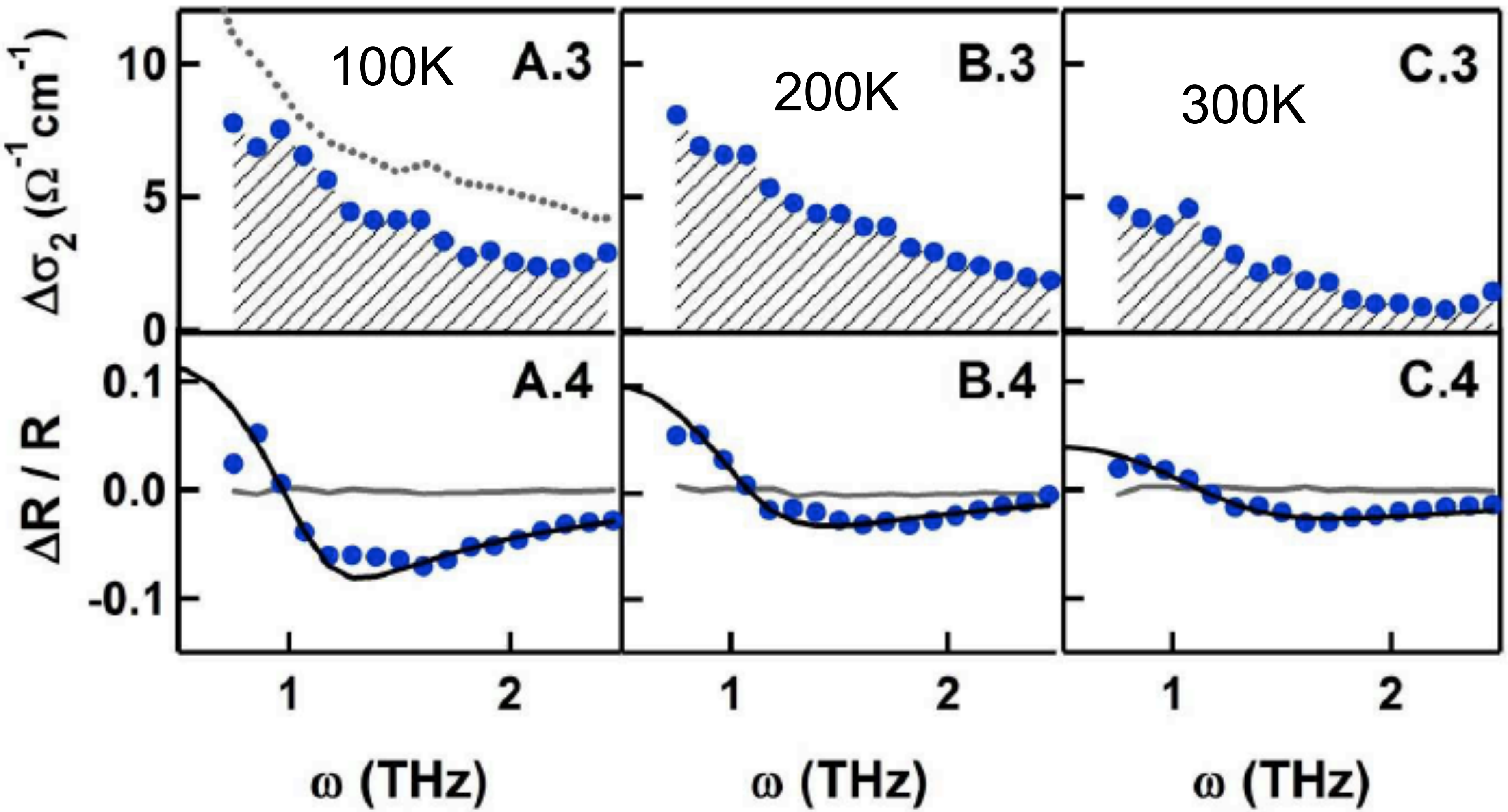


T. Tajima et al., Physical Review Letters 86, 500 (2001)



Above Tc : $YBCO_{6.45}$ 1 ps after excitation

Dotted grey : increase of sigma2 when cooling below Tc at equilibrium



Revealing the light-induced structural changes: Non-Linear Phononics of YBCO

(time-resolved X-ray and theory)

Roman Mankowsky

Alaska Subedi

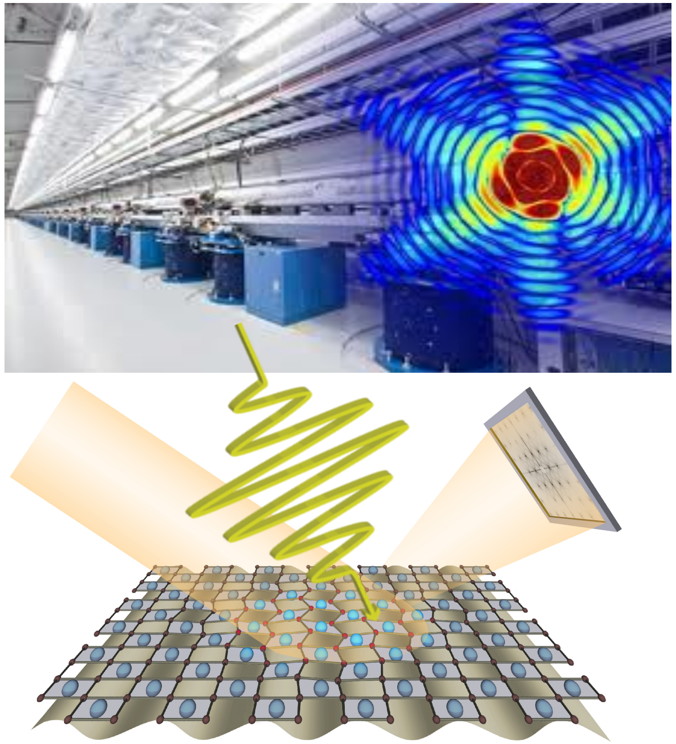
LETTER

doi:10.1038/nature13875

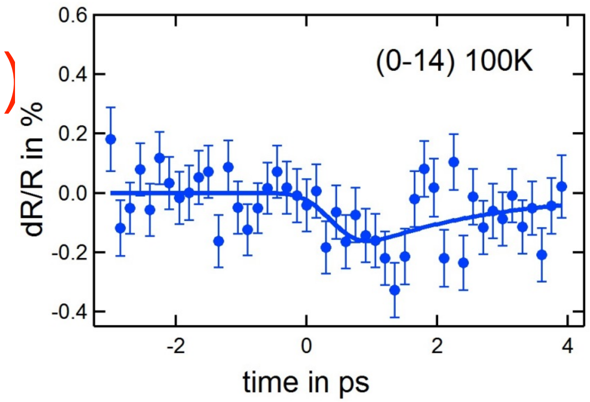
Nonlinear lattice dynamics as a basis for enhanced superconductivity in $YBa_2Cu_3O_{6.5}$

R. Mankowsky[1,2,3]*, A. Subedi[4]*, M. Först[1,3], S. O. Mariager[5], M. Chollet[6], H. T. Lemke[6], J. S. Robinson[6], J. M. Glownia[6], M. P. Minitti[6], A. Frano[7], M. Fechner[8], N. A. Spaldin[8], T. Loew[7], B. Keimer[7], A. Georges[4,9,10] & A. Cavalleri[1,2,3,11]

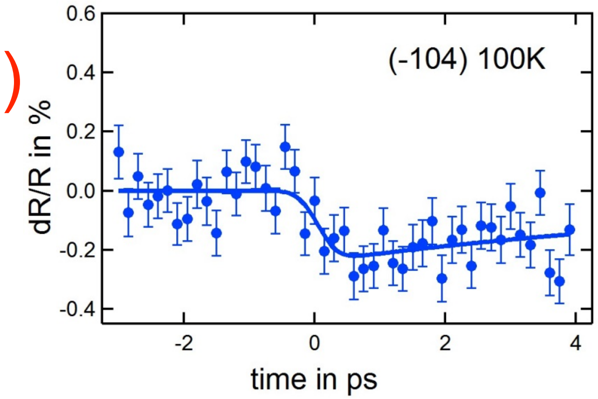
Time-resolved measurements of 4 Bragg peaks



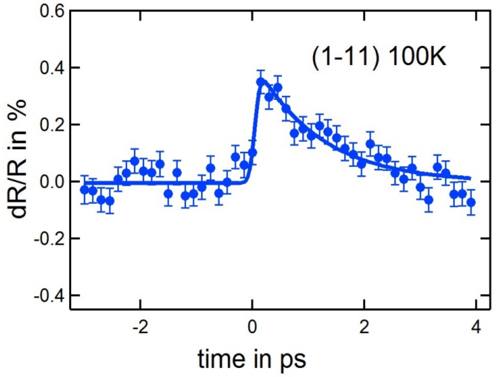
(0,-1,4)



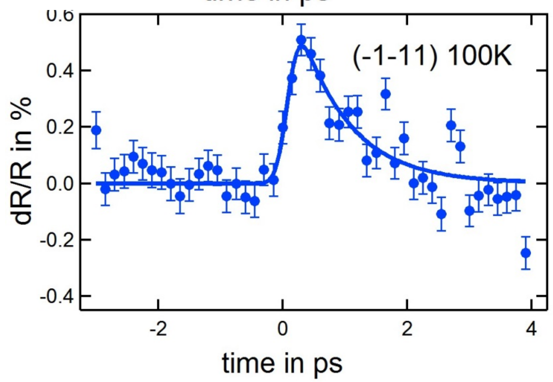
(-1,0,4)



(-1,1,1)

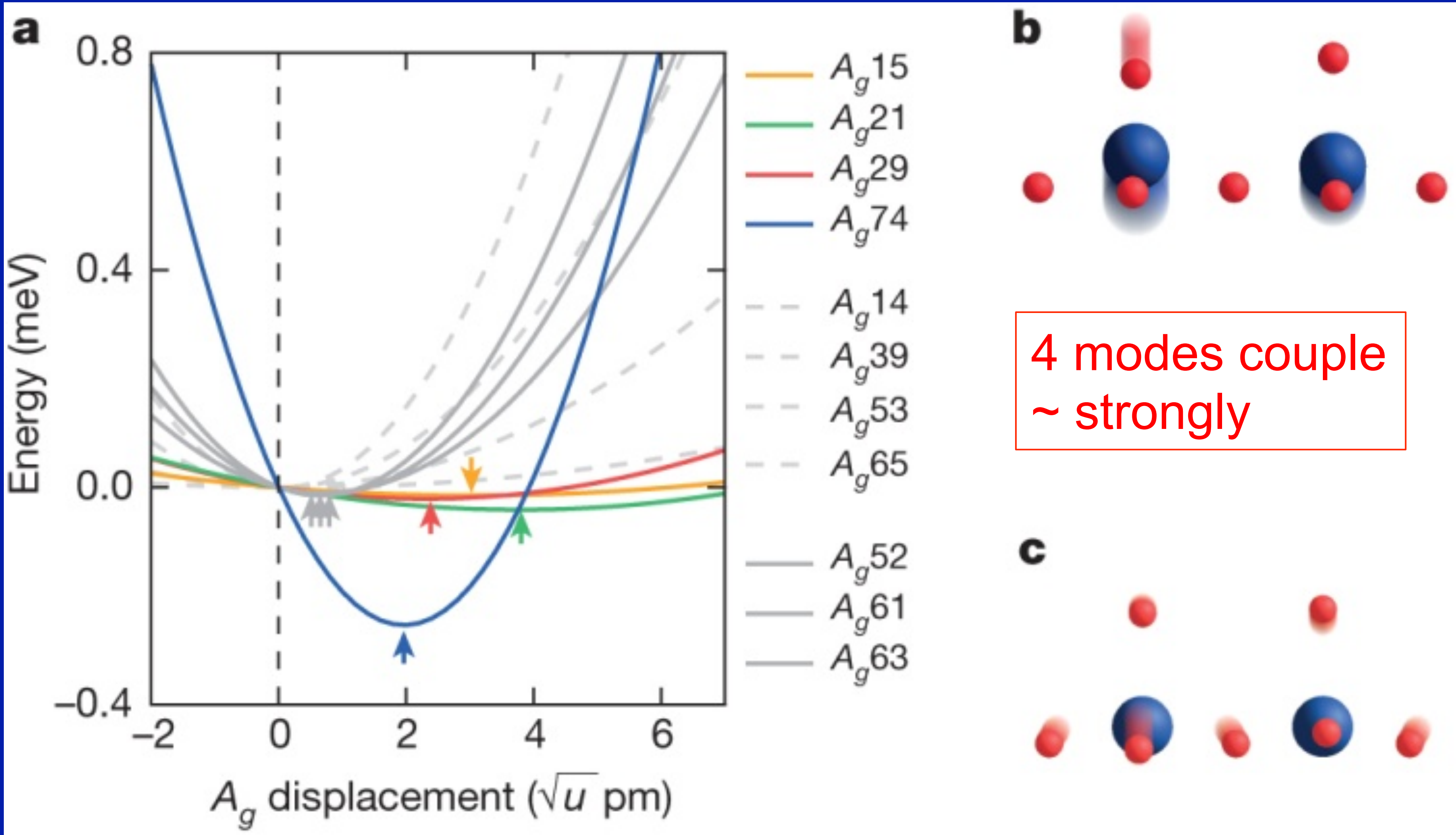


(1,-1,1)



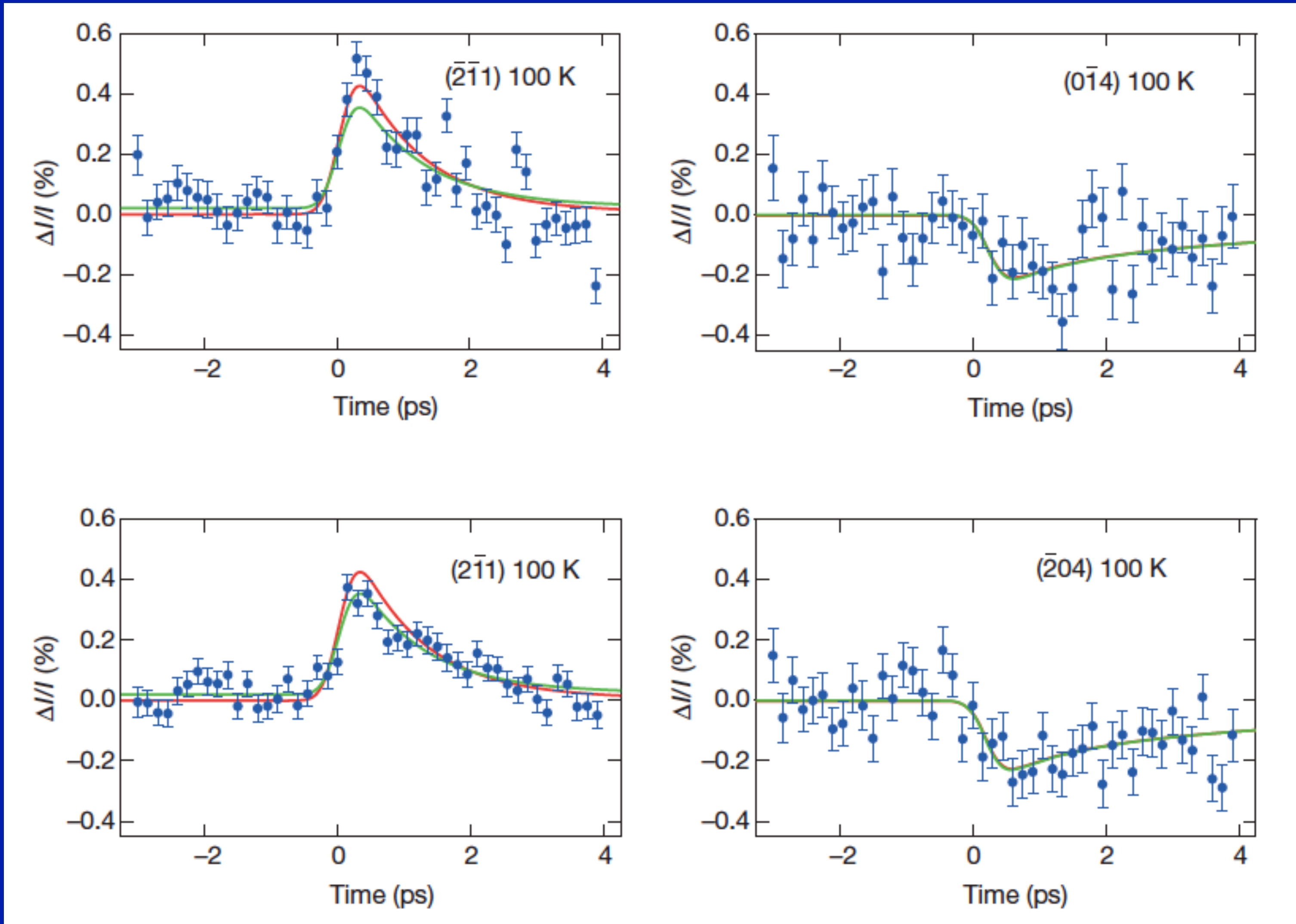
Slide courtesy A.Cavalleri ;R. Mankowski et al. *Nature* (2014)

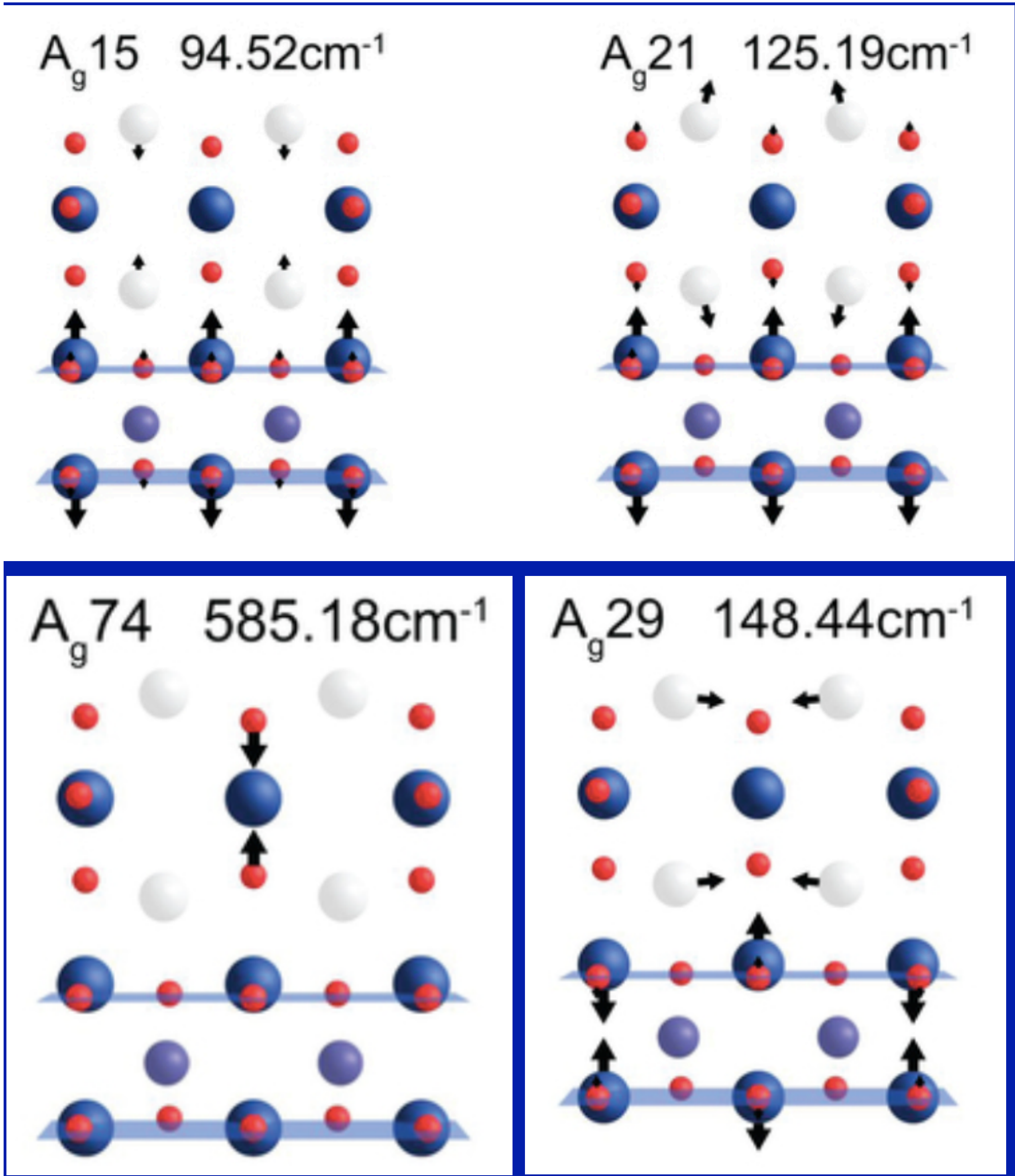
A zoo of phonons…



4 modes couple ~ strongly

Fit of experiment to theory:
1 overall amplitude (and 2 decay constants)





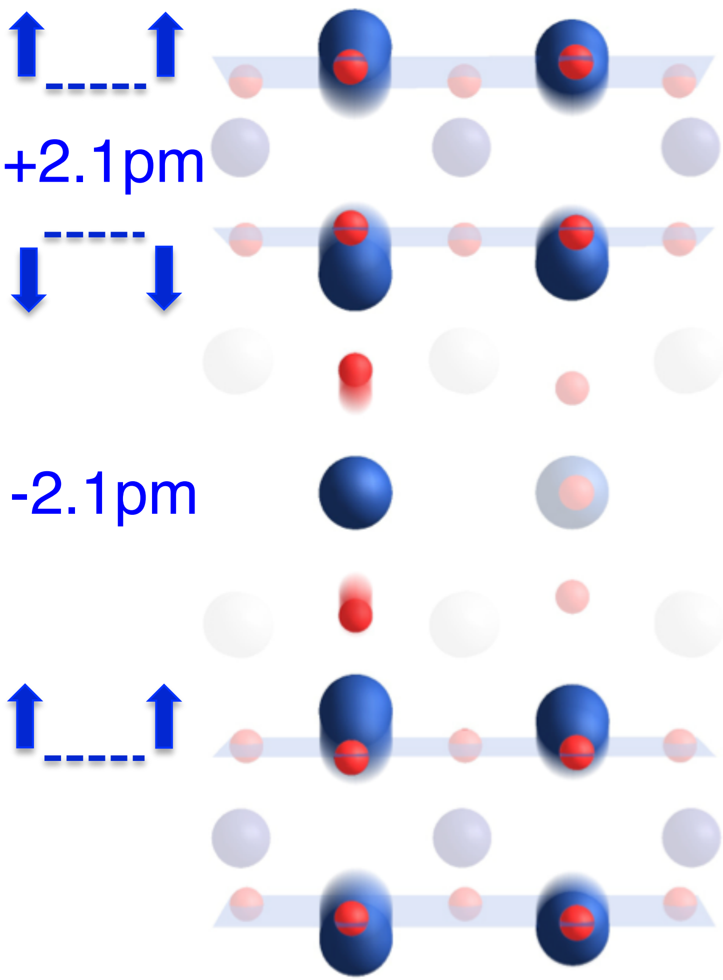
- Buckling of planes INCREASES
- Apical oxygen distance DECREASES slightly (~pm)
- Staggered motion of planes:

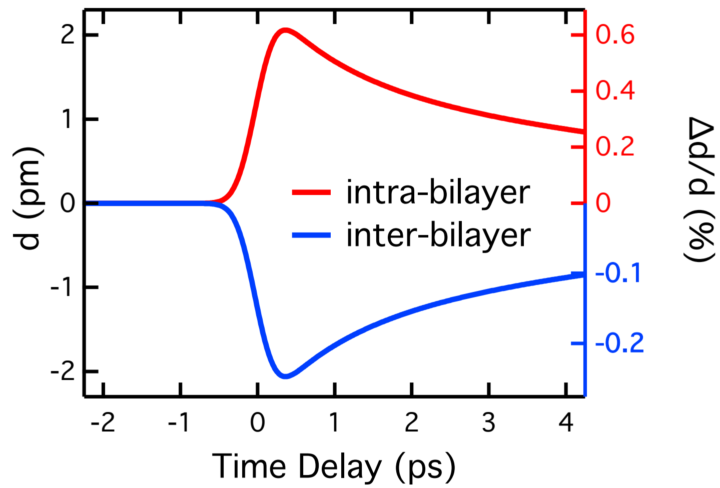
intra-bilayer distance increases

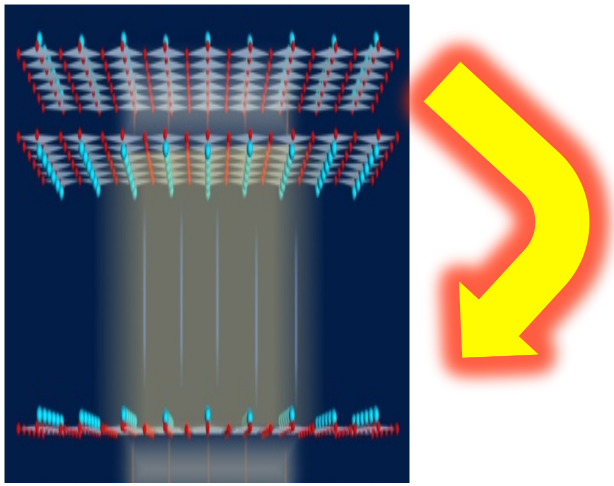
Inter bilayer decreases

Possible mechanisms from observed/calculated transient structural changes

1) Staggered motion of the planes



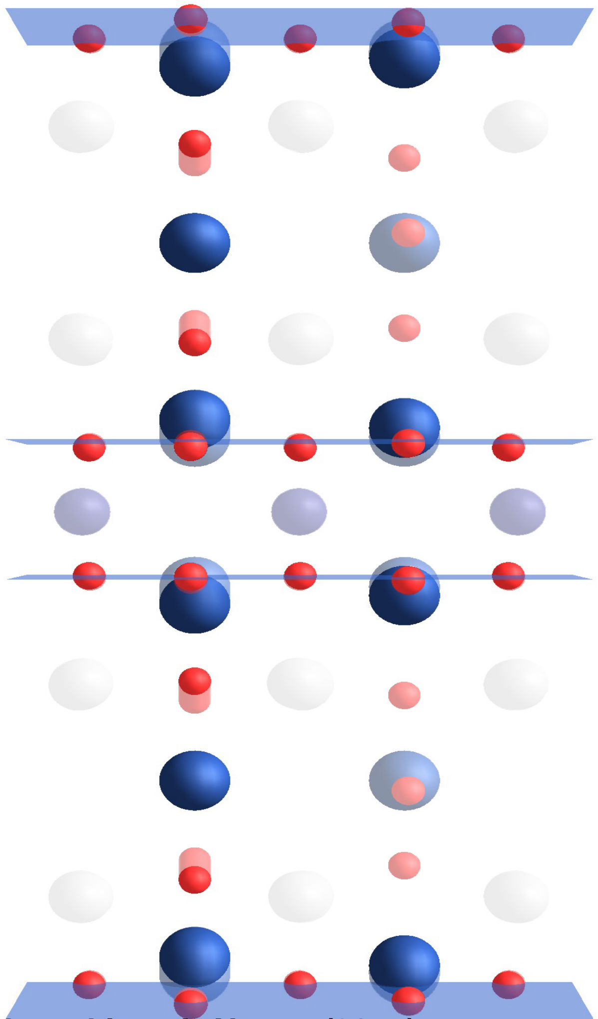


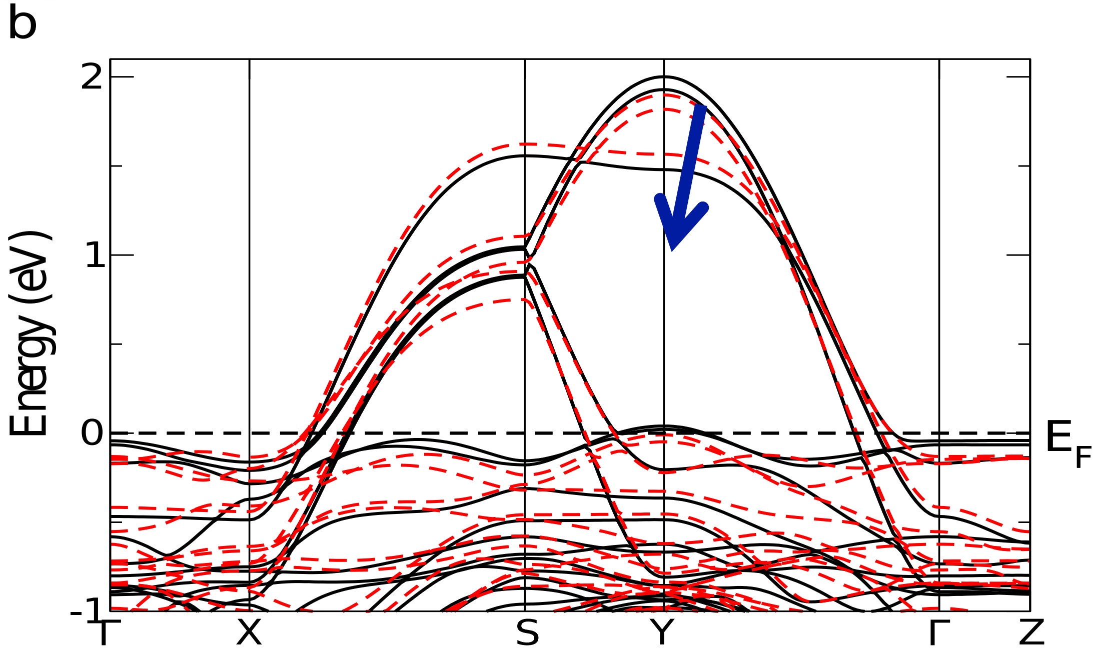


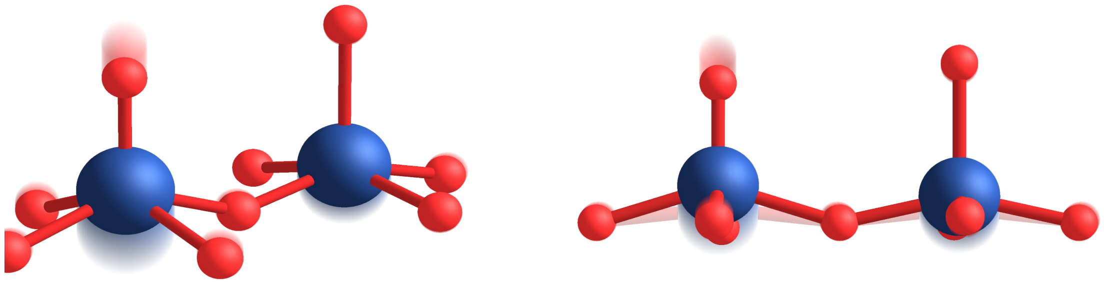
Slide by courtesy of A.Cavalleri

2) Changes of in-plane electronic structure:

(caution: from LDA) O-deficient chain band moves lower in energy



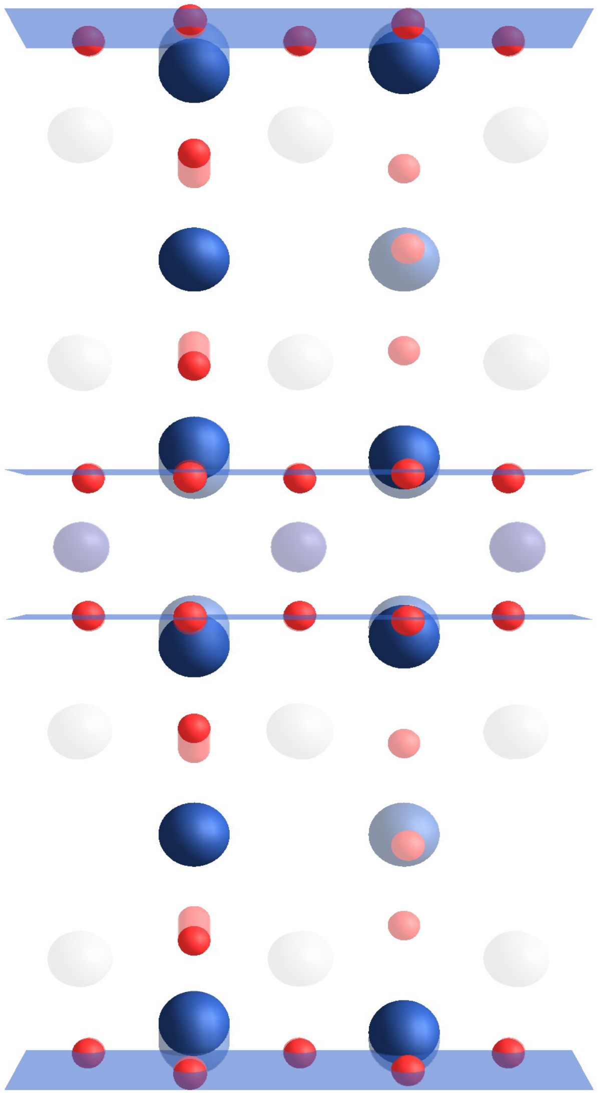


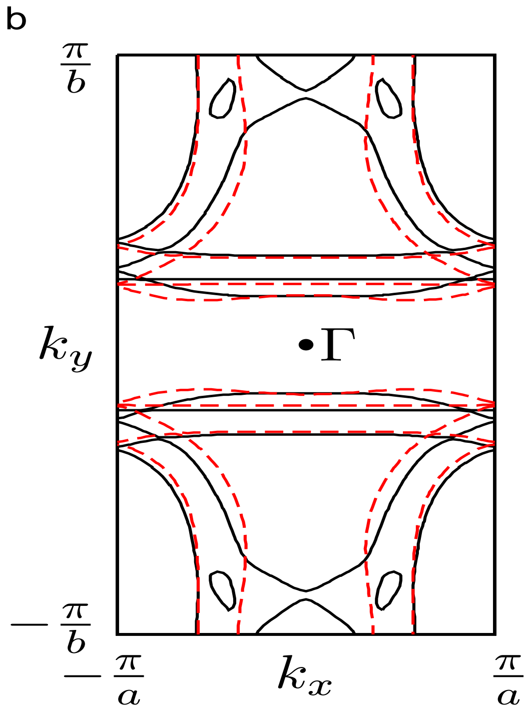


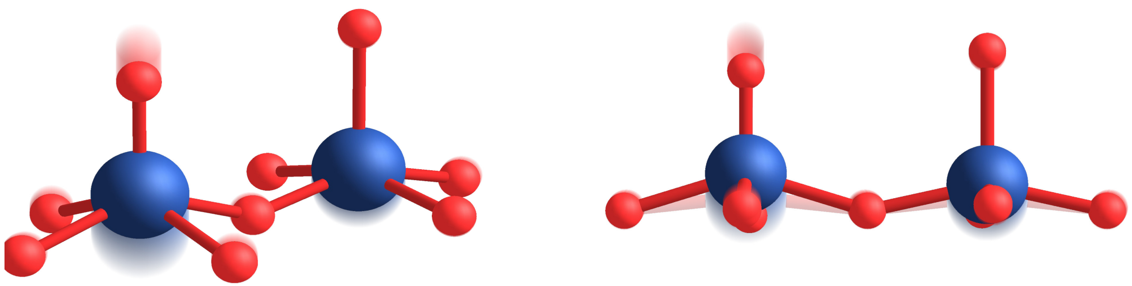
R. Mankowski et al. *Nature* (2014)

Slide by courtesy of A.Cavalleri

LDA calculations show more x^2-y^2







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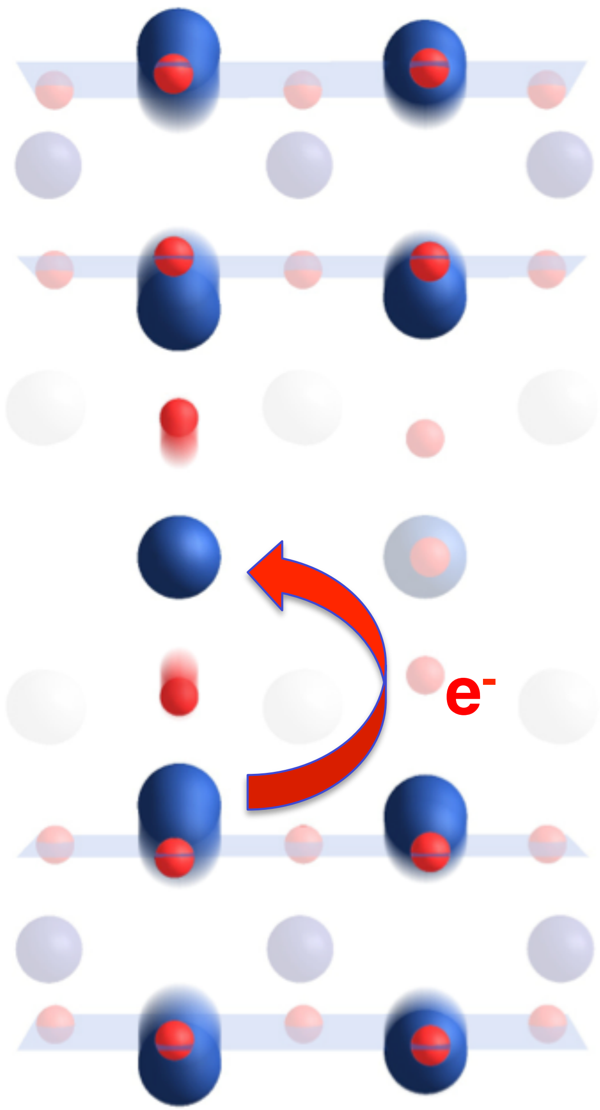
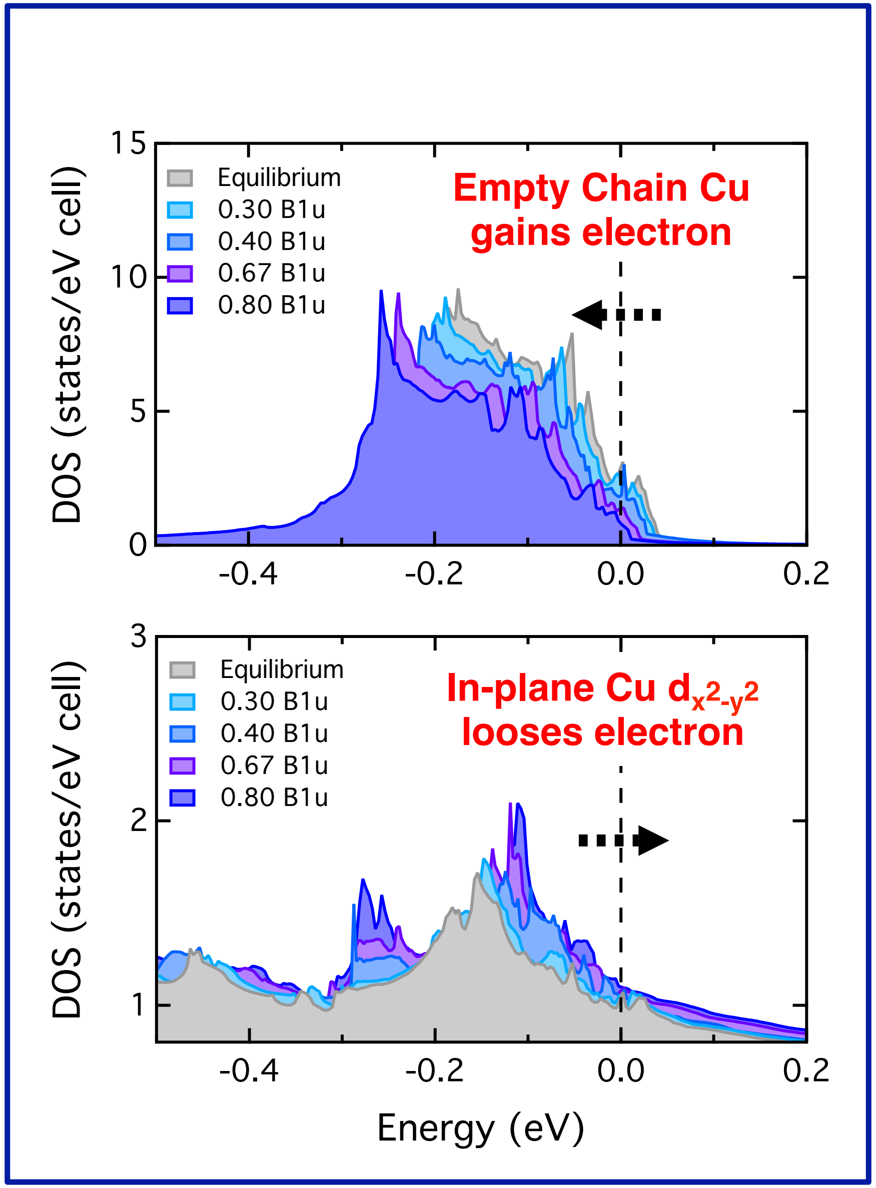
Charge transfer from the planes to the chains



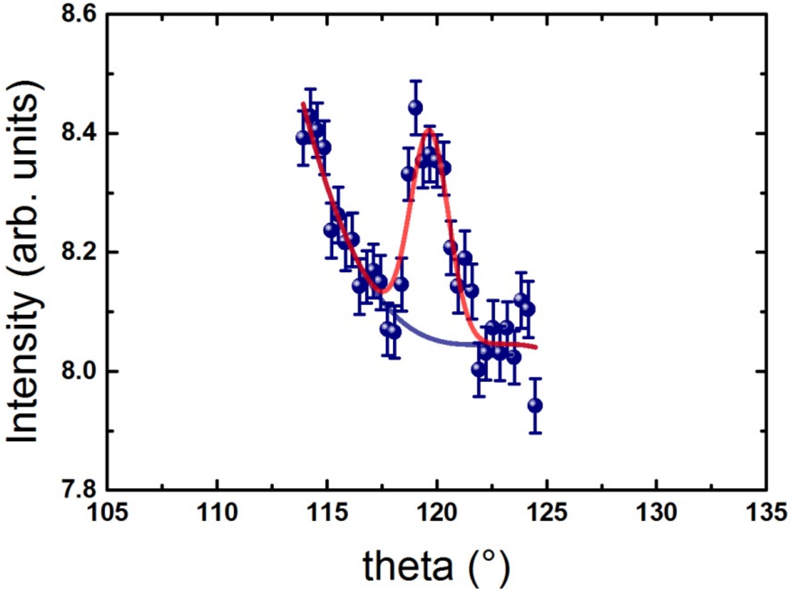


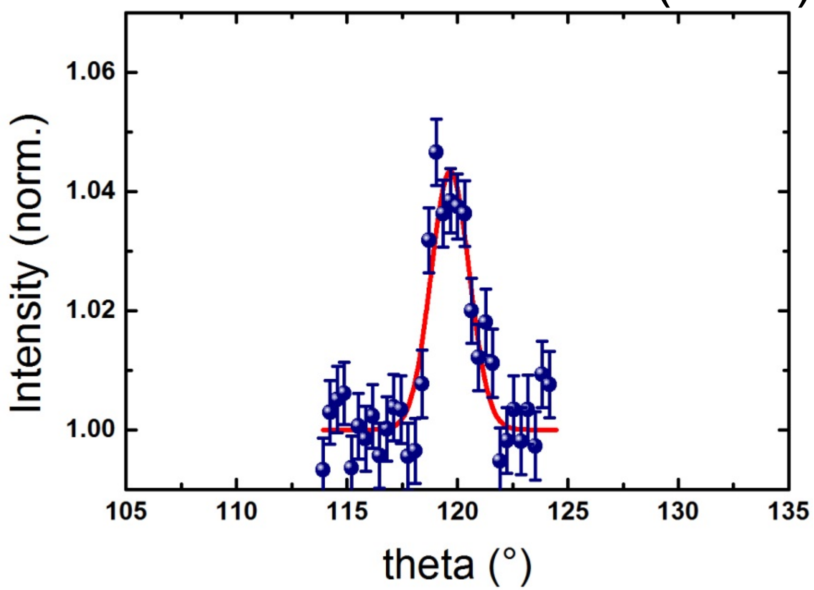
See also UBC EELS Nature Communications

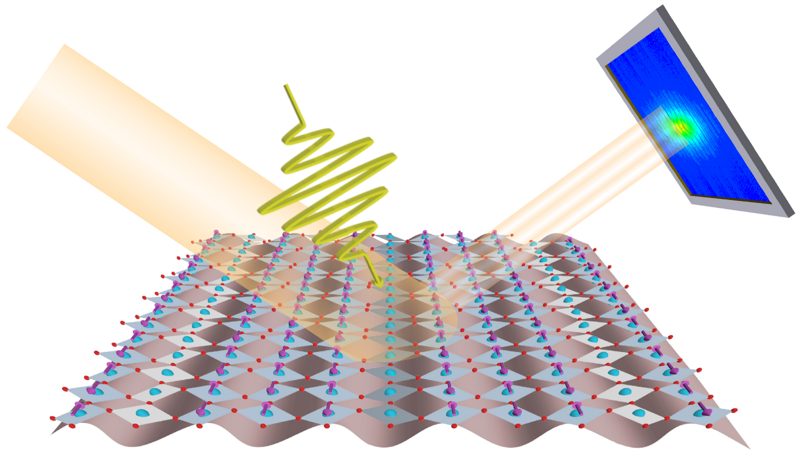
Slide by courtesy of A.Cavalleri

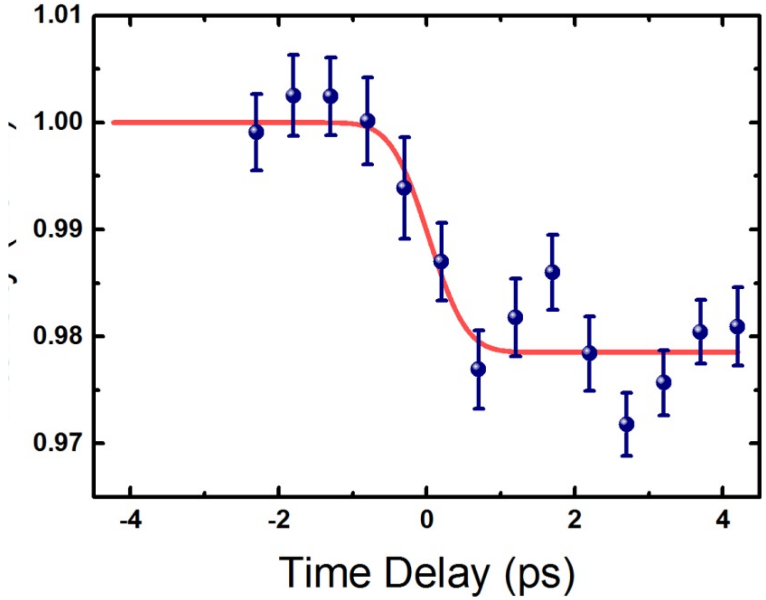
3) Charge order is melted ($YBCO_{6.6}$)

Foerst et al. PRB 90 184514 (2014)









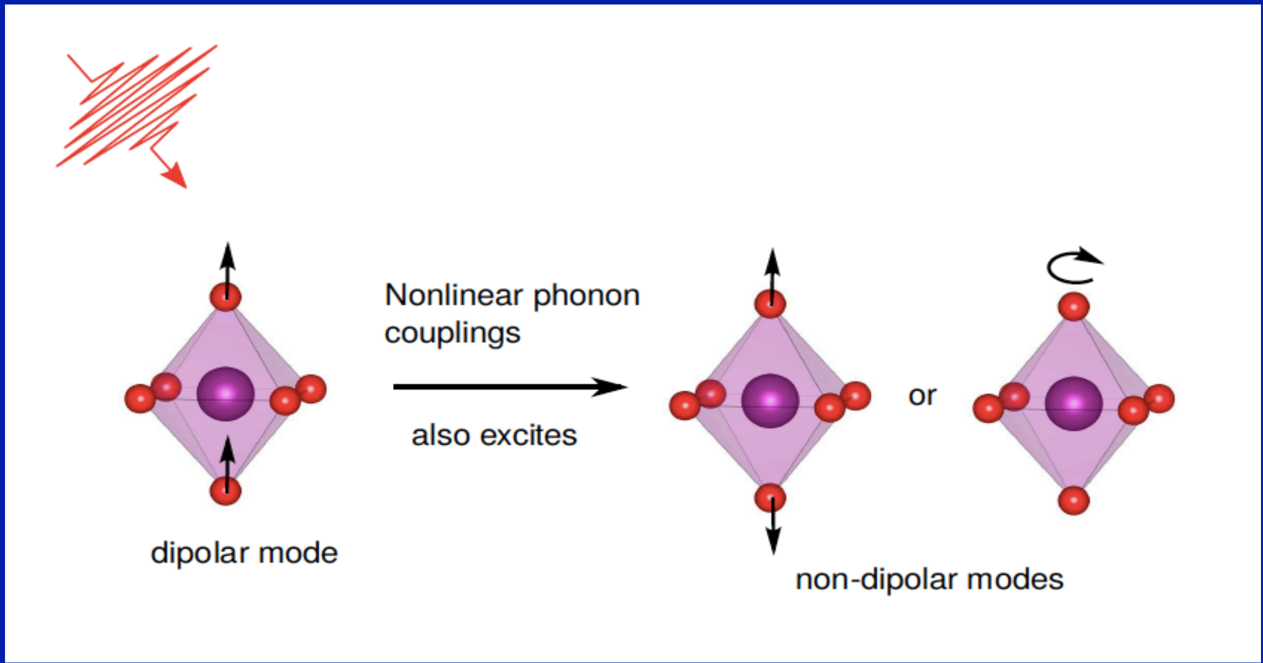
Slide by courtesy of A.Cavalleri

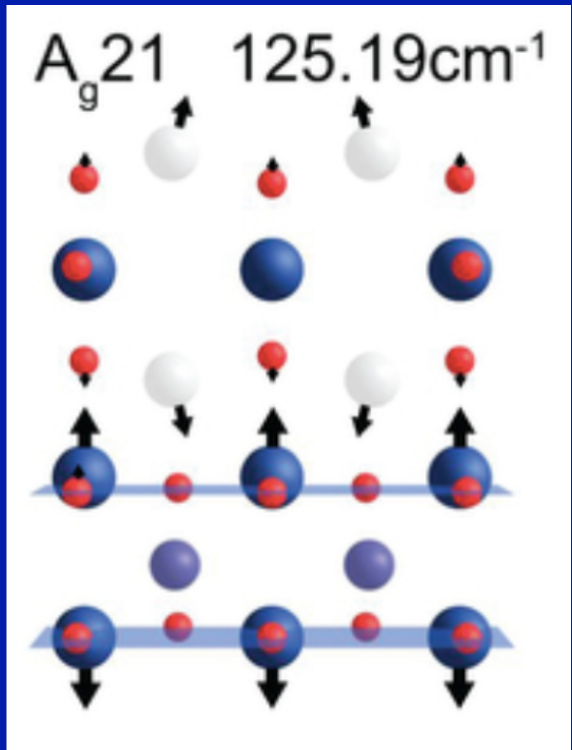
Other possible explanations…

- New driven (Floquet) state by direct coupling of the pumped mode to electronic structure
- Effective cooling of phase fluctuations
- Other …?

Take-Home Message: Selective Control of Quantum Materials through Non-Linear Phononics

Qualitative idea: Först et al. Nature Phys 7, 854 (2011)
Microsopic theory: Subedi, Cavalleri and AG, PRB 89 22031R (2014)





Transient structure of driven YBCO:
Mankowsky et al. Nature 516, 71 (2014)