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Time-resolved Diffuse Scattering: phonon spectoscopy with ultrafast x rays

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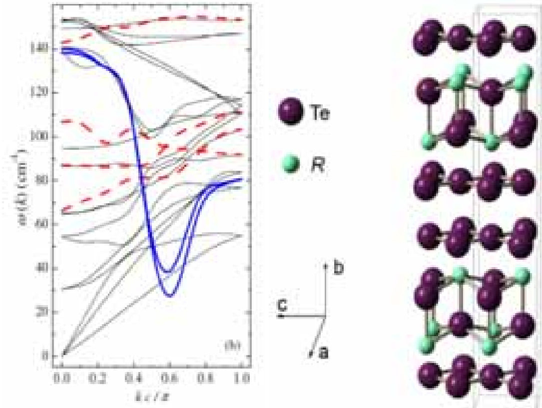
Science at the Hard X-ray Diffraction Limit: Ultrafast Science… Cornell, June 20–21, 2011

phonons play defining role in materials properties

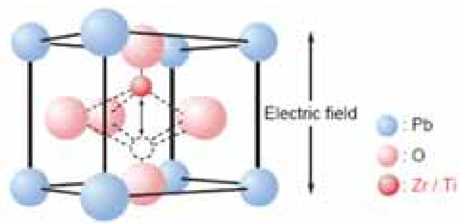
PULSE

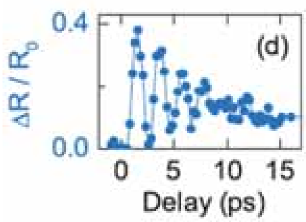
Electrical and thermal conductivity

materials with coupled degrees of freedom

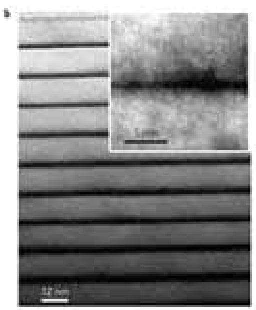


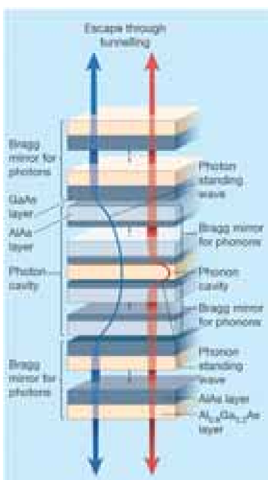
Ferroelectricity

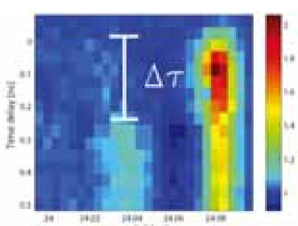




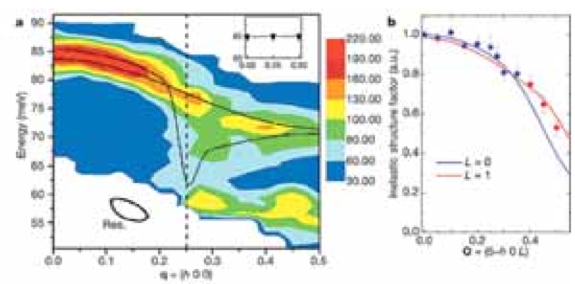
Acoustic phonon "engineering"





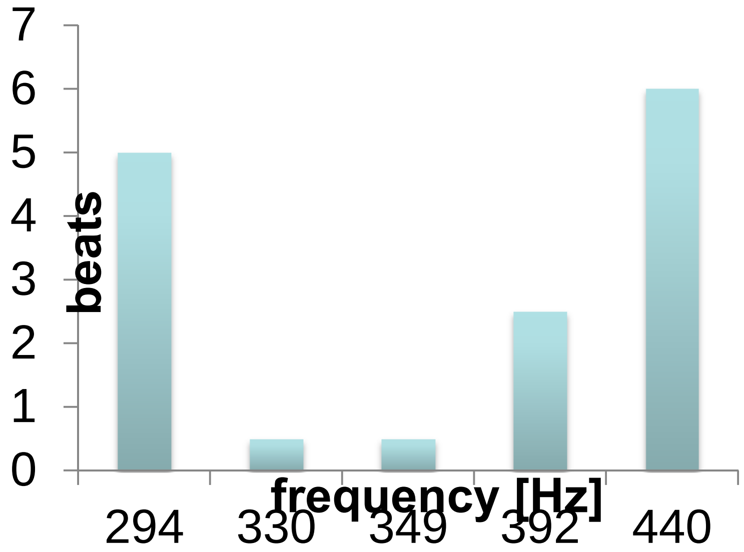


Superconductivity



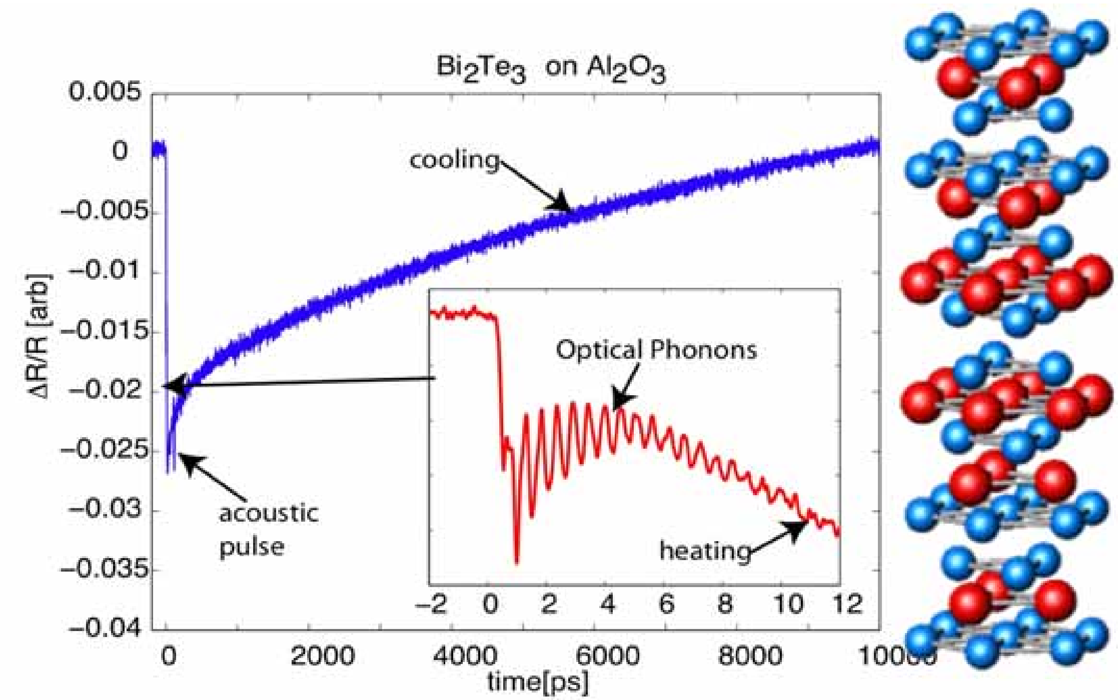
Ultrafast Tickle and Probe

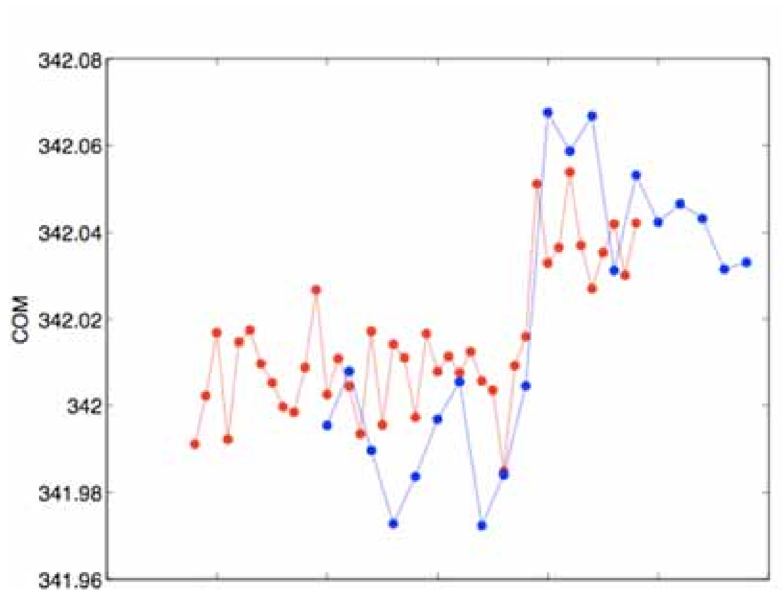
Why do physics in the time domain?



Spectra alone are sometimes inadequate

Separation of time-scales

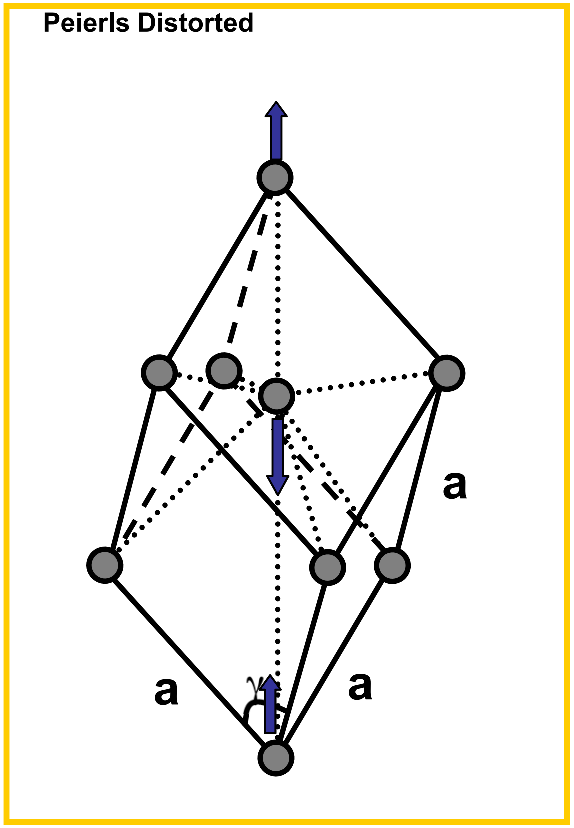


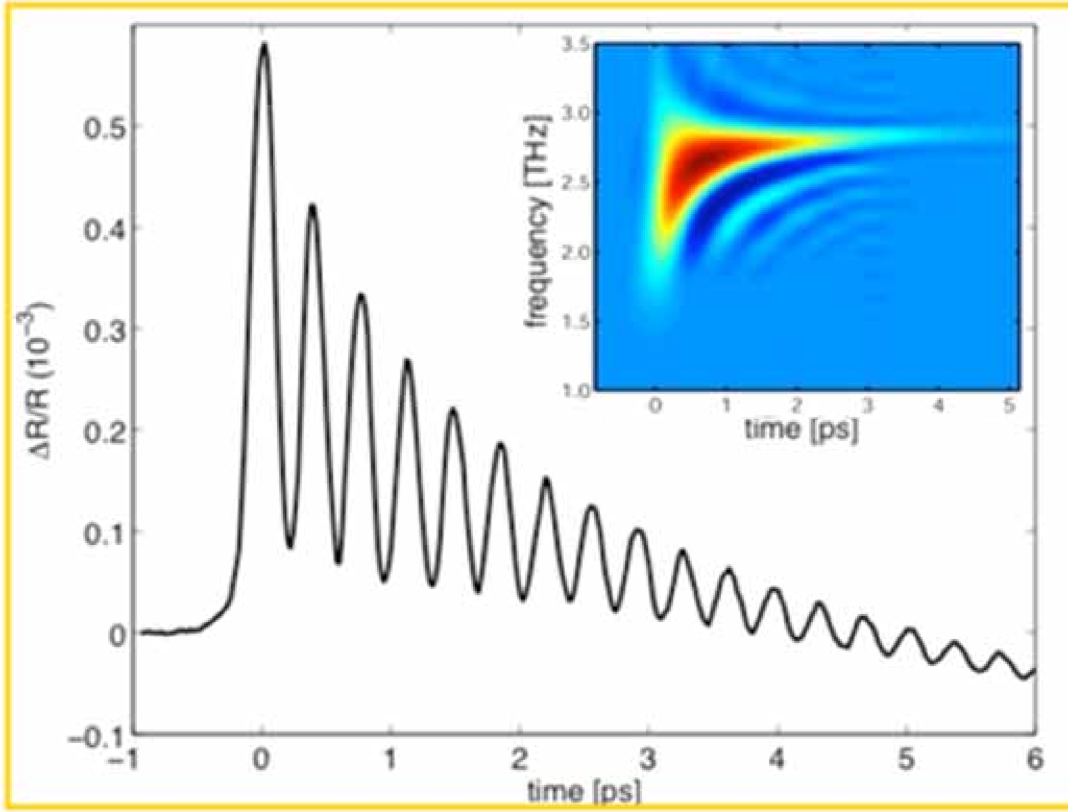


Sheu *et al.* unpublished

Trigo *et al.* unpublished

Example: Photoexcited bismuth (all optical experiments)

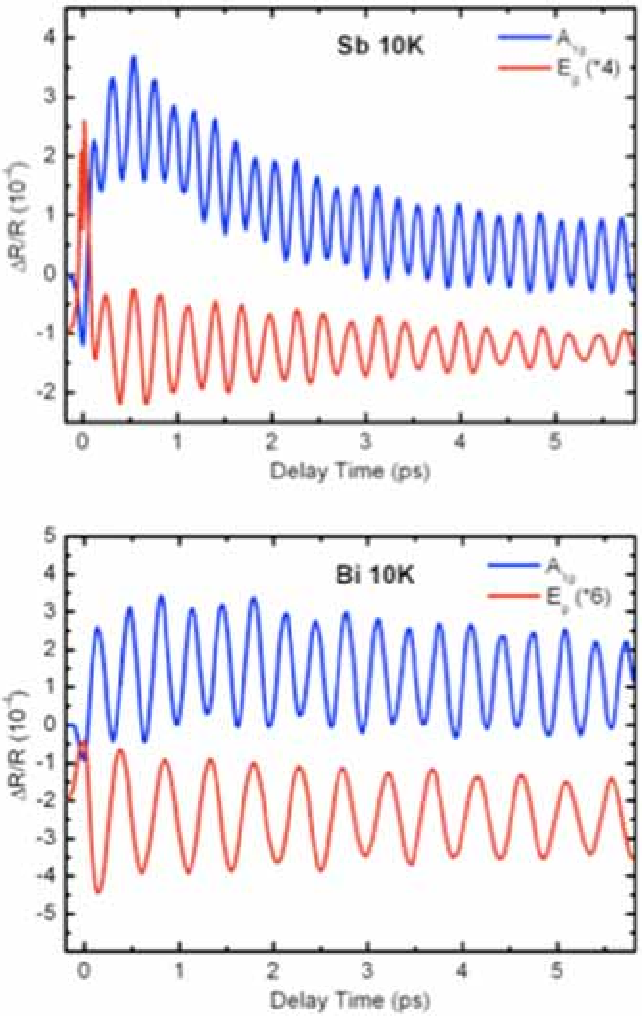


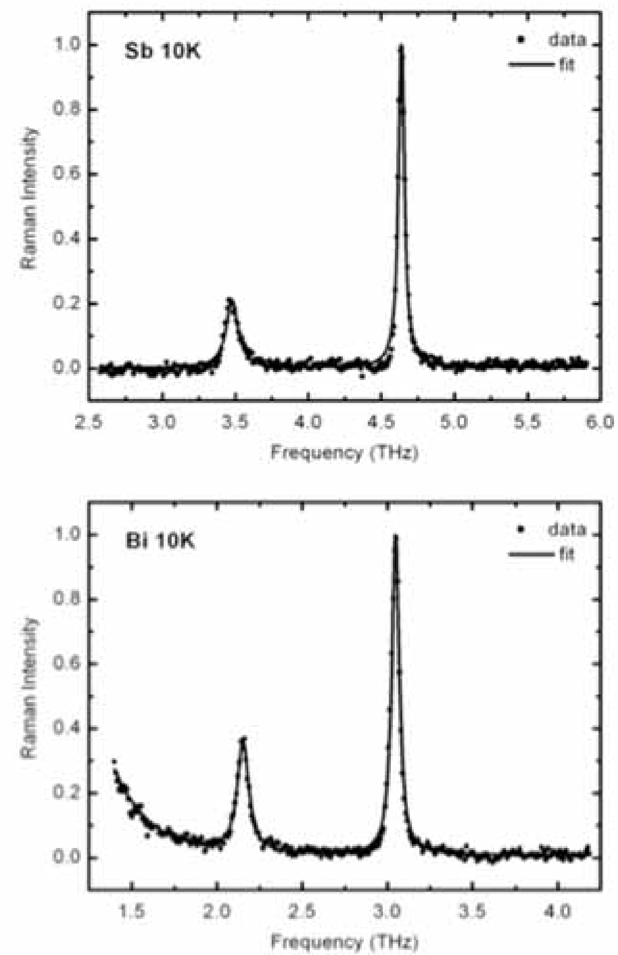


Coherent A_{1g} Mode is strongly softened and chirped.

Murray *et al.* PRB 72, 060301 (R) 2005.

Low excitation (tickle regime)
Pump-probe vs. Spontaneous Raman

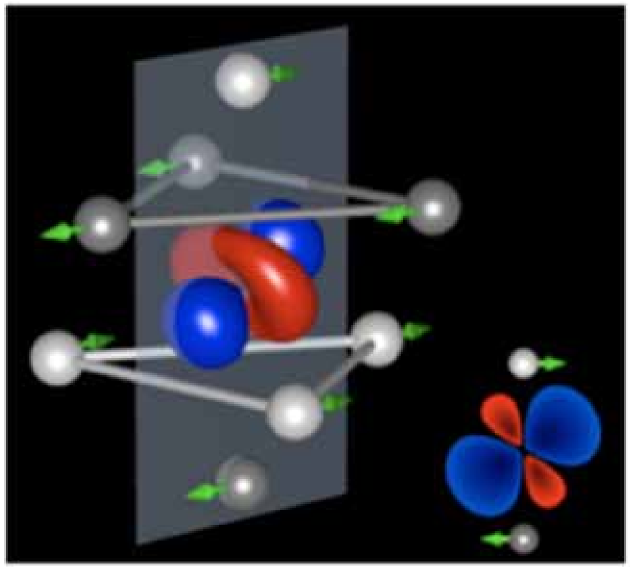


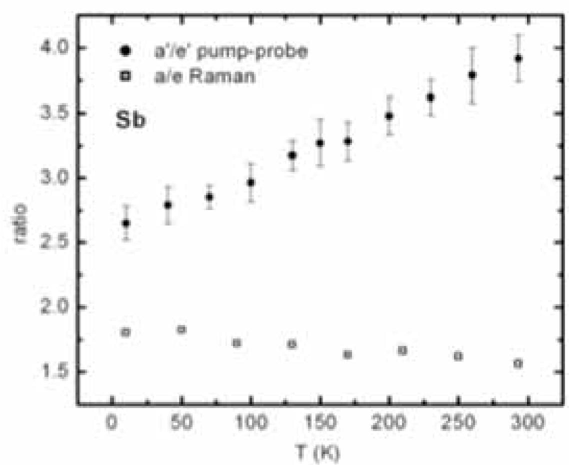


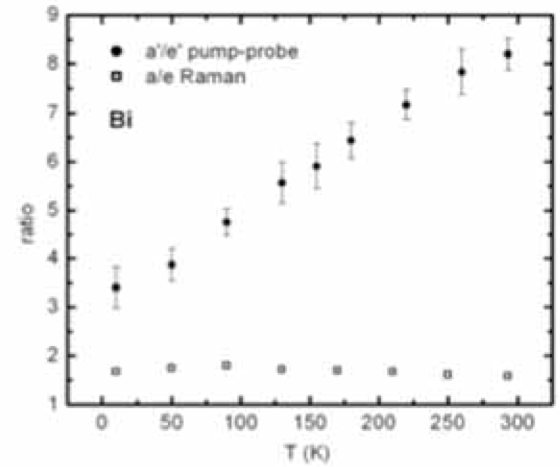
Collaboration with Roberto Merlin, UM

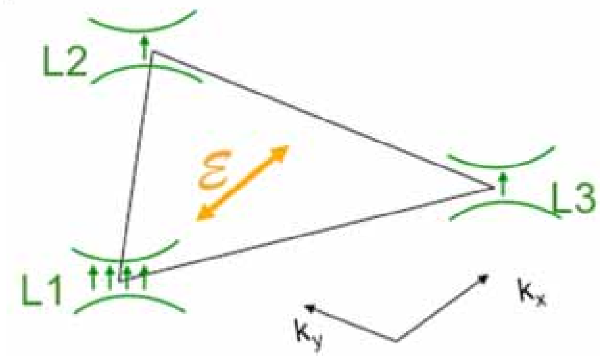
Jian Chen, Jinjing Li, unpublished

Subtle difference between the two…







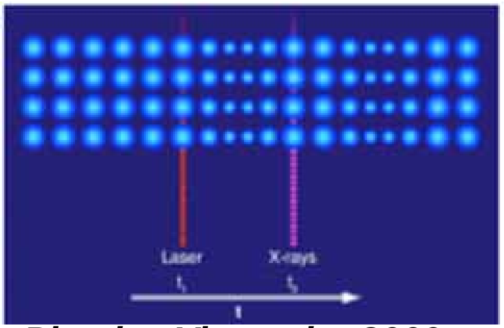


$$F(t)=\begin{cases} F_0 e^{-\Gamma t} & t \ge 0 \\ 0 & t < 0 \end{cases}$$

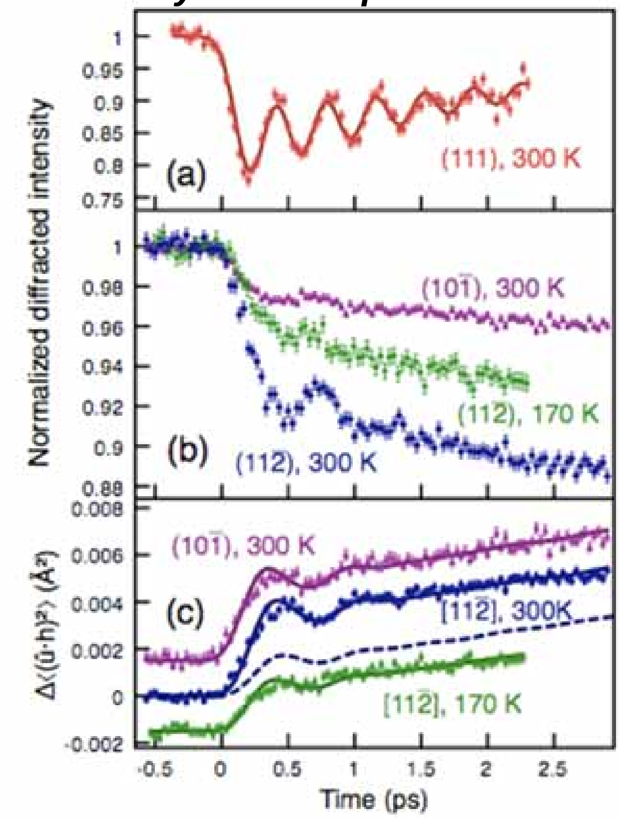
$$Q(t)=\int_{-\infty}^{t}\frac{\sin[\Omega(t-\tau)]}{\Omega}e^{-\gamma(t-\tau)}F(\tau)d\tau=\frac{F_0 e^{-\Gamma t}}{\Omega^2+(\Gamma-\gamma)^2}-\frac{F_0 e^{-\gamma t}\cos(\Omega t+\varphi)}{\Omega\sqrt{\Omega^2+(\Gamma-\gamma)^2}}$$

Jian Chen, Jinjing Li, unpublished

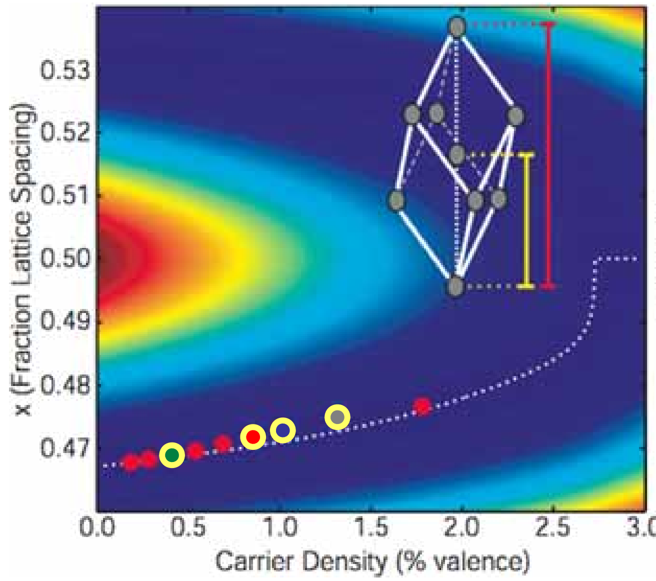
Electronic Softening in Bi by femtosecond X-ray Diffraction

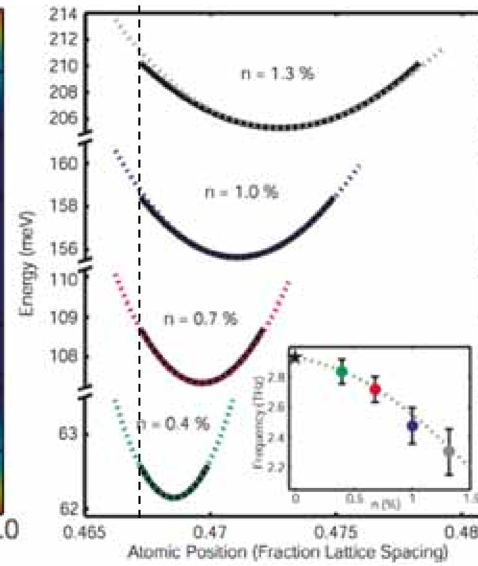


Reis *Physics Viewpoint 2009*



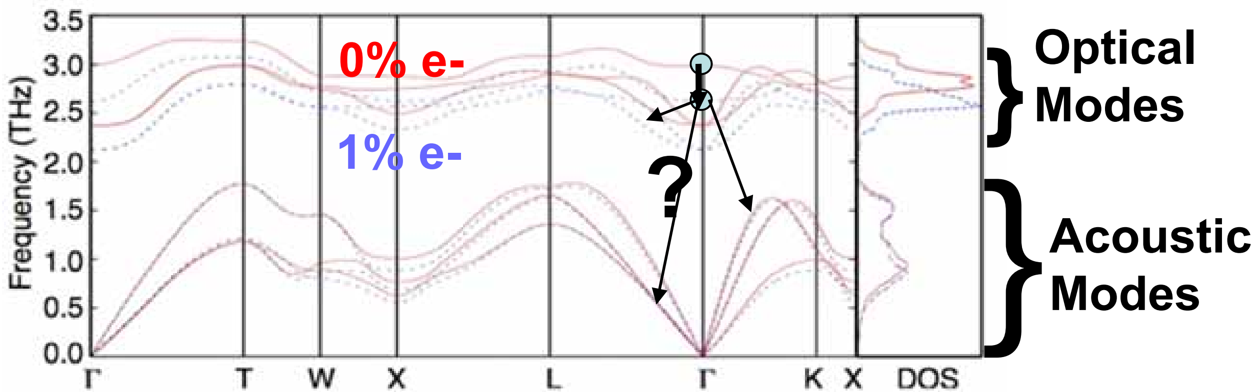
Johnson *et al.* PRL 2009.





D. M. Fritz et al. Science 315, 2007.

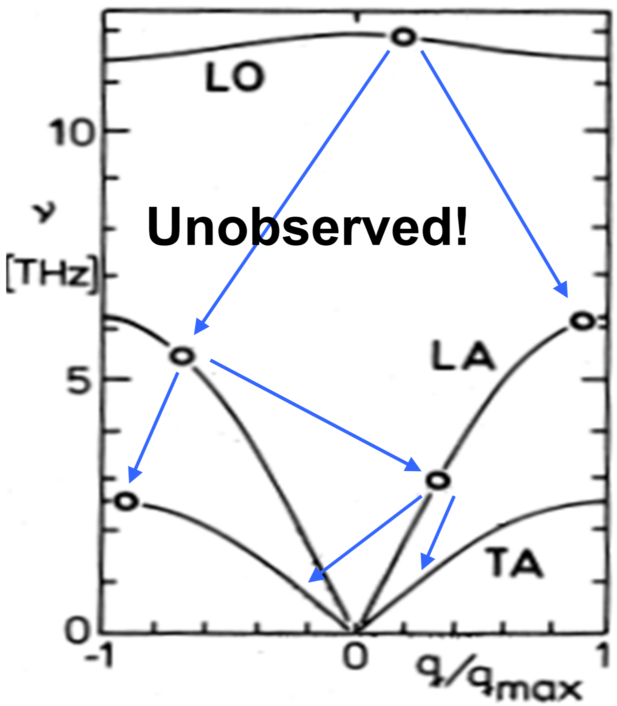
DFPT calculation



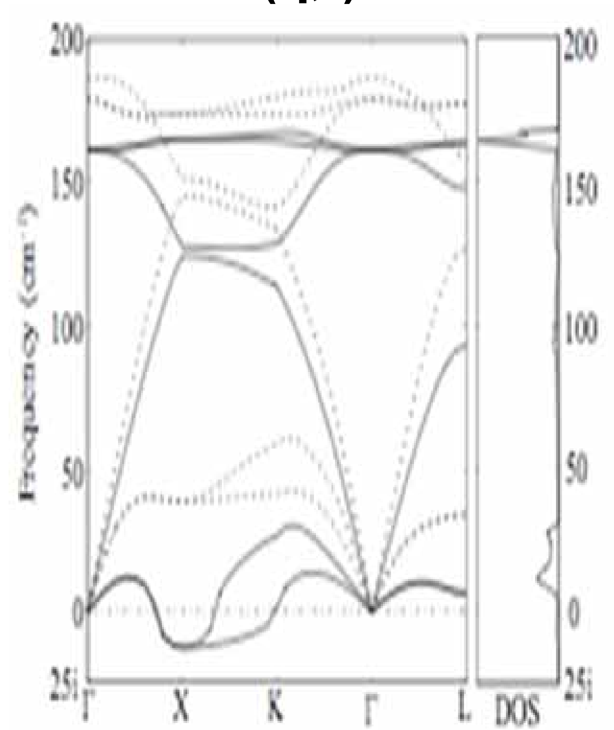
Murray *et al.* PRB 75 2007.

Momentum and Time-resolved Phonon Spectroscopies virtually nonexistent

n(q,t)

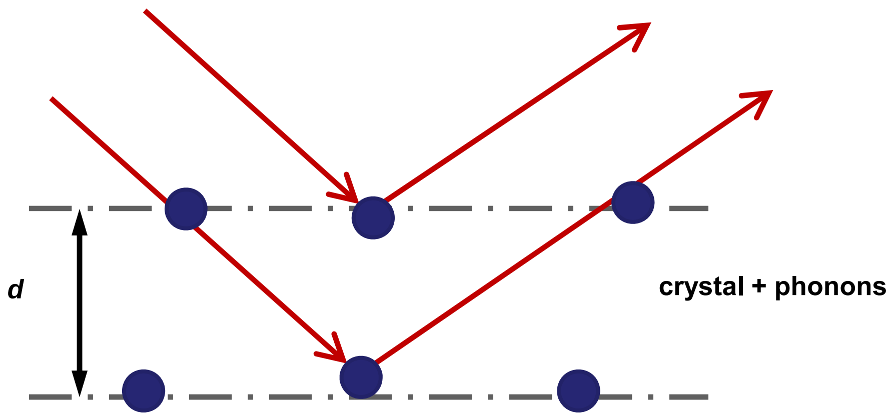


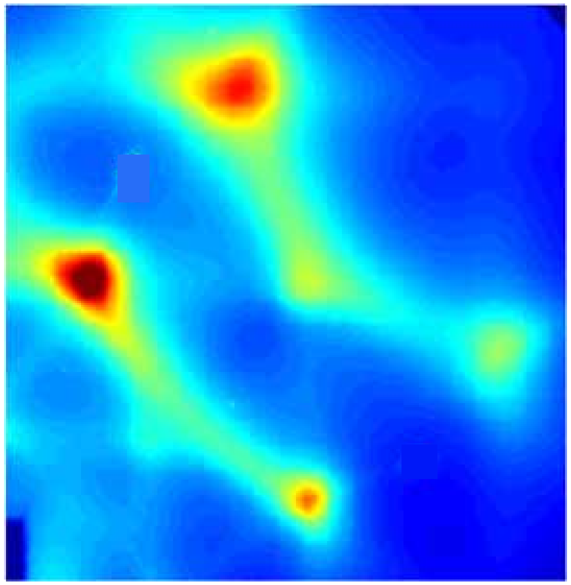
ω(q,t)



phonon-phonon and electron-phonon coupling, interatomic forces.

x-ray diffuse scattering:
measure deviations from average structure

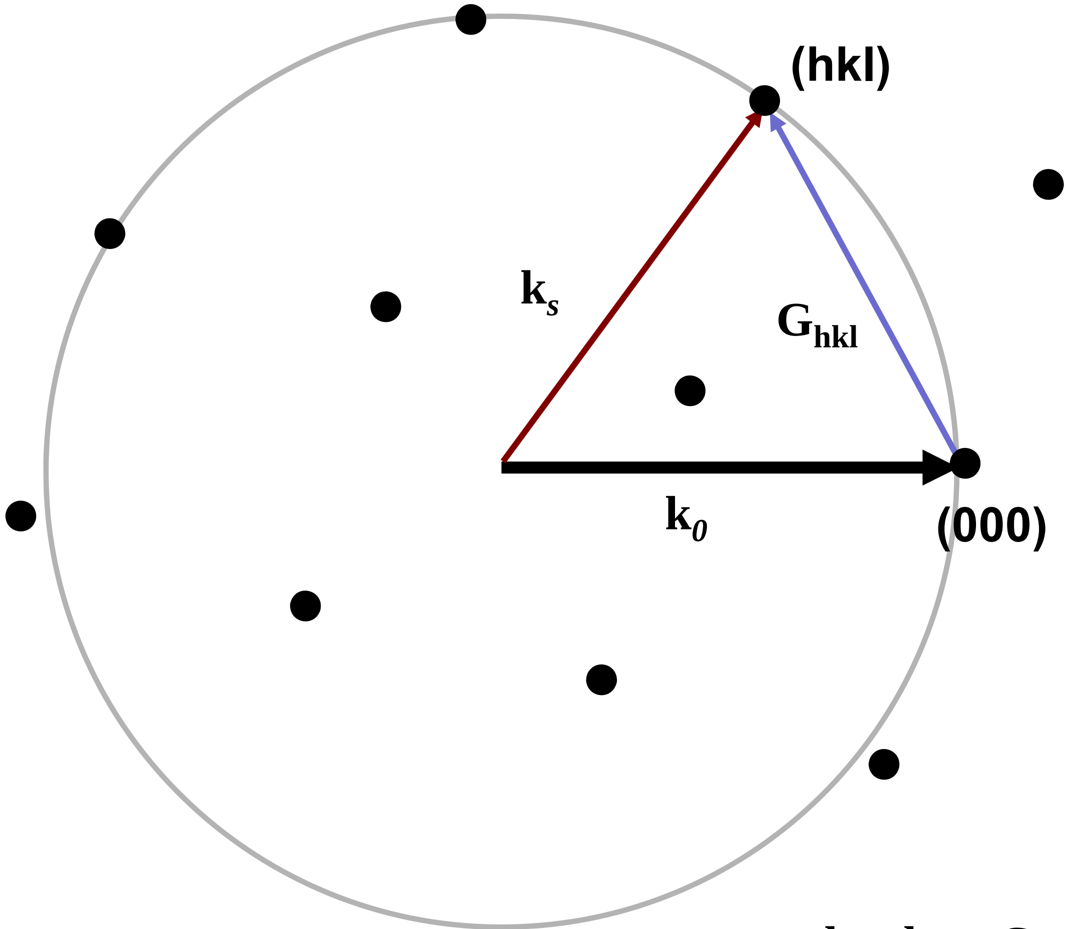




Very weak, we need bright x-ray pulses!

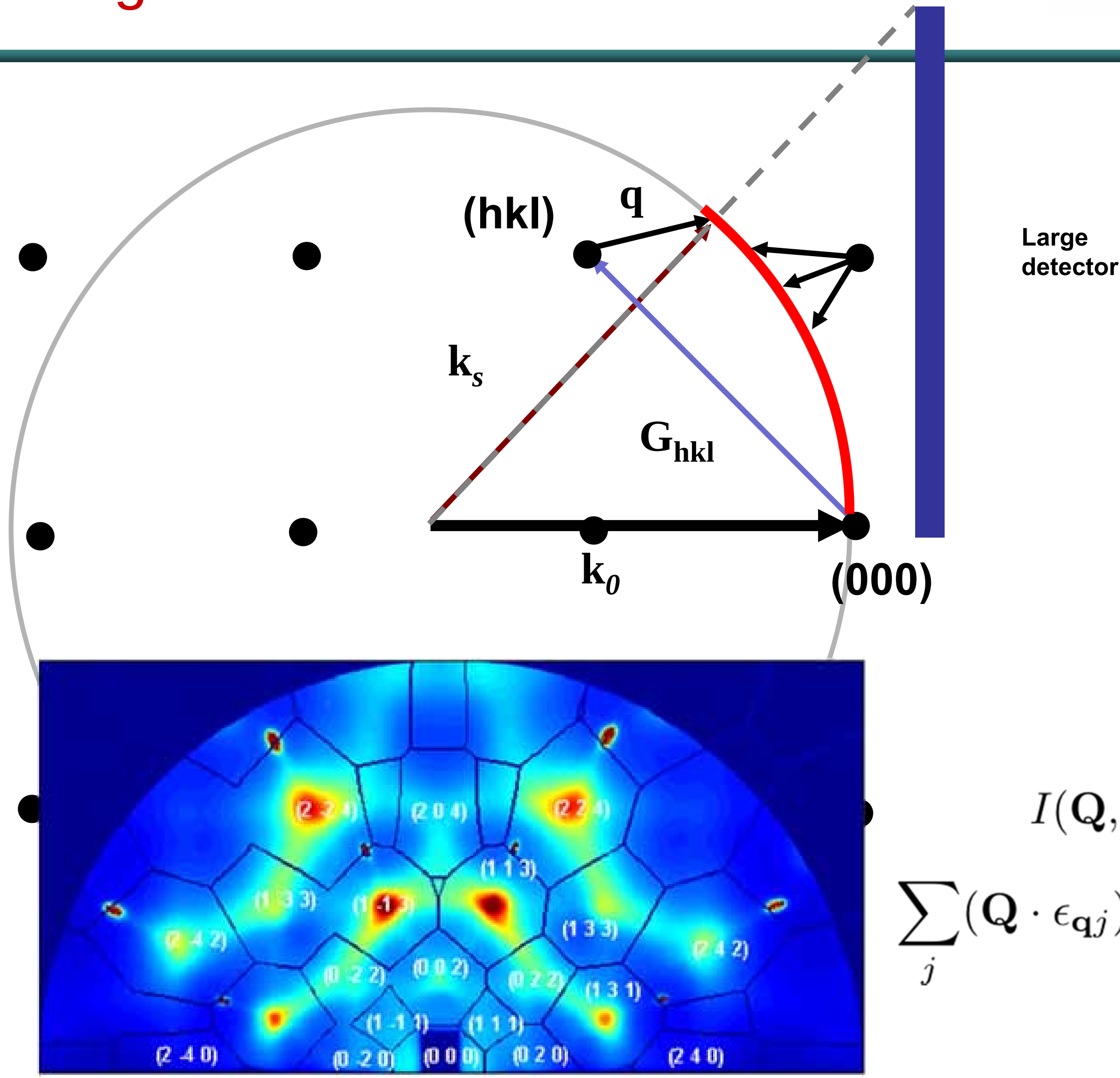
Example, diffuse scattering by thermal phonons

Bragg scattering



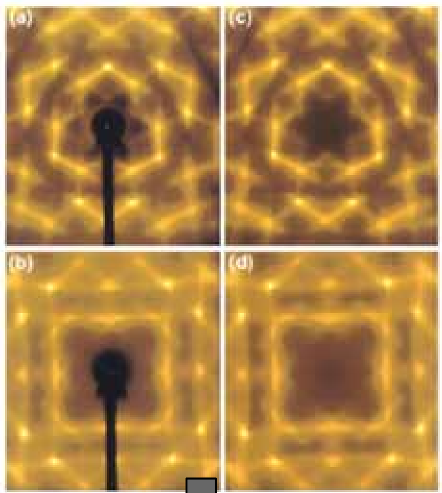
$$\mathbf{k}_s - \mathbf{k}_0 = \mathbf{G}_{hkl}$$

Diffuse scattering

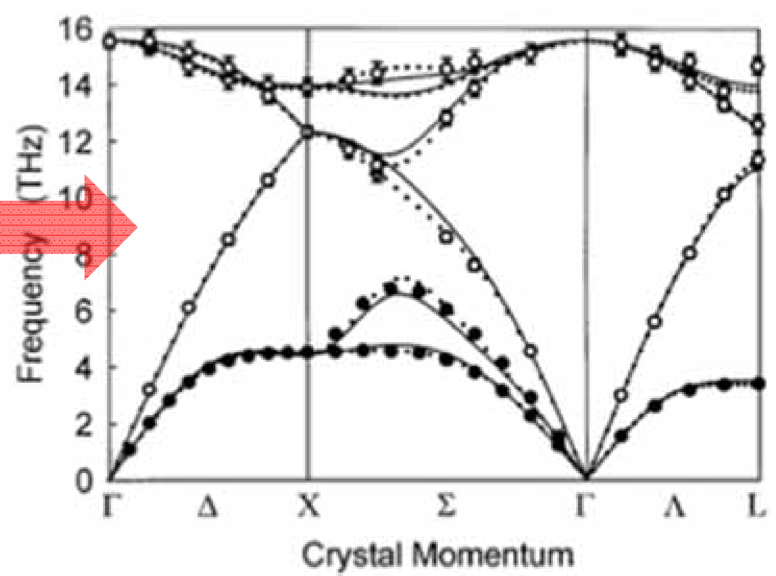


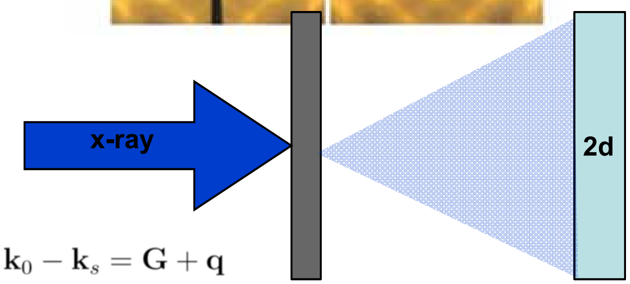
$$I(\mathbf{Q},t) \propto \sum_j (\mathbf{Q}\cdot\epsilon_{\mathbf{q}j})^2 \frac{2n_{\mathbf{q}j}+1}{\omega_{\mathbf{q}j}}$$

Phonon Dispersion from TDS and limitations

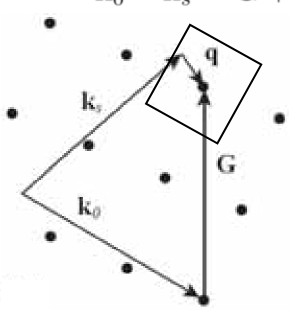


?





$\mathbf{k}_0 - \mathbf{k}_s = \mathbf{G} + \mathbf{q}$

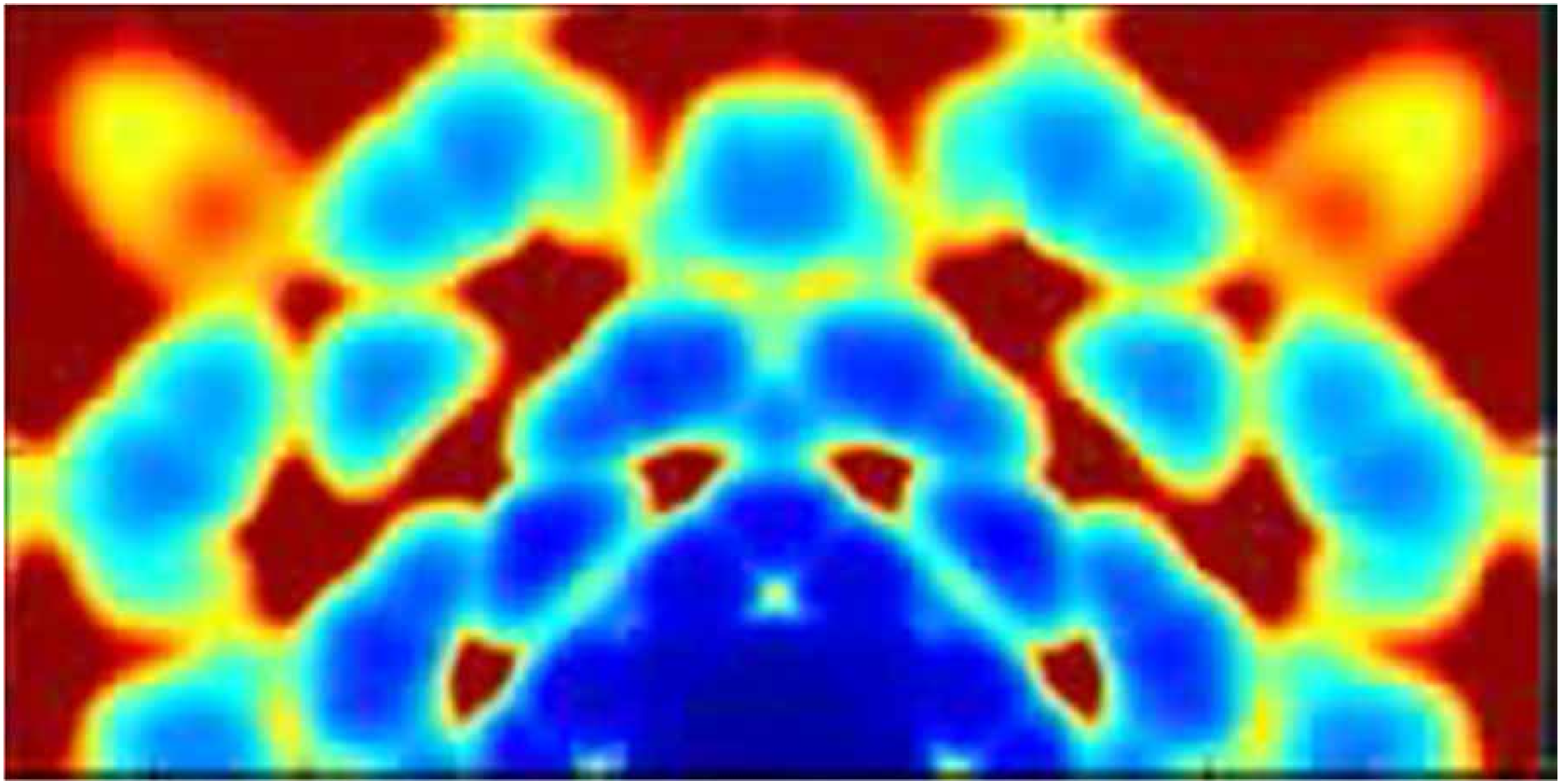


Joynson, Phys. Rev. 94, 851 (1954)…
…M. Holt et al., PRL 83, 1999.

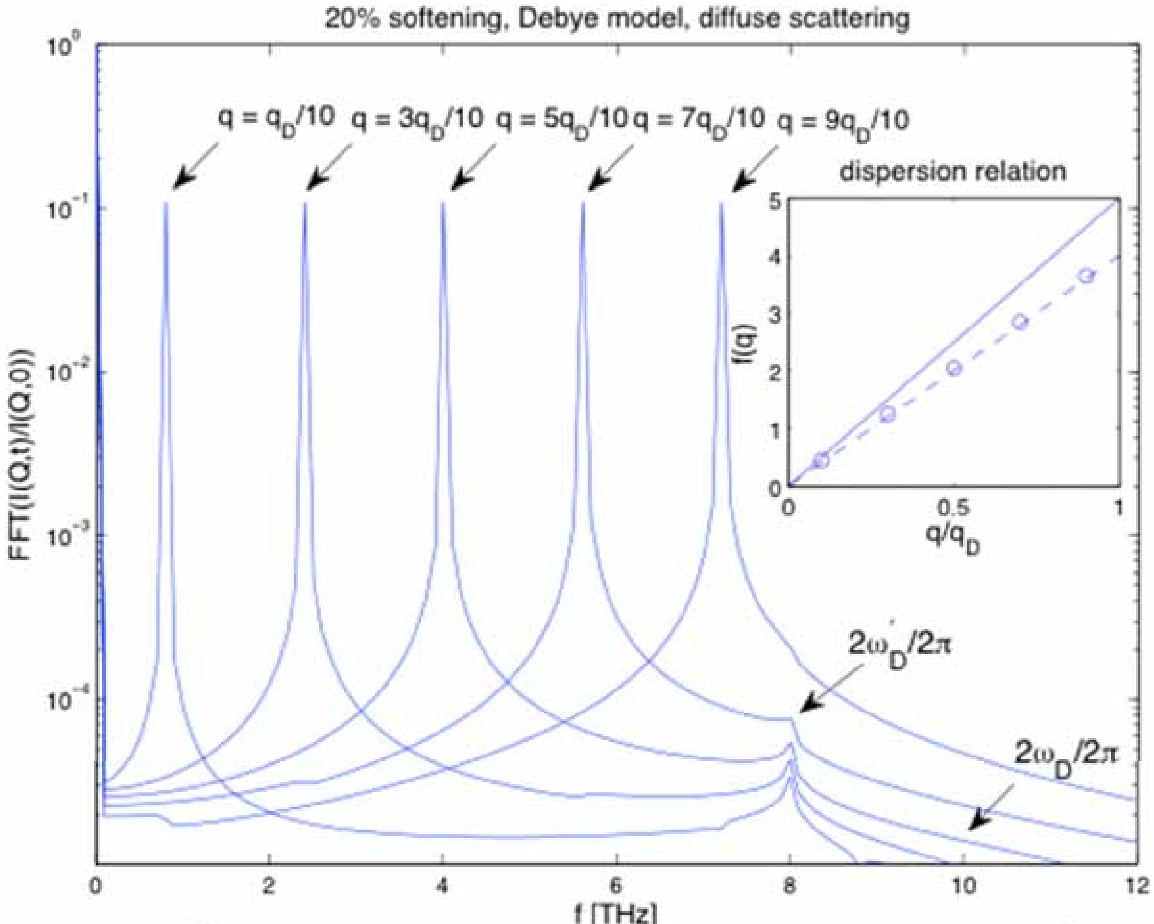
$$I(\vec{Q},t) \propto (\vec{Q}\cdot\vec{\epsilon}_{\lambda,\vec{q}})^2 \frac{(2n_{\lambda,\vec{q}}(t)+1)}{\hbar\omega_{\lambda,\vec{q}}},$$

TDS: Limited to simple cases (# fit parameters low) and have a constraint (assumes Bose-Einstein distribution)

Simulation of InP impulse softening of TA by 20%



Fourier transform of I(q,t) yields phonon dispersion (excited state)

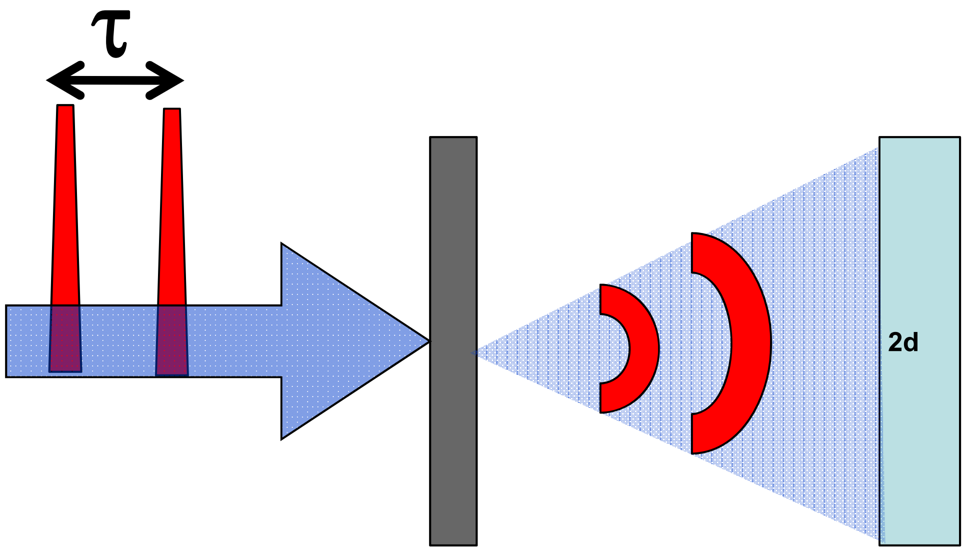


$$< u_q(t)^2 >= \frac{1}{2} < u_q(0)^2 > \left(\alpha \left(1 + \cos(2\alpha\omega(q)t)\right) + \frac{1}{\alpha}\left(1 - \cos(2\alpha\omega(q)t)\right) \right),$$

$$< u^2(t) >= \frac{9k_BT}{2M\omega_D^2} \left(\left(1 + \frac{1}{\alpha^2}\right) + \frac{\sin(2\alpha\omega_D t)}{2\alpha\omega_D t} \left(1 - \frac{1}{\alpha^2}\right) \right).$$

Hillyard, Reis and Gaffney PRB 77, 195213 (2008).

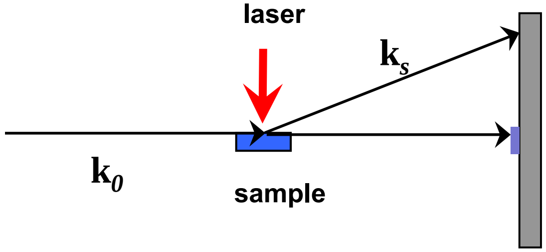
Alternatively, can use multiple coherent pulses

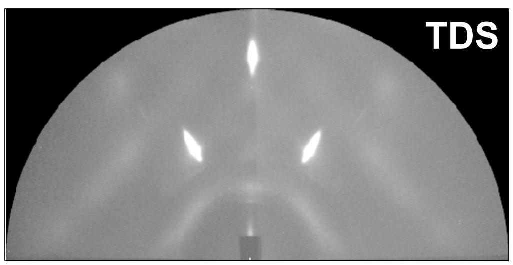


If you want to go really crazy, multiple colors, incidence angle, stimulated x-ray (electronic) Raman selected w & q…

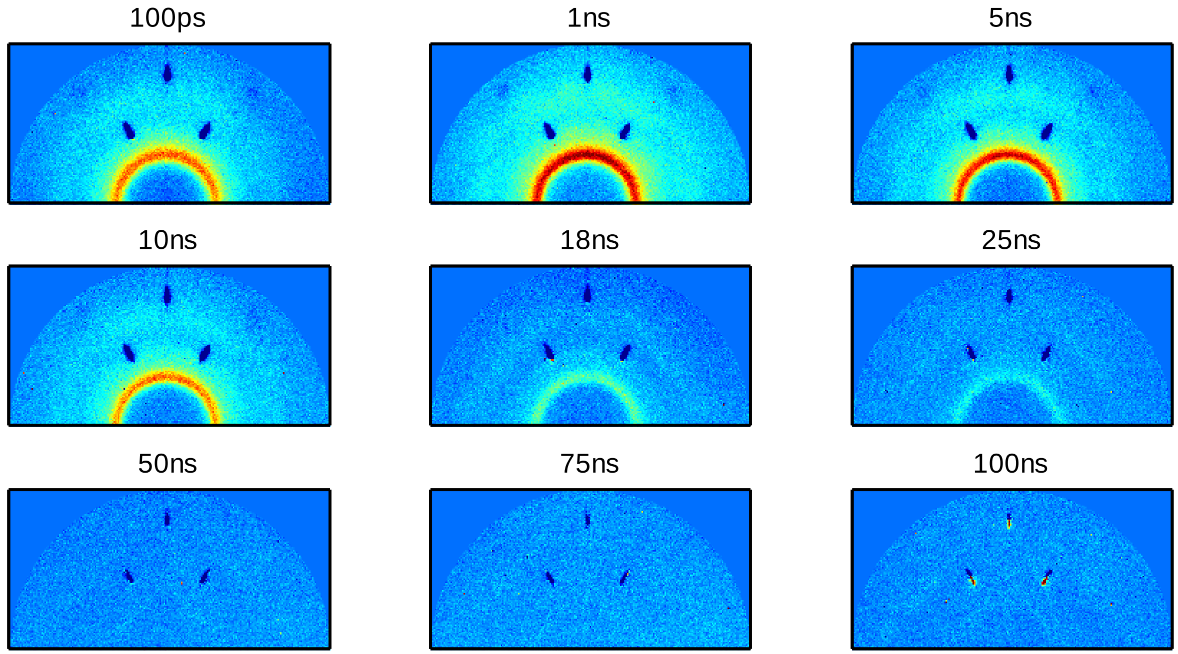
Can we do x-ray 4-wave mixing, ala K. Nelson or S. Mukamel?

Synchrotron data limited by time-resolution





Laser-melt and epitaxial regrowth of Bi on Sapphire



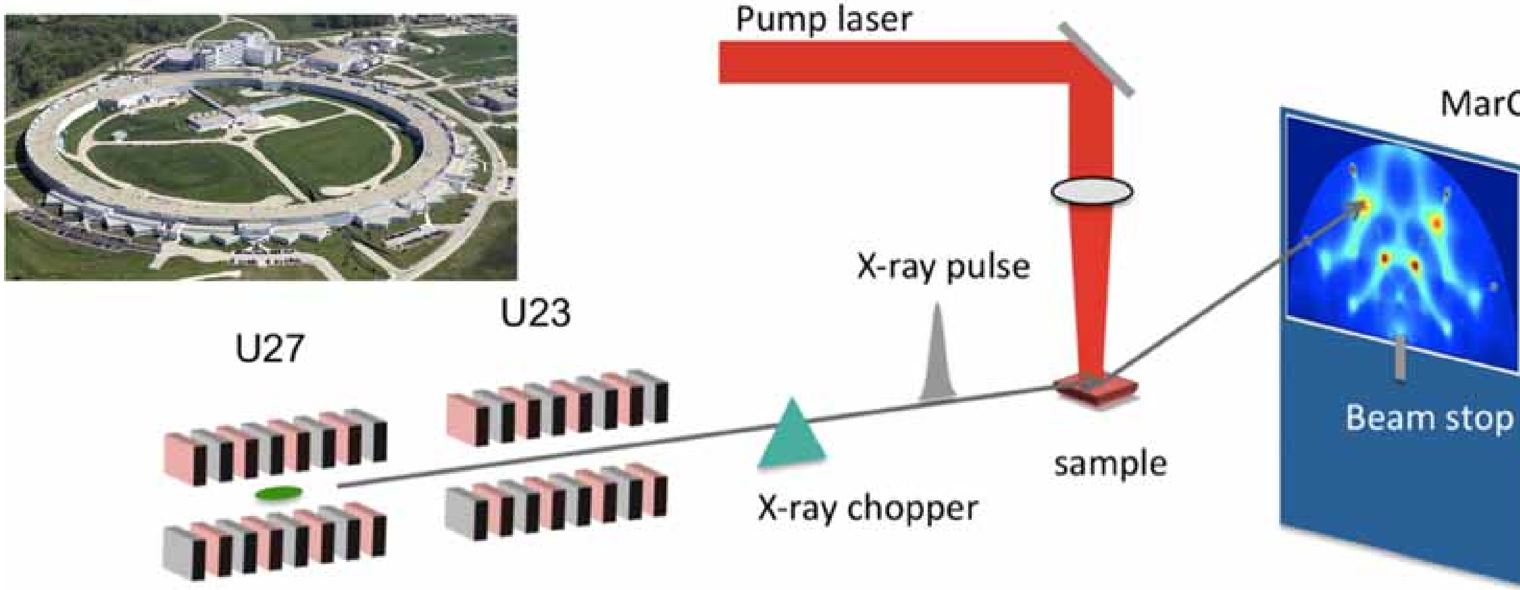
Average of 5 shots

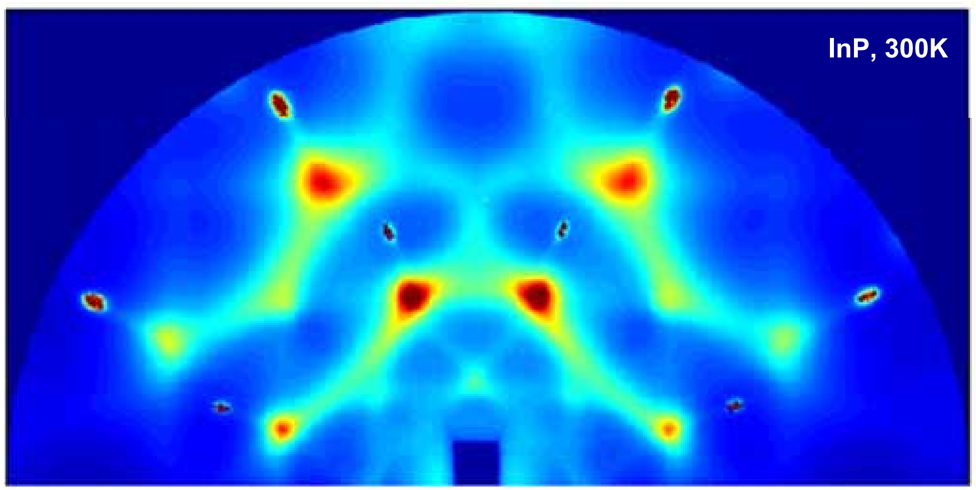
BioCARS beamline at APS
~1% of LCLS photons/pulse but 100ps

Trigo et al., unpublished

Time-resolved x-ray diffuse scattering with 100ps resolution

Advanced Photon Source





15 keV x-rays

~ 100 single x-ray Pulses

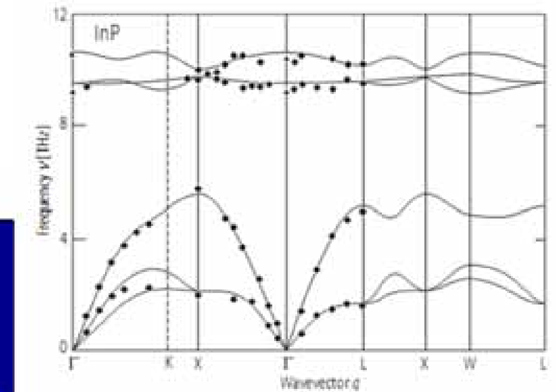
Equivalent to a single LCLS shot!

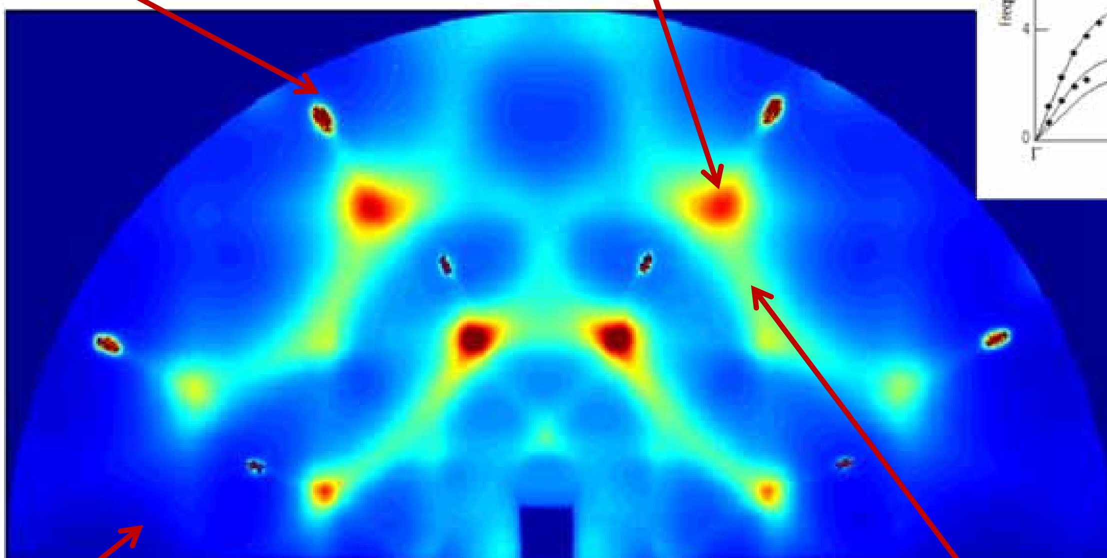
Trigo et al. Phys. Rev. B, 82(23):235205, 2010.

Primarily TDS

"Bragg Rods" from surface scattering

bright spots due to acoustic phonons near **zone center**





p-Polarization forbids scattering at 90°

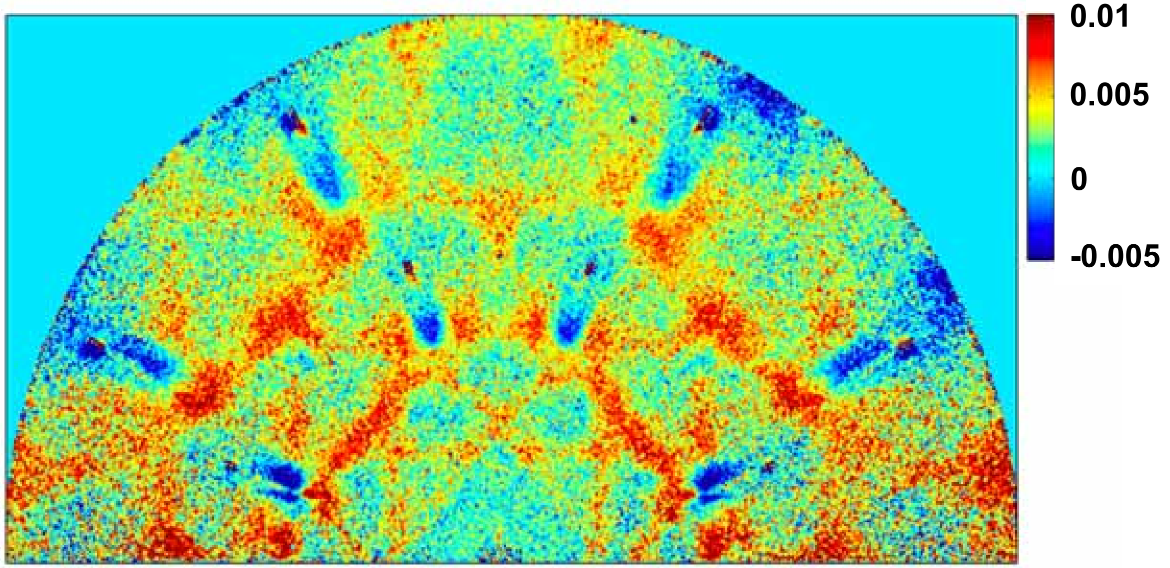
Beam stop shadow

High-symmetry directions, where acoustic branches are "soft".

Trigo et al. Phys. Rev. B, 82(23):235205, 2010.

But, more than heating

[I(400ps) −I(100ps)] / I(off)



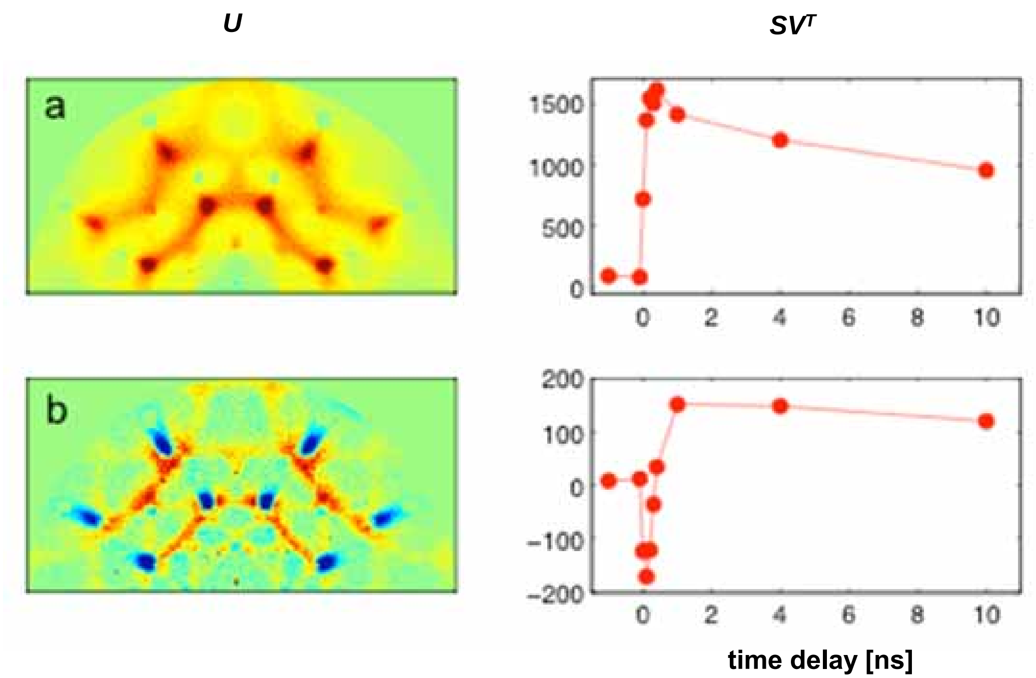
If processes were only thermal, $$\frac{\partial I(t)}{\partial t} \propto \Delta n \rightarrow \frac{1}{I_0}\frac{\partial I(t)}{\partial t} \propto \Delta T = \text{const}$$

Trigo et al. Phys. Rev. B, 82(23):235205, 2010.

Interpretation by Singular Value Decomposition

Similar to equilibrium image

Positive and negative differential scattering



Sharp raise + exponential decay

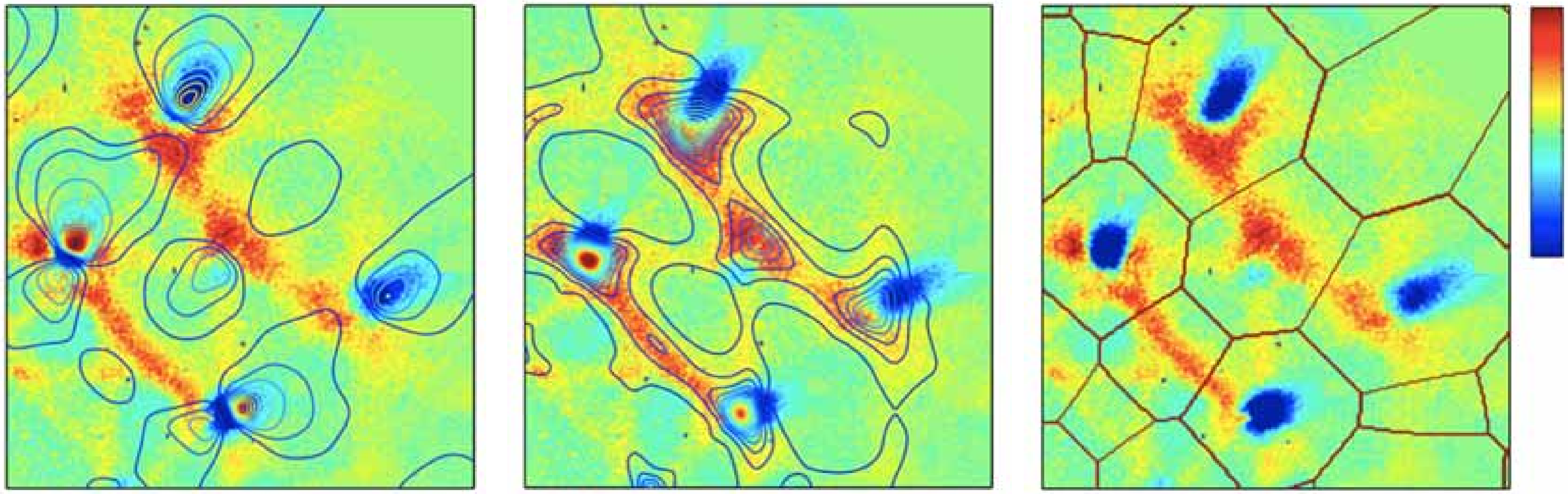
Delayed

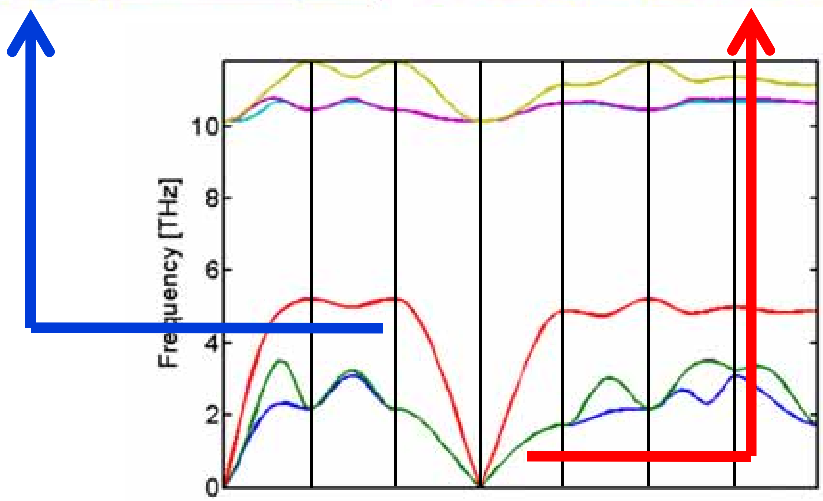
Complex dynamics in the phonon populations due to the anharmonic coupling between modes

Trigo et al. Phys. Rev. B, 82(23):235205, 2010.

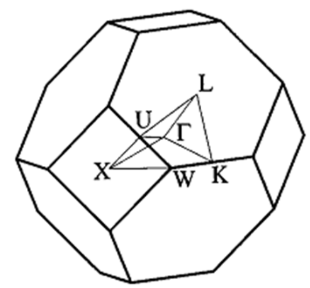
Contribution from acoustic phonon branches

LA TA



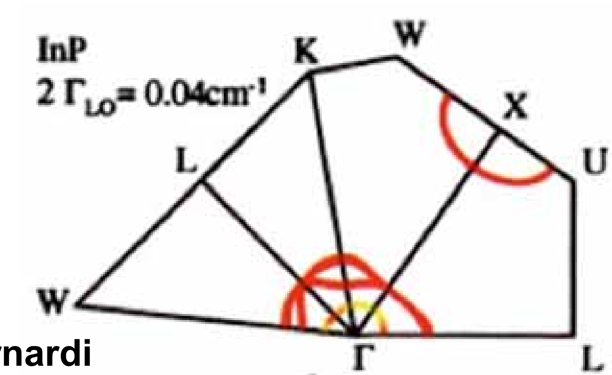


Brillouin zones



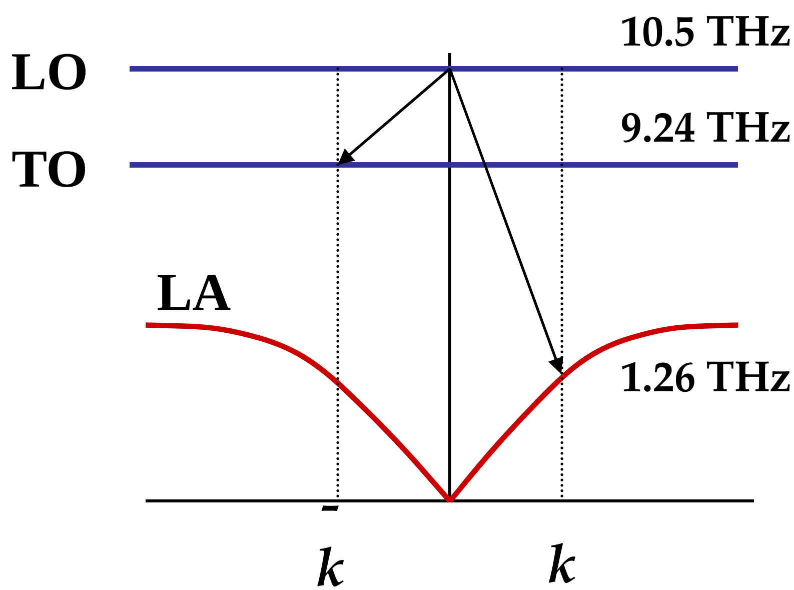
Trigo et al. Phys. Rev. B, 82(23):235205, 2010.

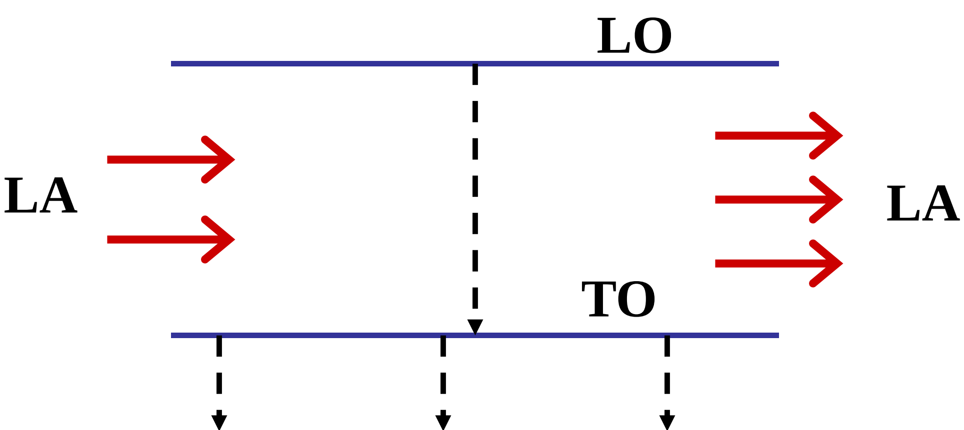
Can we modify optical phonon lifetime?



Debernardi

Decay channel:
LO → TO + LA





Stimulated decay and emission of TO + LA

Chen, Khurgin, Merlin, APL 80 2901, 2002.

Will study anharmonic decay at LCLS this fall!

Acknowldegements

Marino Trigo, Jian Chen, Mason Jian, Matthias Fuchs, Mike Kozina, Shambhu Ghimire, Vinayak Vishwanath, PULSE, SLAC, Stanford

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Yu-Miin Sheu,

Los Alamos

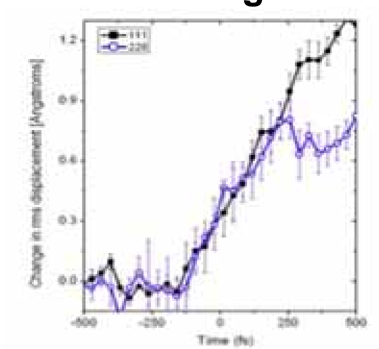
Tim Graber, Robert Henning,

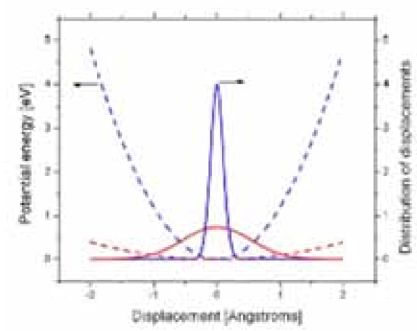
CARS, U. Chicago

Supported by the U.S. Department of Energy, Office of Basic Energy Science

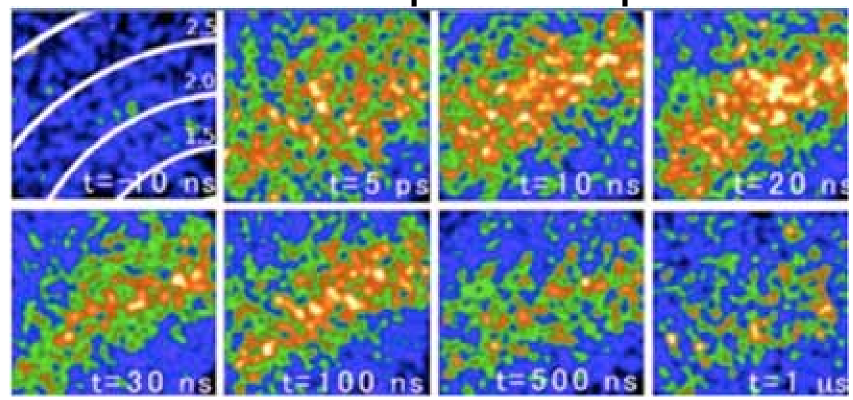
Lattice instabilities

Diffraction data consistent with complete softening

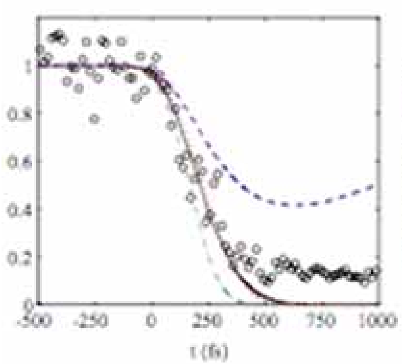


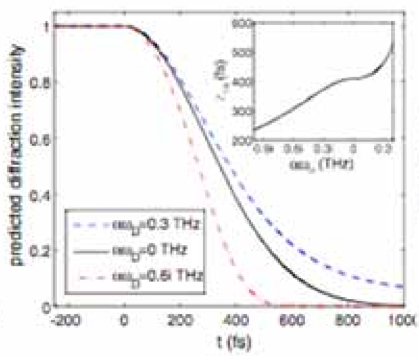


formation of a nonequilibrium liquid

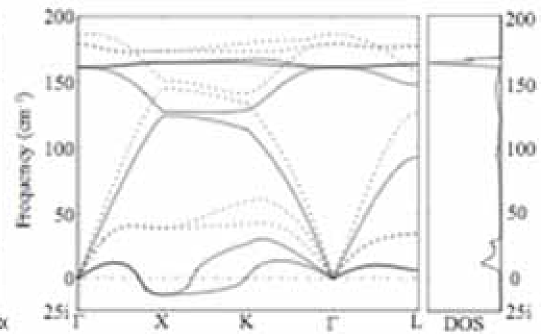


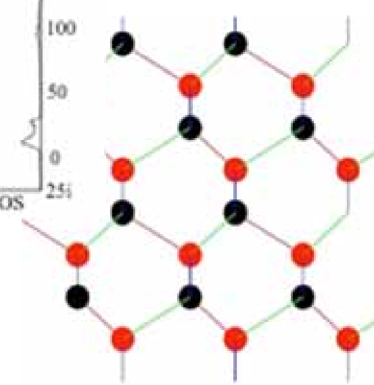
Model assuming uniform softening
Gives similar results to inertial dynamics





DFPT predicts instability first develops at X point





LCLS beamtime this fall to study Ultrafast diffuse scattering will measure evolution of interatomic forces and look for instability

 Lindenberg *et al.,* Science 308, 2005;Gaffney *et al.*, PRL 95, 2005; Hillyard et al., PRL 98, 2007.