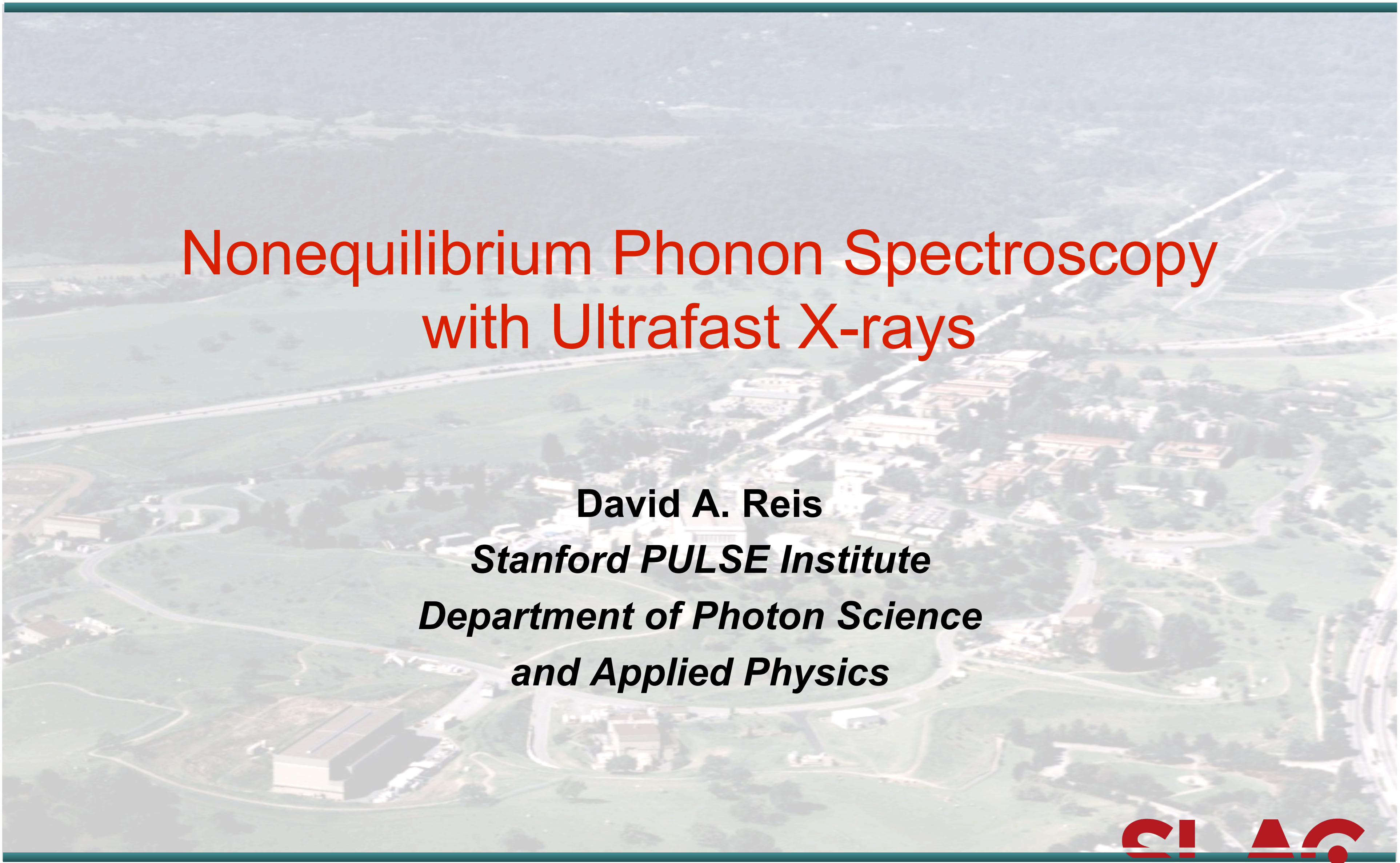
PULSE STANFORD



Nonequilibrium Phonon Spectroscopy with Ultrafast X-rays

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Why (or perhaps when) ultrafast x rays?

- Want to measure inelastic x-ray scattering in time-domain:
  S(q,t)–structure and dynamics

- More information: than optical measurements (epsilon, time or freq. domain Raman…), primarily measure q=0 or density of states (2nd-order or impurity). Q-coverage.

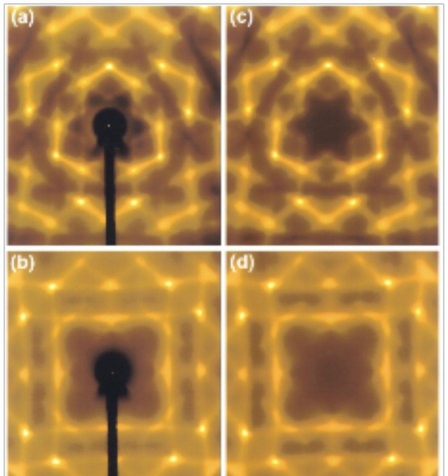
- Time-domain often necessary when states not accessed through equilibrium, short lifetime, rare events. inhomogeneous/homogeneous broadening, etc.

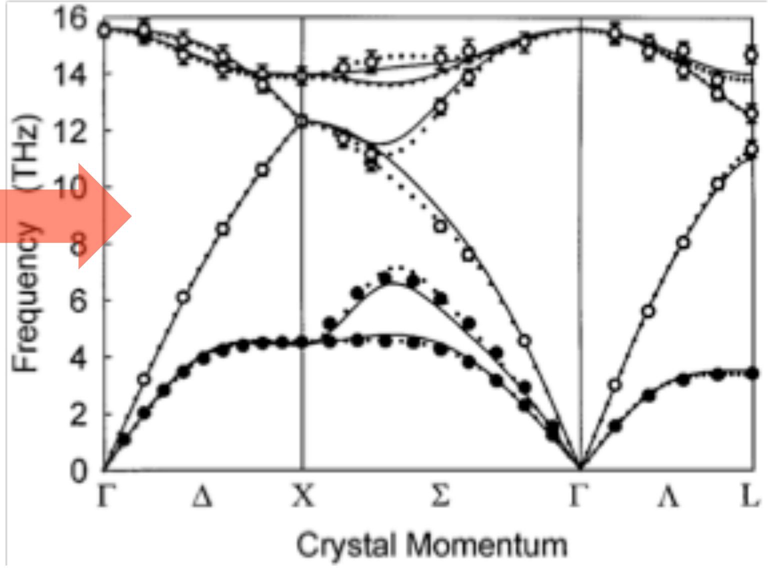
- Not only ultrafast: highest spectral resolution often easier in time-domain. strength/weakness: particularly sensitive to coherence, lifetime

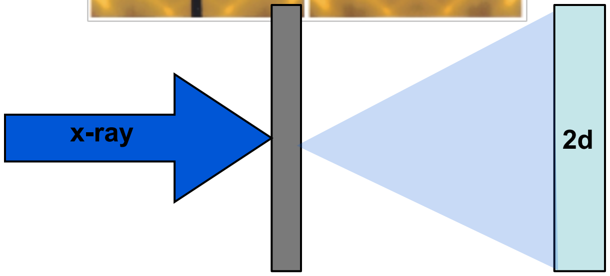
- Can't use neutrons: give dispersion and line-widths but lack possibility for ultrafast time-resolution.

- Complements ultrafast photoemission and electron diffraction

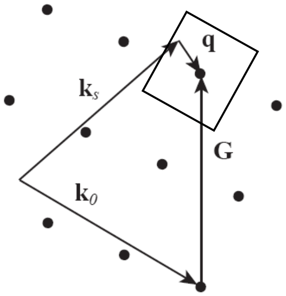
Phonon Dispersion from TDS and limitations







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



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

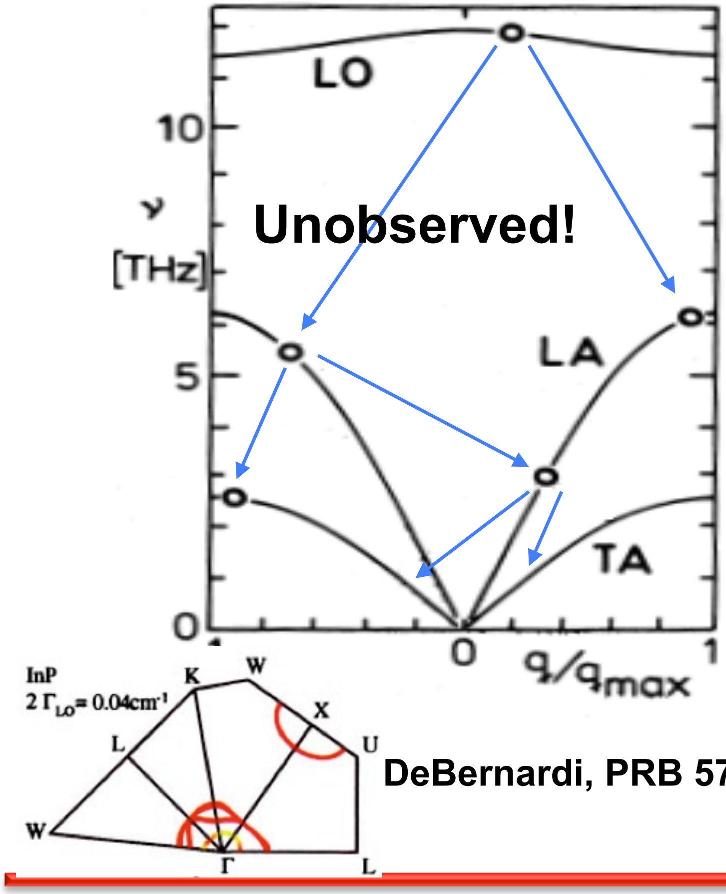
TDS: Limited to simple cases (# fit parameters low) and have a constraint (assumes Bose-Einstein distribution)

Momentum and Time-resolved Phonon Spectrsoscopy

phonon-phonon and electron-phonon interactions,
anharmonic decay, interatomic forces, phase transitions…

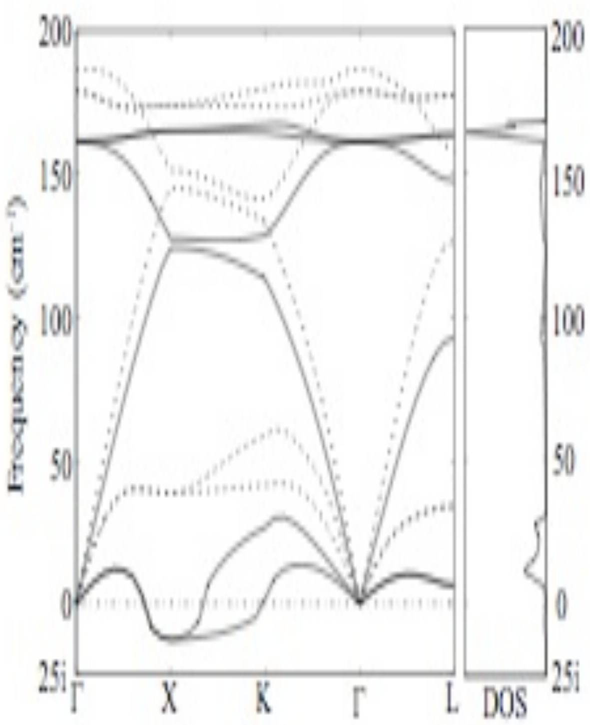
$n(q,t)$

$\omega(q,t)$

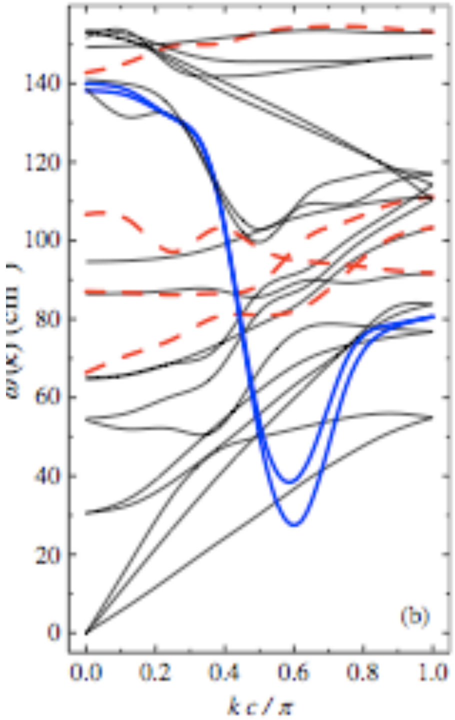


Unobserved!

DeBernardi, PRB 57, 1998

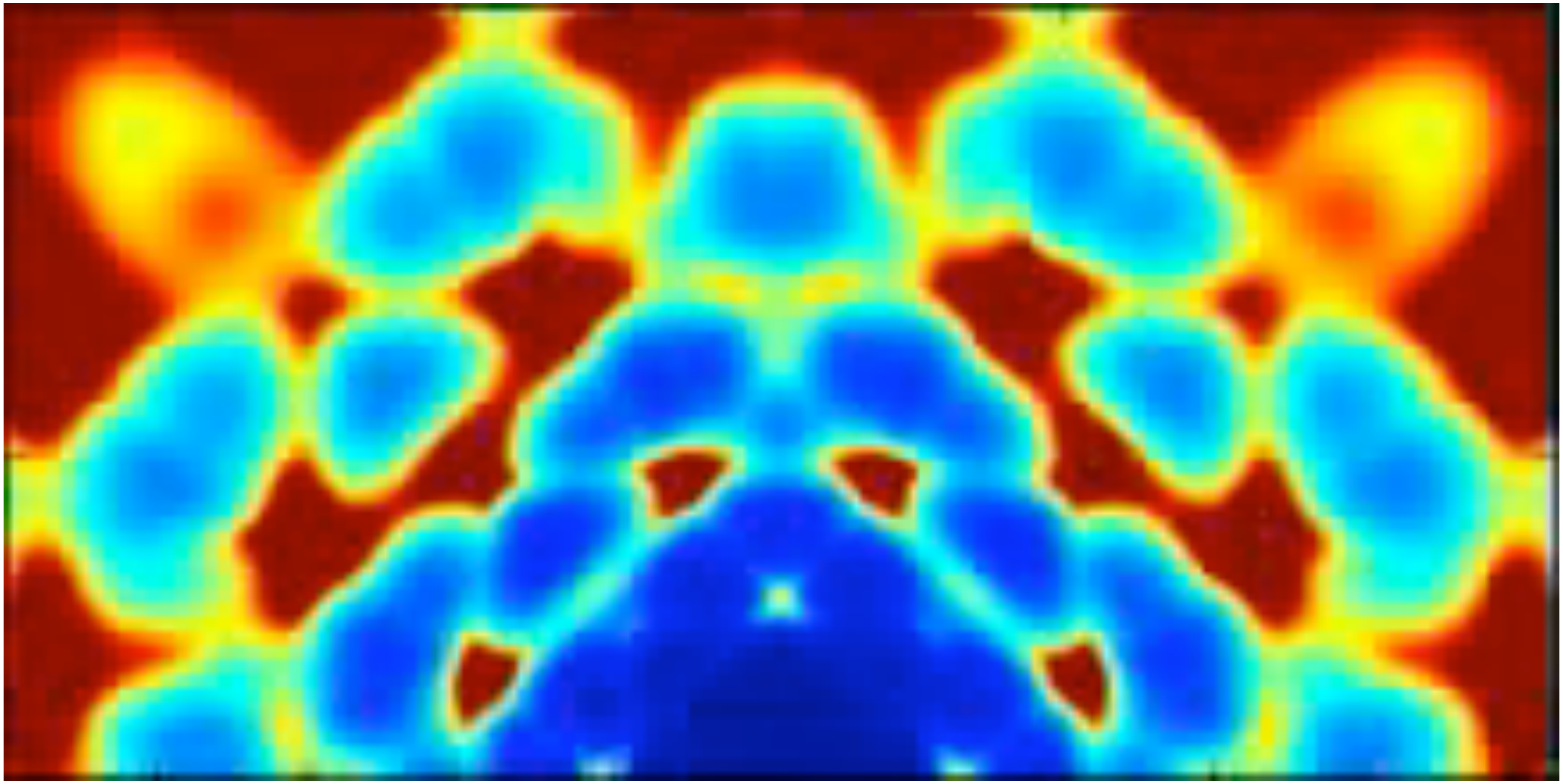


Hillyard et al., PRL 98, 2007

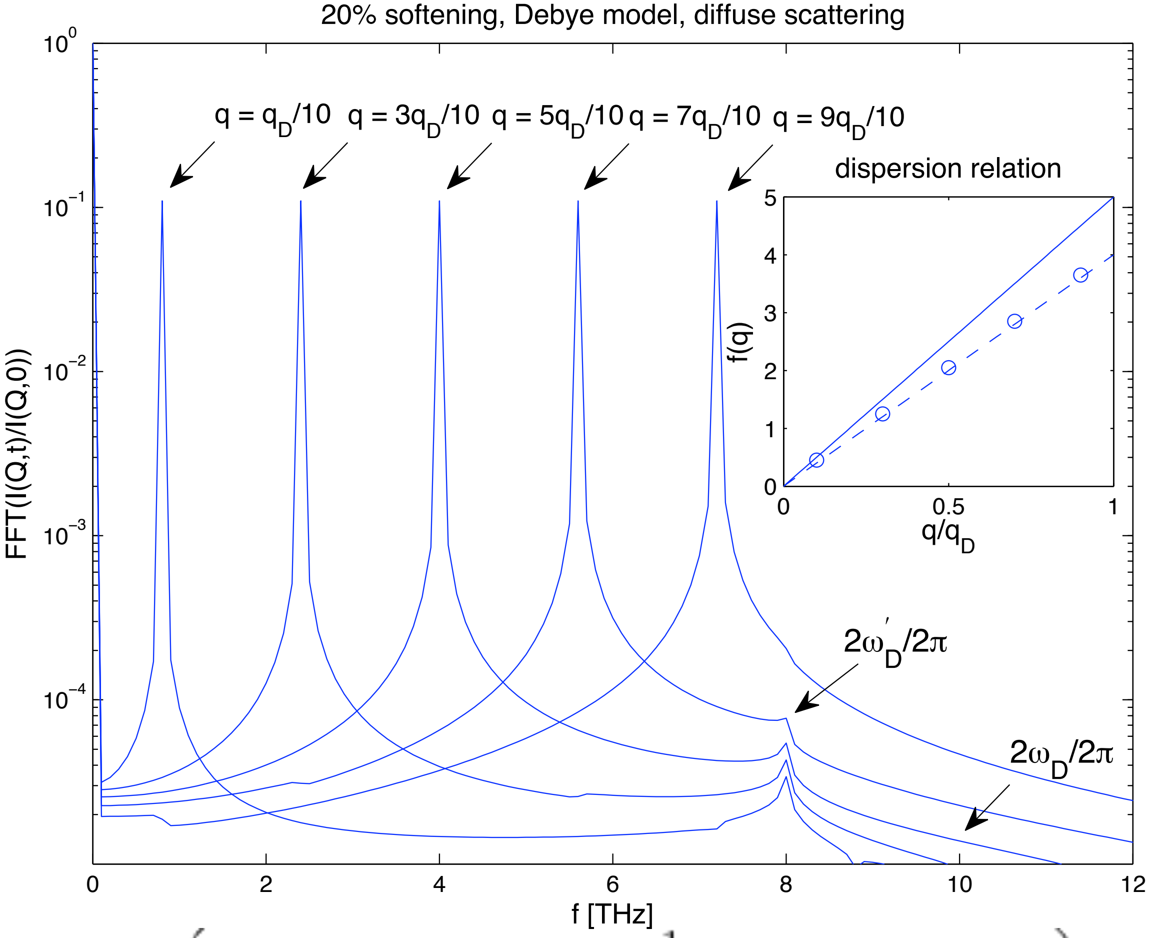


Lavagnini et al., PRB 78, 2008.

Simulation of InP impulse softening of TA by 20%



Extracting Excited State Dispersion (zero damping).


20% softening, Debye model, diffuse scattering

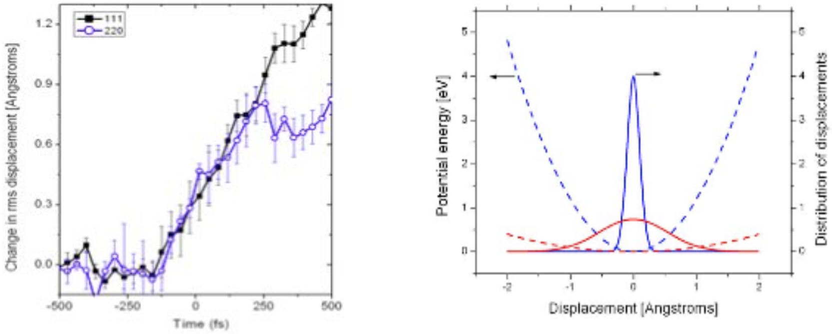
$$< u_q(t)^2 > = \frac{1}{2} < u_q(0)^2 > \left(\alpha\left(1+\cos(2\alpha\omega(q)t)\right) + \frac{1}{\alpha}\left(1-\cos(2\alpha\omega(q)t)\right)\right),$$

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

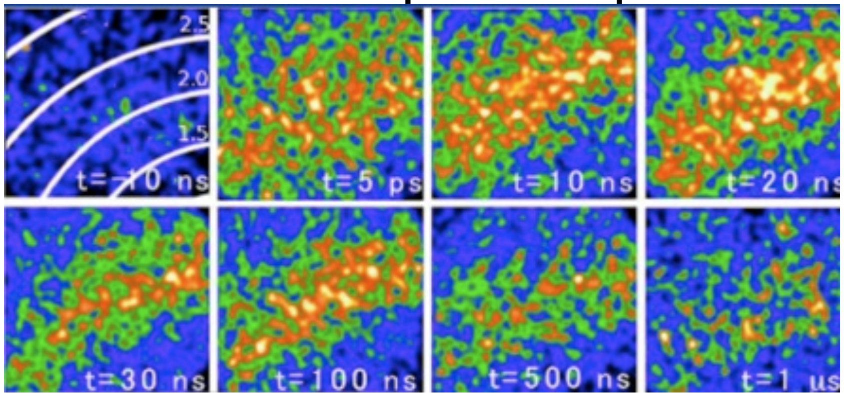
Analysis of nonthermal melting in InSb (DW factor) Hillyard, Reis and Gaffney PRB 77, 195213 (2008).

electronically-driven disorder in InSb

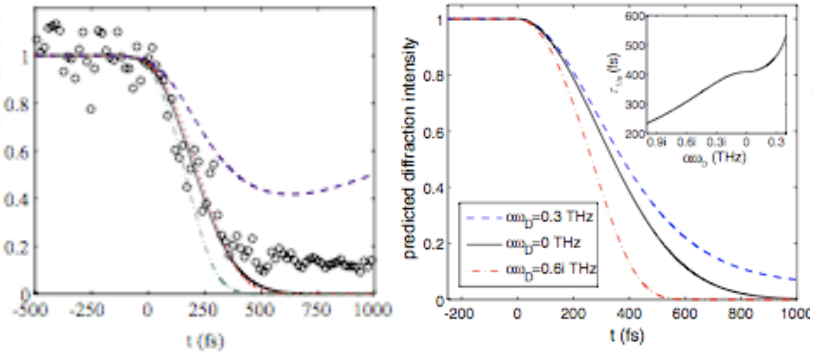
Diffraction data consistent with complete softening



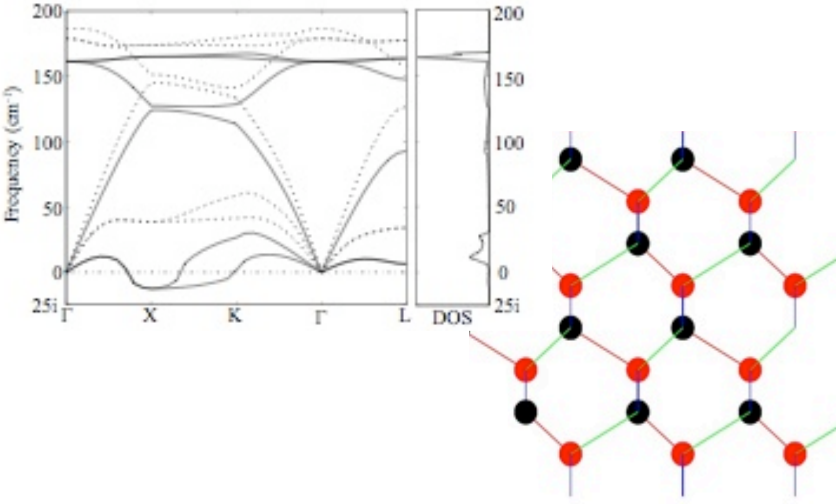
formation of a nonequilibrium liquid



Model assuming uniform softening
Gives similar results to inertial dynamics

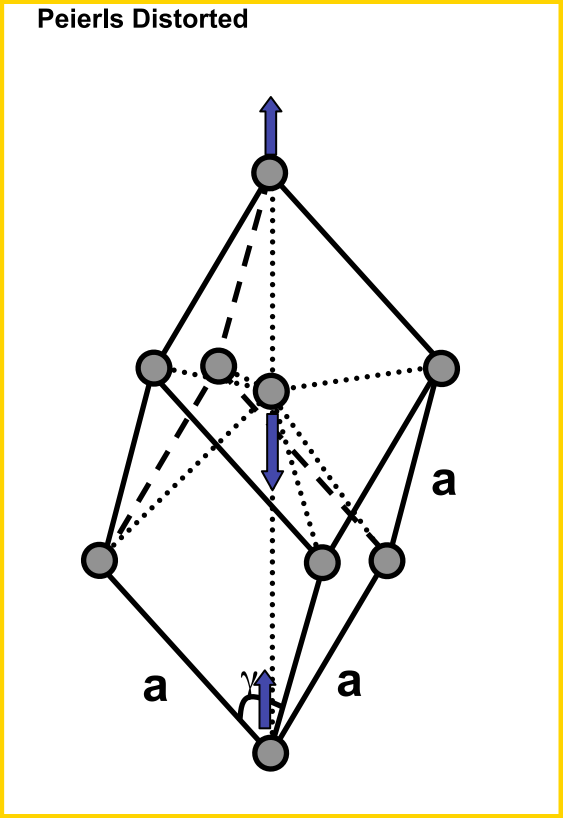


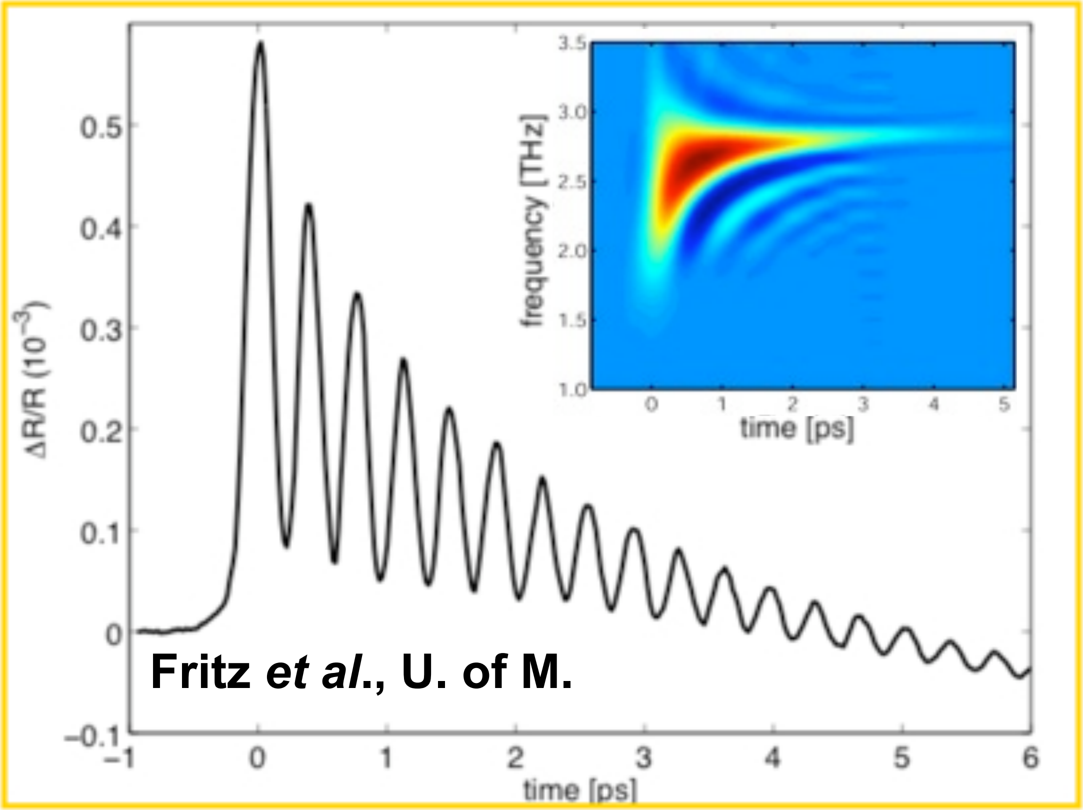
DFPT predicts instability first develops at X point



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

Example: Photoexcited bismuth (all optical experiments)





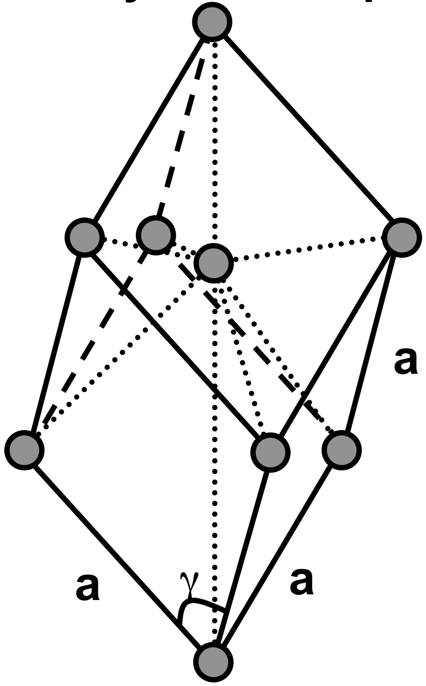
Fritz *et al*., U. of M.

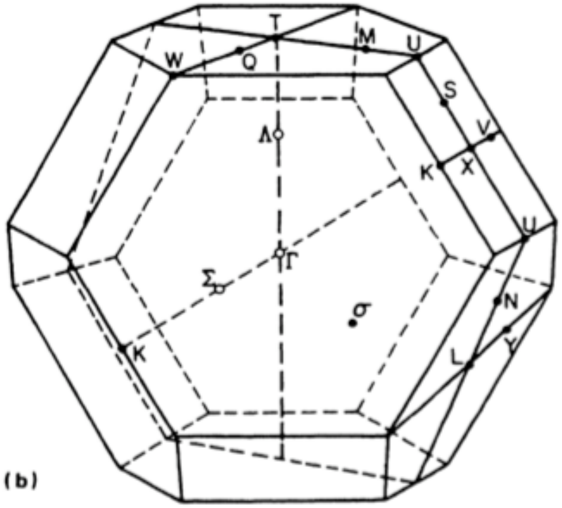
Coherent A_{1g} Mode is strongly softened and chirped.

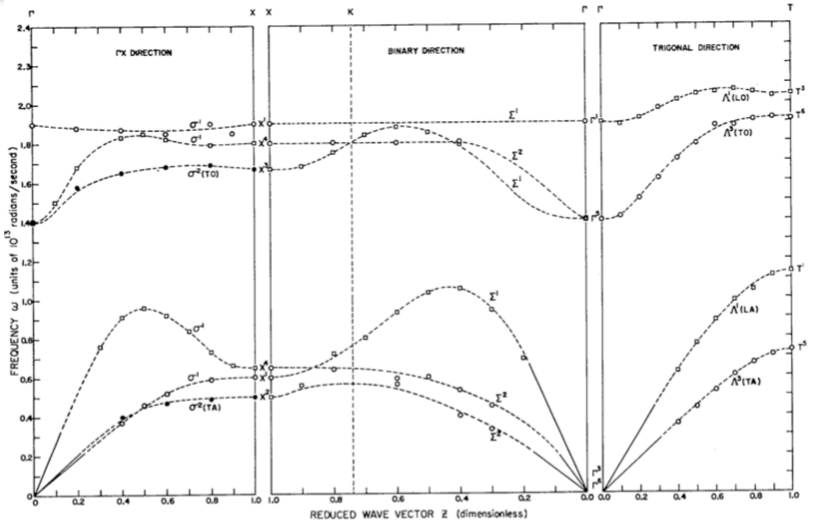
anharmonicity or electronic softening? Fahy and Reis PRL 93 109701, 2004; Murray *et al.* PRB 72, 060301 (R) 2005

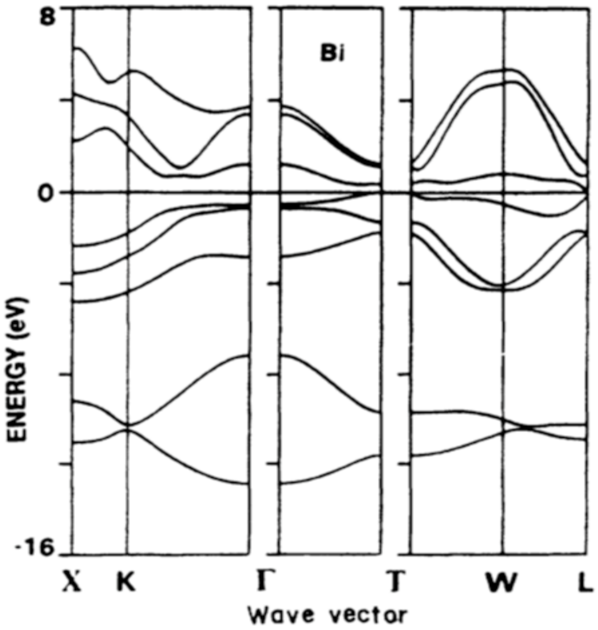
bismuth lattice dynamics, band structure

- •Prototypical Peierls distorted structure.
- •2 atom/cell (rhombohedral distortion of simple cubic)
- •(indirect) semimetal (holes @ T, elect. at L)
- •distortion along c-axis sensitive to carrier density and thus photoexcitation.



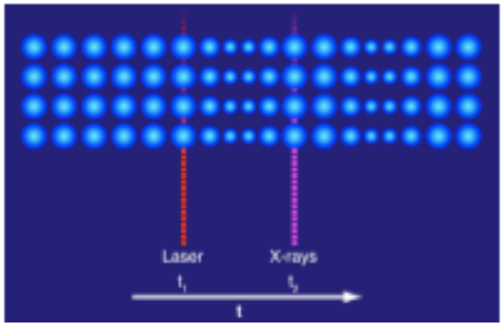




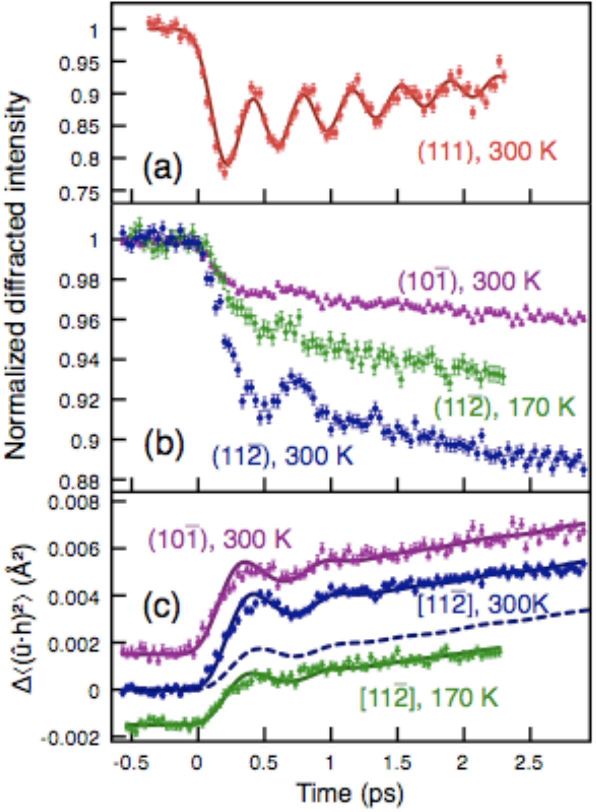


Phonon Dispersion (neutron): Smith 1967, Los Alamos Rep., Elect. Structure (DFT), Gonze 1990 PRB

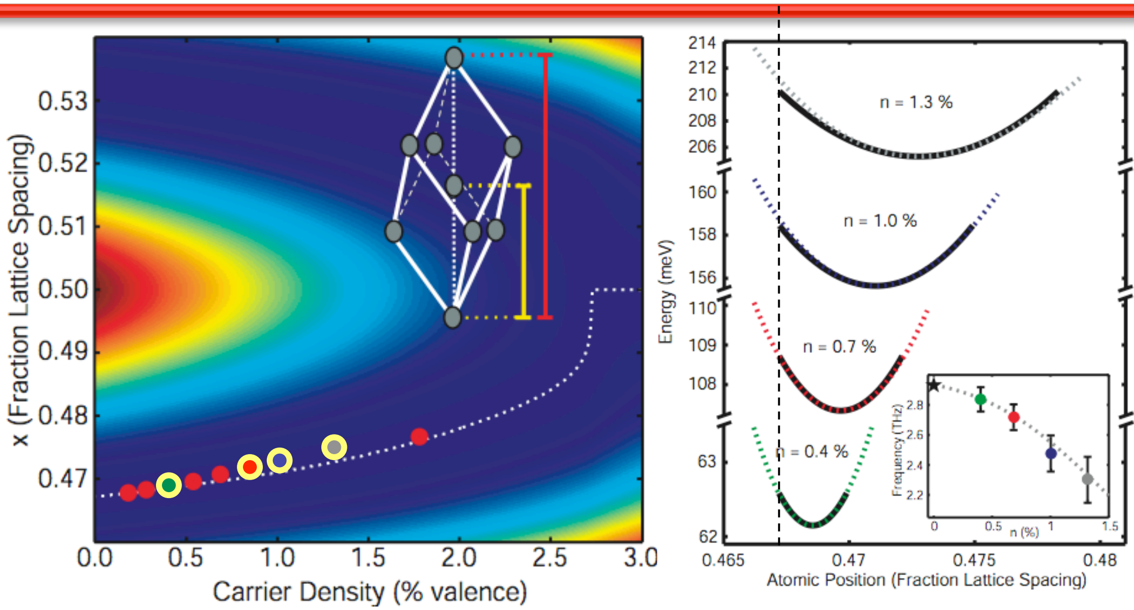
Electronic Softening in Bi
State-of-the-art femtosecond X-ray Diffraction



Reis *Physics Viewpoint 2009*

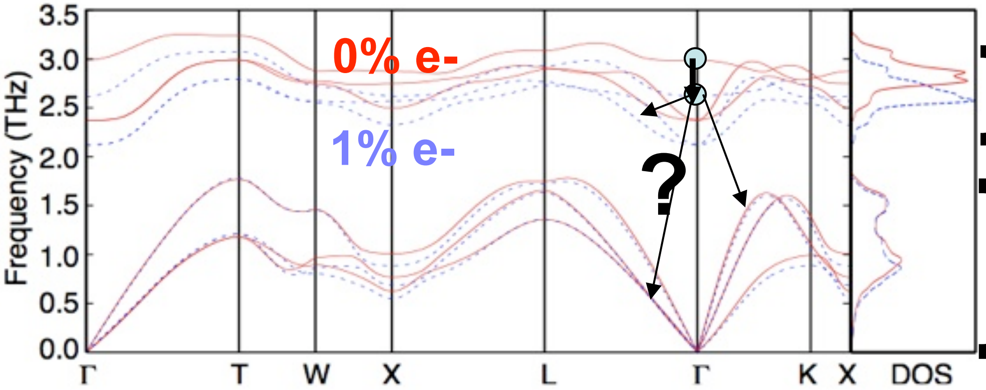


Johnson *et al.* PRL 2009.



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

DFPT calculation

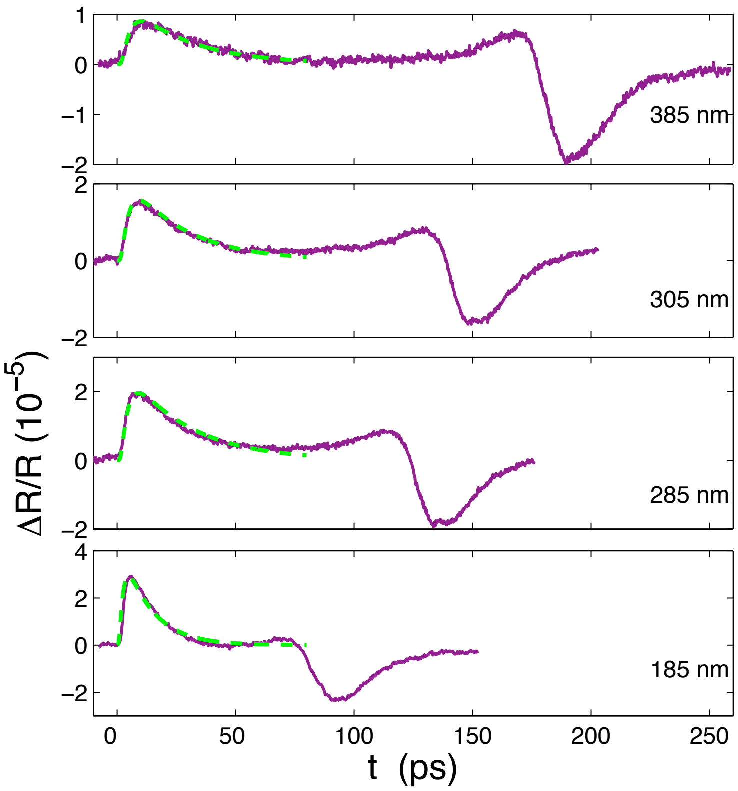


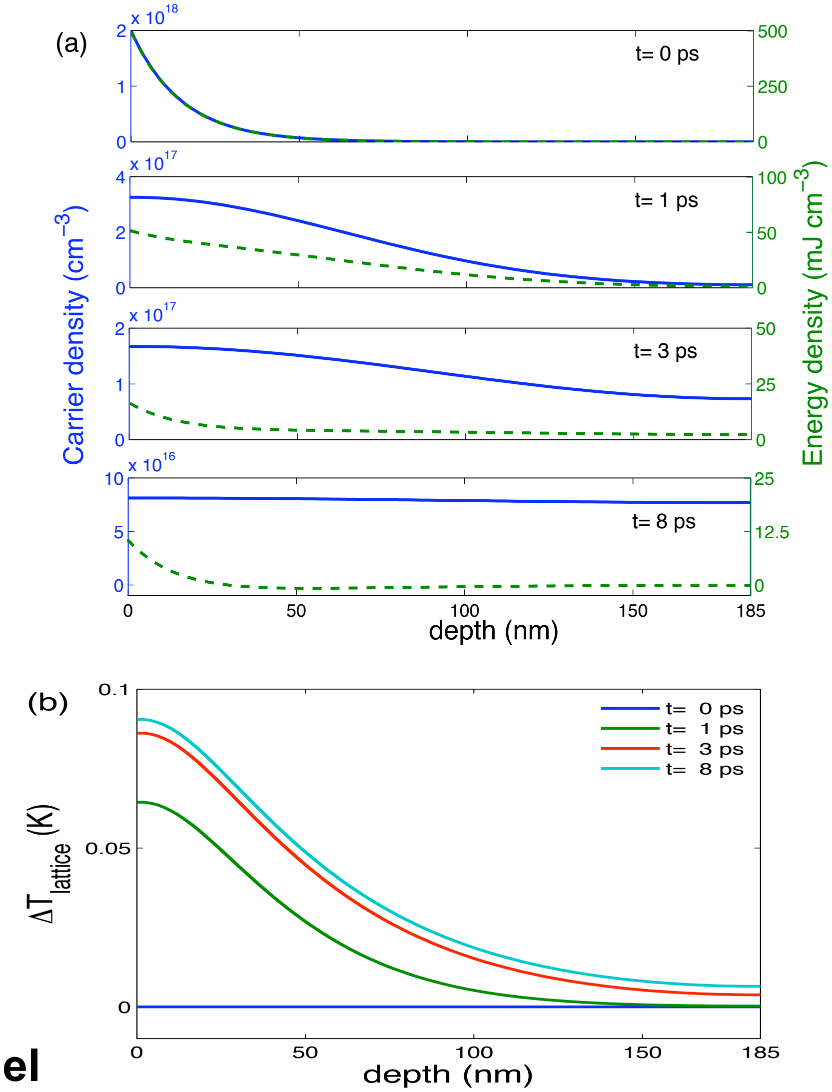
Optical Modes

Acoustic Modes

Murray *et al.* PRB 75 2007.

photoexcited carrier dynamics





"Fast" Ambipolar Diffusion
"Fast" lattice heating (phonon emission)
"Slow" recombination
Consistent with 2-chemical potential model

Sheu et al., 2010 responding to referees

Outstanding questions

•experimental determination of excited state potential (other than along A1g coord).

•transient high symmetry metalic state?

•Anharmonic decay channels and rates (A1g and lattice thermalization)

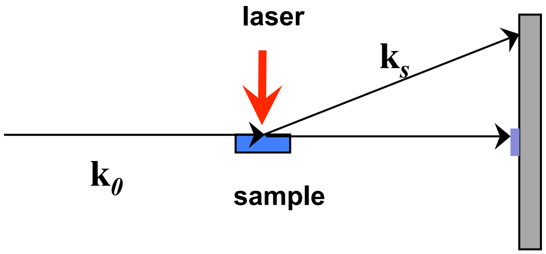
•Electron/hole distributions, details of thermalization and recombination

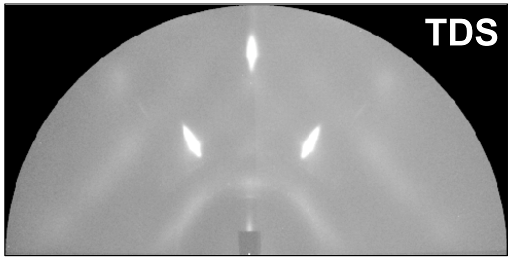
•Nature of extremely fragile (with temperature) Eg coherence (not shown)

•etc.

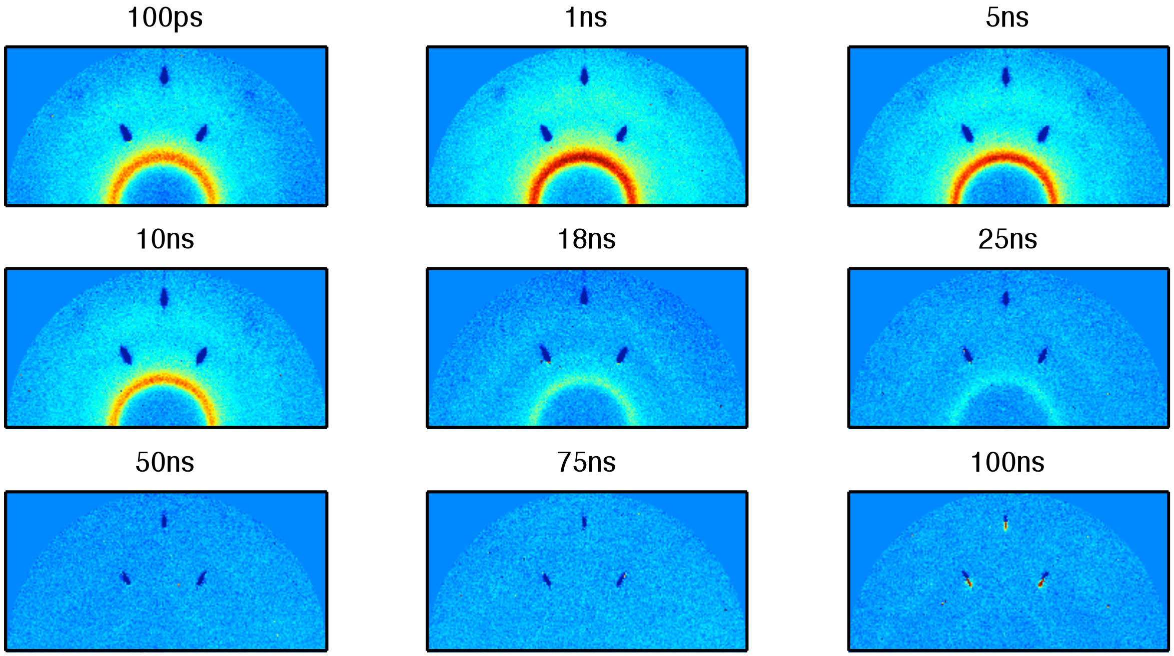
5 LCLS shifts in November on XPP (Dave Fritz et al.)

Synchrotron data limited by time-resolution





Laser-melt and epitaxial regrowth of Bi on Sapphire



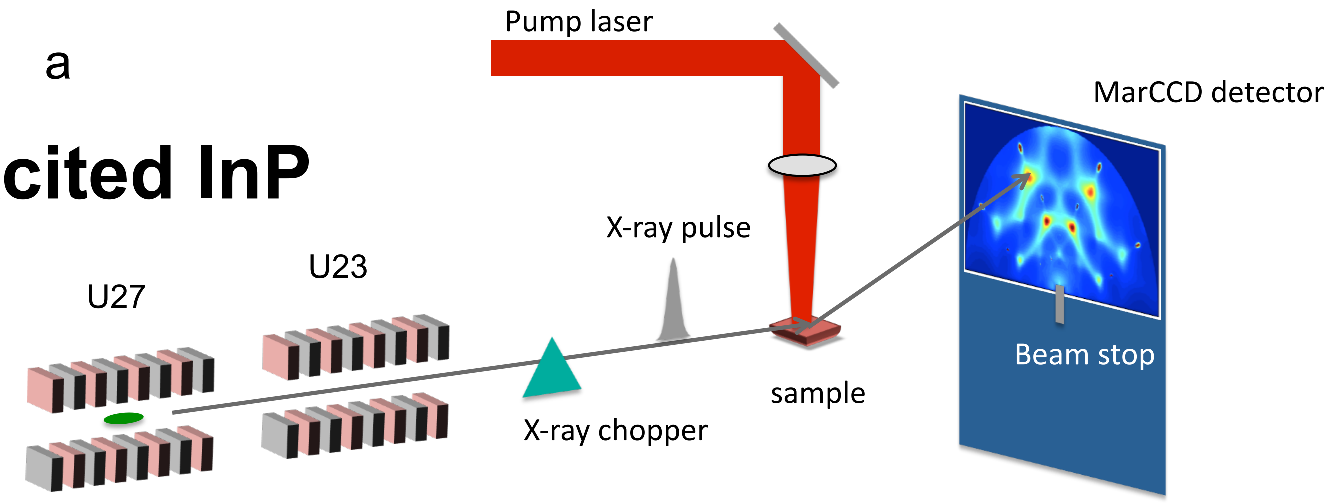
Average of 5 shots

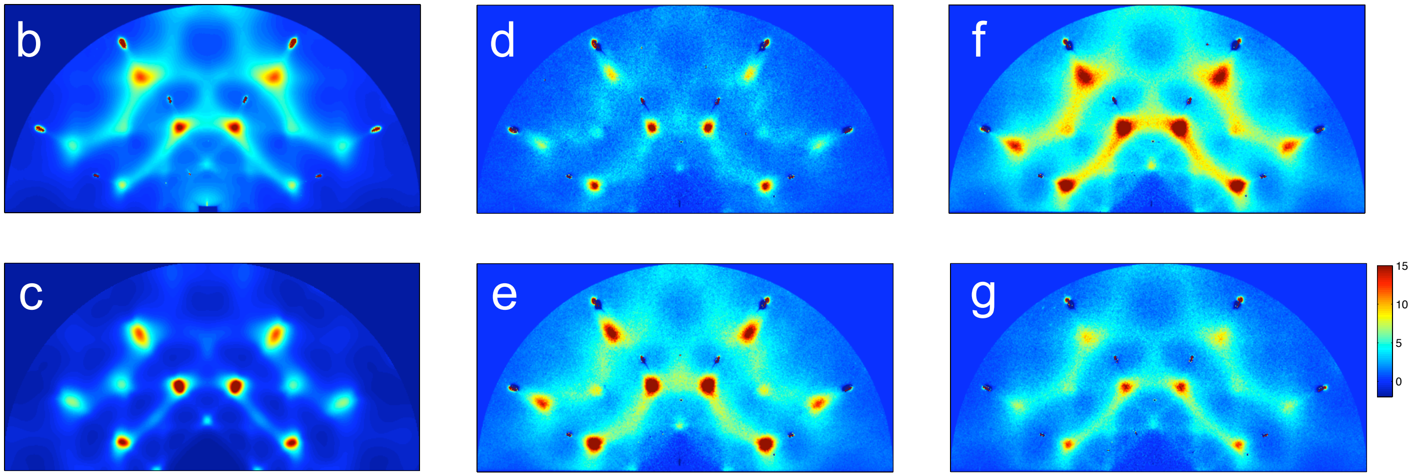
BioCARS beamline at APS
~1% of LCLS photons/pulse but 100ps

Trigo et al., unpublished

Time-resolved Diffuse Scattering Images of Nonequilibrium Phonons

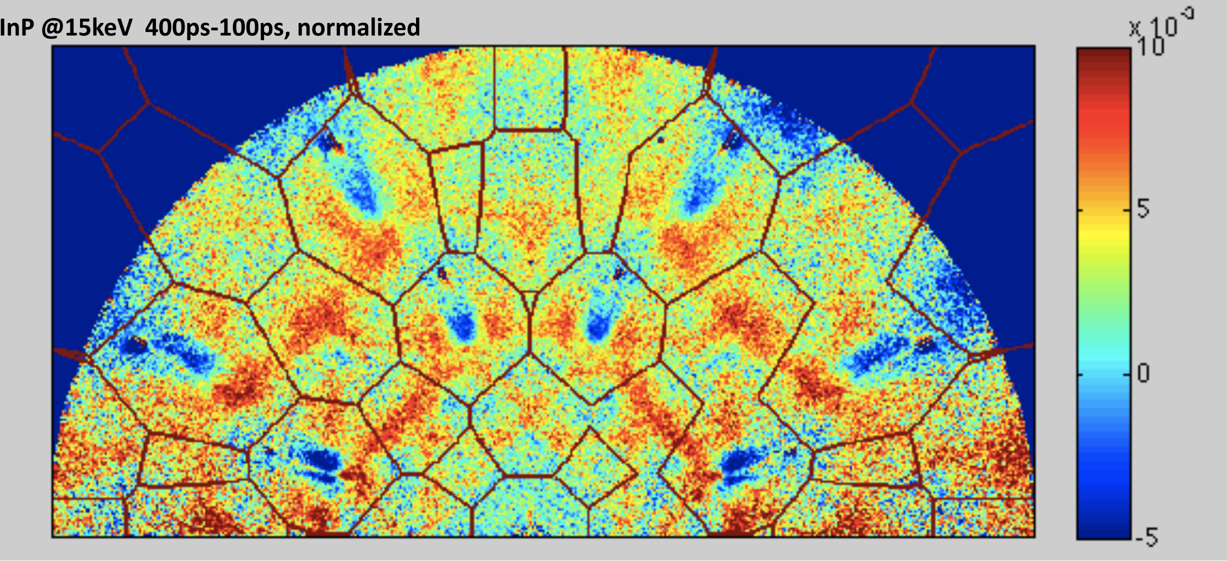
Photoexcited InP

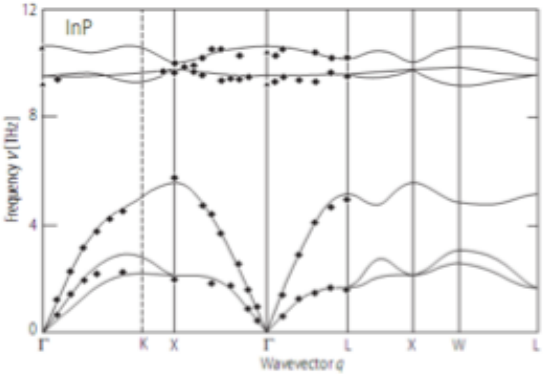


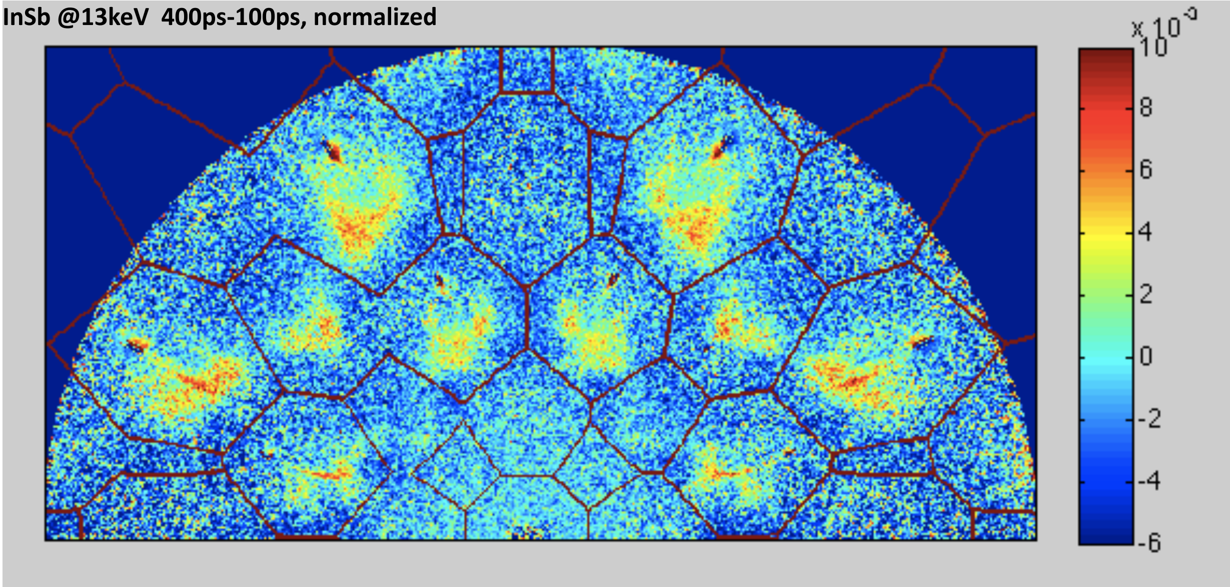


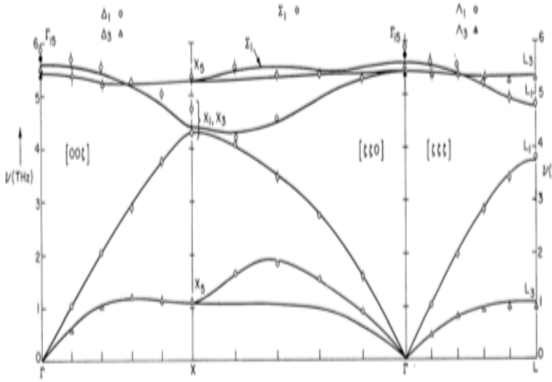
Trigo et al., submitted 2010

Time-resolved Diffuse Scattering Images of Nonequilibrium Phonons in InP and InSb


InP @15keV 400ps-100ps, normalized




InSb @13keV 400ps-100ps, normalized



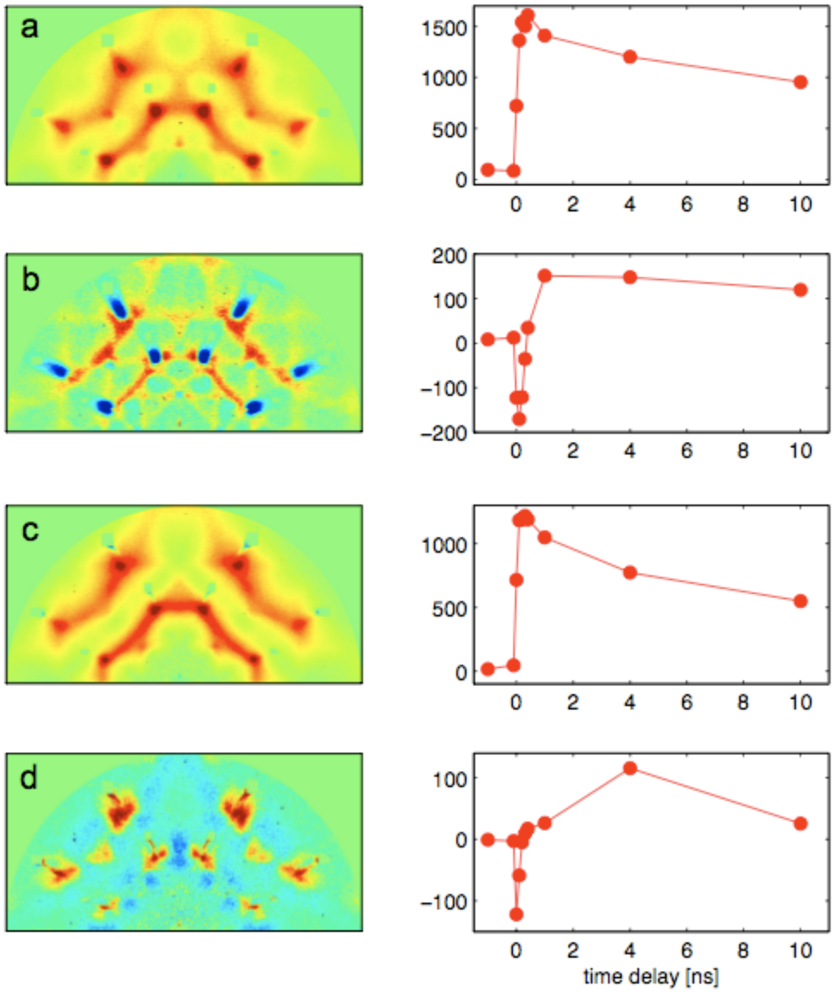
Trigo et al., (2010) unpublished.

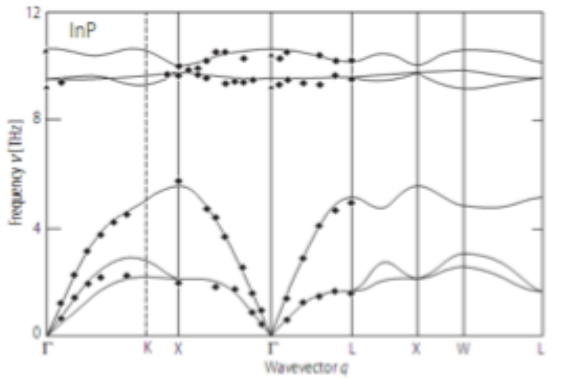
Trigo et al., submitted 2010

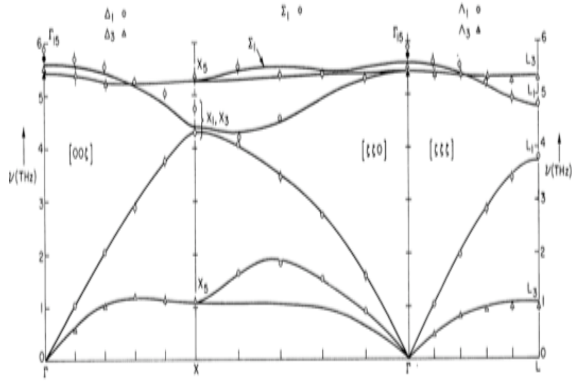
Singular value decomposition

InP

InSb





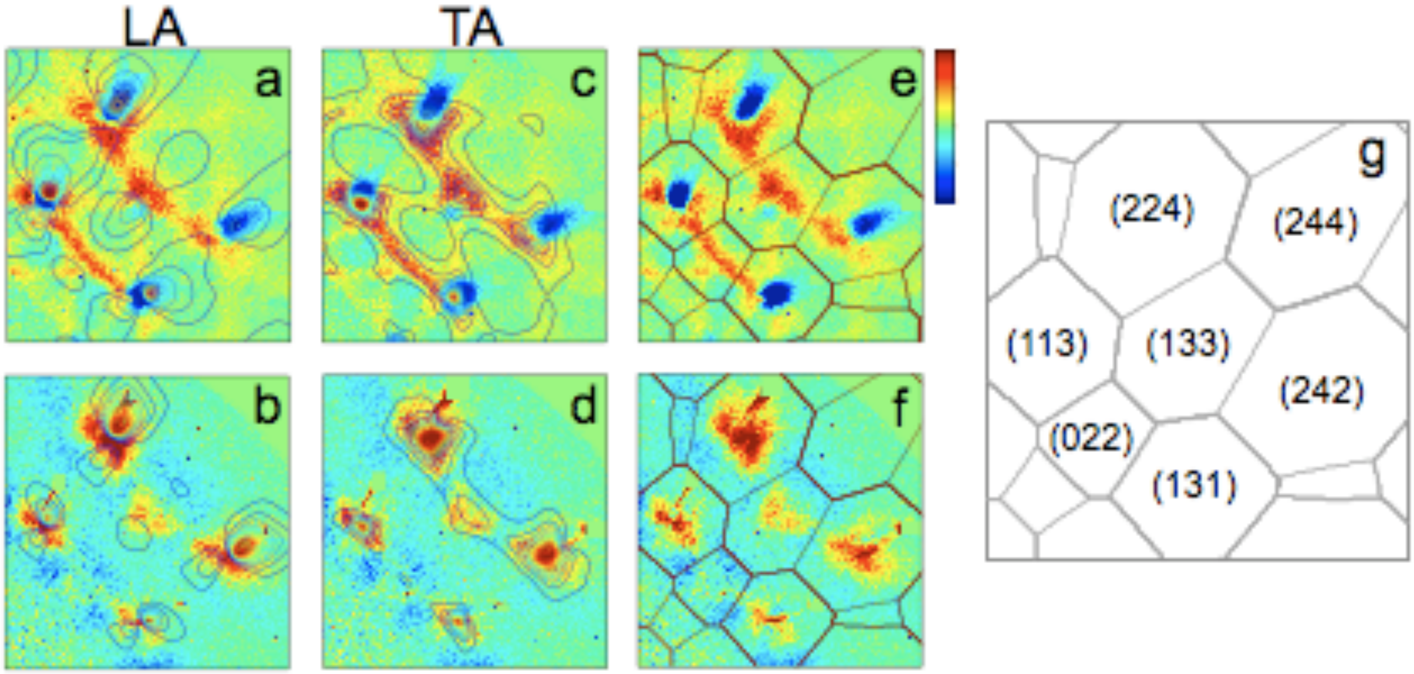


Trigo et al., submitted 2010

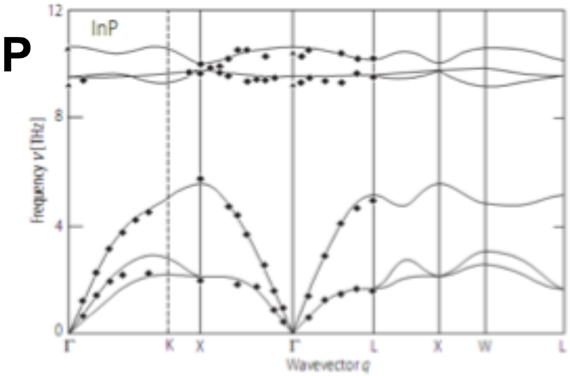
correlation with TA/LA branches

InP

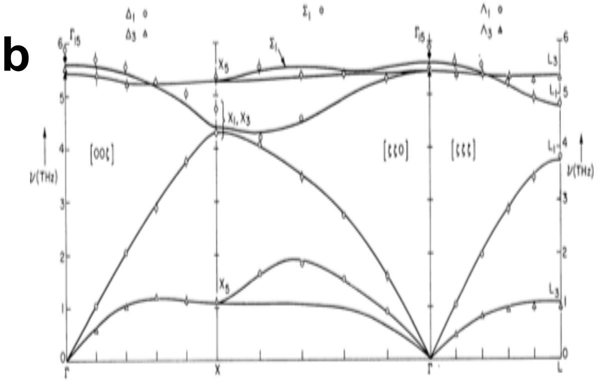
InSb



InP



InSb



Trigo et al., submitted 2010

Summary

- time-resolved x-ray scattering can be powerful tool for studying transient nonequilibrium dynamics.

- Progress on understanding of excited state in bismuth, but many fundamental questions remain.

- Surprising long-time nonequilibrium phonon dynamics found in photoexcited polar semiconductors.

- require intense short x-ray pulses (ala LCLS) and access to beamtime.

- much of discussion also applicable to electronic processes, resonance…

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