Condensed Matter Experiments Enabled by Picosecond X rays

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FOCUS Center and Department of Physics

University of Michigan

Acknowledgments

Students

(past) Adrian Cavalieri, David Fritz, Soo-Heyong Lee,

Jian Chen,Yu-Miin Sheu, Mariano Trigo,

Post Docs

Shambhu Ghimire, Emily Peterson

APS Sector 7 Staff Scientists

B. Adams, Dohn Arms, Eric Dufresne, Eric Landahl, Don Walko

SPPS collaboration

(~13 institutions/50 scientists, J. Hastings spokesperson,

A. Lindenberg, K. Gaffney, P. Hillyard, et al.)

and…

P. Bucksbaum, R. Clarke, M. DeCamp,

R. Goldman, R. Hegde, B. Lings, R. Merlin, E. Murray, S. Fahy, M. Reason, V. Stoica,

M. Swan, J. Wahlstrand, J. Wark,

…et al.

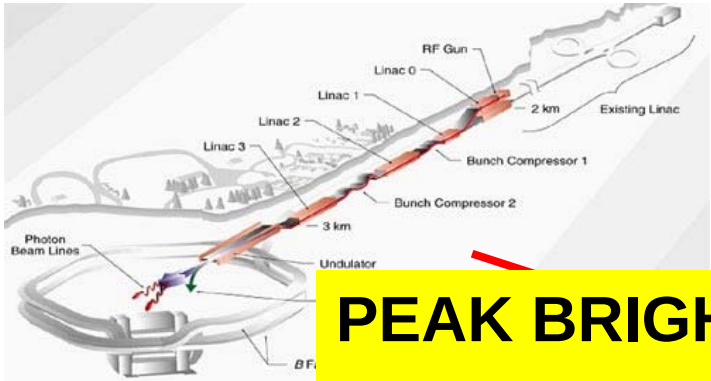
DOE BES AMOS Program

and Stanford PULSE Center

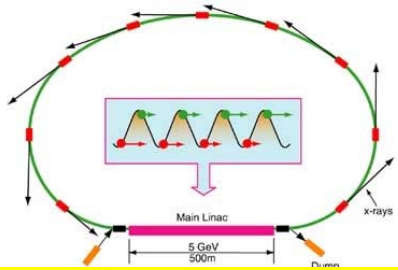
NSF

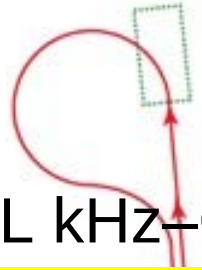
FOCUS PFC

A new Era of Ultrafast X-ray Sources



LCLS: 12





ERL kHz–GHz

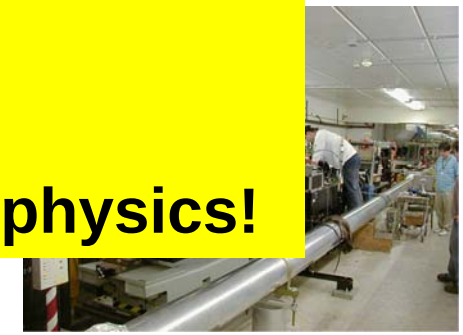
PEAK BRIGHTNESS ISN'T EVERYTHING:

Want combination of

- **short pulse**
- **tunability**
- **repetition rate**
- **accessibility**
- **average number of photons**
- **focusibility, etc…**

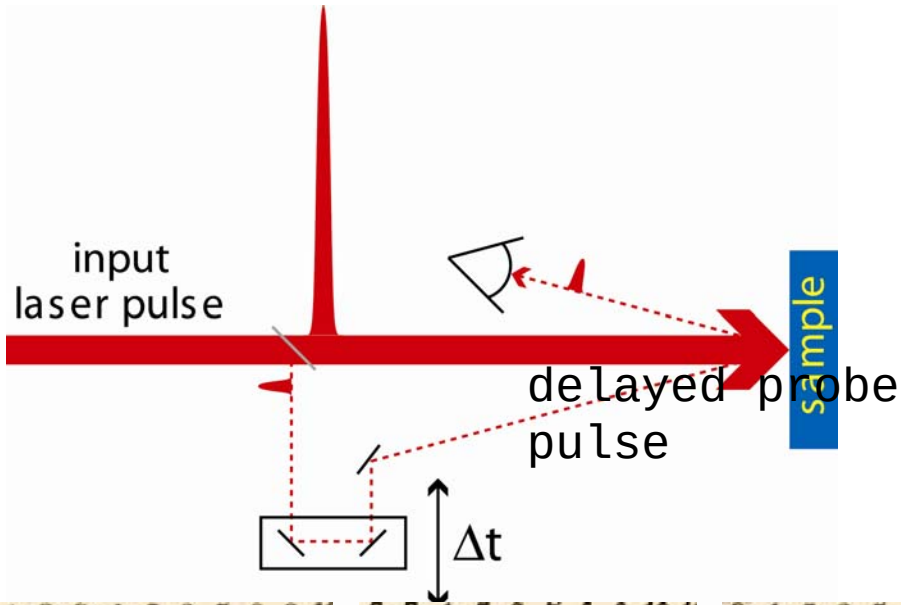
Flexible source will enable new physics!

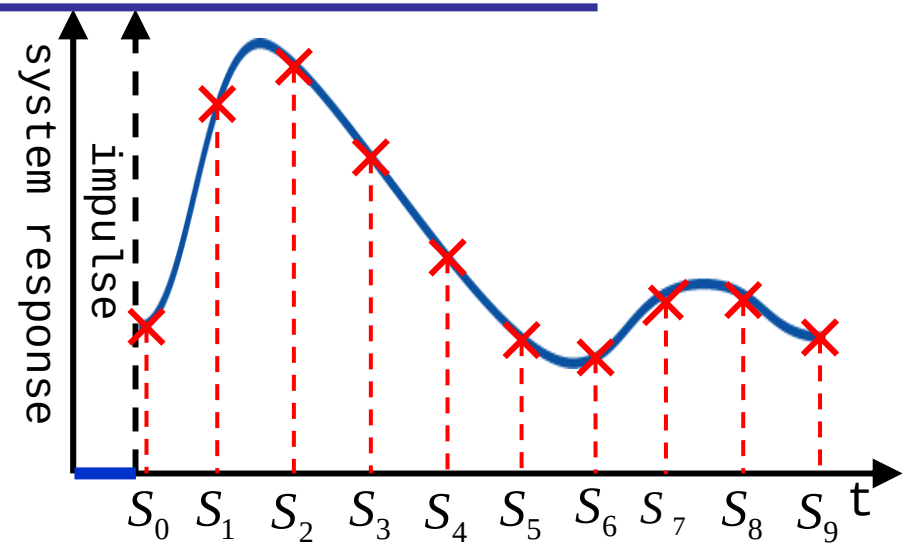
APS: MHz (ps upgrade 120–1kHz, 6.5MHz?)

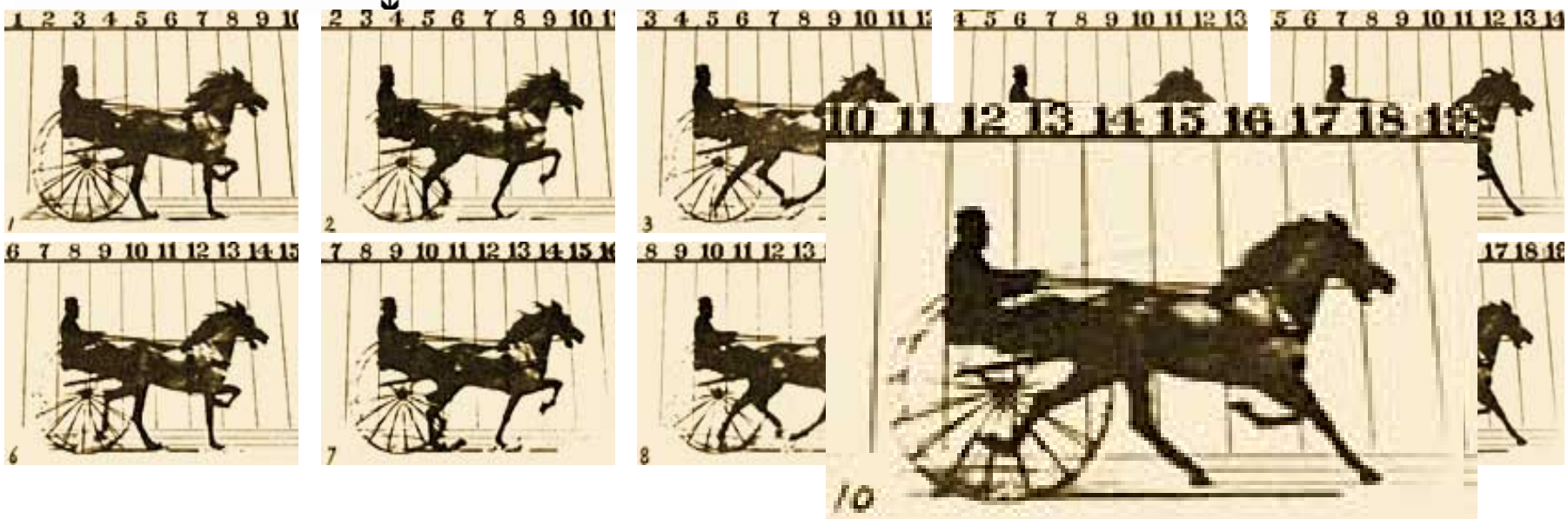


SPPS 10Hz

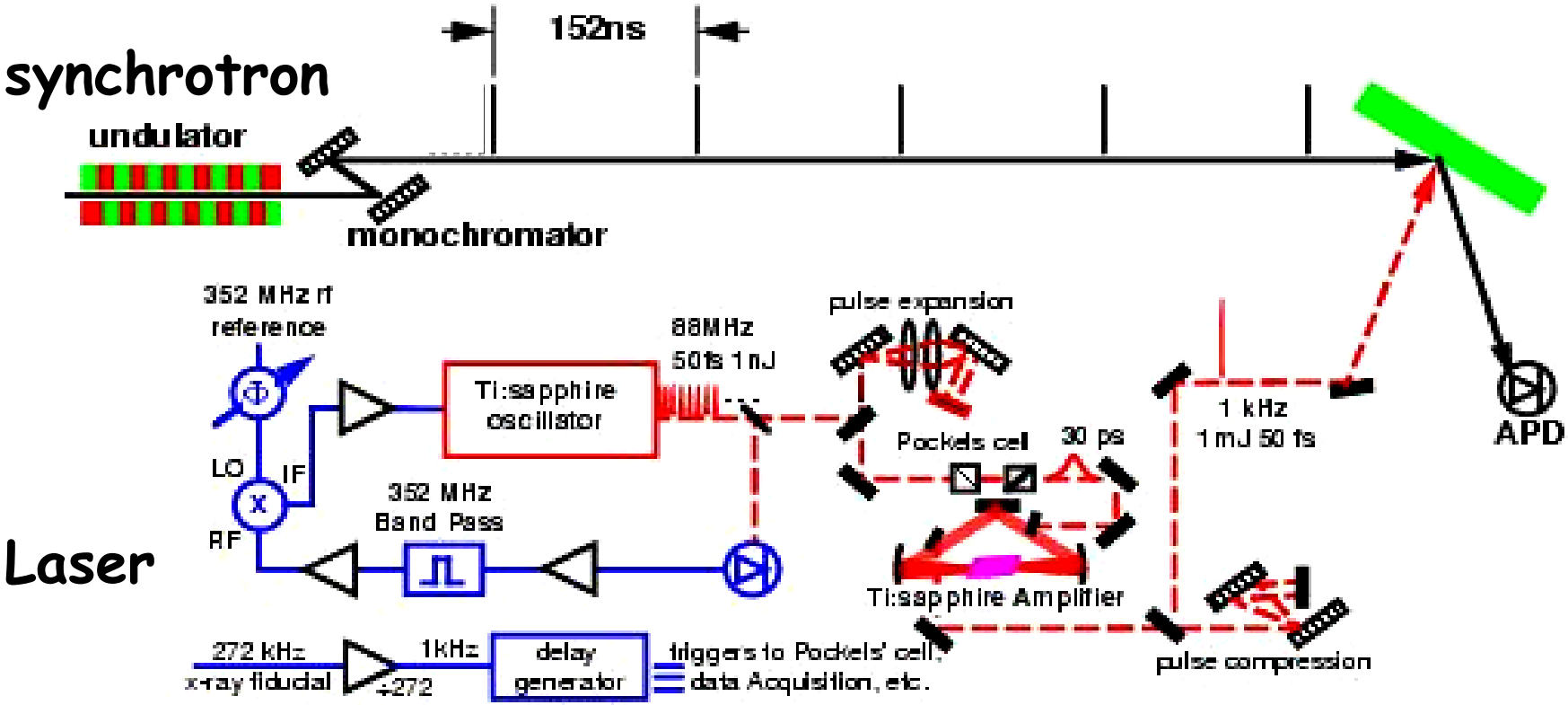
Pump-probe technique



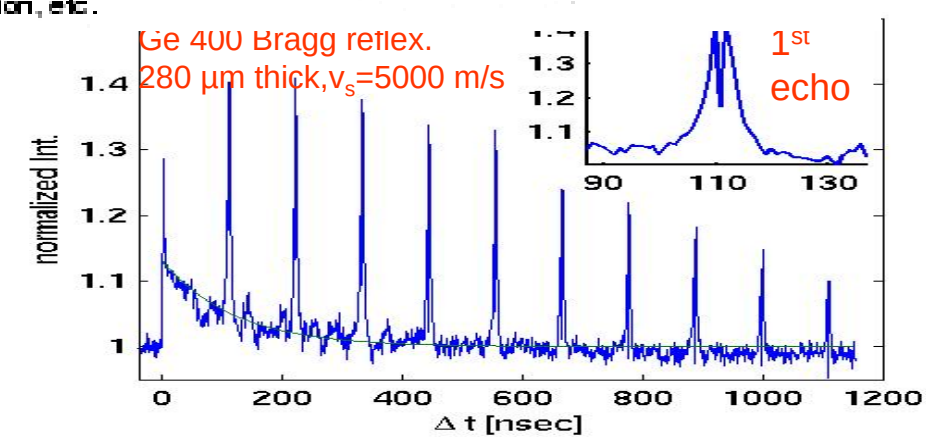




Pump-Probe at synchrotron

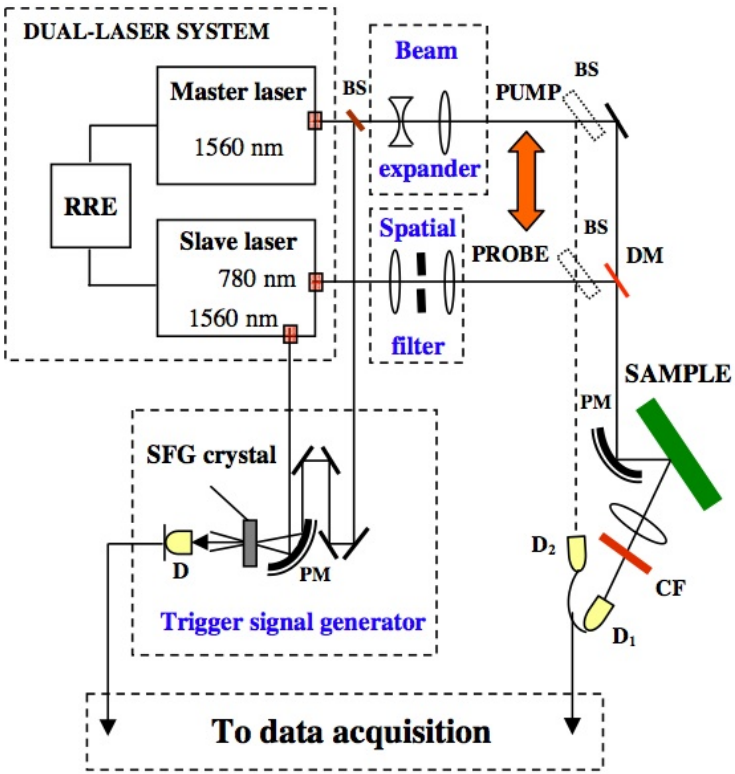


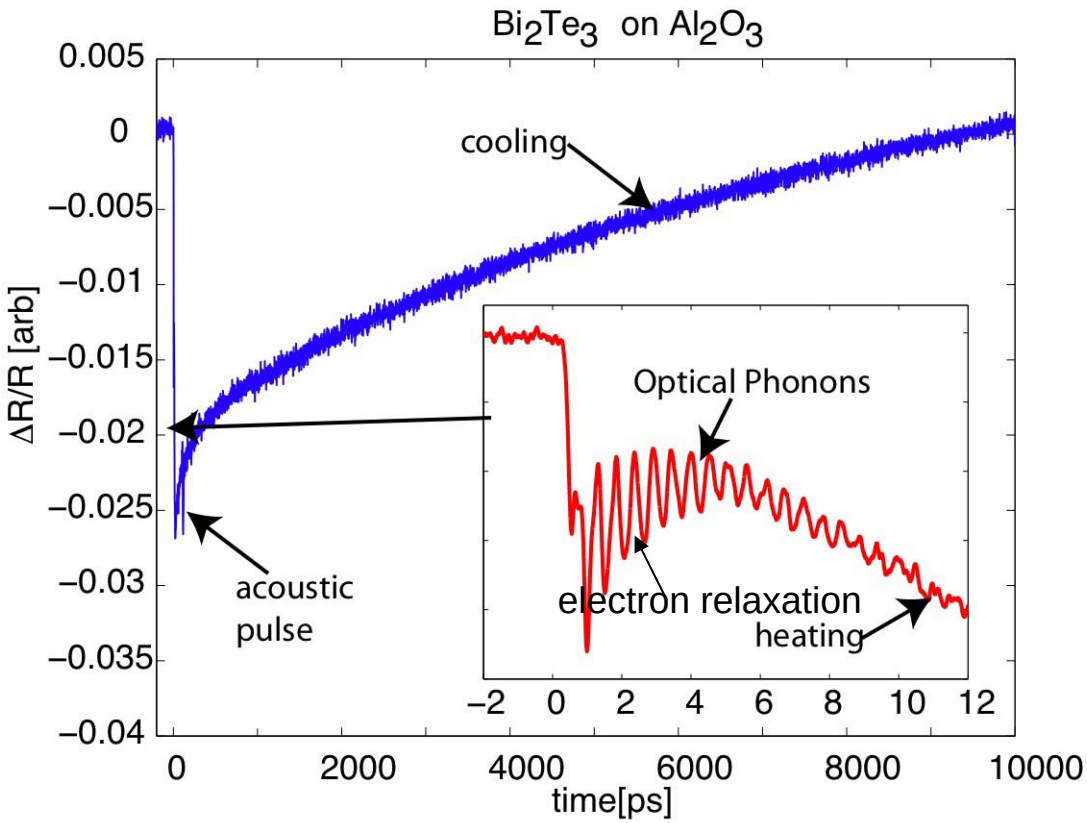
- Resolution limited by the bunch duration (or the timing jitter)
- Arbitrary pump-probe delay (NOT limited by bunch separation)



Solid-State Dynamics occur over vast time scales

Coherent and incoherent excitations in thin-film thermoelectrics

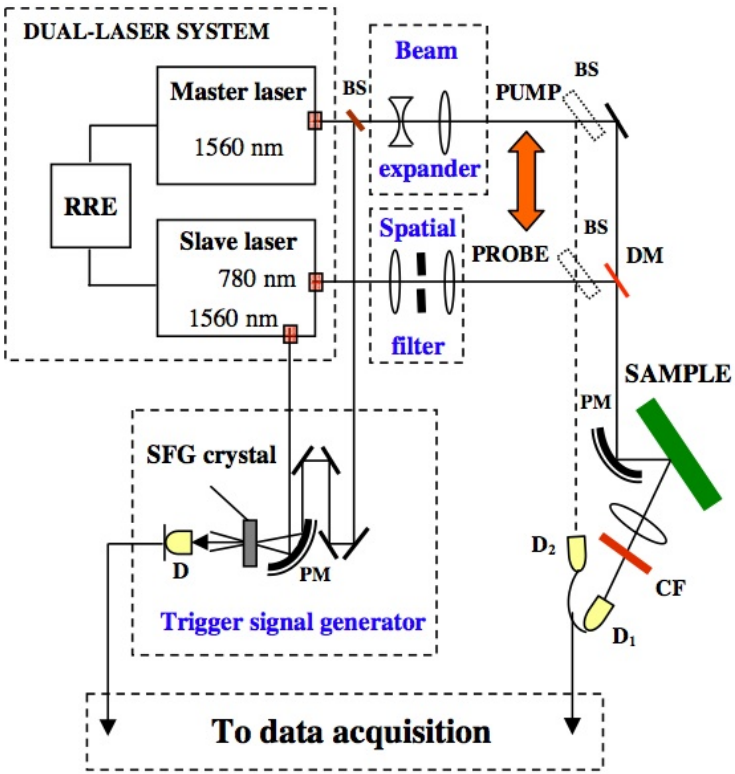


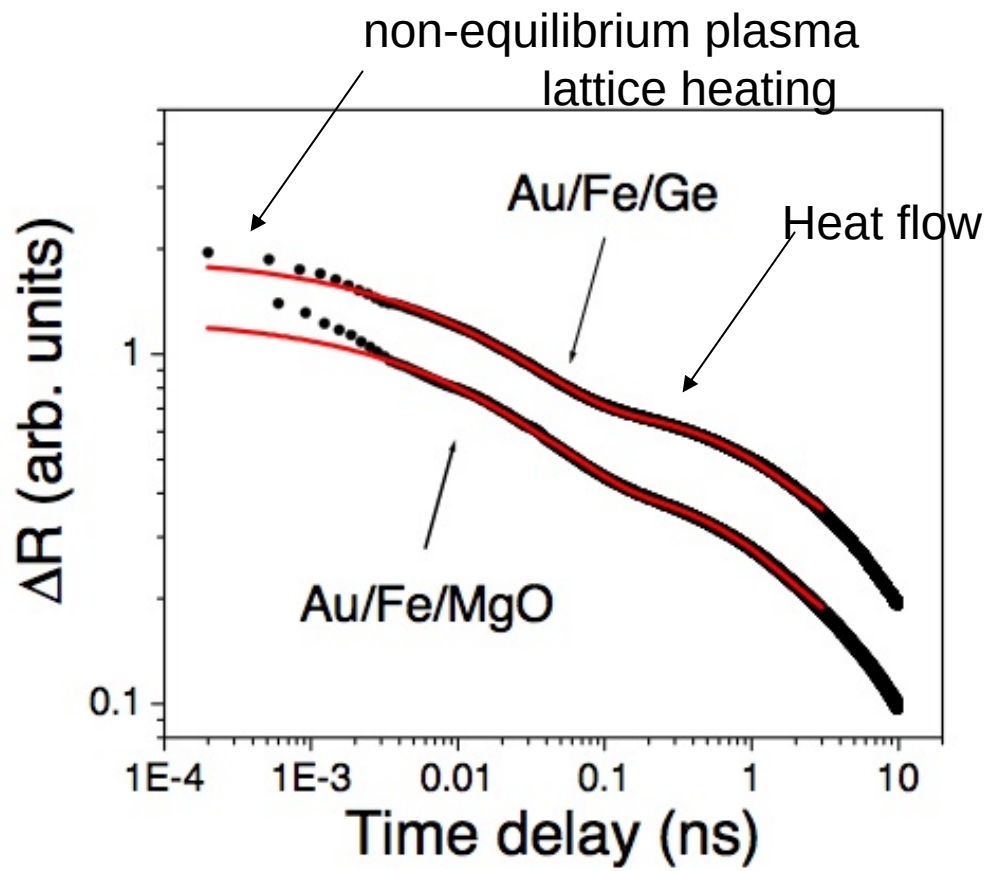


Y. Sheu, V. Stoica, R. Clarke, C. Uher and D. Reis unpublished

Solid-State Dynamics occur over vast time scales

Thermal transport in thin metal films

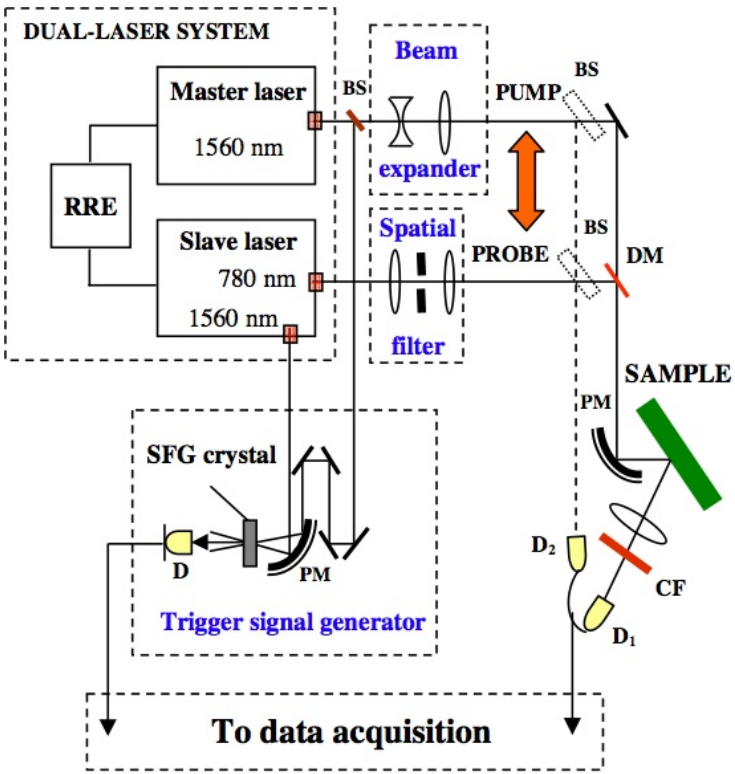


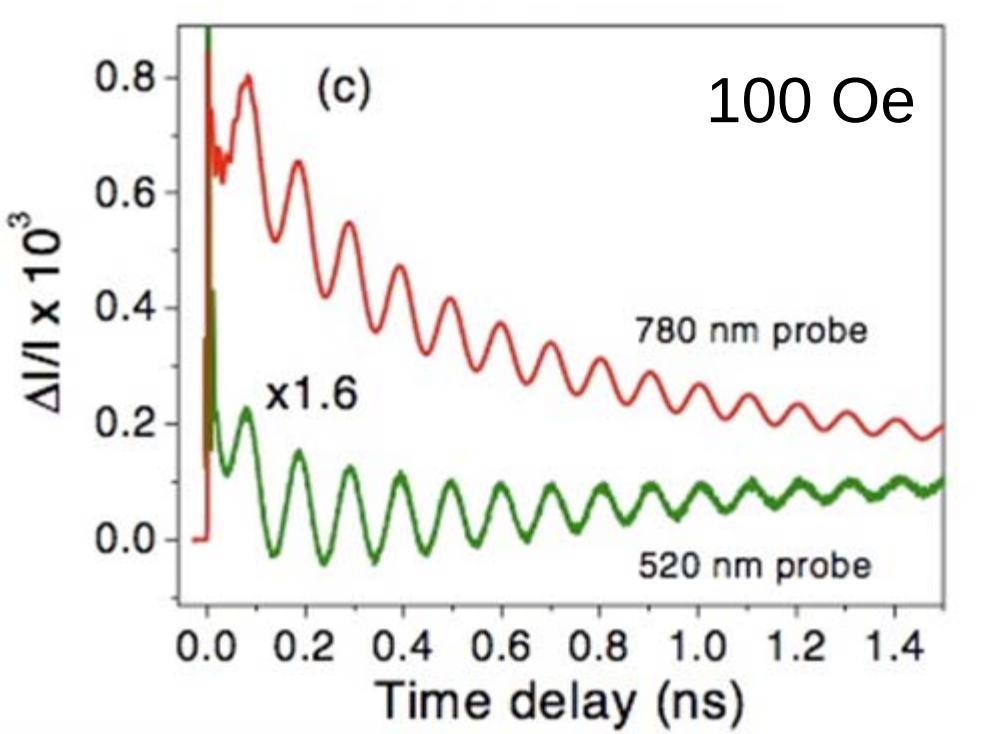


V. Stoica, Y. Sheu, D. Reis, and R. Clarke, "Wideband detection of transient solid-state dynamics using ultrafast fiber lasers and asynchronous optical sampling," Opt. Express 16, 2322-2335 (2008).

Solid-State Dynamics occur over vast time scales

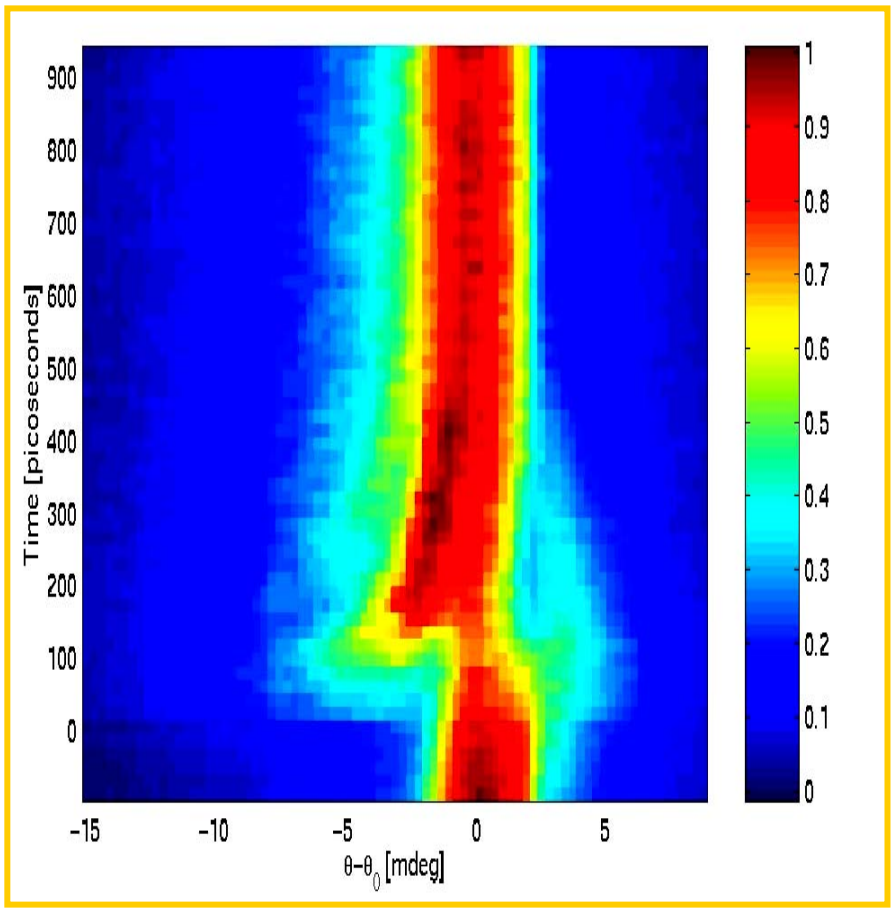
Magnitization Dynamics of Fe/Ge films

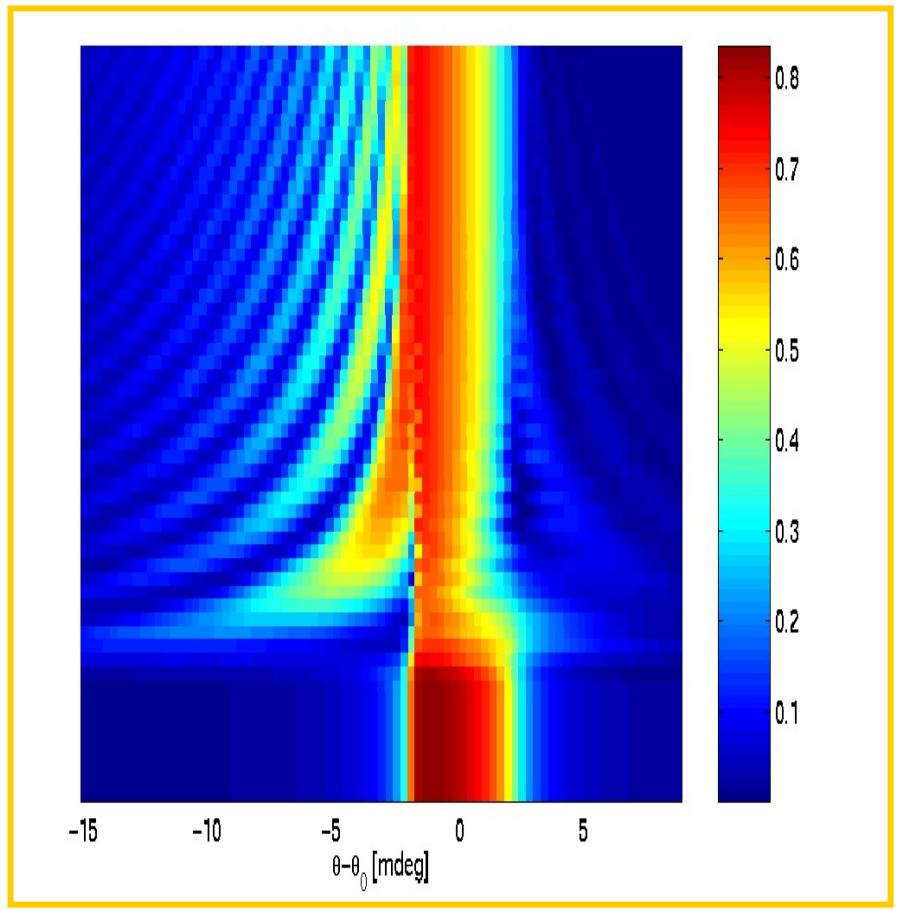




V. Stoica, Y. Sheu, D. Reis, and R. Clarke, "Wideband detection of transient solid-state dynamics using ultrafast fiber lasers and asynchronous optical sampling," Opt. Express 16, 2322-2335 (2008).

Time-resolved Bragg Diffraction: Coherent Acoustic Phonons



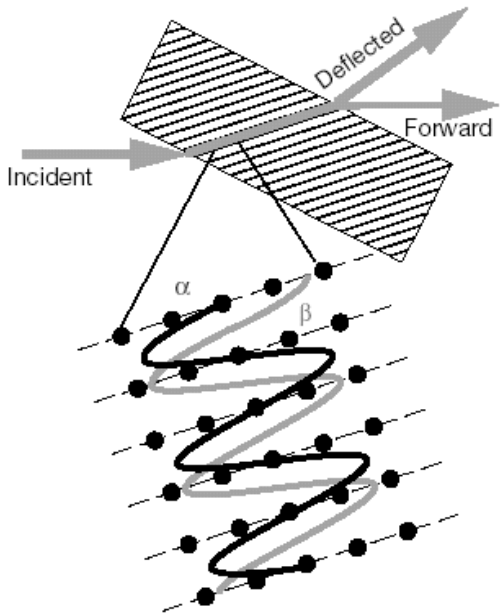


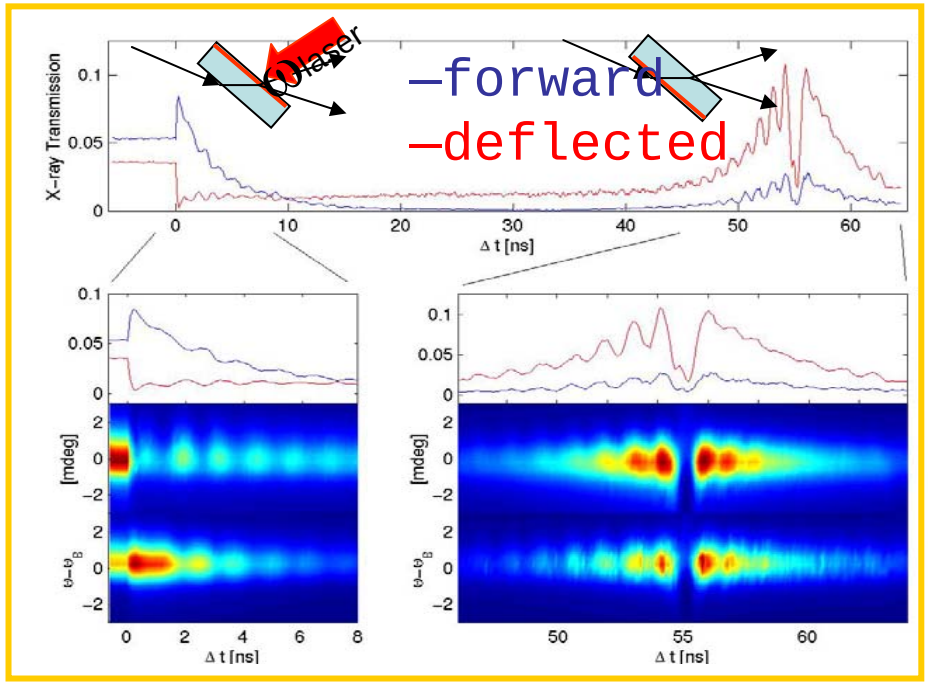
experiment: InSb 111, 10mJ/cm^2

simulation: 100ps & 1.25mdeg conv.

Reis *et al.* Phys Rev. Lett.(86) 2001

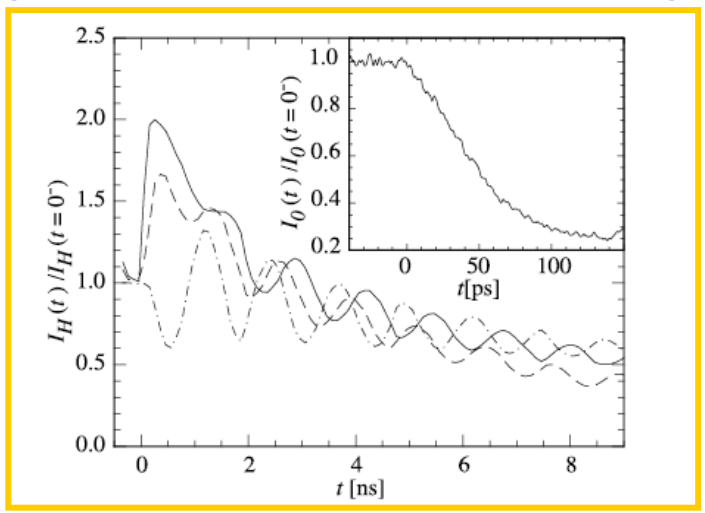
Coherent Control of Pulsed X-ray Beams

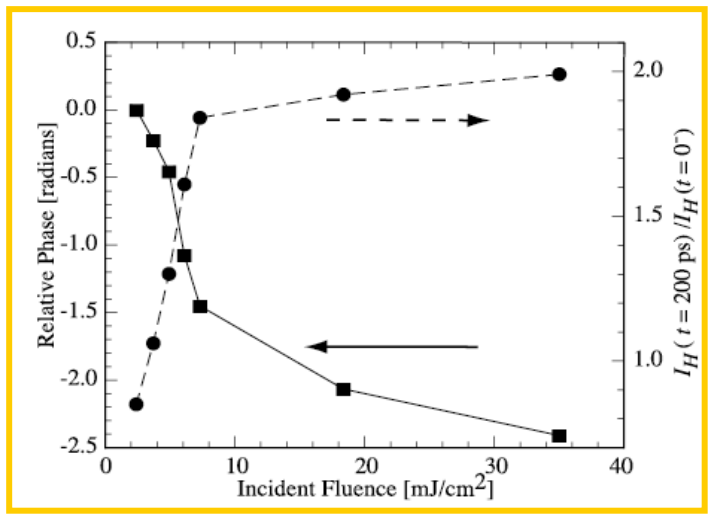




M.F. DeCamp *et al.*, Nature 413, 825 2001

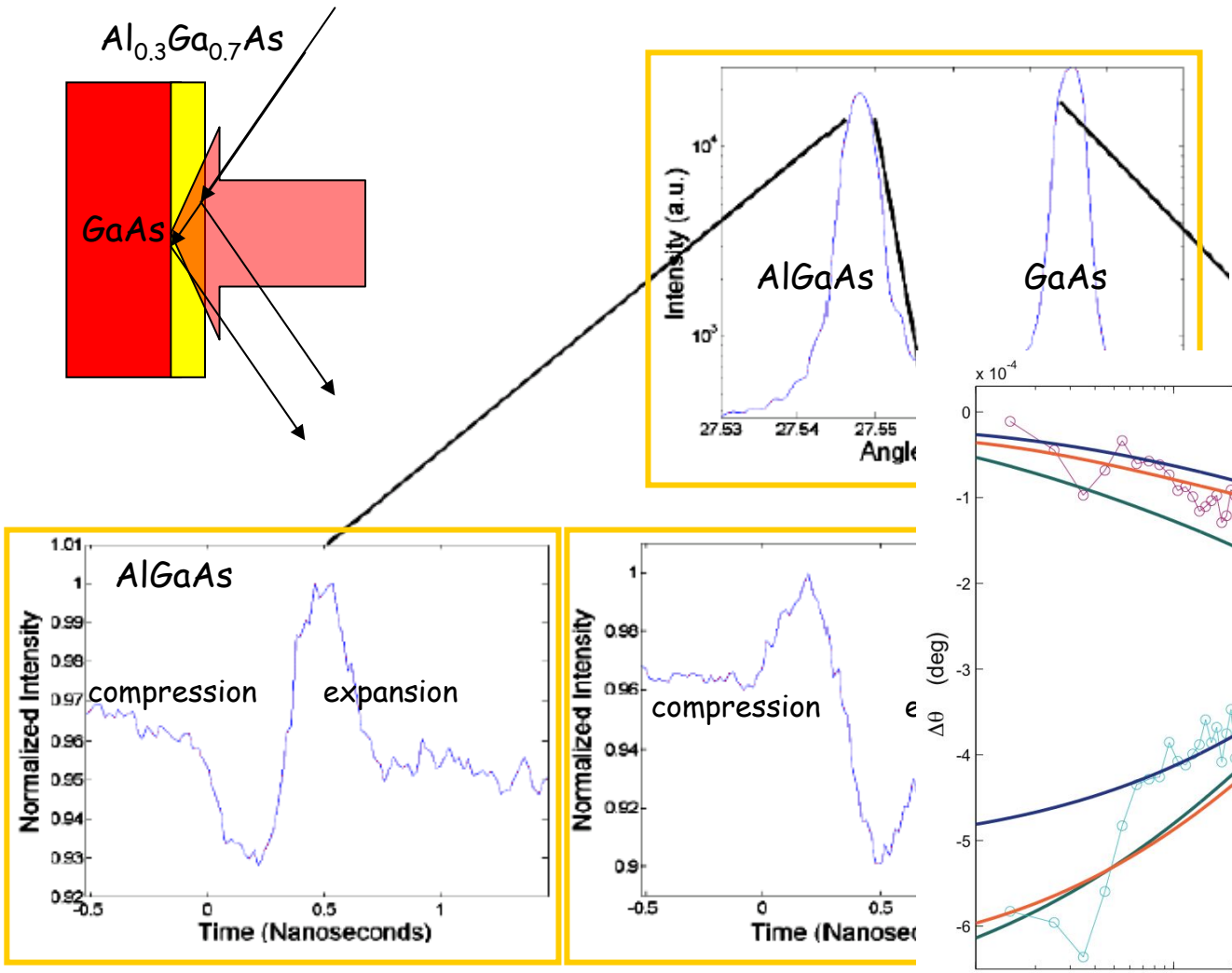
Ambipolar diffusion of dense e-h plasma



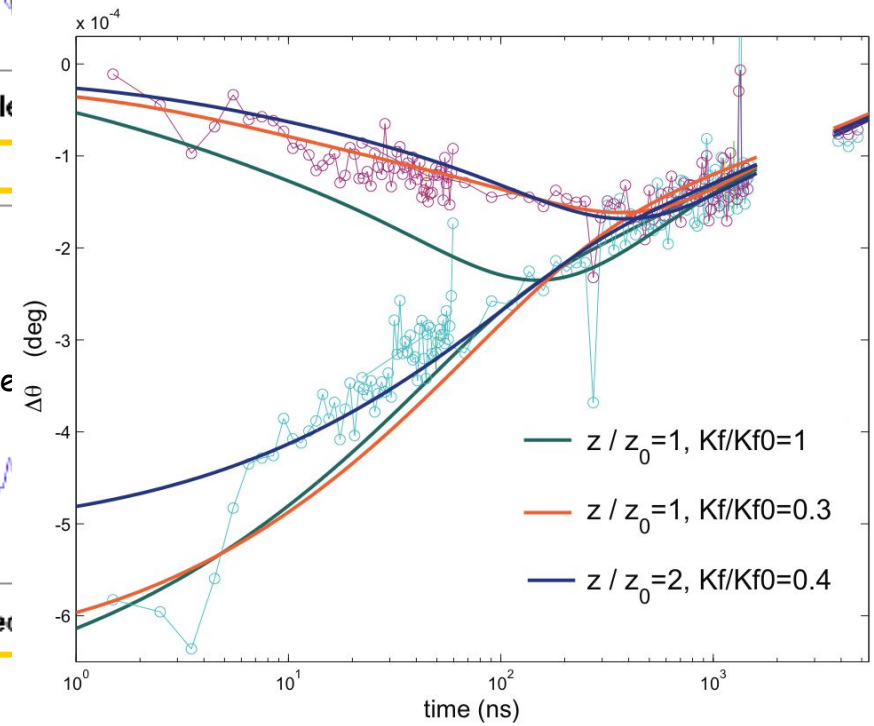


M.F. DeCamp *et al.*, Phys. Rev. Lett. 91, 165502, 2003.

Electronic, Thermal and Mechanical Properties of thin films and interfaces

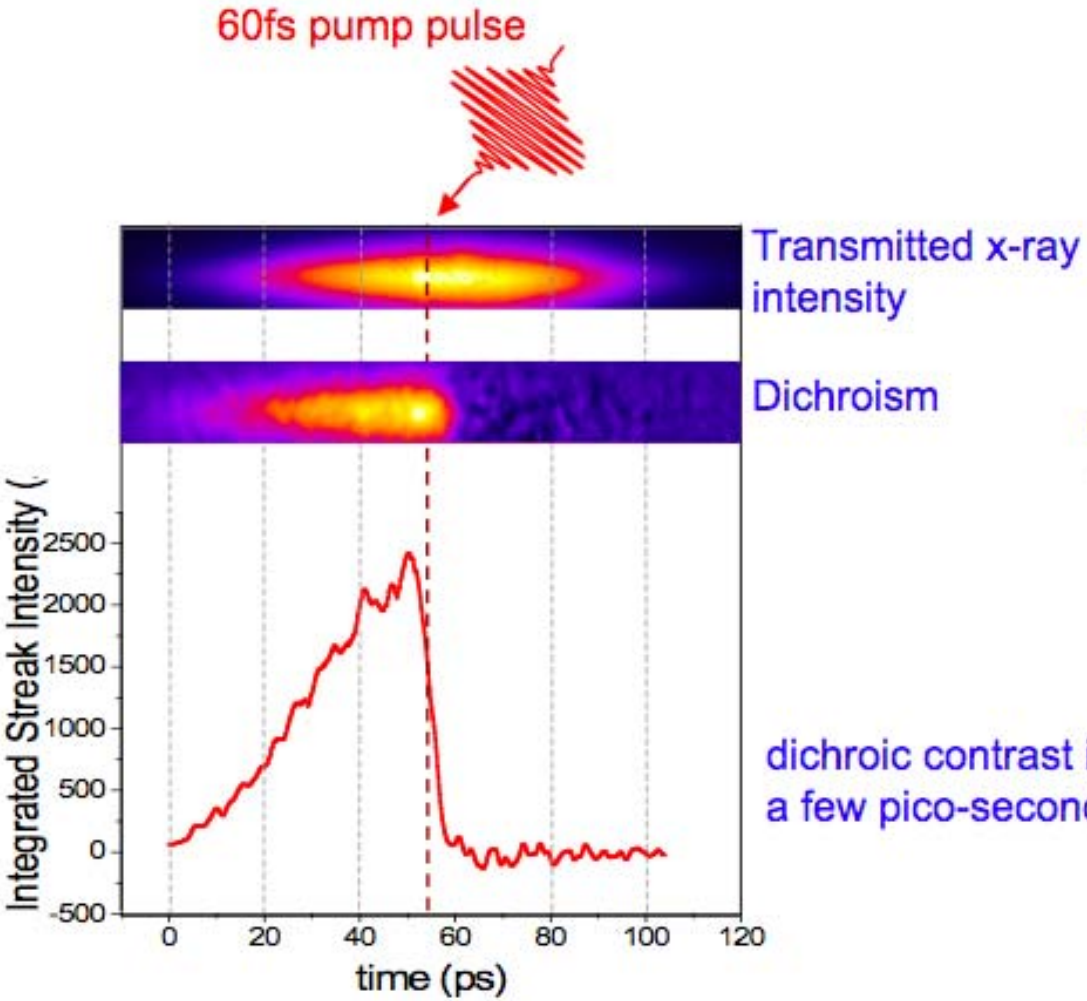


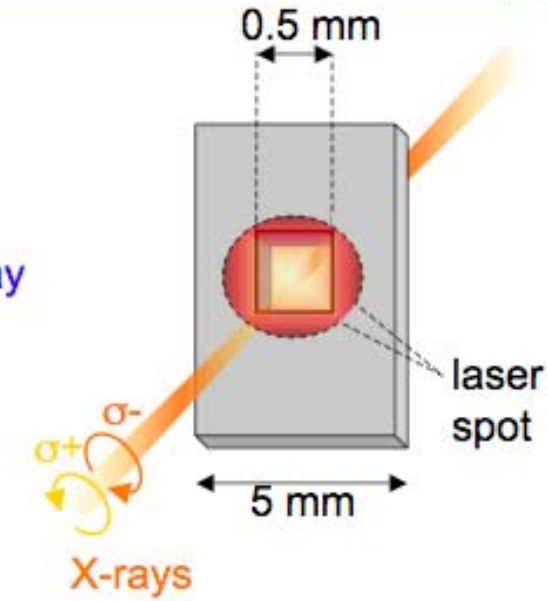
- Deformation potential and thermal expansion.
- Free carrier absorption
- Electron-phonon
- Boundary resistance (Kapitza)
- thermal conductivity and



S.H. Lee *et al.*, PRL 95, 246104 (2005); Sheu et al. PRB with referees (2008)

Time-Resolved XMCD at Fe L_3 Edge



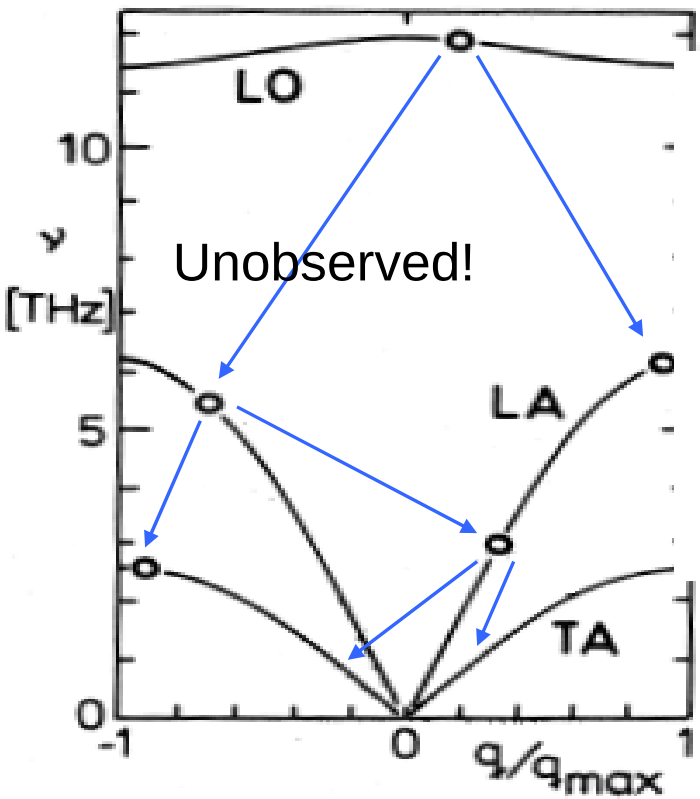


dichroic contrast is lost in a few pico-seconds

A. Scholl

Ultrafast Vibrational Spectrocsopy of Throughout Brillouin zone

A Motivation: anharmonic decay of phonons
(wavevector resolved!)



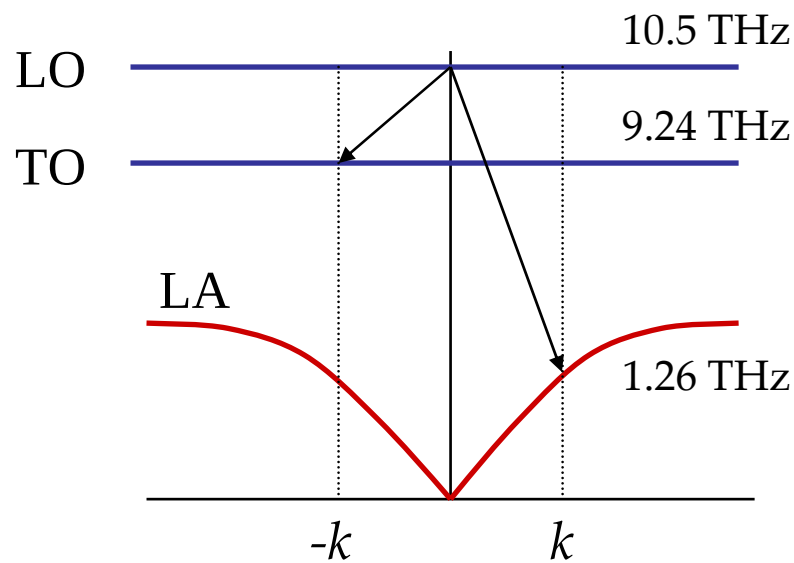
QuickTime™ and a
TIFF (LZW) decompressor
are needed to see this picture.

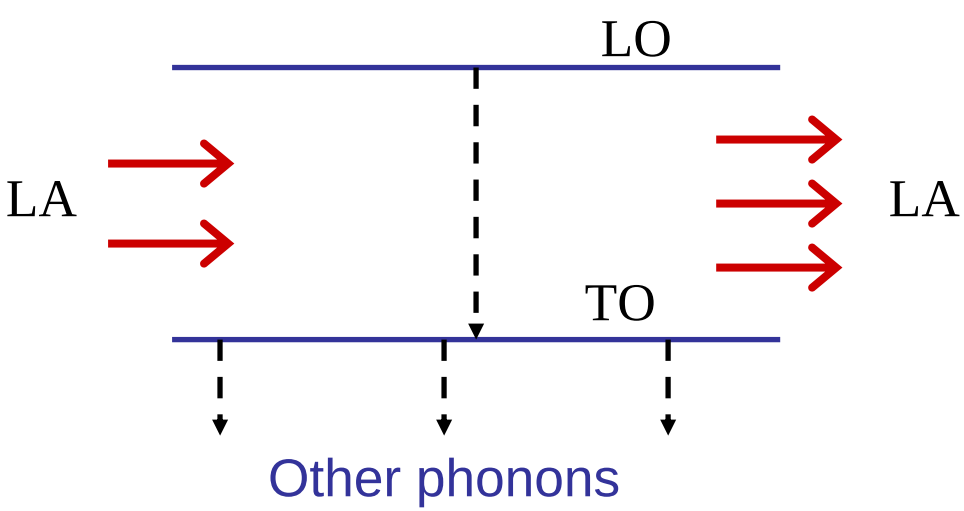
Holt et al, PRL 83, 1999

One Motivation: Optical-phonon lifetime of InP

InP:

Decay channel:
LO → TO + LA

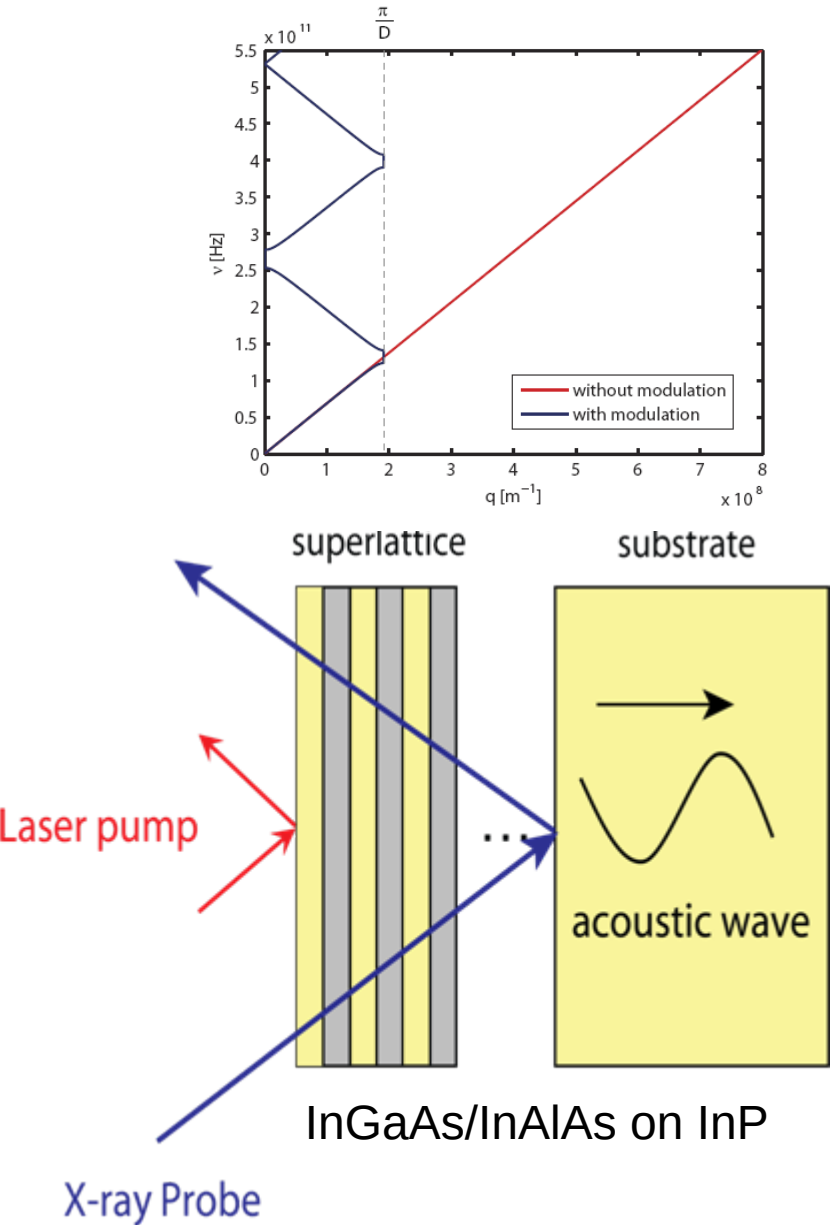




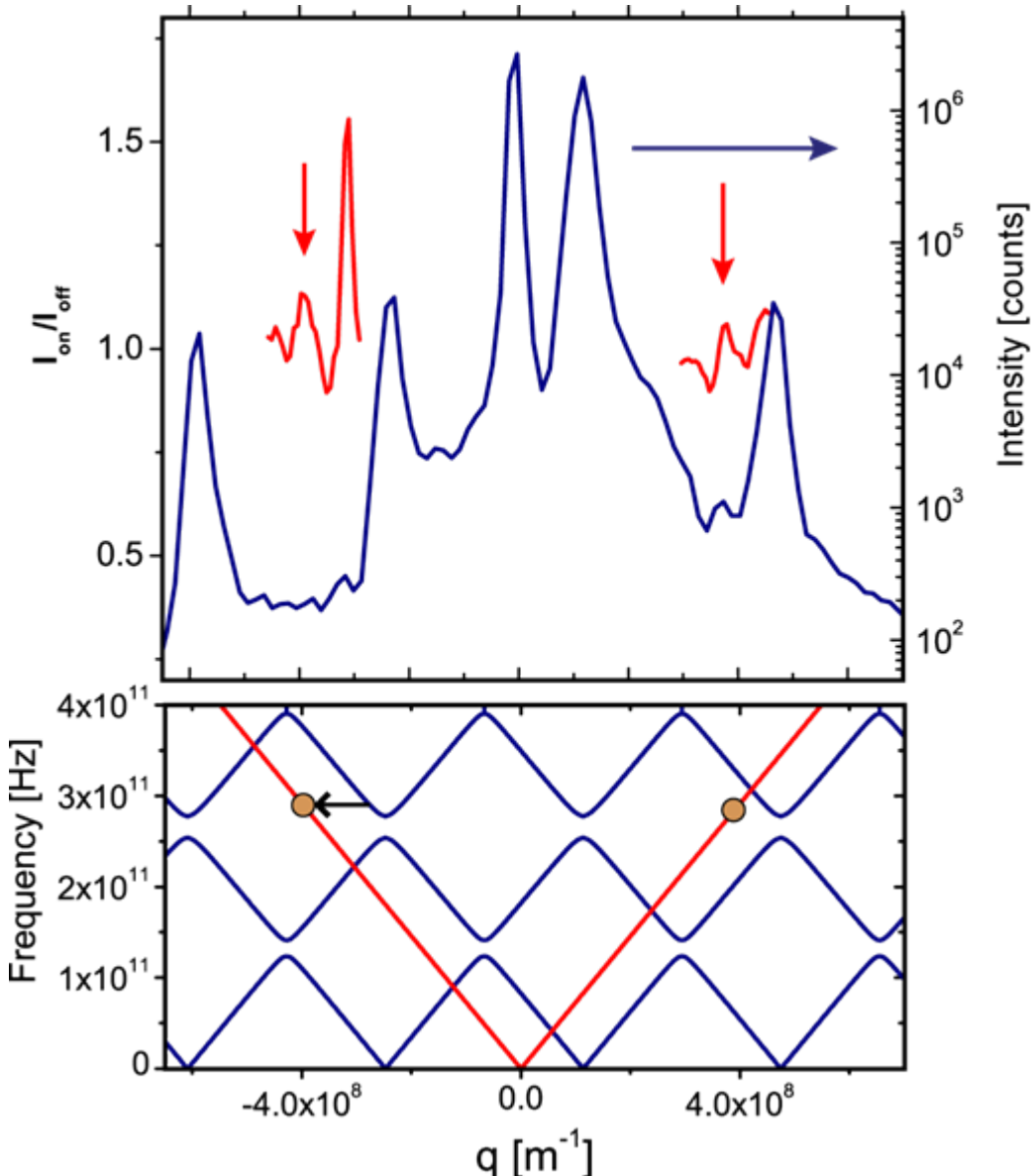
Stimulated decay and emission of TO + LA

Future work…

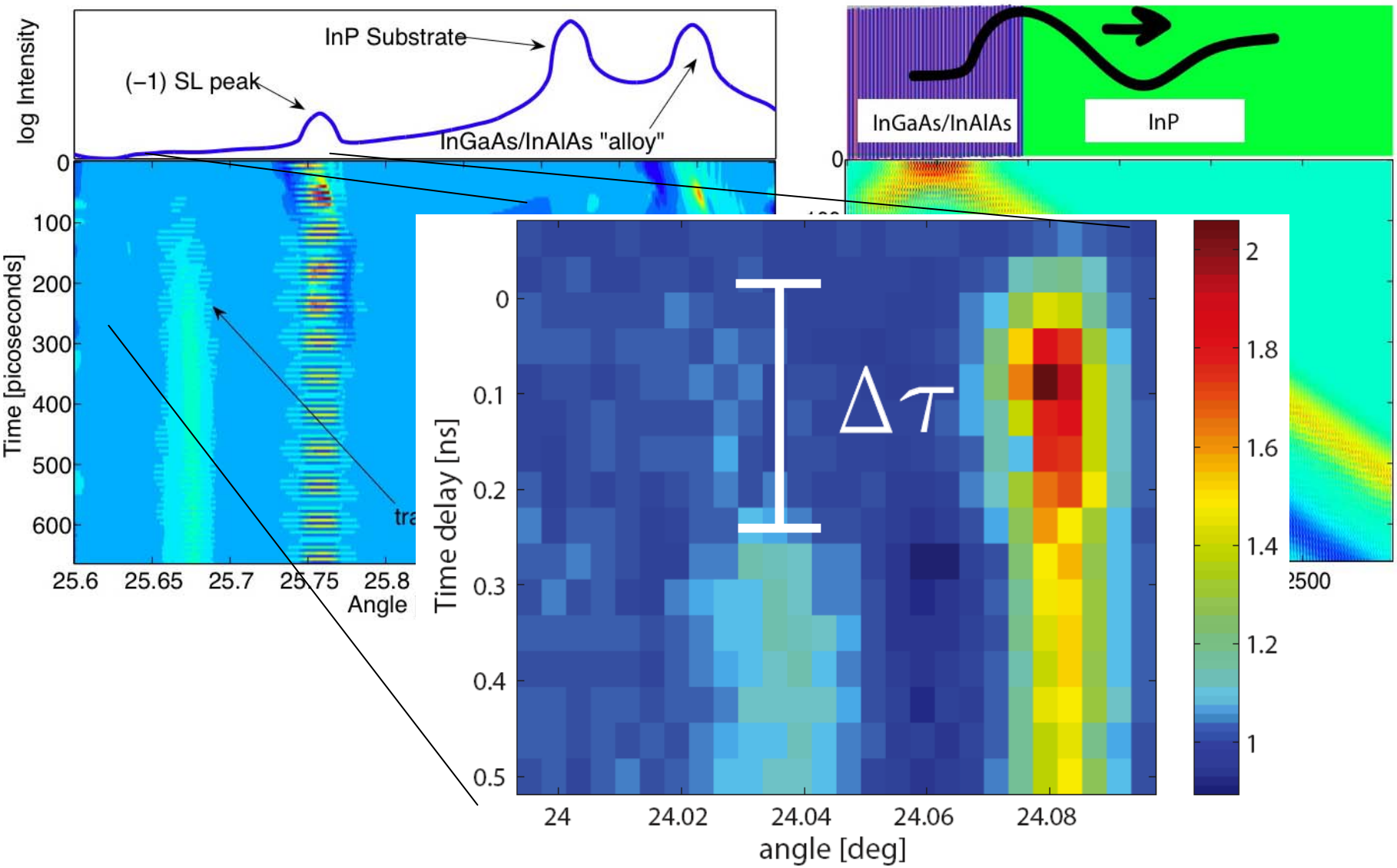
Coherent zone *unfolded* phonons: X-ray diffraction experiments (100ps)



InGaAs/InAlAs on InP

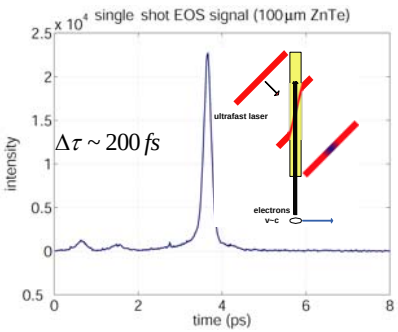


Coherent zone *unfolded* phonons: X-ray diffraction experiments (100ps)

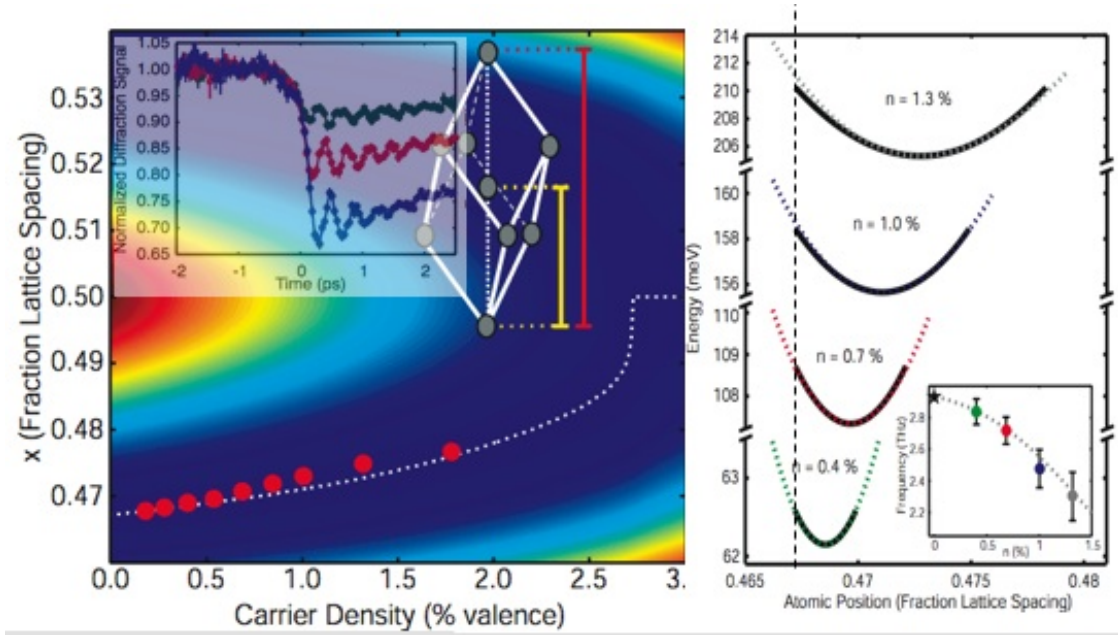


State-of-the-art in ultrafast x-ray science

Measurement of the Interatomic Potential:
Enabled by ultafast x rays and random sampling technique

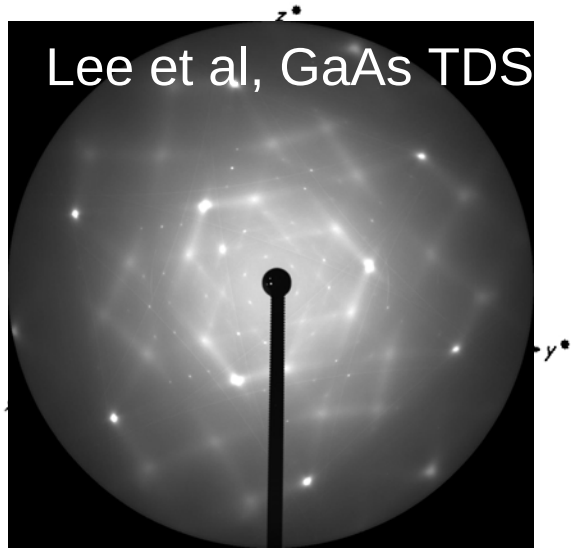


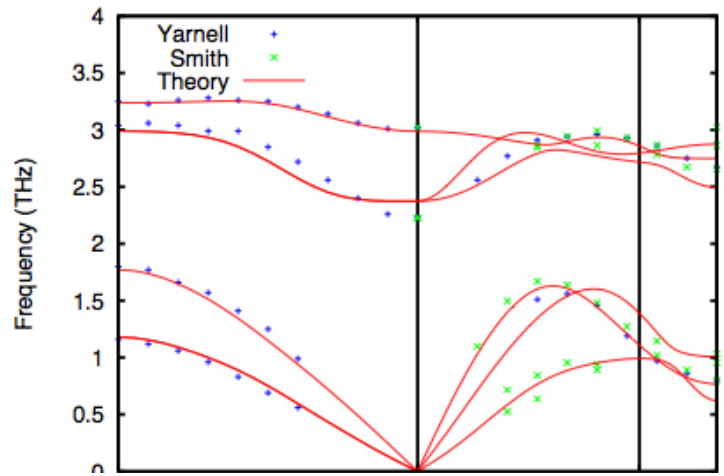
and a
ompressor
is picture.

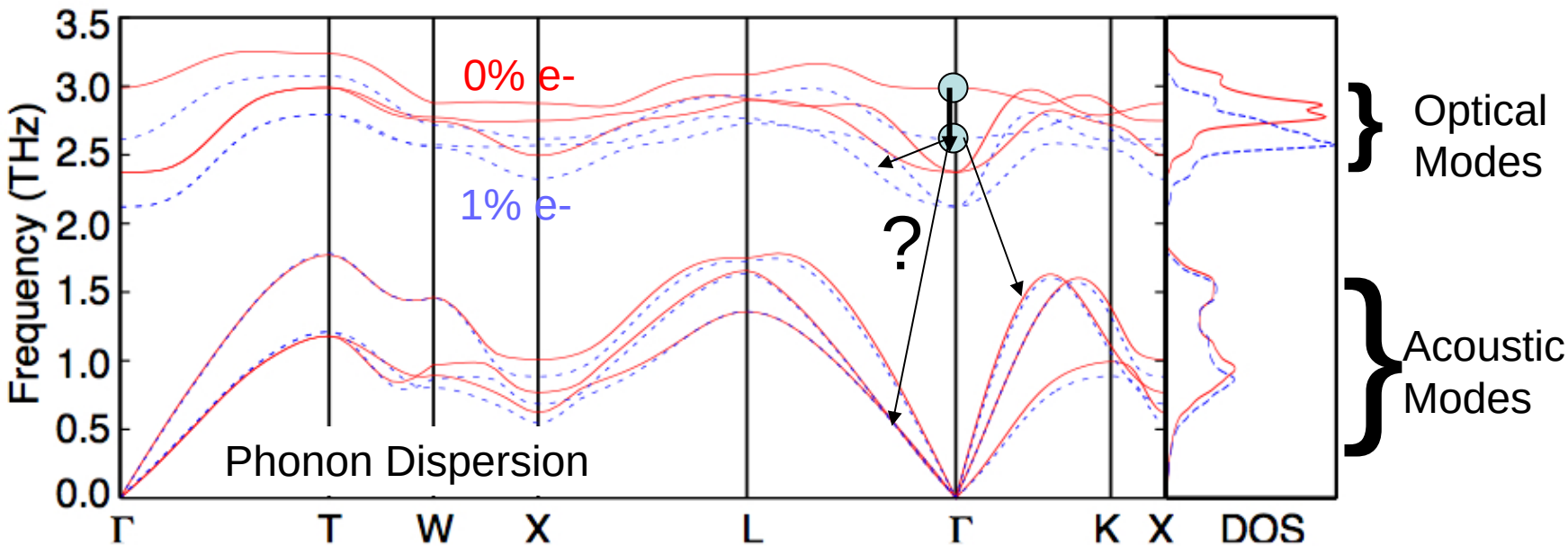


A. Cavalieri et al., Phys. Rev. Lett. 94 144801, 2005 D. M. Fritz et al. Science, 315:633–636, 2007.

Photoexcited carriers change the full vibrational spectra of Bi



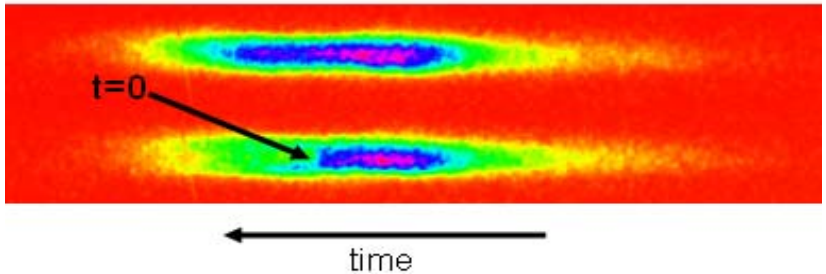


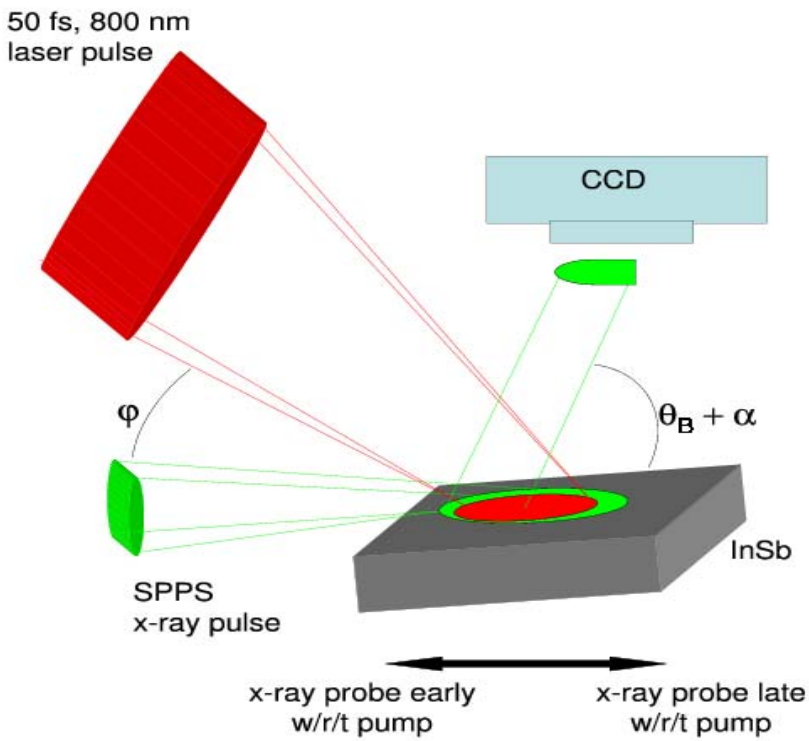


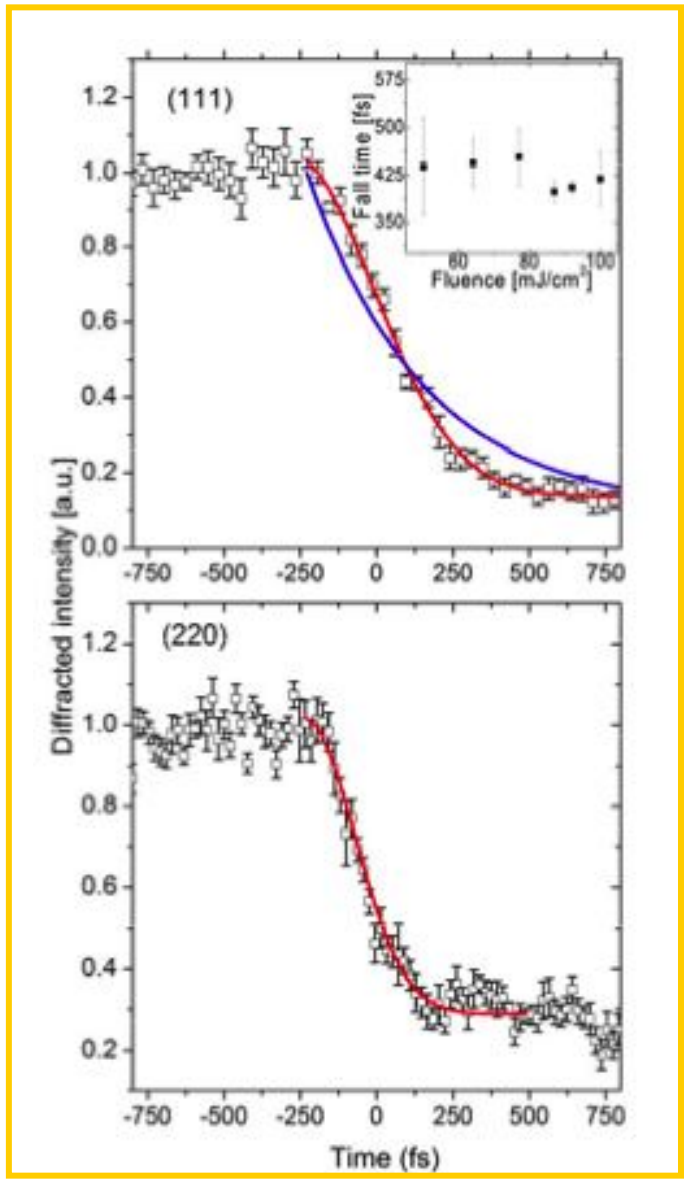
Eigenstates of the Interatomic Forces

Murray *et al.* PRB 75 2007.

Observation of Ultrafast Melting?

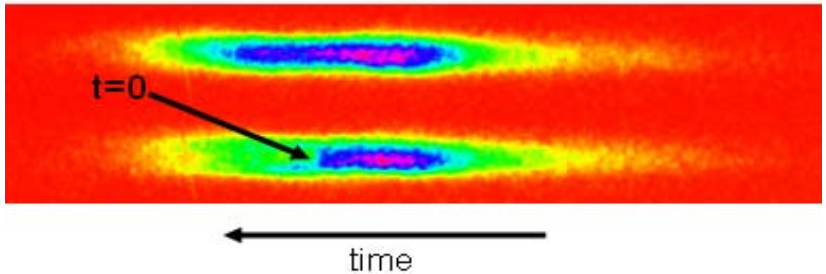


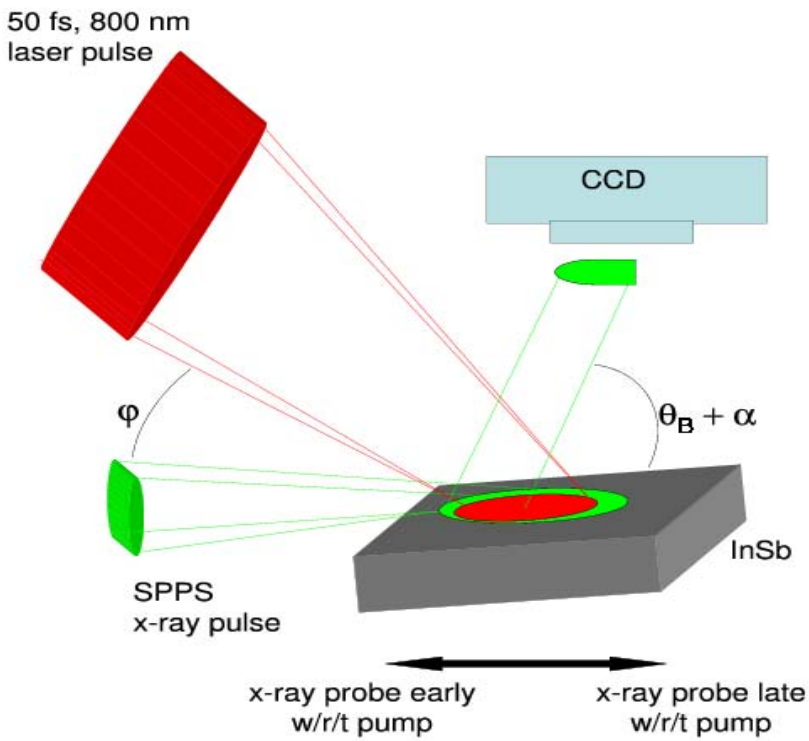


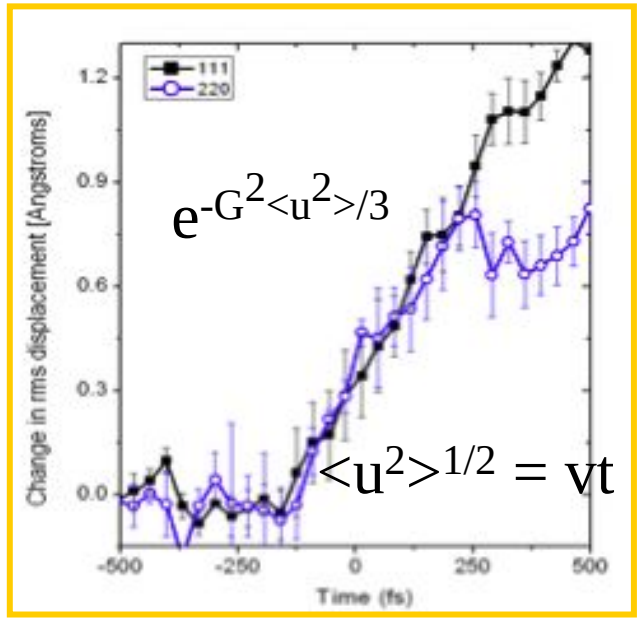


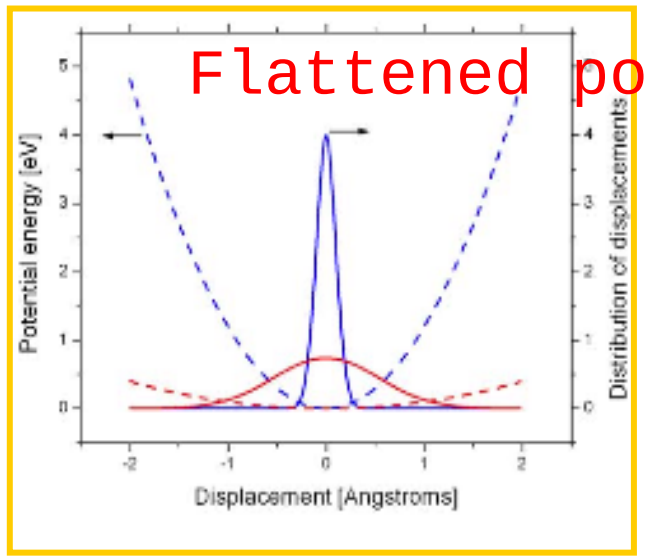
Lindenberg *et al.,* Science 308, 2005;
Gaffney *et al.*, PRL 95, 2005; Hillyard et al., PRL 98, 2007.

Observation of Ultrafast Melting?







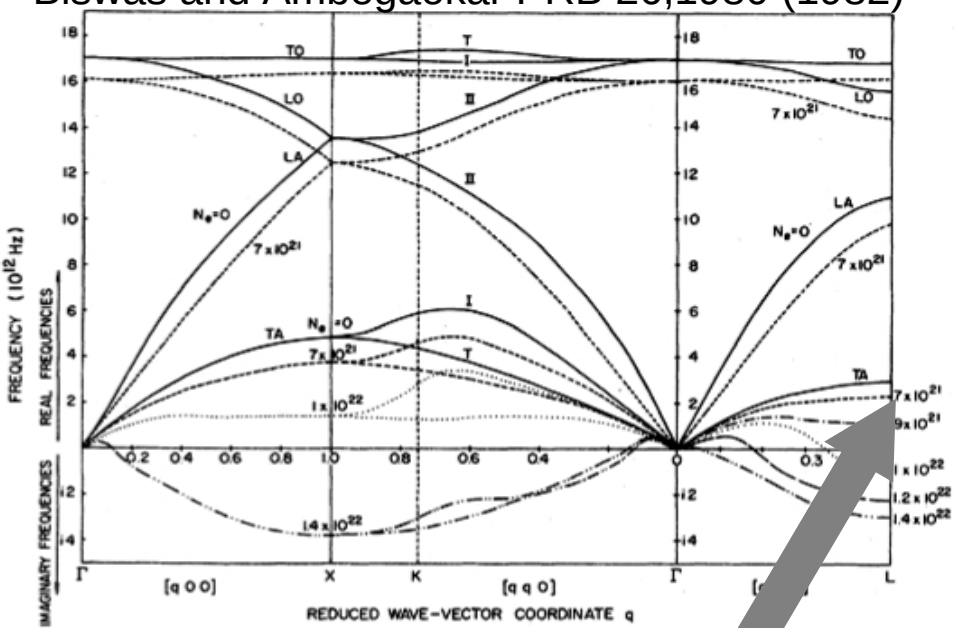


Lindenberg *et al.,* Science 308, 2005;
Gaffney *et al.*, PRL 95, 2005; Hillyard et al., PRL 98, 2007.

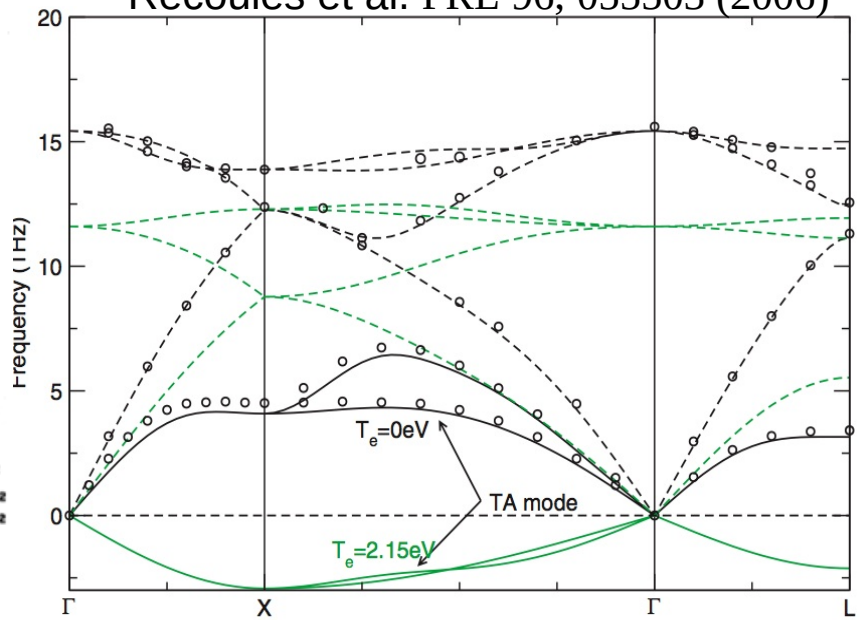
Theoretical Predictions of Ultrafast Melting

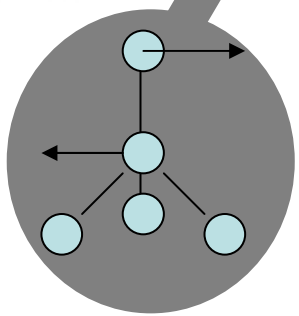
Bond-charge model and DFPT for Silicon

Biswas and Ambegaokar PRB 26,1980 (1982)



Recoules et al. PRL 96, 055503 (2006)





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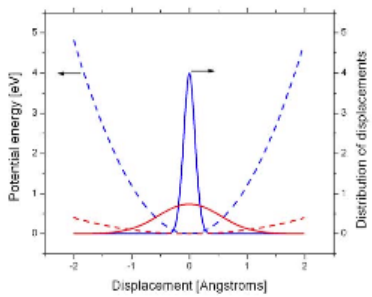
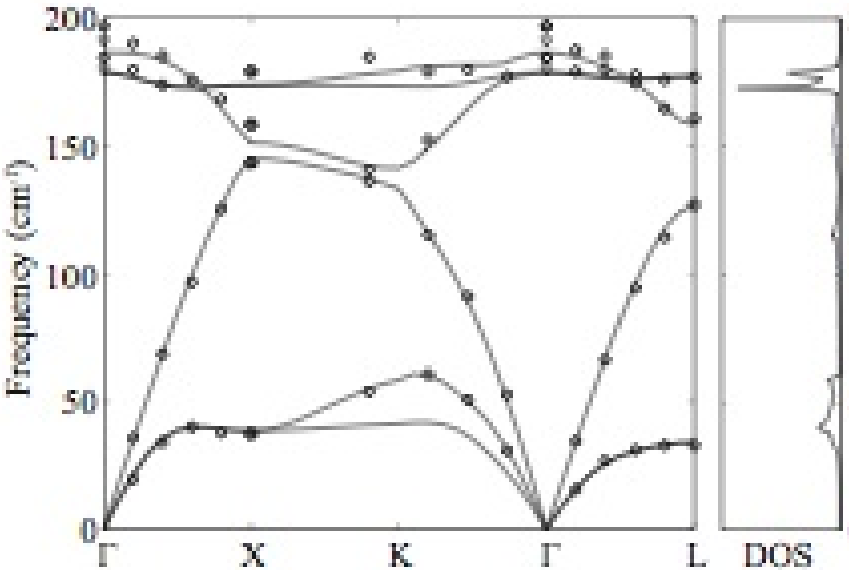
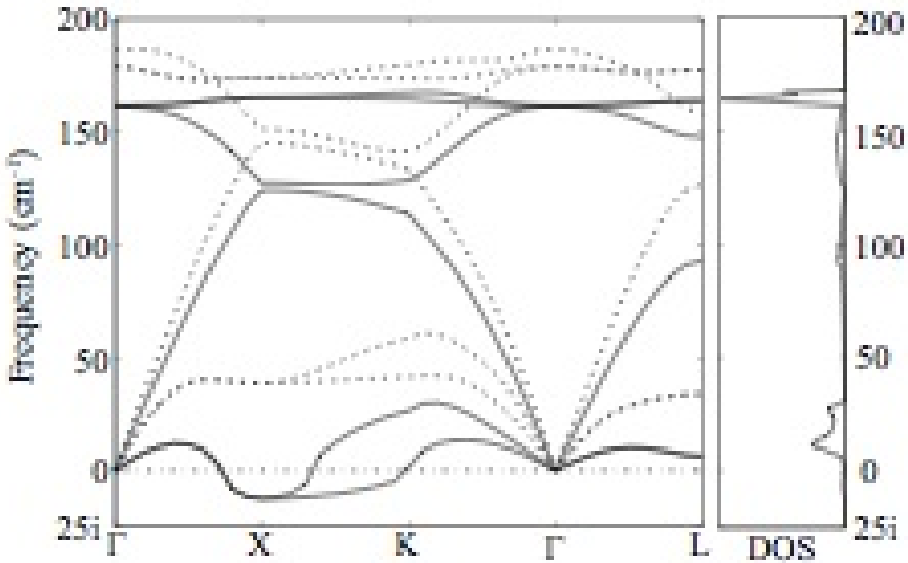
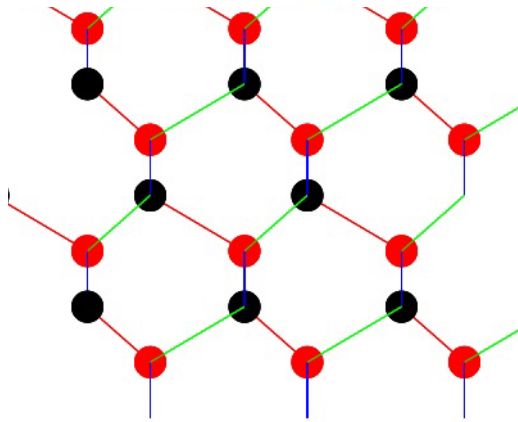


Theoretical Predictions of Ultrafast Melting

InSb goes unstable at the first @ the X point







P.B. Hillyard, D.A. Reis and K.J. Gaffney, submitted PRB (2008)