Ultrafast laser-matter interactions

D. A. Reis

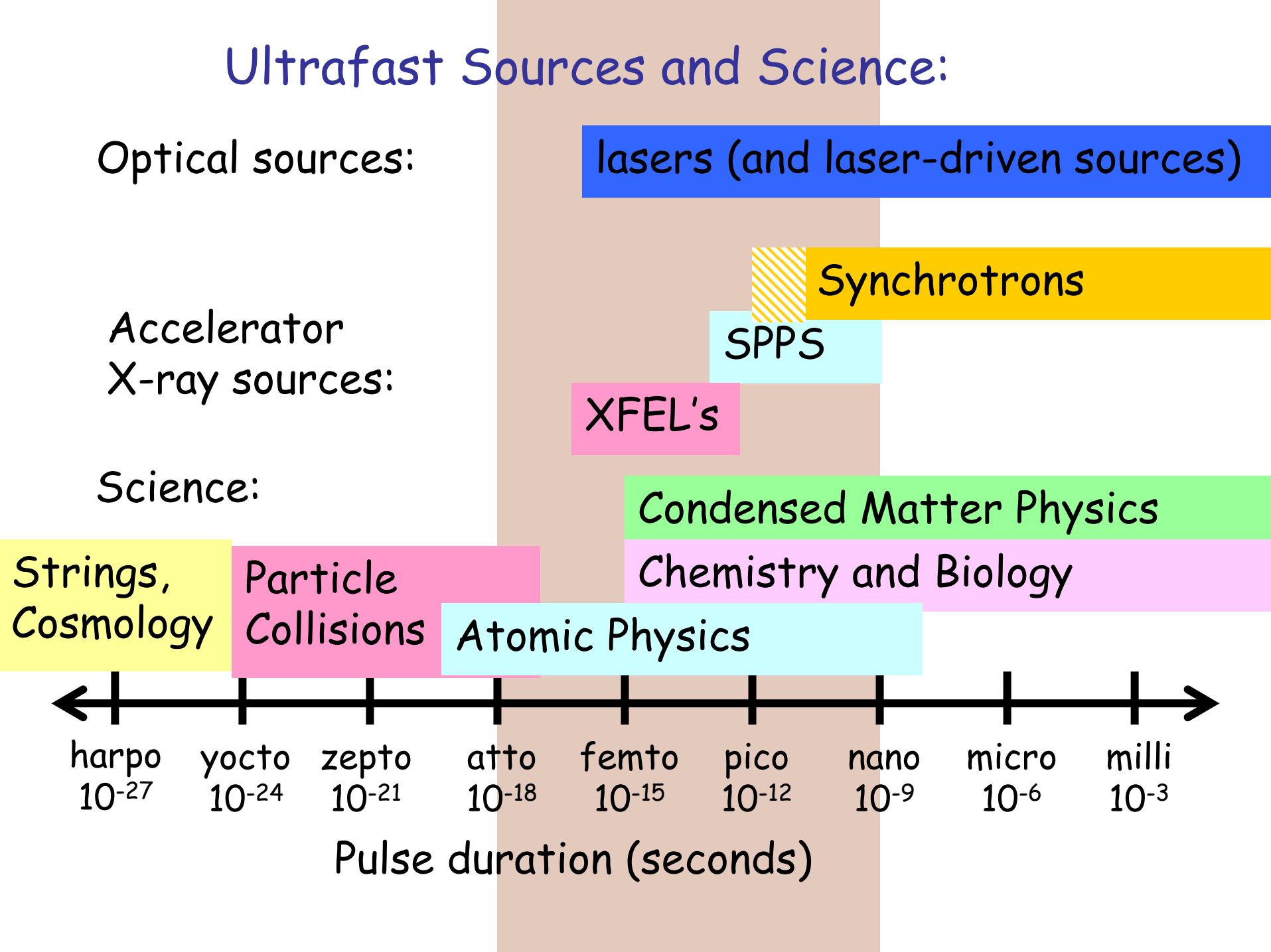
FOCUS Center and Department of Physics, University of Michigan, Ann Arbor, MI 48109

Cross-Cut Review:
Science Requiring the Pulsed Structure of the APS X-ray Beam
Wednesday, January 26, 2005

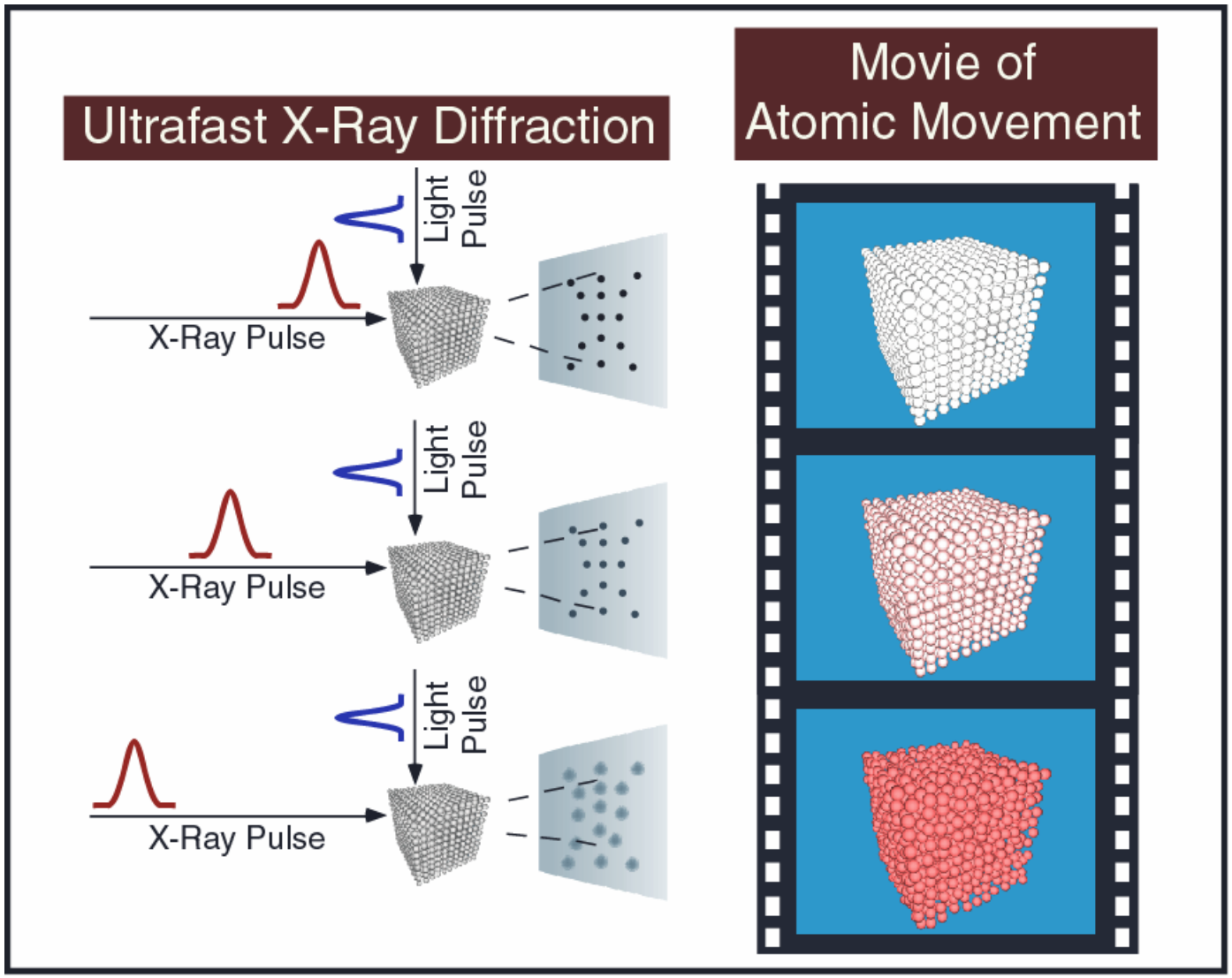
Sub-Picosecond Pulse Source

FOCUS

Ultrafast Sources and Science:



Hard X-rays can probe structure
Ultrafast x-rays can probe it faster: dynamics



Stuff moves in 0.01-10 ps

Source: LCLS – The First Experiments; graphic from C. Siders

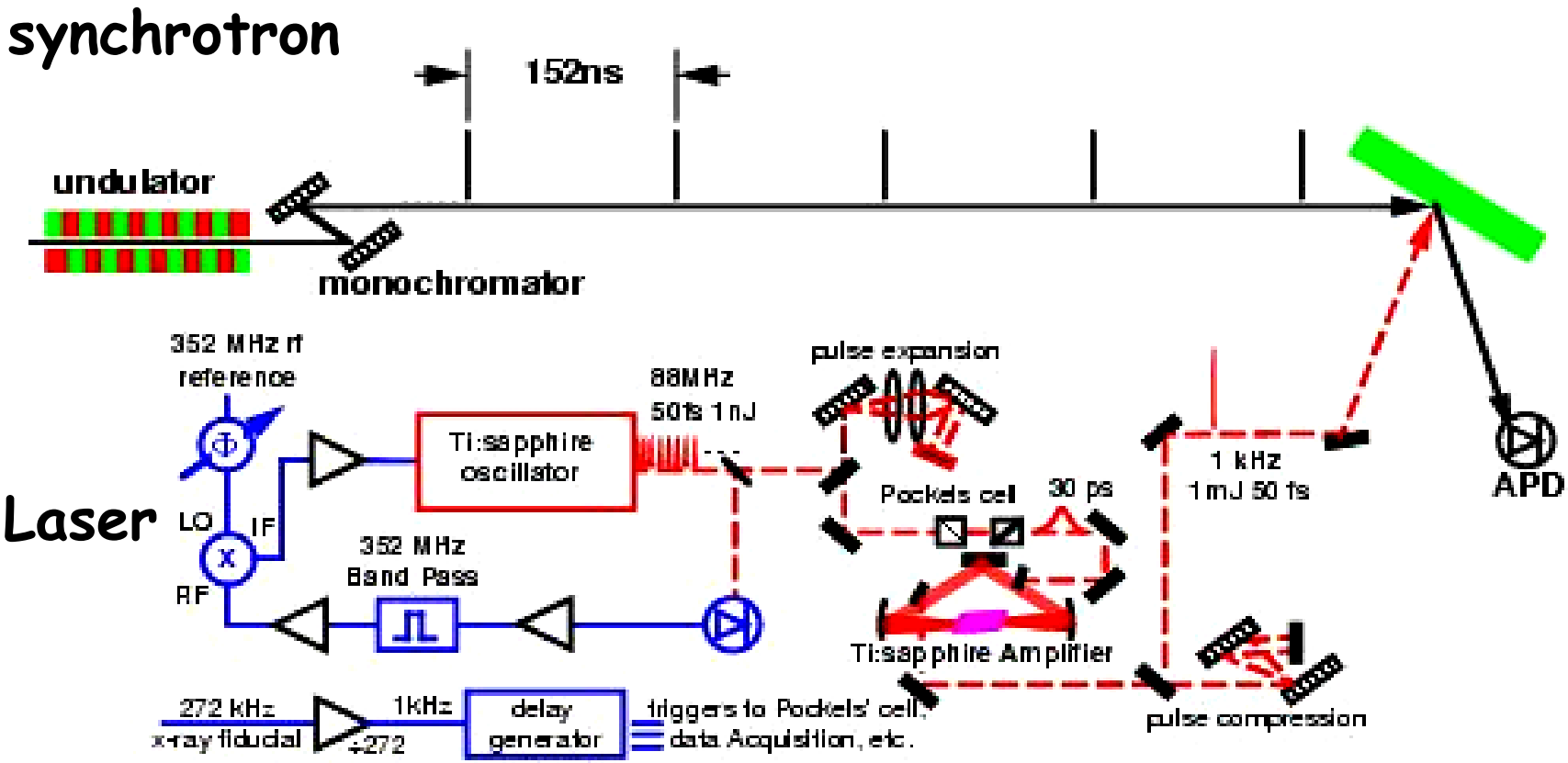
Laser-Matter Interactions, opportunities for time-domain studies

Dynamics in Condensed Matter:
- Coherent excitations
- Nonequilibrium electron and phonon dynamics
- Phase transitions and domain reversals
- Nucleation, growth and phase separation

Atomic and Molecular Physics:
- Understand strong-field effects on inner-shell processes
- Coherent control of molecular processes
- Structural dynamics & phase transitions in isolated targets

Chemical and Biological Dynamics:
- Resolve the fastest time-scale motions of atoms and molecules in order to monitor biological and chemical reactions in real time
- Follow structural evolution correlated to fundamental processes of life and chemistry across multiple timescales
- Explore broad range of molecular dynamics and structural transitions and molecular signaling and energy transduction.

Laser-pump–X-ray-probe

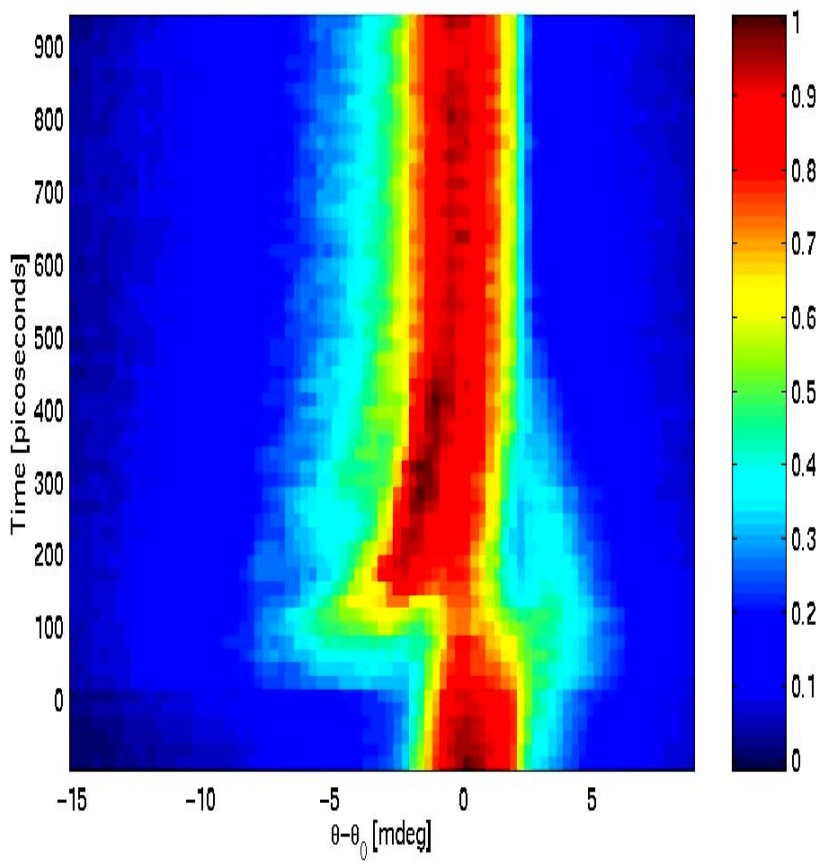


- Resolution limited by the bunch duration (or the timing jitter)
- Arbitrary pump-probe delay (NOT limited by bunch separation)

Time-resolved Bragg Diffraction: Coherent Acoustic Phonons

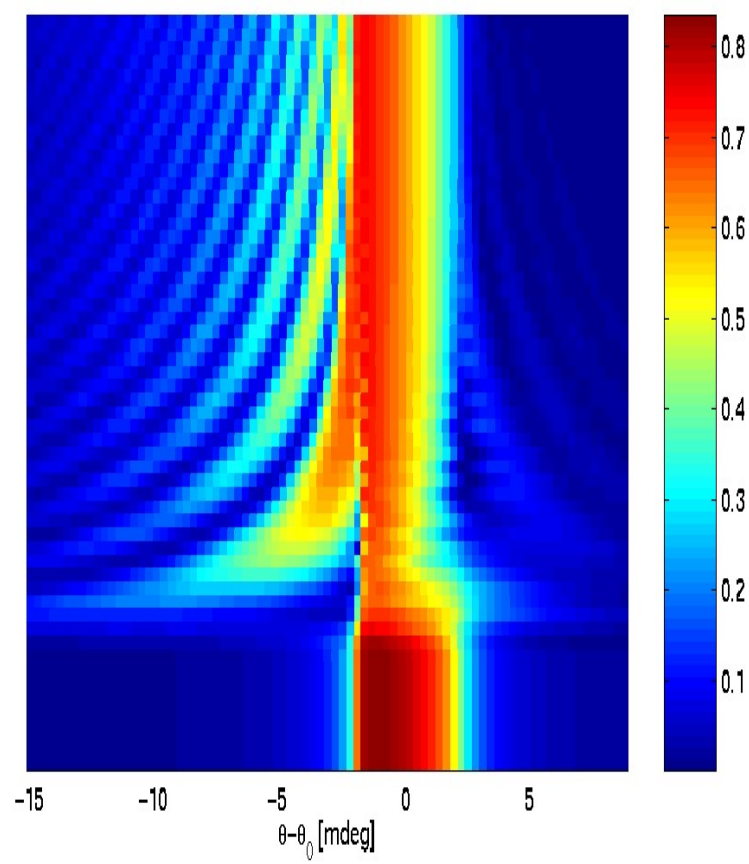
Laser Pump/X-ray Probe

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



Impulsive Strain Generation

(Thomsen *et al.* Phys Rev. B (24) 1986.)



experiment: InSb 111, 10mJ/cm^2

Time-resolved Bragg Diffraction: Coherent Acoustic Phonons

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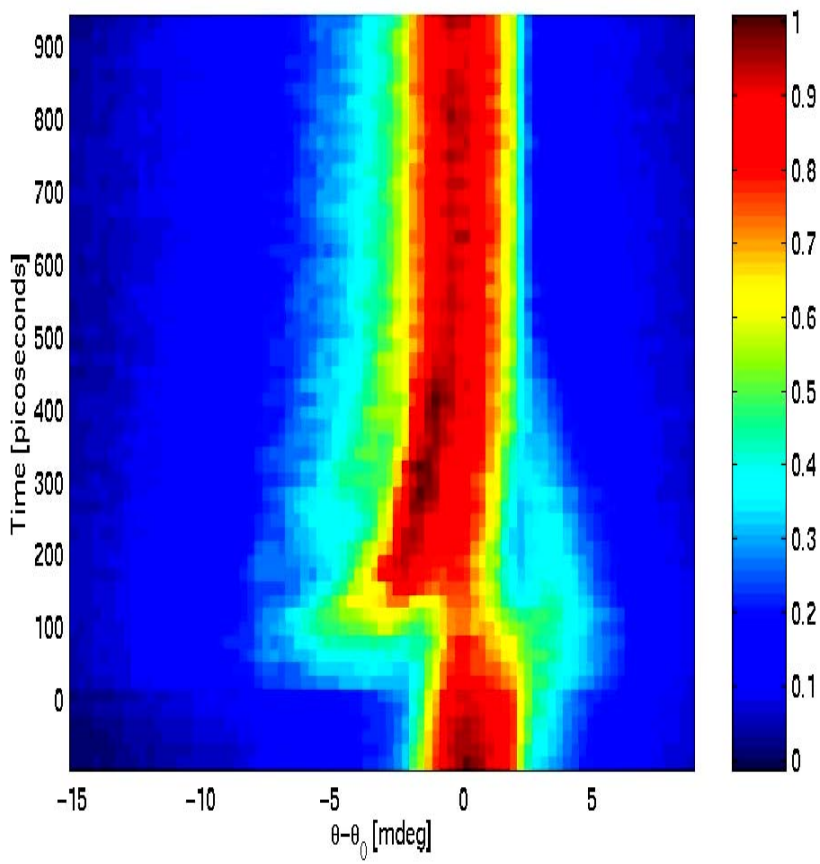

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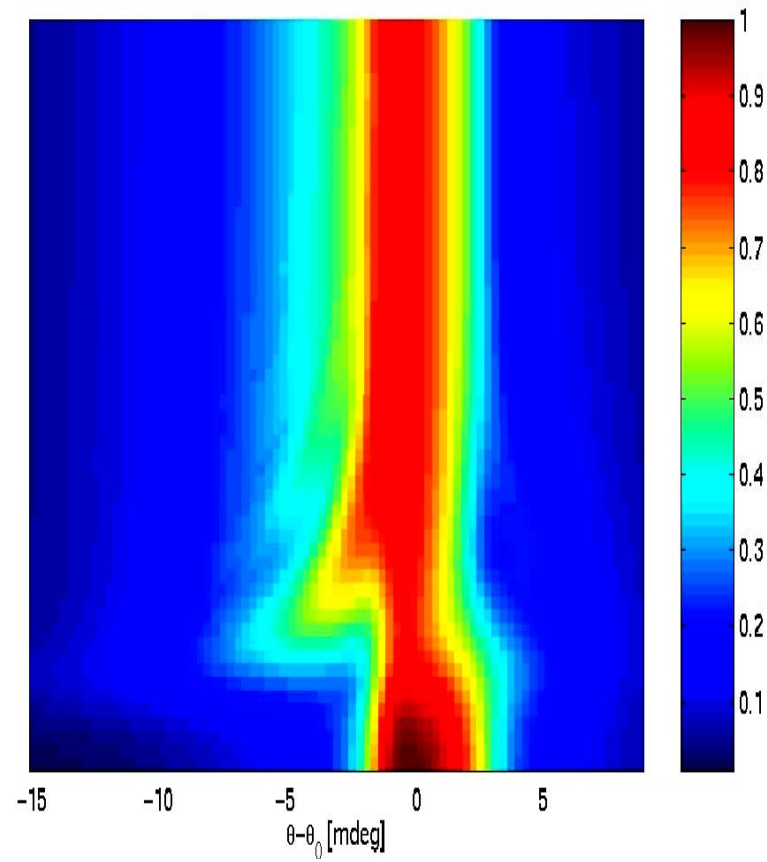
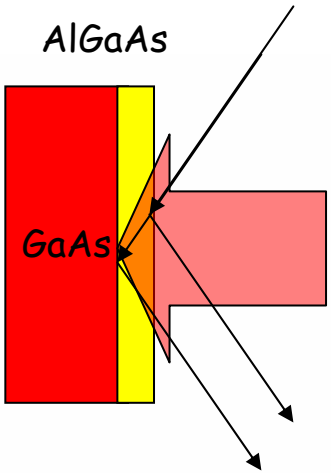
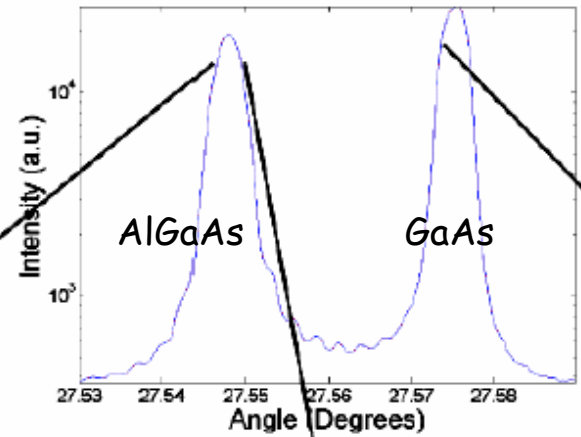


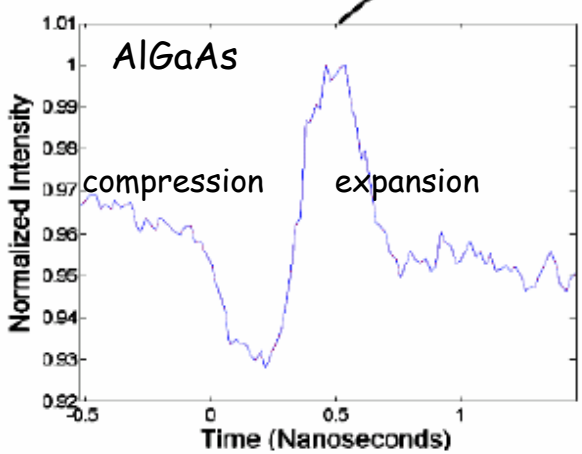
simulation: 100ps & 1.25mdeg conv.

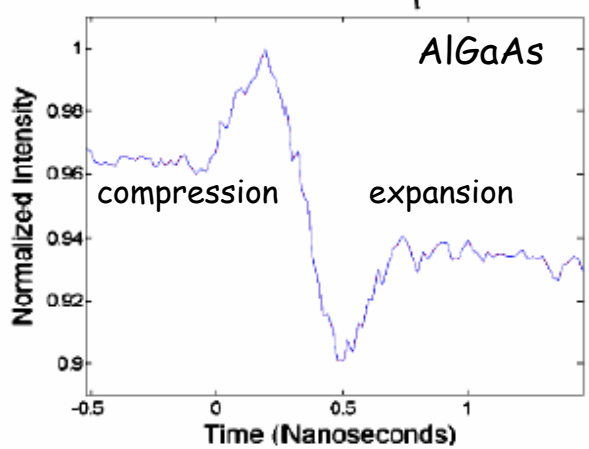
Strain propagation across heterostructure boundaries

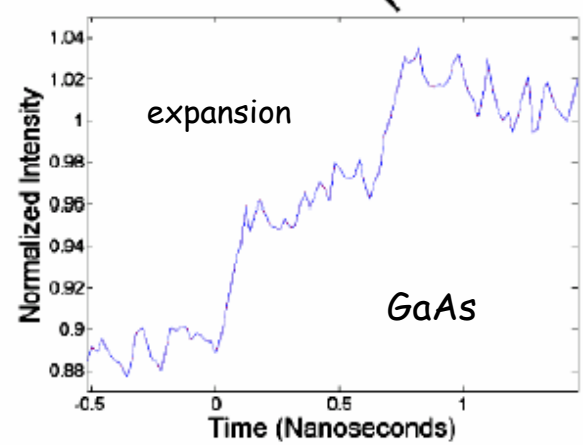




- Electron-phonon coupling and transport
- Boundary resistance (Kapitza), thermal conductivity etc.
- Superlattices and folded phonons

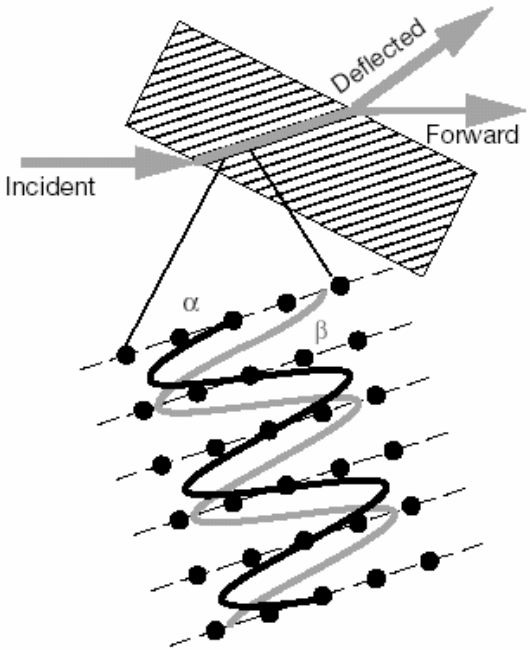


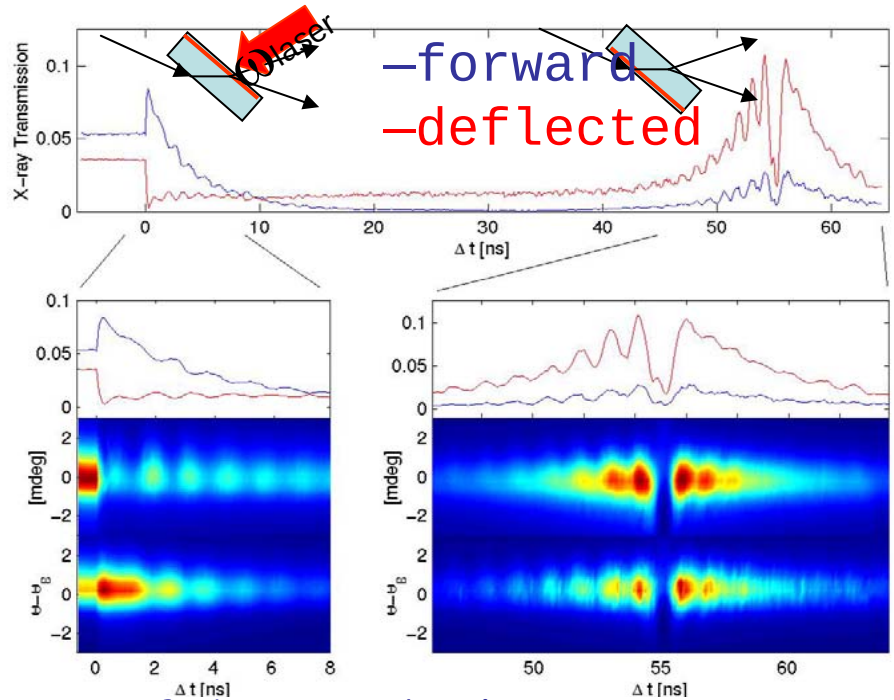




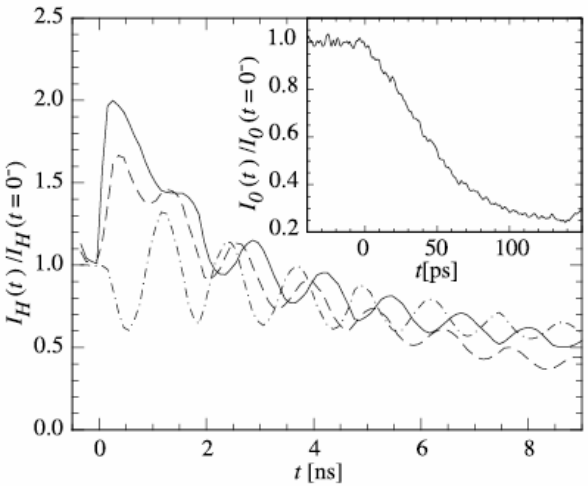
Sample by R. Goldman and R. Hegde

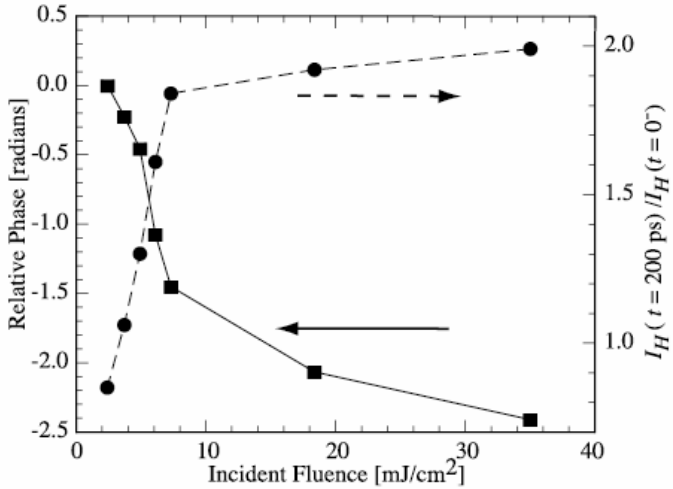
Coherent Control of Pulsed X-ray Beams





Ambipolar diffusion of dense e-h plasma

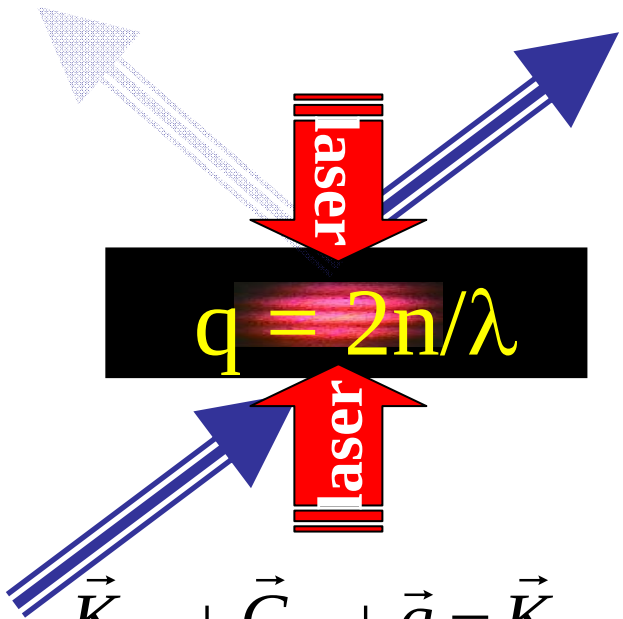




M.F. DeCamp *et al.*, Nature 413, 825 2001, Phys. Rev. Lett. 91, 165502, 2003.

fs X-ray switch

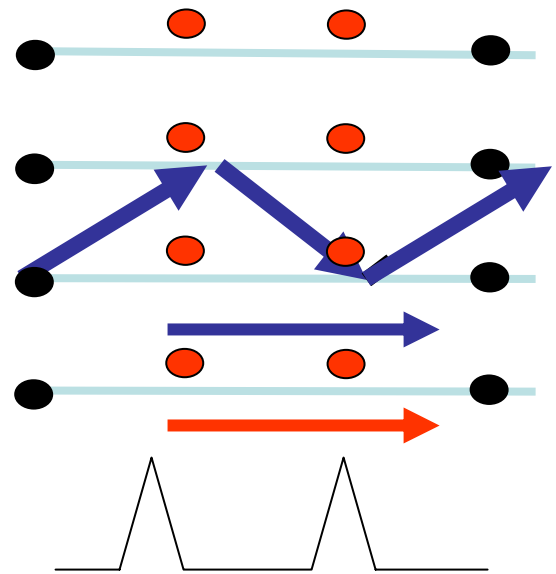
resonant Raman scattering



$$\vec{K}_{0\alpha} + \vec{G}_H + \vec{q} = \vec{K}_{H\beta}$$

$$\vec{K}_{0\beta} + \vec{G}_H - \vec{q} = \vec{K}_{H\alpha}$$

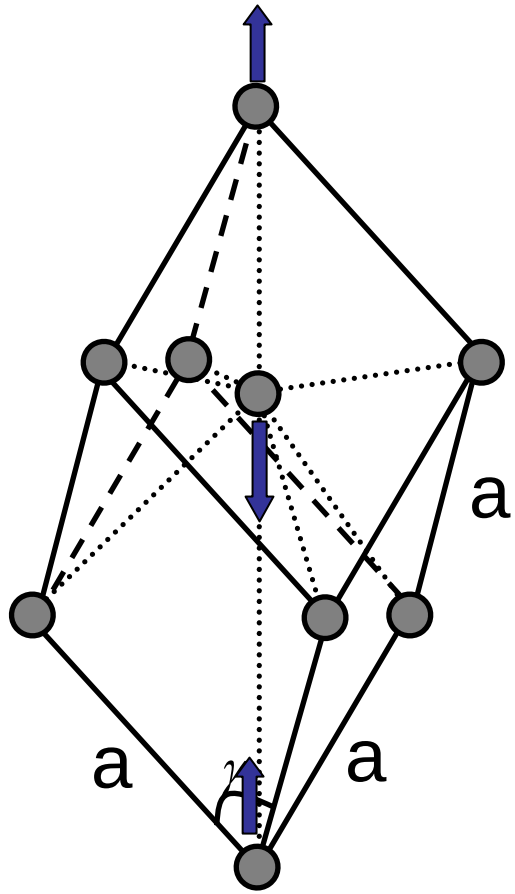
velocity matched Pendellösung



V (*x-ray*) = V_g (*laser*)

Dynamics of high amplitude coherent phonons in photo-excited bismuth

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



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

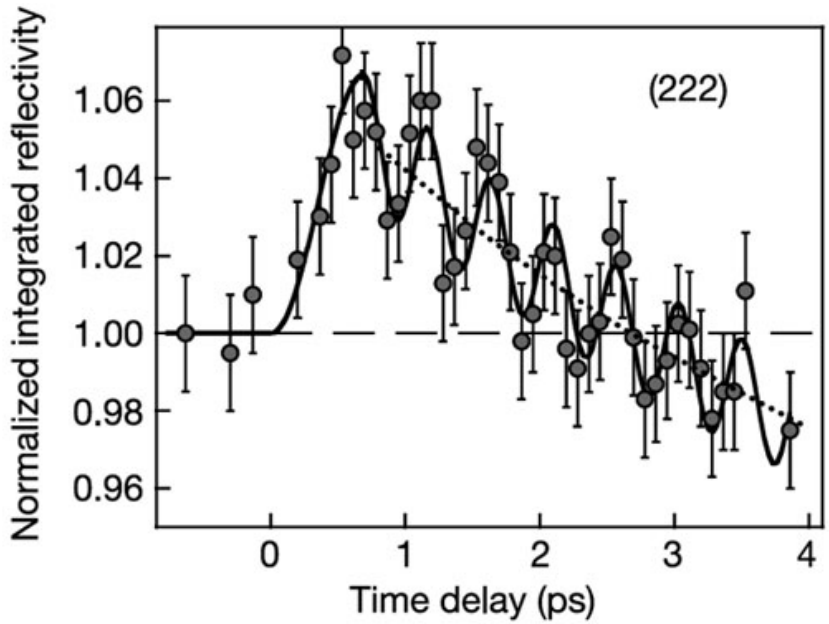
anharmonicity or electronic softening? (Fahy and Reis PRL 93 109701, 2004)

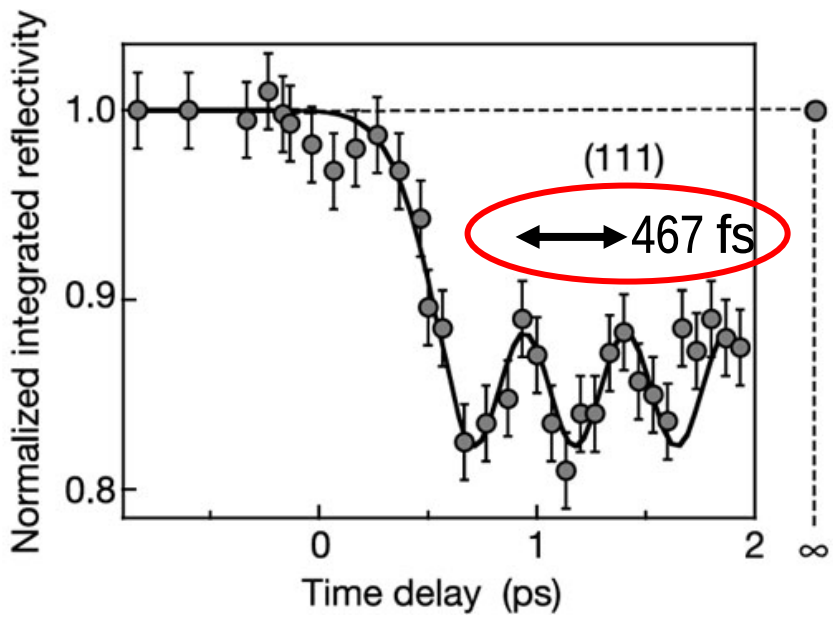
Laser plasma x-rays: Time resolution <1ps, but not many photons

Femtosecond X-ray measurement of coherent lattice vibrations near the Lindemann stability limit

Klaus Sokolowski-Tinten*, Christian Blome*, Juris Blums*, Andrea Cavalleri†, Clemens Dietrich*, Alexander Tarasevitch*, Ingo Uschmann‡, Eckhard Förster‡, Martin Kammler§ Michael Horn-von-Hoegen* & Dietrich von der Linde*

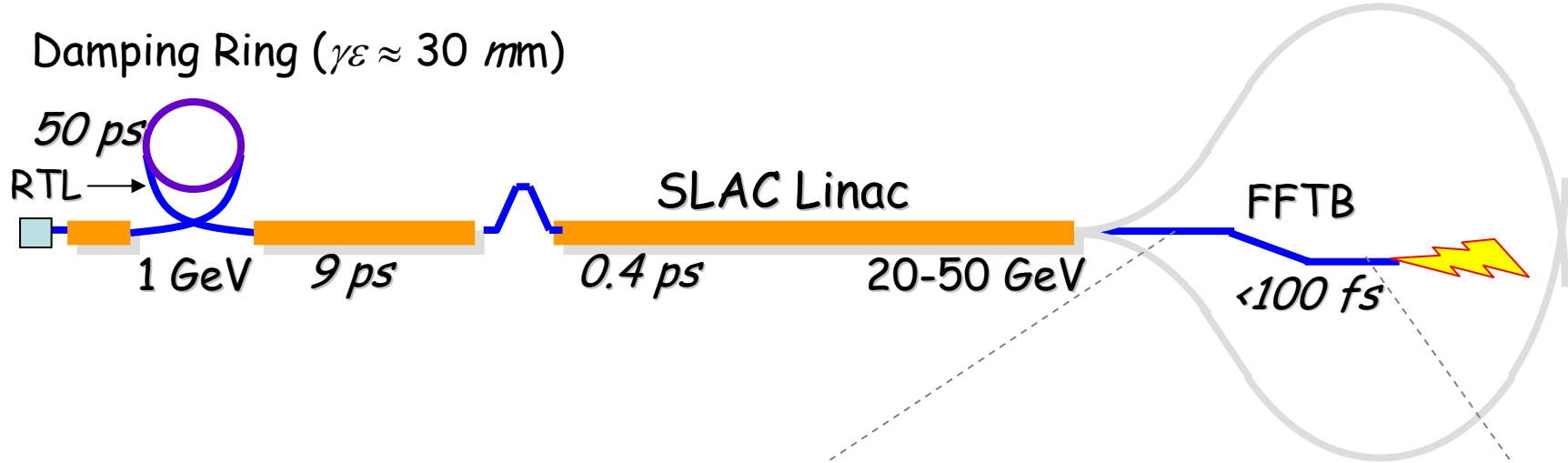
Nature, **422**,p. 287 (2003)

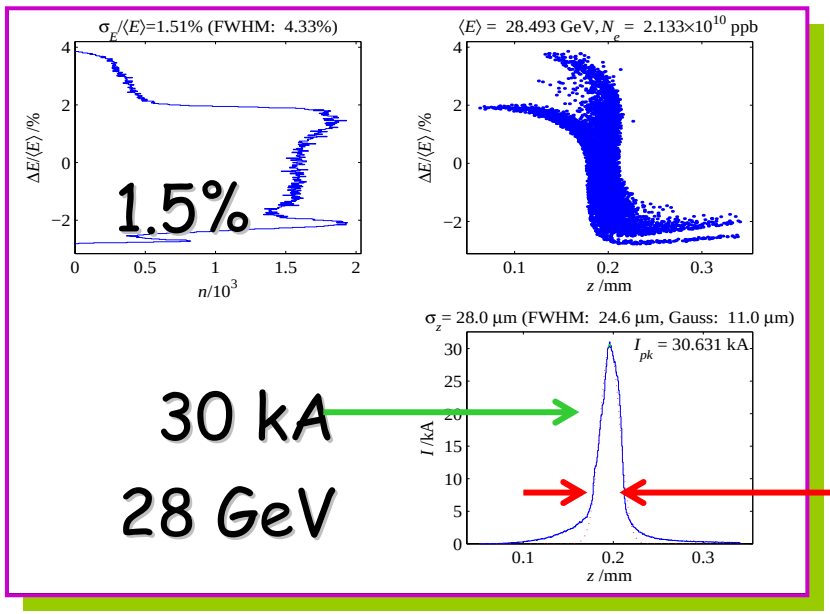


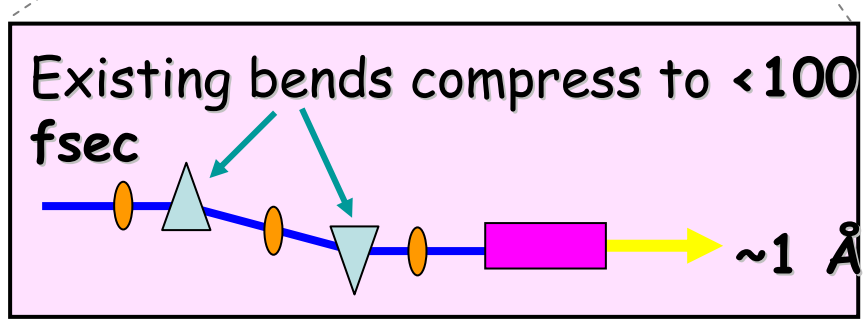


The Sub-Picosecond Pulsed Source (SPPS)

Short Bunch Generation in the SLAC Linac





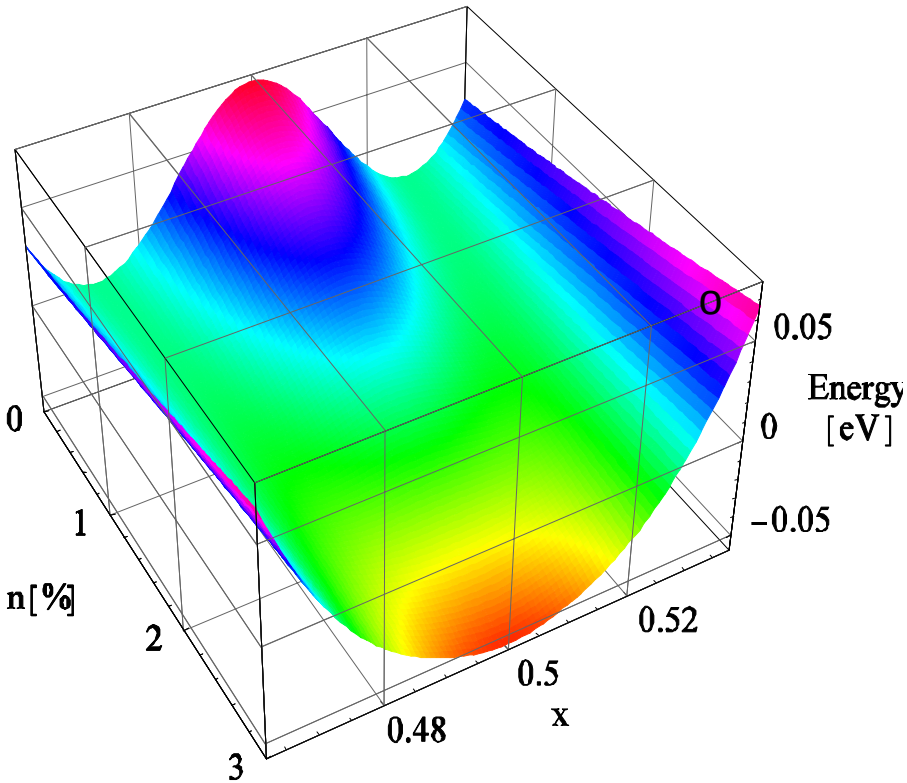


80 fsec FWHM

J. Hastings *et al.*

Short pulses lots of γ's, but 10 Hz and limited beamtime

Electronic Softening and reversal of Peierls distortion (density functional calculation)



Murray *et al.*, *etc.*

Sokolowski-Tinten *et al.*

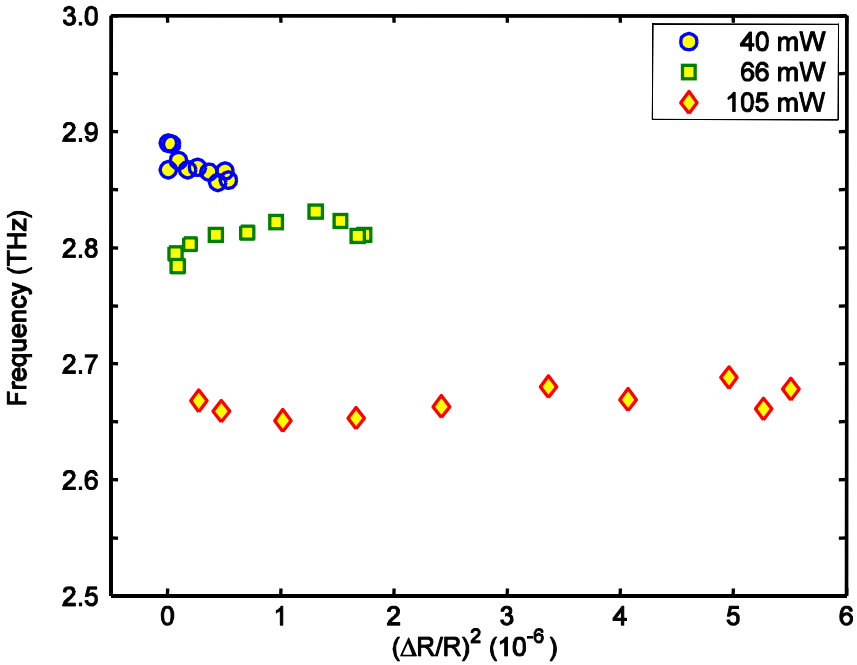
QuickTime™ and a
TIFF (LZW) decompressor
are needed to see this picture.

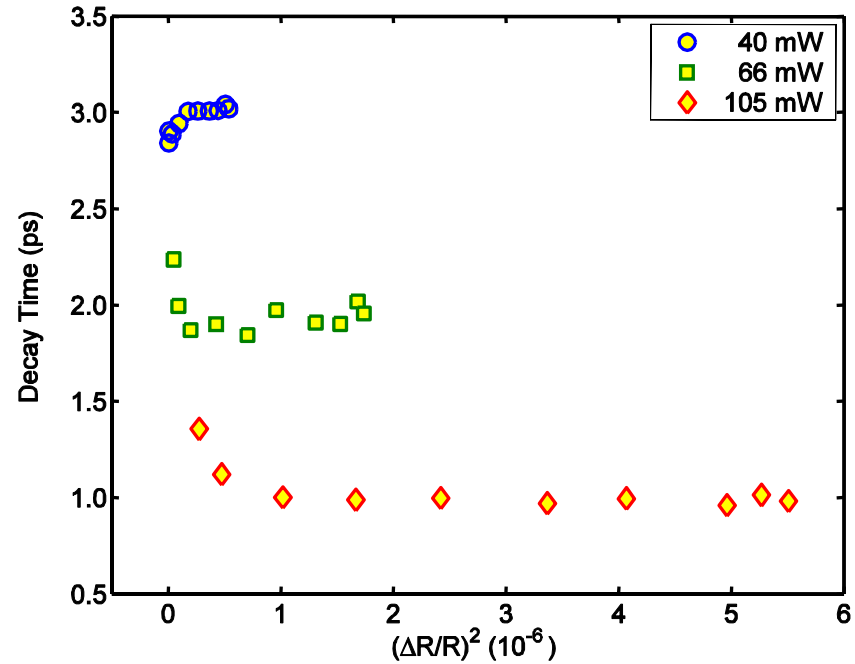
Promotion of electrons into the conduction band alters the atomic equilibrium position and the restoring force.

Murray *et al.*, Cork

Optical probes distinguish:

Electronic softening from anharmonicity (using coherent control)





X-rays probes will measure:

- Anharmonicity (true measure of atomic displacements),
- density dependent properties (diffusion, thermal conductivity, etc.),
- phonon decay modes and rates (via diffuse scattering),
- role of optical phonon in phase transitions, domains reversals …

Future needs and desires

- Source and Detectors
  - –High rep-rate Picosecond (or shorter) x-rays. (all experiments shown would benefit from higher temporal resolution).
  - –Choppers that can isolate single bunch in SOM (>1kHz) and/or fast single photon >0-dim detectors.
  - –High average power, high repetition rate lasers (8 MHz?)
- Facilities and Management
  - –Expand SAC for additional ultrafast representation.
  - –Time-domain proposal review committee.
  - –Active user recruitment, visiting scientists, post-doc/student support.
  - –Maintain time-resolved compatible SOM!

Michigan X-ray Group

A. Cavalieri[1], D. Fritz[1], S. Lee[1], M.F. DeCamp[2]

P.H. Bucksbaum[1], R. Merlin[1], R. Clarke[1],D. A. Reis[1]

[1]FOCUS Center and Department of Physics, Univ. of Michigan

[2]Chemistry Department, MIT

MHATT-XOR

E. Dufresne, E. Landahl, D. Arms, D. Walko, B. Adams

And collaborators

S. Fahy, E. Murray, J. Wahlstrand, J. Wark, B. Lings, *et al.*

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Sub-Picosecond Pulse Source

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