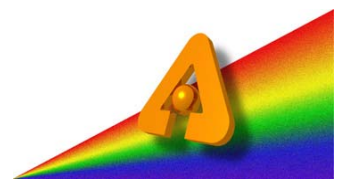
Why do we want 1ps x-rays at APS?

D. A. Reis
U. Michigan/FOCUS
L. Chen and L. Young
ANL
...et al.



APS Strategic Planning Meeting

Time-Domain Workshop Summary and Recommendations

Linda Young (ANL) Atomic & Molecular Physics

Lin Chen (ANL) Chemistry & Biology

David Reis (UMich) Condensed Matter Physics

Stephen Milton (ANL) Accelerator Physics

Workshop Chairs

September 2-3, 2004, The Abbey, Fontana, Lake Geneva Area, WI

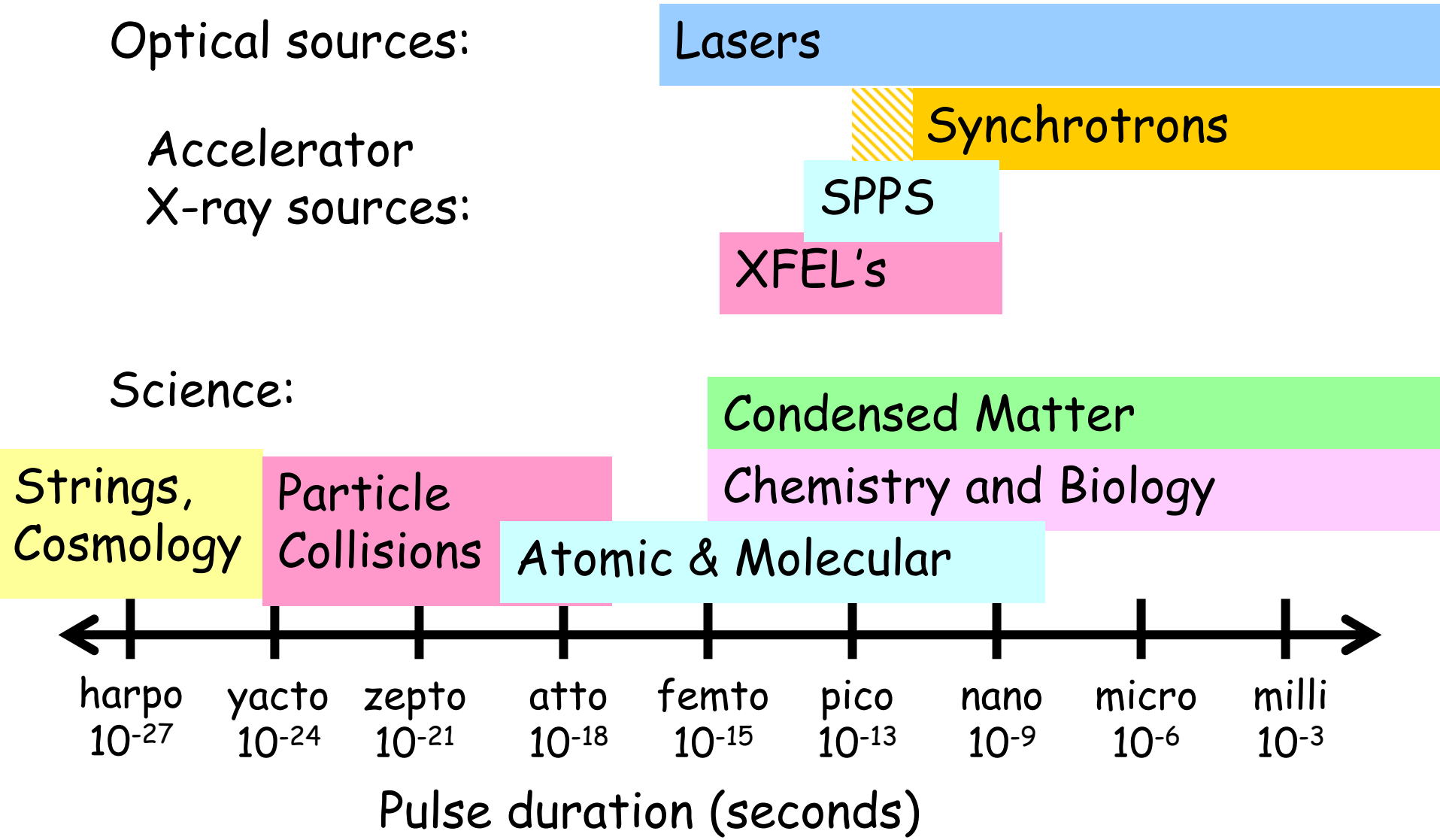
WORKSHOP ON TIME DOMAIN SCIENCE USING X-RAY TECHNIQUES

Workshop Summary and Recommendations

1. Facilitate and enhance experiments on timescales ≥ 100 ps

2. Develop short time capabilities of high-flux ≈1 ps pulse at APS

Ultrafast Sources and Science:



Scientific Challenges & Opportunities

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.

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

X-ray facilities: toward ultrafast & ultraintense

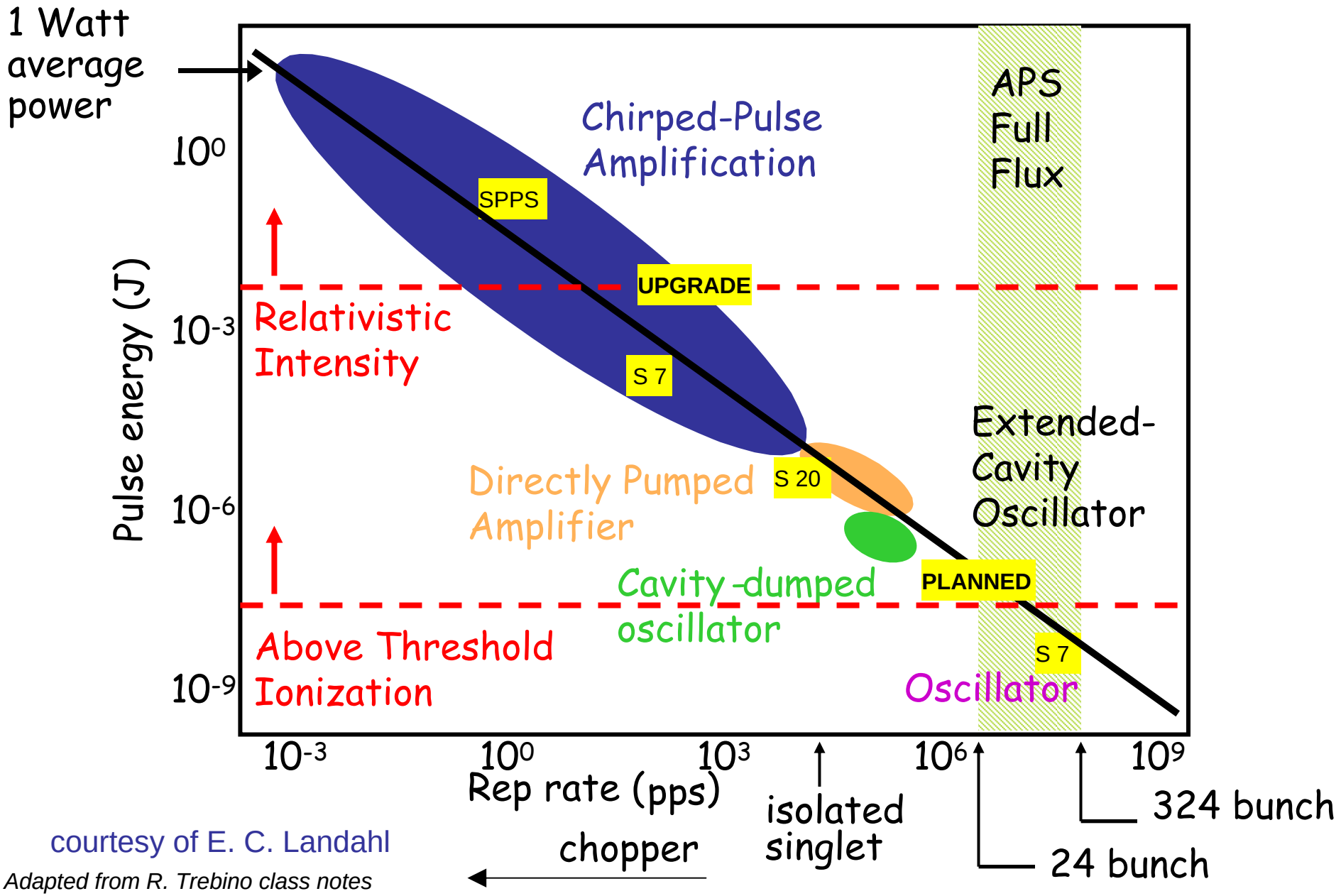
Single bunch specs

10^8 x-rays **100 ps @ ≤6.5 MHz**

≈10^7 x-rays **80 fs @ ≤10Hz**

10^{12} x-rays **230 fs @ ≤120Hz**

Femtosecond-laser pulse energy vs. repetition rate



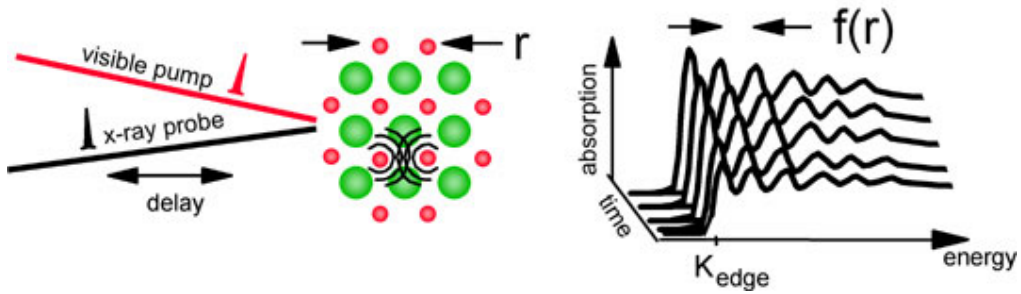
courtesy of E. C. Landahl

Adapted from R. Trebino class notes

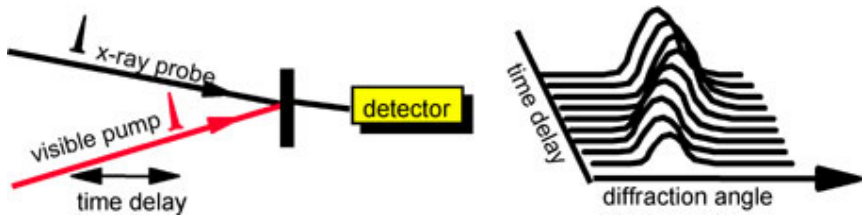
Ultrafast x-ray science

LUX website

Time-resolved EXAFS, NEXAFS, surface EXAFS



Time-resolved x-ray diffraction

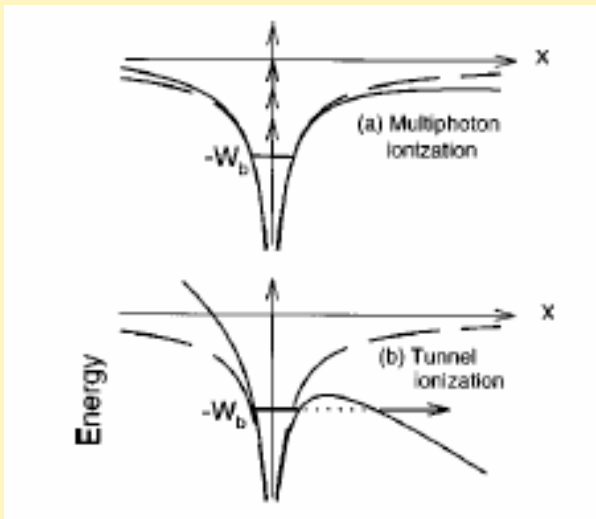


Laser-pump/x-ray probe techniques central
Short pulses automatically yield high intensities
1 mJ/100 fs/(0.1 mm)^2 $\approx 10^{14}$ W/cm^2 $\approx$ 3 V/Å

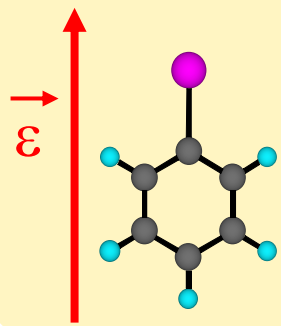
AMO - new opportunities from 100 ps - 1ps

- ***Understand strong-field effects on inner-shell processes***
- ***Coherent control of molecular degrees of freedom***
- ***Structural dynamics & phase transitions in isolated targets***

Perturbed x-ray spectra



Aligned molecules



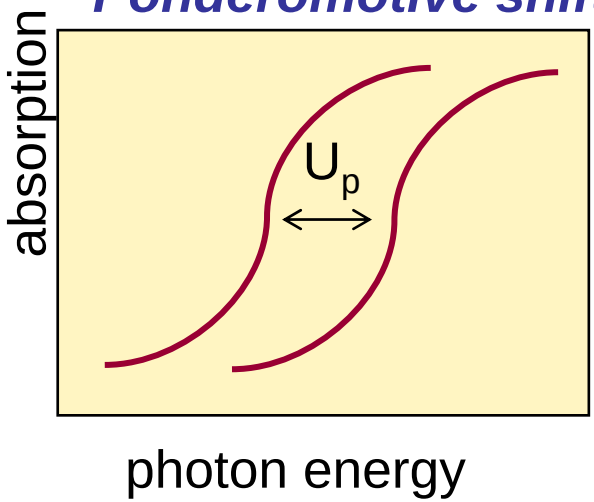
Strong field effects on inner-shell processes

- ***X-ray photoionization is fairly well understood in the weak-field limit***
- ***Understand changes to x-ray processes in presence of strong laser fields***
- ***Theoretical predictions***

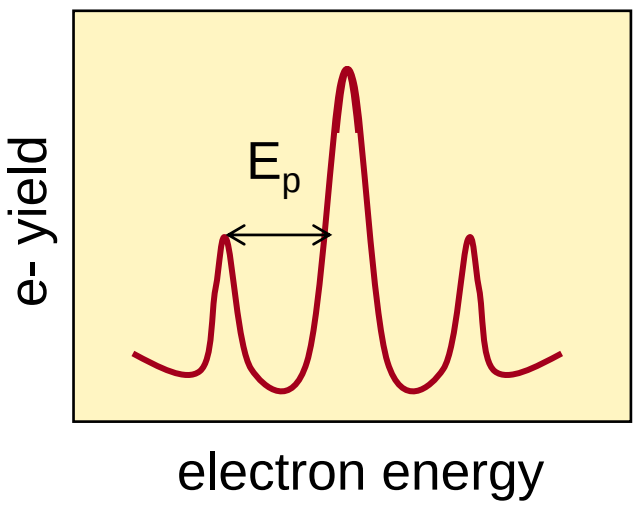
  ponderomotive shift in threshold -> absorption spectrum

  free-free transitions in continuum -> electron spectra

Ponderomotive shift



Electron satellites



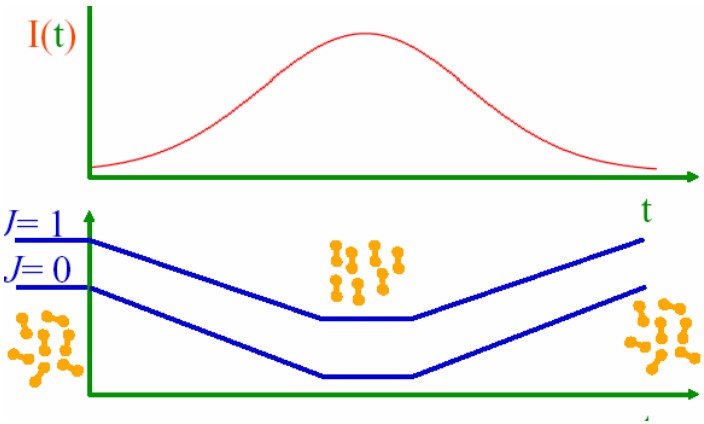
Coherent control: Aligning molecules

Motivations

- *Stringent tests of photoionization and photodissociation*
- *Study behavior of molecules as fcn of strength of aligning potential*
- *Aligned molecules aid single biomolecule structure determination (Hajdu et al)*
- *Test coherent control techniques to optimize alignment*

Adiabatic alignment

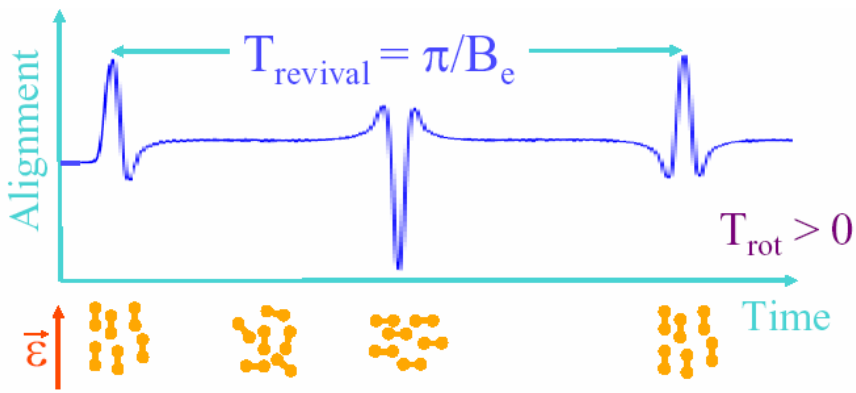
$\tau_{laser} > \tau_{rot}$



Friedrich & Hershbach, PRL 74, 4623 (95)

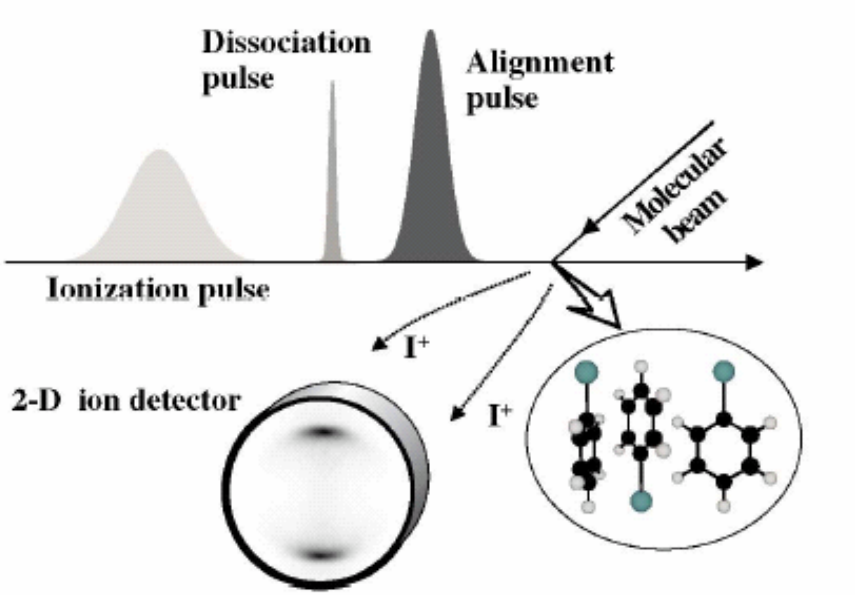
Field-free alignment

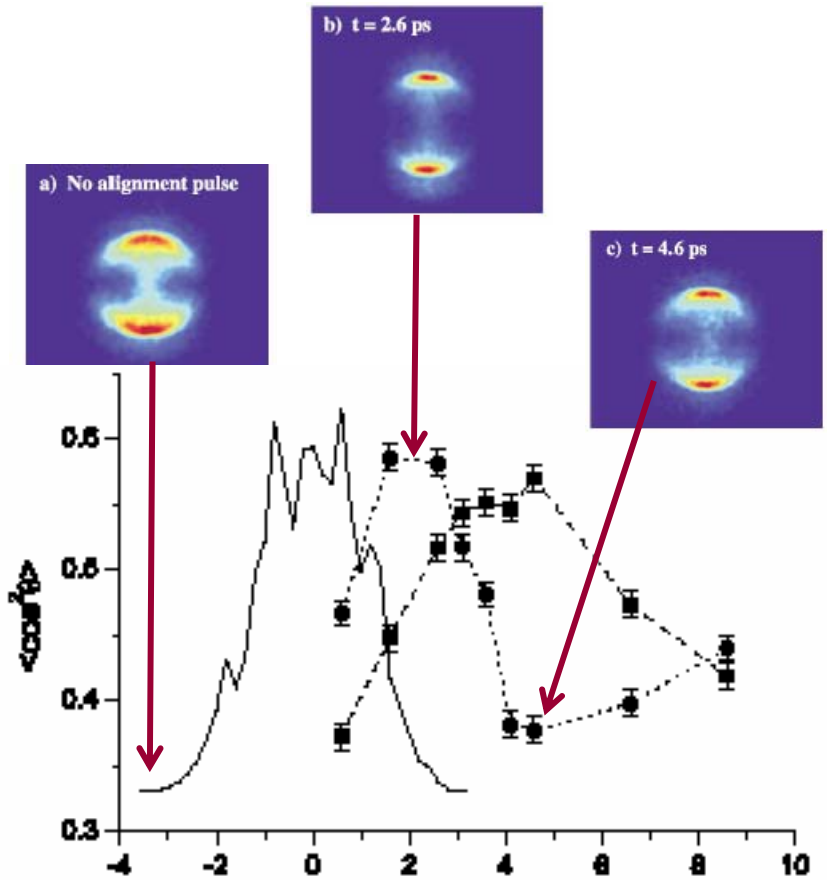
$\tau_{laser} < \tau_{rot}$



T. Seideman, PRL 83, 4971 (99)

Field-free alignment with optical probe





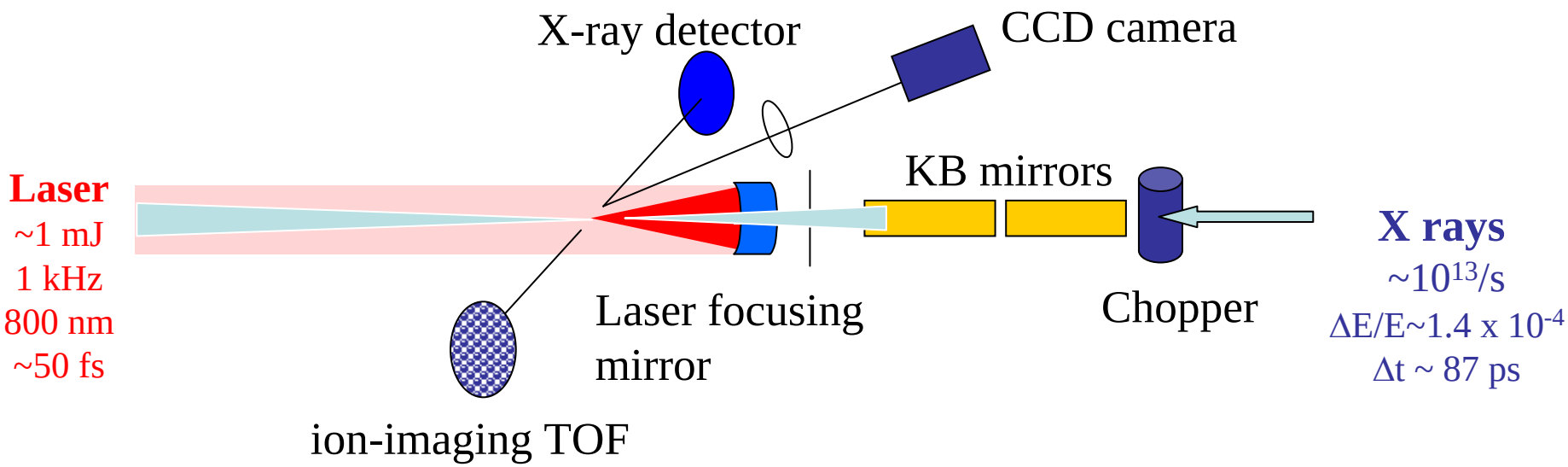
Peronne et al.
Phys. Rev. Lett. **91**, 043003 (2003)

High intensities mandate microfocus

Strong-field perturbed spectra: ~10^{15} - 10^{13} W/cm^2 (E ~ 9 - 0.9 V/Å)

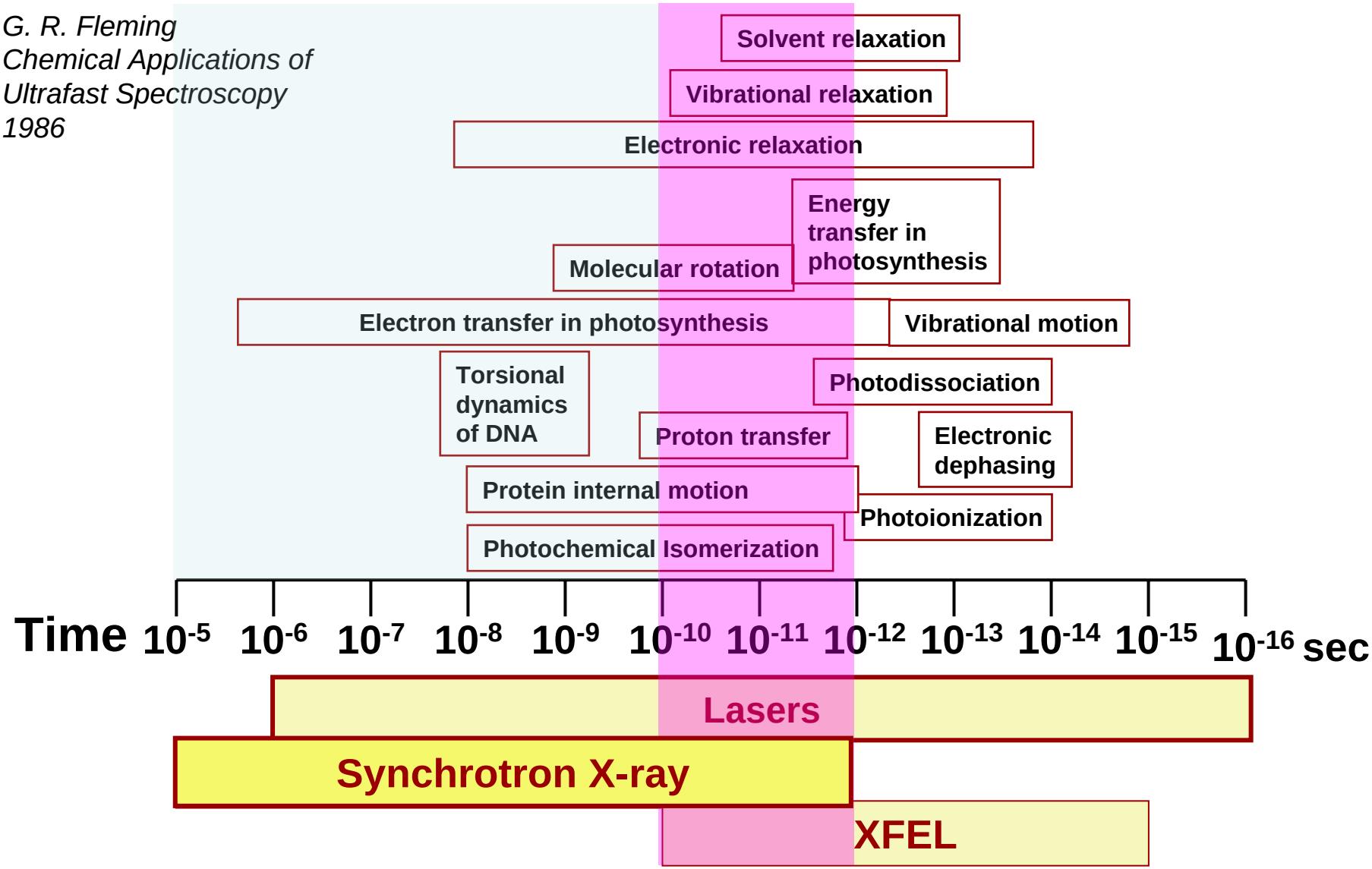
Molecular alignment : ~10^{13} - 10^{11} W/cm^2

Coherent control: ~10^{9} W/cm^2

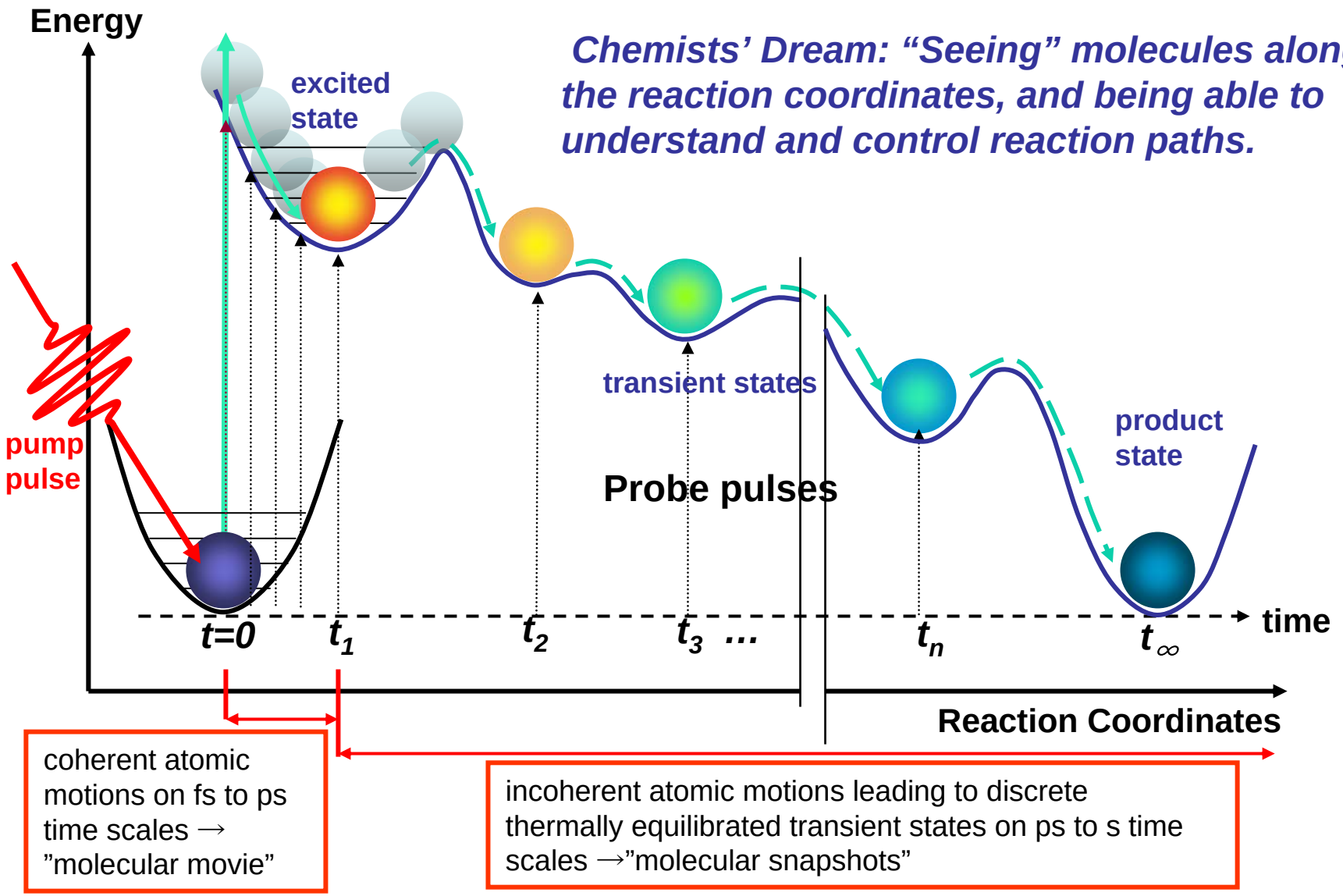


Multiple Temporal Scales in Chemical Sciences

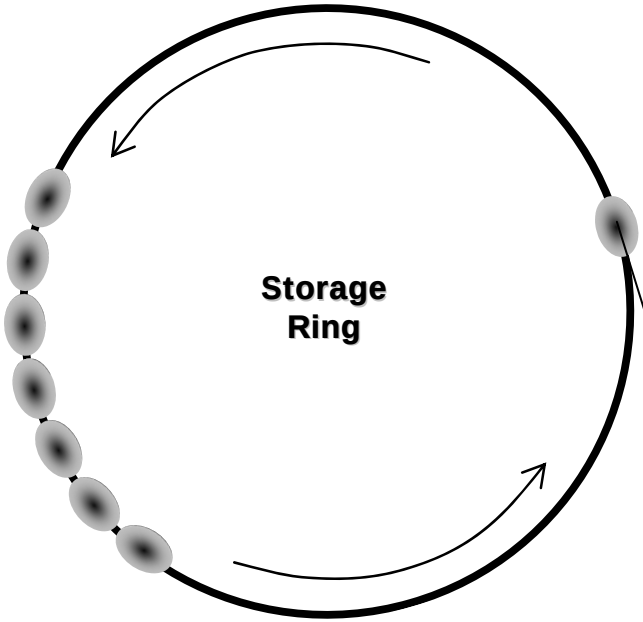
G. R. Fleming
Chemical Applications of
Ultrafast Spectroscopy
1986

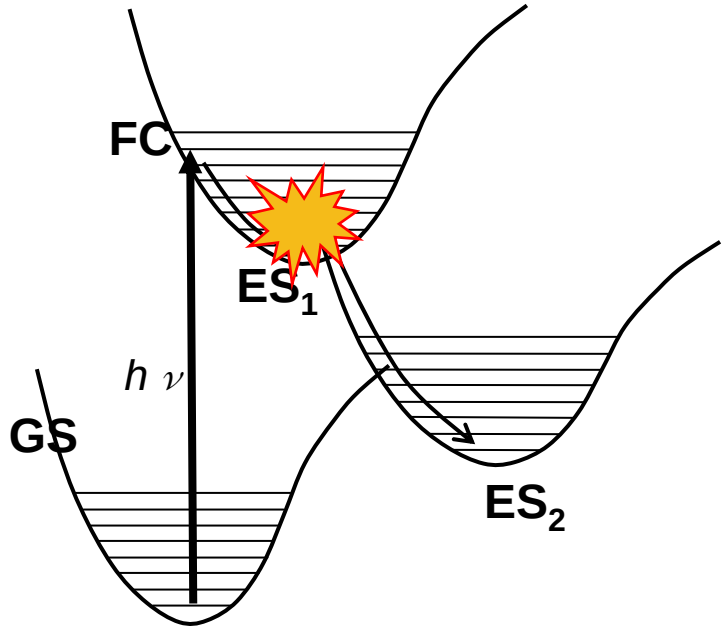


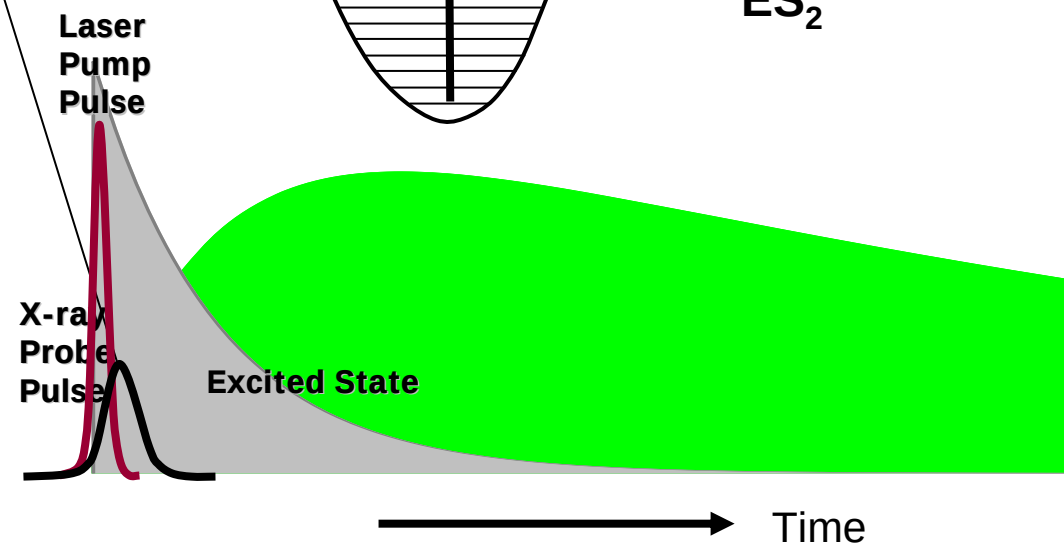
Launching Chemical Processes Synchronously with Laser Pulses



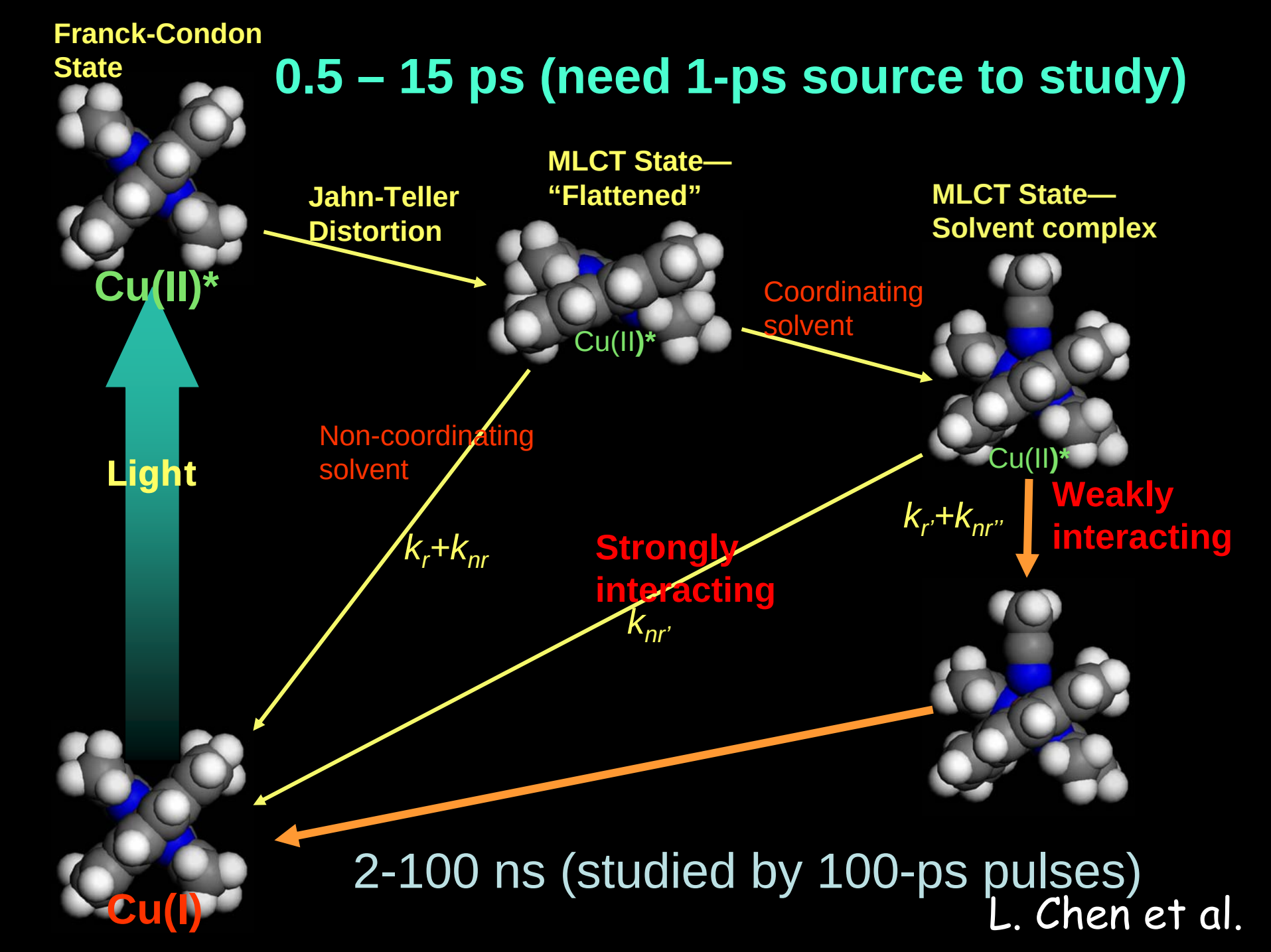
Capturing Excited State Structures with Pulsed X-rays





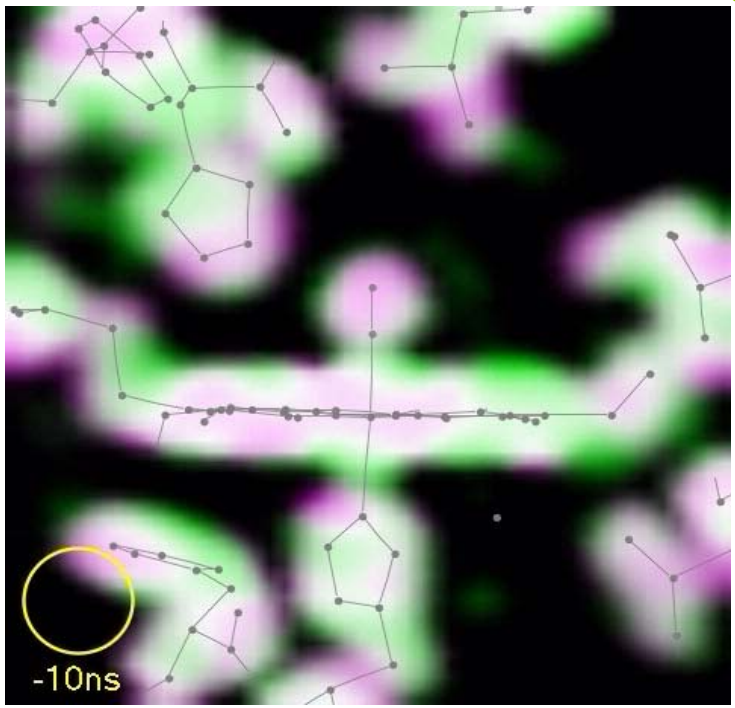


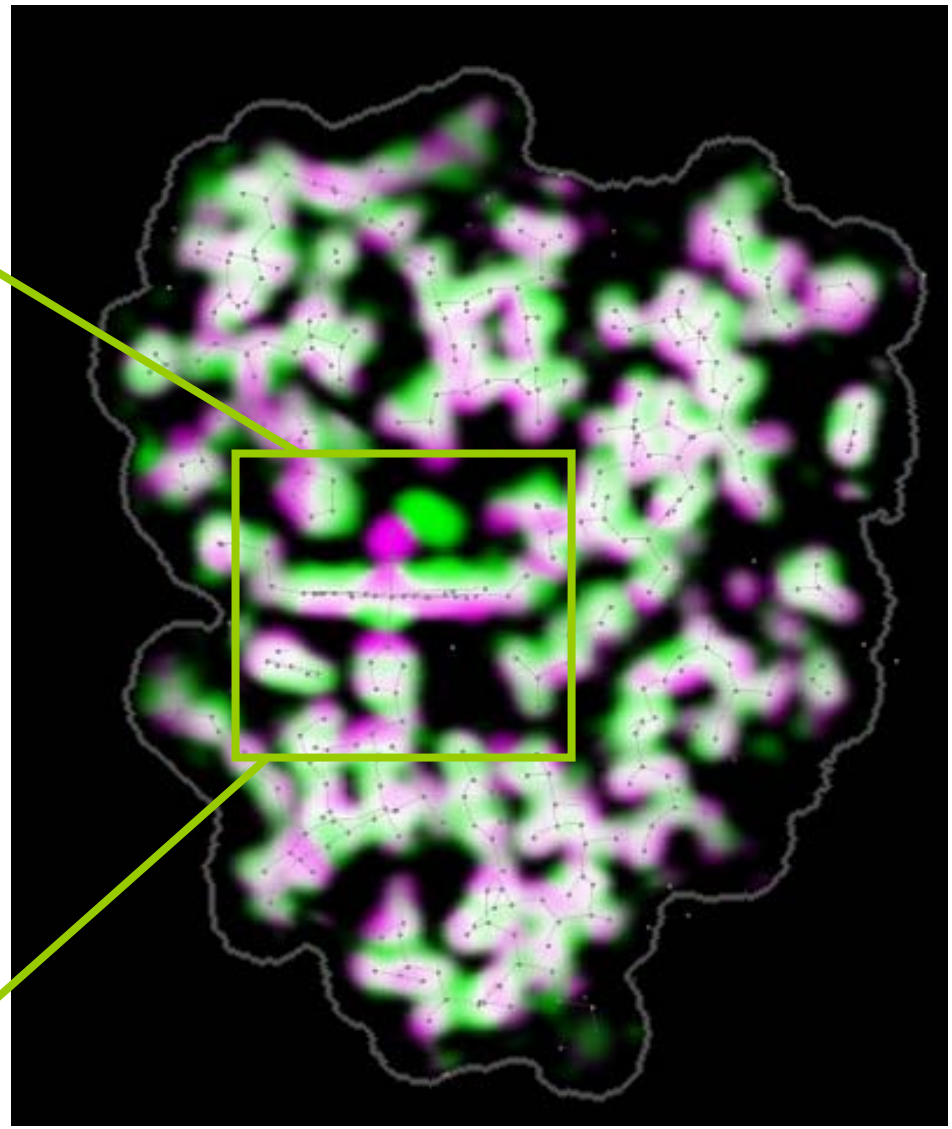
Laser pulse pump
X-ray pulse probe
Intrinsic time resolution:
30 – 100 ps fwhm


Franck-Condon State
0.5 – 15 ps (need 1-ps source to study)
Jahn-Teller Distortion
MLCT State—"Flattened"
MLCT State—Solvent complex
Cu(II)*
Coordinating solvent
Cu(II)*
Cu(II)*
Light
Non-coordinating solvent
k_r+k_{nr}
$k_{r'}+k_{nr''}$
Weakly interacting
Strongly interacting
$k_{nr'}$
2-100 ns (studied by 100-ps pulses)
Cu(I)
L. Chen et al.

Molecular motions in chemical and biological processes take place on multiple time and length scales

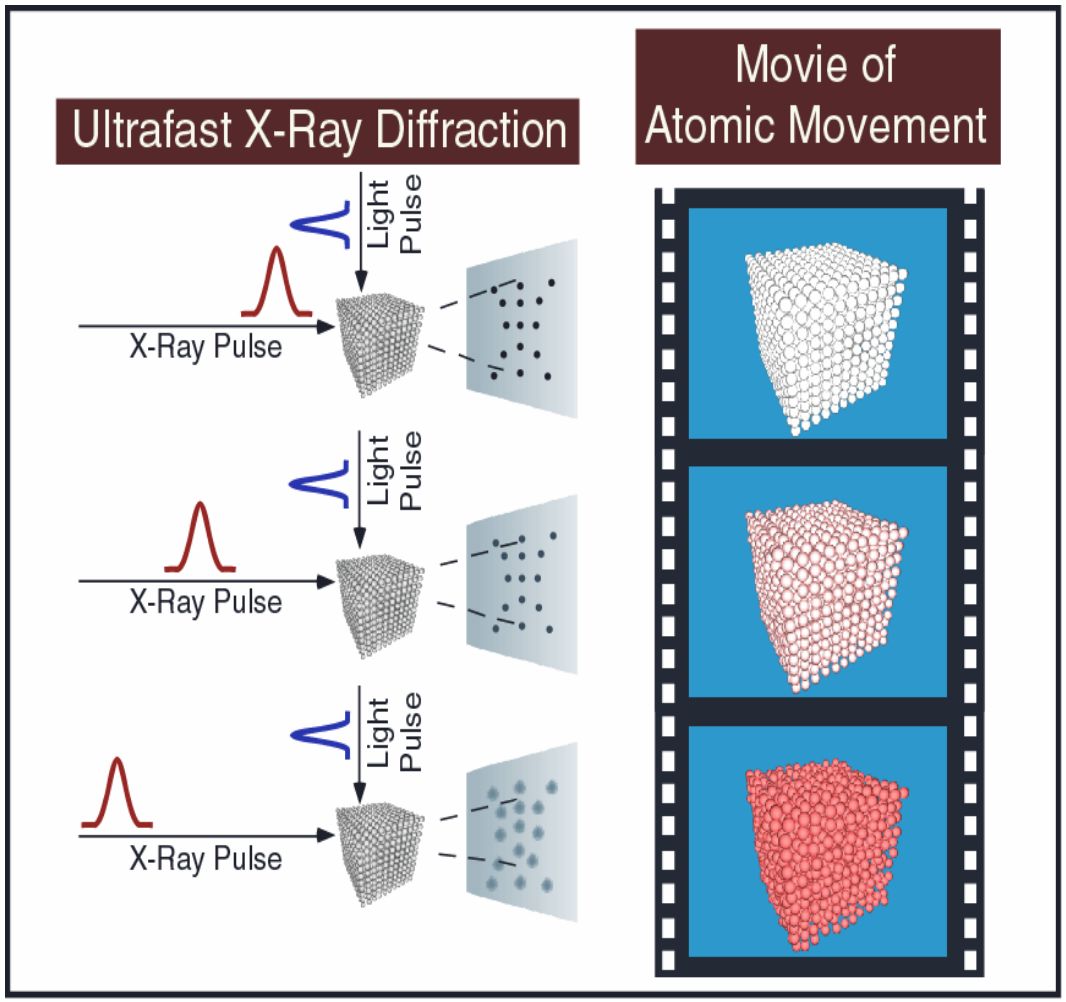
Philip Anfinrud et al.
Photodissociation of *CO* from Myoglobin (ESRF)





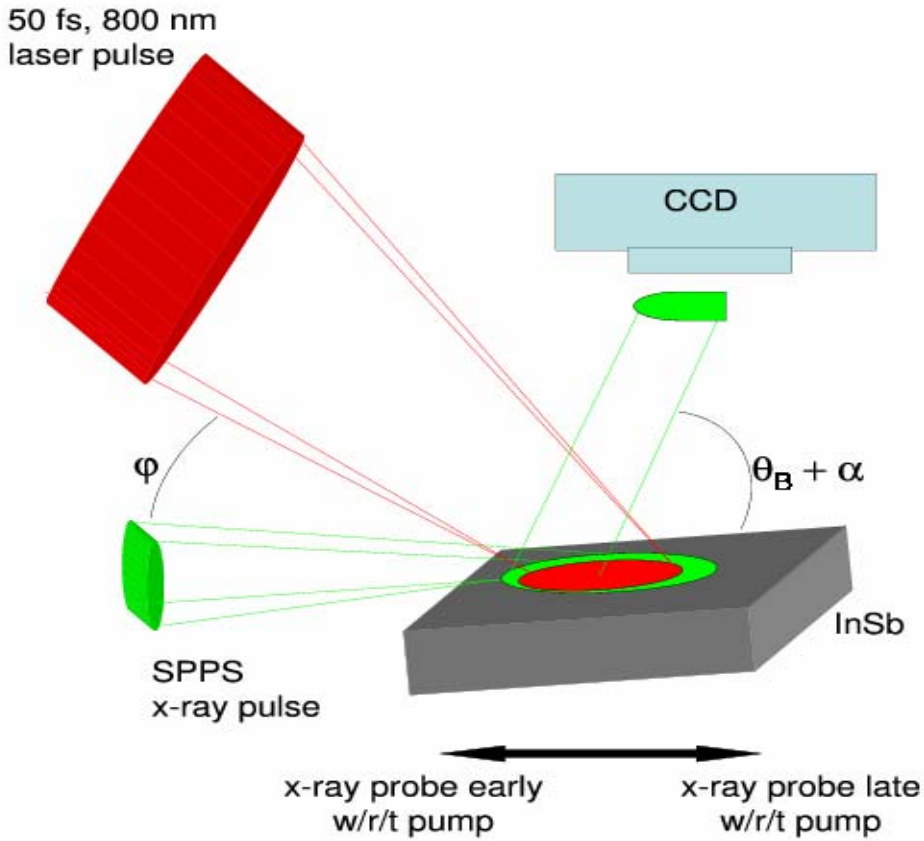
Journal of Structural Biology 147, 235-246 ▪ (2004)

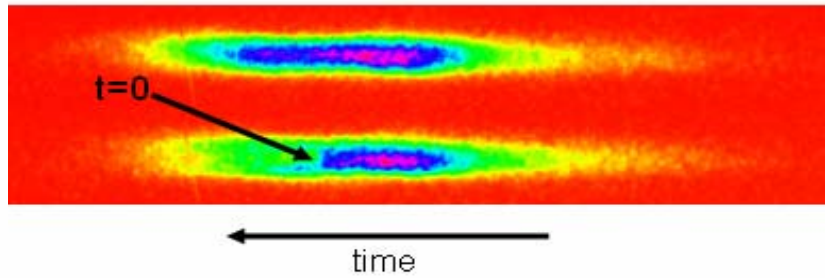
Hard X-rays can probe structure
Ultrafast x-rays can probe it faster: dynamics



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

Single Shot Measurement of Ultrafast Disordering In Solids



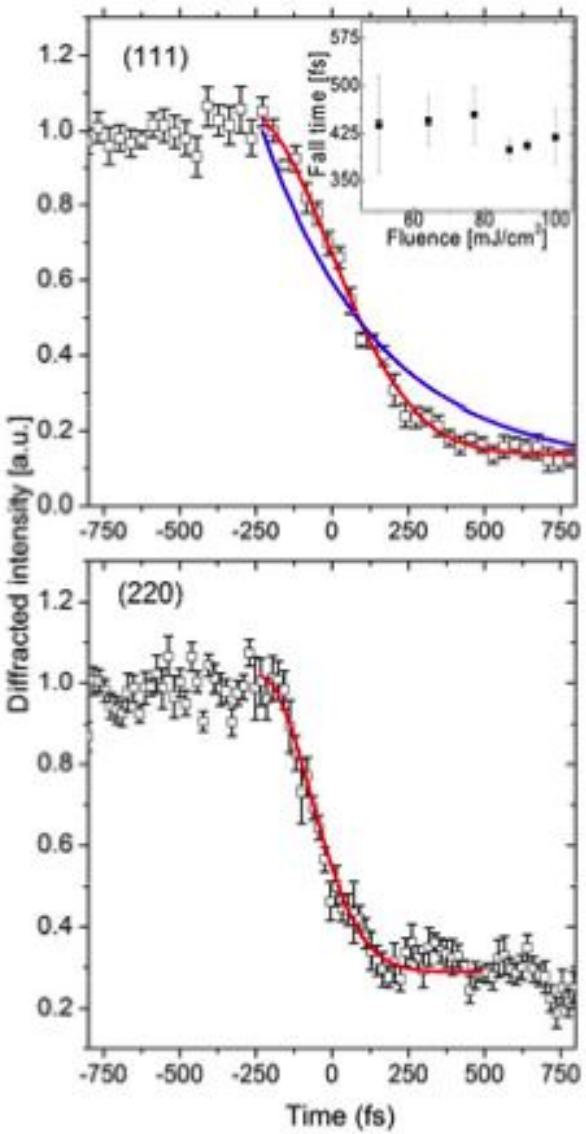


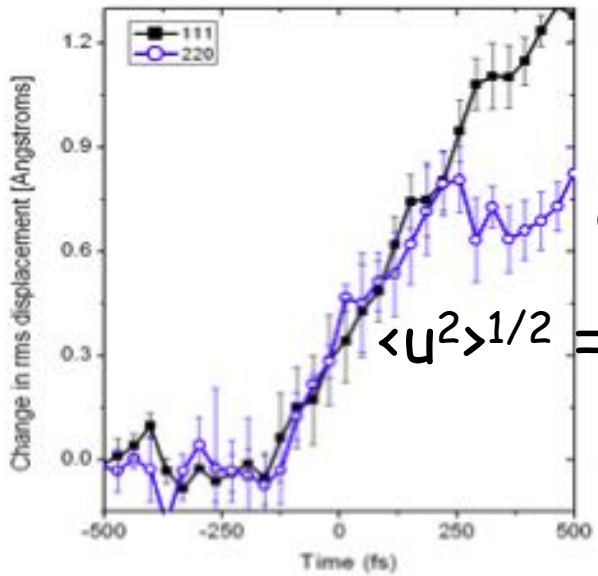
- For ϕ = 24 deg and x-rays grazing: ~18 fs/pixel
- Measures complete time history around t=0 in single shot

Sub-picosecond structural phase transition

A. Lindenberg *et al.*, Science 308, April 15, 2005

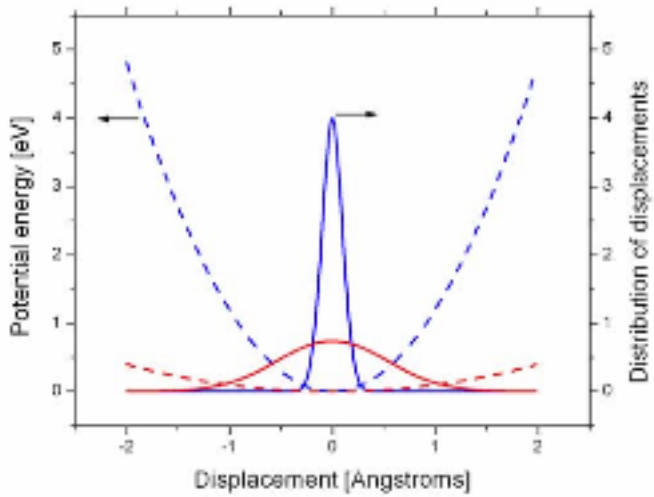
Ultrafast Disordering In Solids



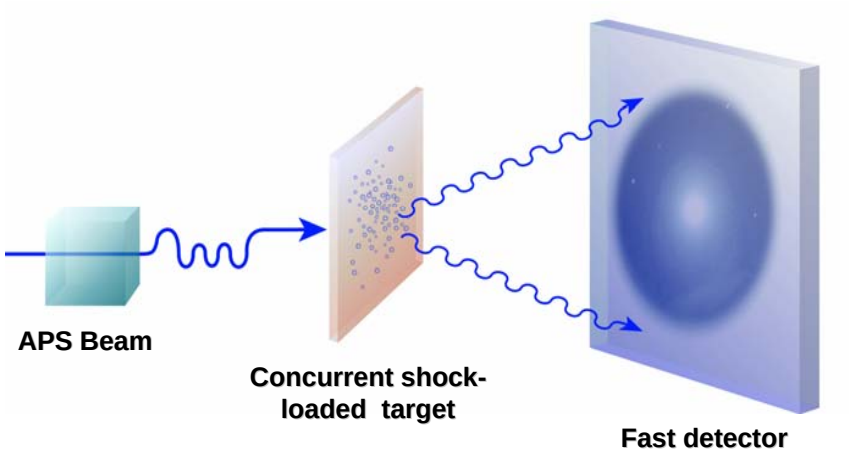


$e^{(-G^2\langle u^2\rangle/3)}$

$\langle u^2\rangle^{1/2} = vt$



Picosecond to sub-picosecond pulse duration enables ultrafast, in situ measurements with the shockwave *frozen* inside the solid



Schematic illustration of experiment

Participants:
LLNL: J. Belak, H. Lorenzana, J. Kinney, R. Lee, R. Hanks, C. May,
Oxford: J. Wark
APS: J. Ilavsky, J. Hessler, W-K, Lee, K. Fezzaa, E. Dufresne, E. Landahl

LLNL LDRD: Ultrafast, in situ probing of shocked solids, H. Lorenzana, Lead PI

Capabilities

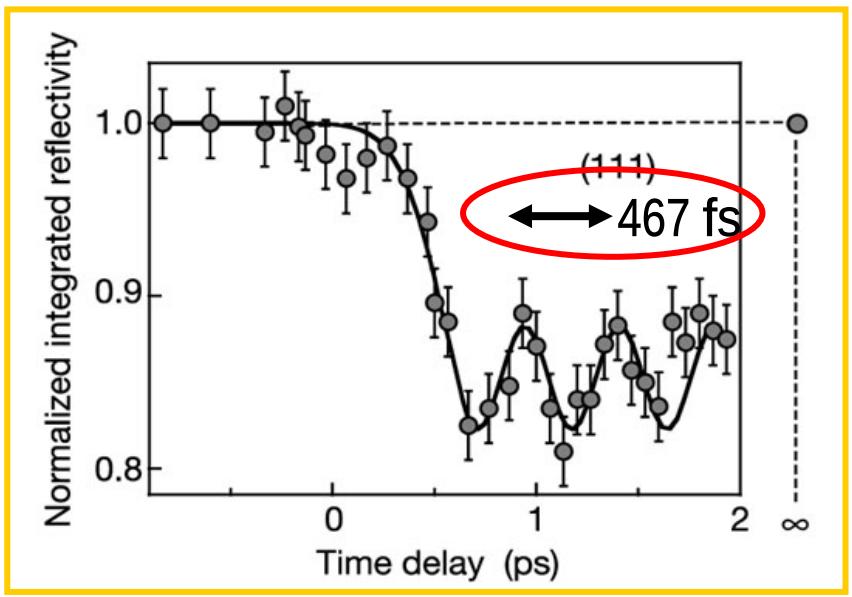
- **Diffraction: dynamic lattice deformation including phase transformation**
- **Small Angle Scattering: Sub-micron void distribution during nucleation and growth of spallation fracture**
- **Diffuse Scattering: Dislocation content and lattice disorder produced by shock loading**

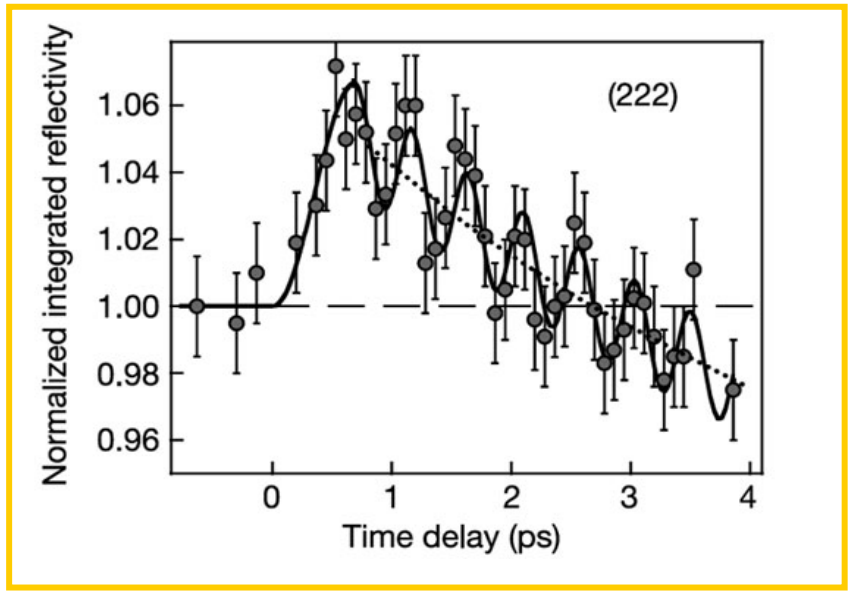
Specifications

- **10's keV photons**
- **10^7 –10^9 photons/pulse**
- **ps to sub-ps pulse width**
- **Highly collimated and monochromatic**
- **Ultra-low background noise**
- **Mini-flyer shockwave loading**

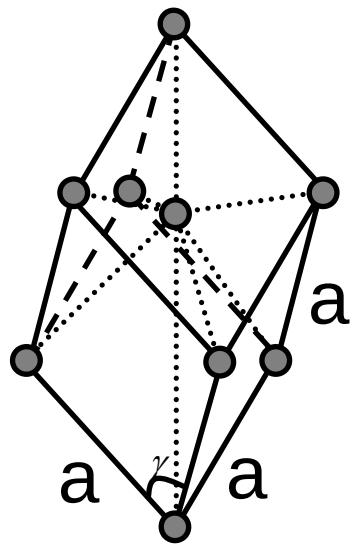
x-ray diffraction from coherent optical phonons in Bi

Sokolowski-Tinten *et al.*, Nature, **422**,p. 287 (2003)

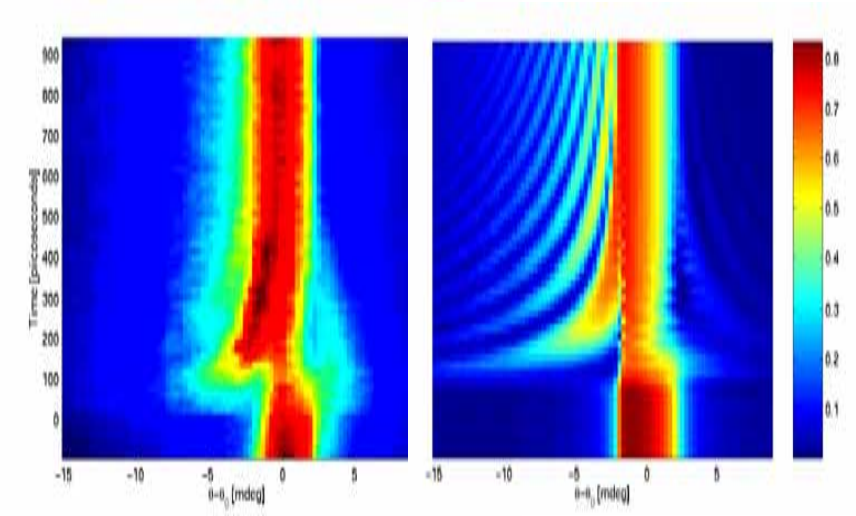




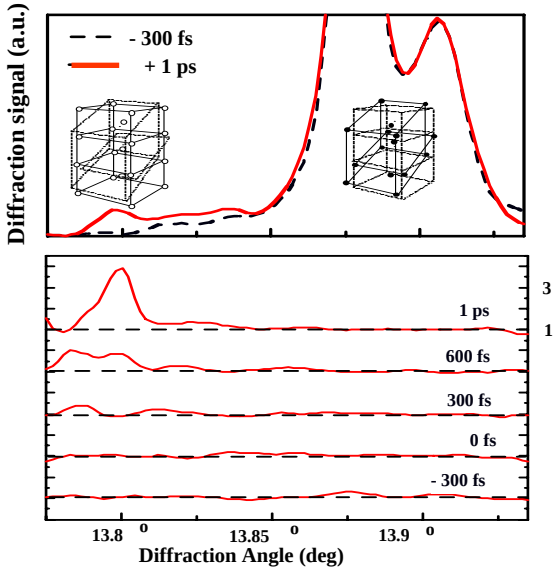
- Strong softening 2.9->2.1 THz
- Large displacements ~15pm
- Low flux, plasma source limited S:N



Diffraction studies near zone-center ▪ modes and symmetry changes

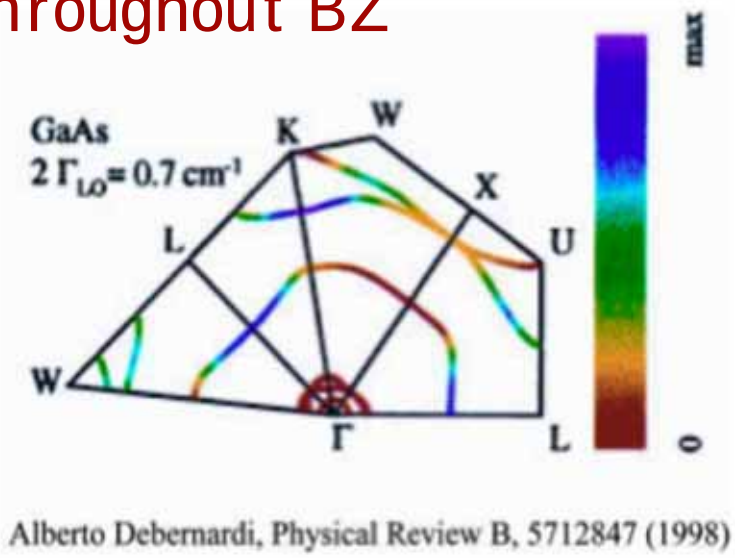


D.A. Reis, *et al,* Phys. Rev. Lett. 86, 3072–3075, 2001.

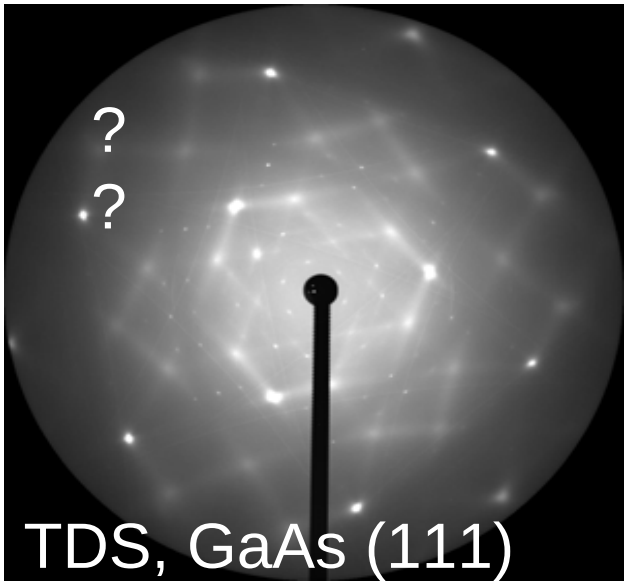


Cavalleri et al. *Phys. Rev. Lett.* 87, 237401 (2001)

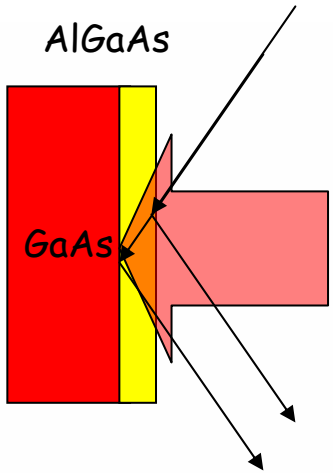
TR x-ray diffuse scattering of nonthermal phonons Throughout BZ

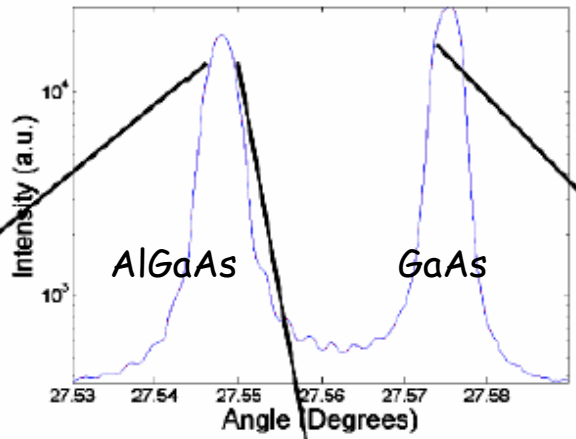


Alberto Debernardi, Physical Review B, 5712847 (1998)

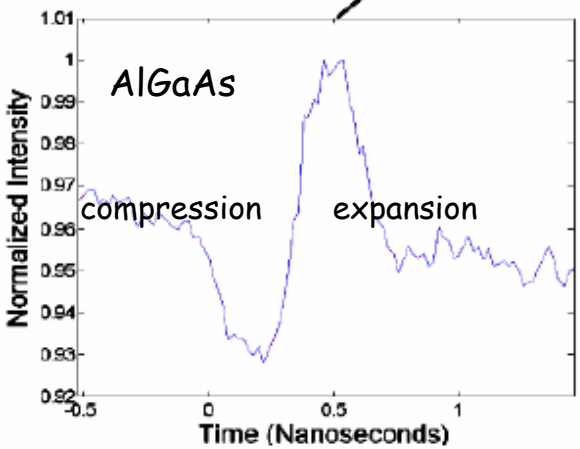


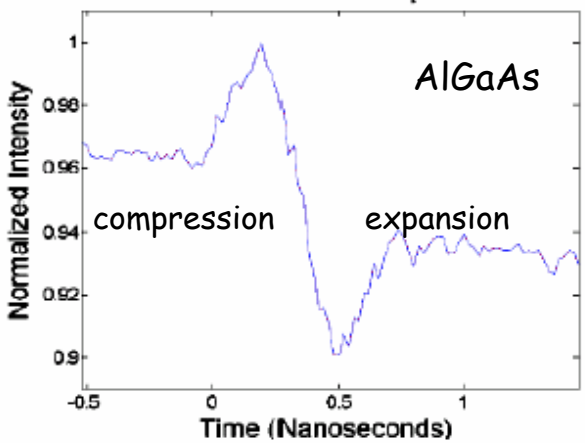
Strain propagation across heterostructure boundaries

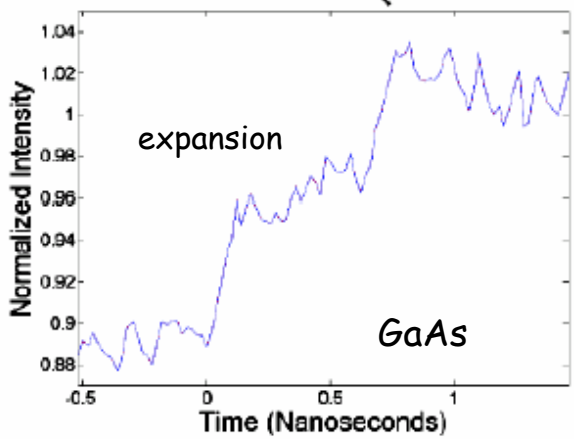




- Electron-phonon coupling and transport
- Boundary resistance (Kapitza), thermal conductivity etc.
- Superlattices and folded phonons







S. Lee, A. Cavalieri, D. Fritz, R. Hegde, R. Goldman and D. Reis

A Challenge to Accelerator Physicists