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Picosecond X-ray Science

Eric Landahl, David Reis, Jin Wang, Linda Young

APS Upgrade Summary

August 11, 2006

- *Current time-resolved science at APS*
  - *New phenomena at 1 ps*
  - *Current x-ray beam parameters*
- *Phase I and II (Sep 07 and Sep 08)*
- *ERL*

APS Strategic Planning Meeting

Time-Domain Workshop Summary and Recommendations

Linda Young (ANL) Atomic & Molecular Physics

Lin Chen (ANL) Chemistry & Biology

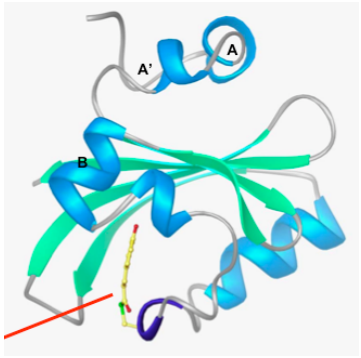
David Reis (UMich) Condensed Matter Physics

Stephen Milton (ANL) Accelerator Physics

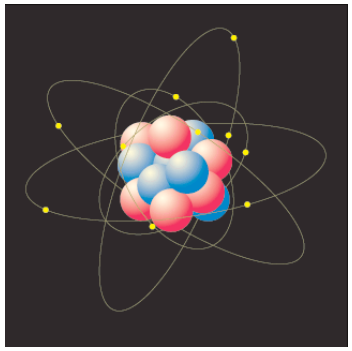
Workshop Chairs

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

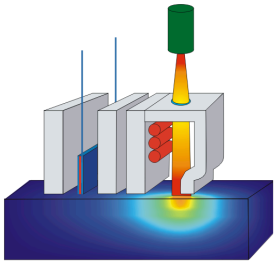
Tracking nuclear motions with x-rays transcends disciplines



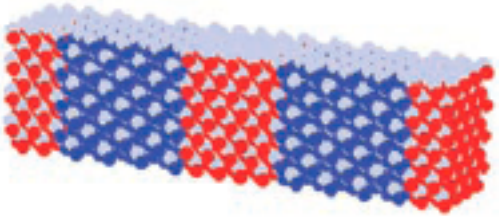
Biology



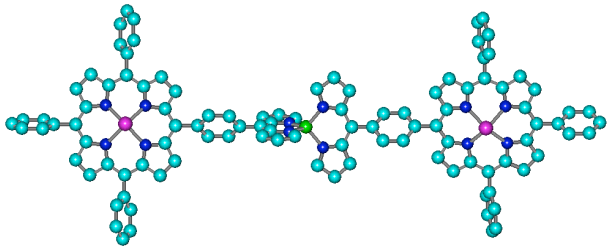
AMO



Materials Science



Condensed Matter



Chemistry

Fundamental Challenge: Understand Structure-Function Relationship on the Atomic Scale : picometers and picoseconds

AMO: coherently controlled atomic & molecular processes

Chemistry: photosynthetic systems, molecular machines

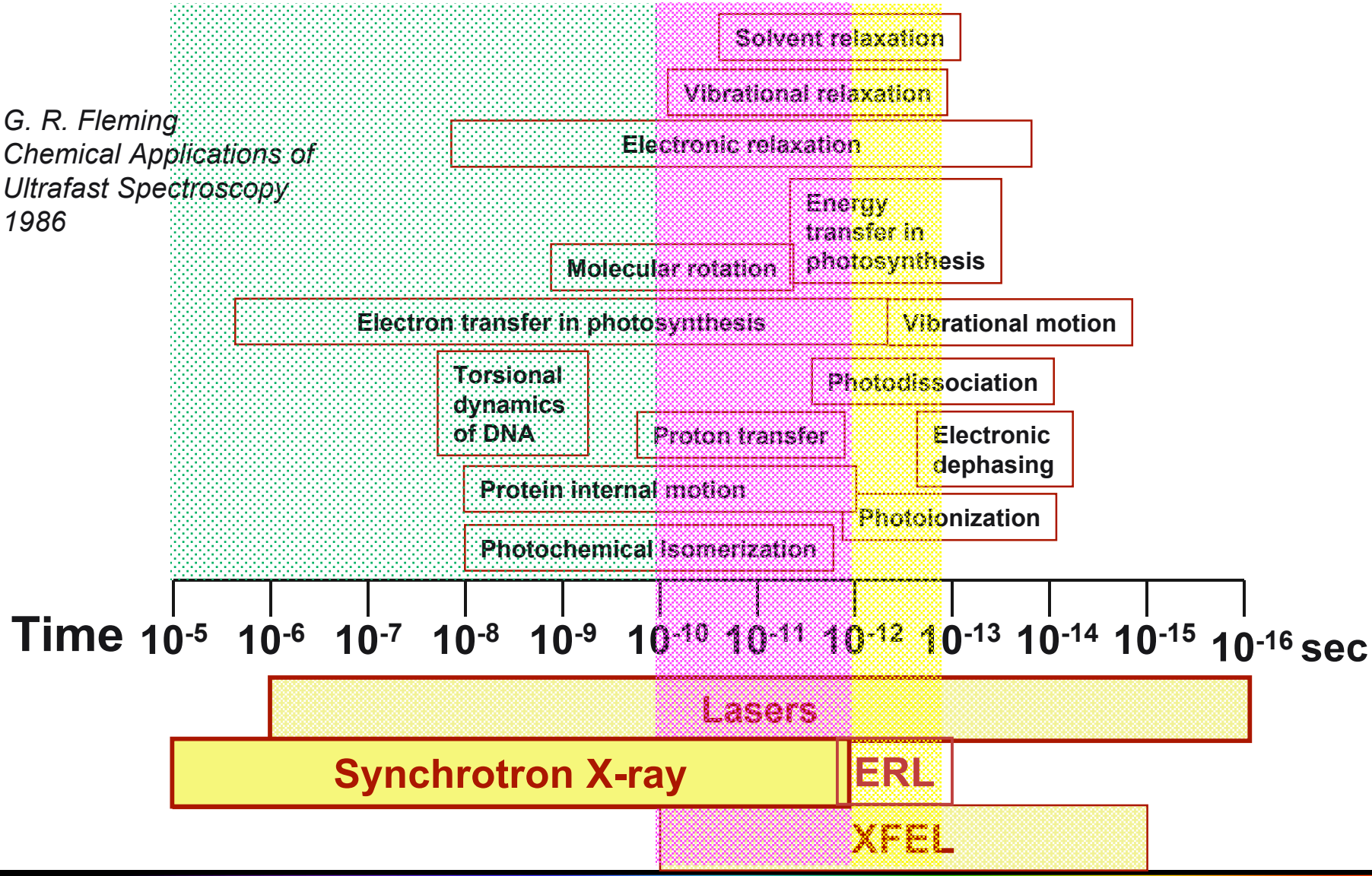
Condensed Matter: phase transitions (structural & magnetic)
strain propagation - coherent phonons
shock phenomena

Materials Science: engineered nanostructure devices
magnetic structures

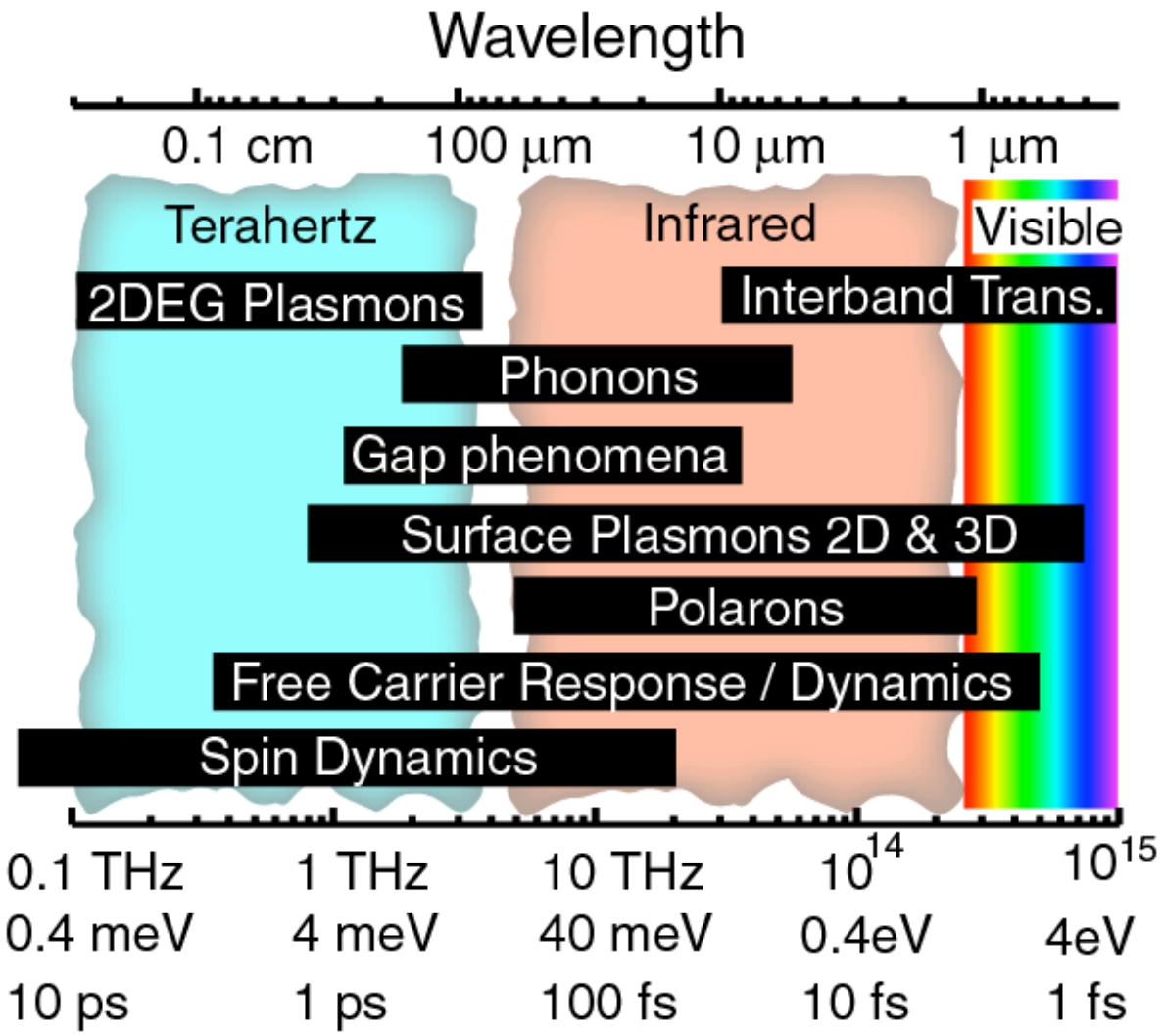
Biology: photoactive proteins

Multiple timescales in chemical processes

G. R. Fleming
Chemical Applications of
Ultrafast Spectroscopy
1986



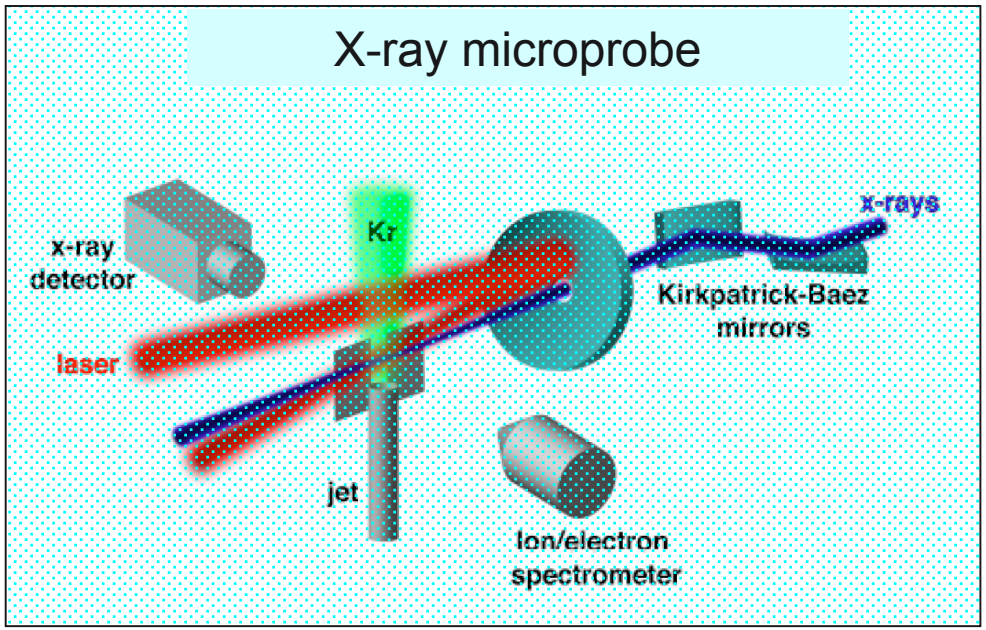
Multiple timescales in condensed matter



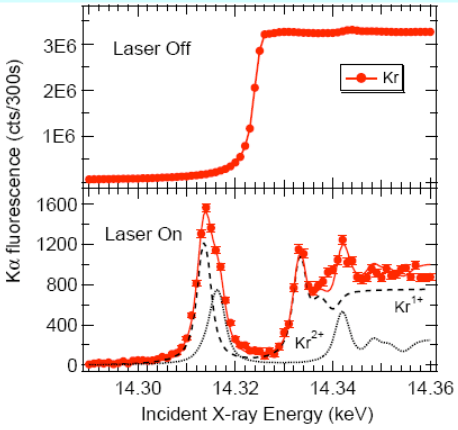
From Toni Taylor's presentation @ Cornell ERL workshop Jun 06

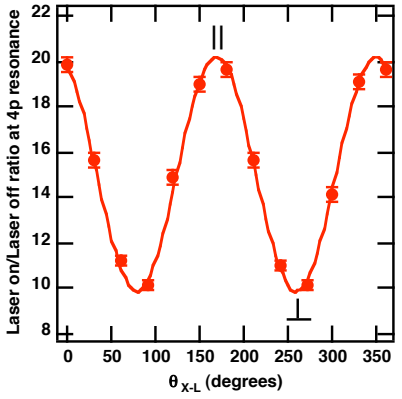
AMO: *Krypton - orbital alignment in strong-field ionization*

Motivation: Understand and control atoms/molecules in strong laser fields



Time-resolved resonant, polarized x-ray absorption

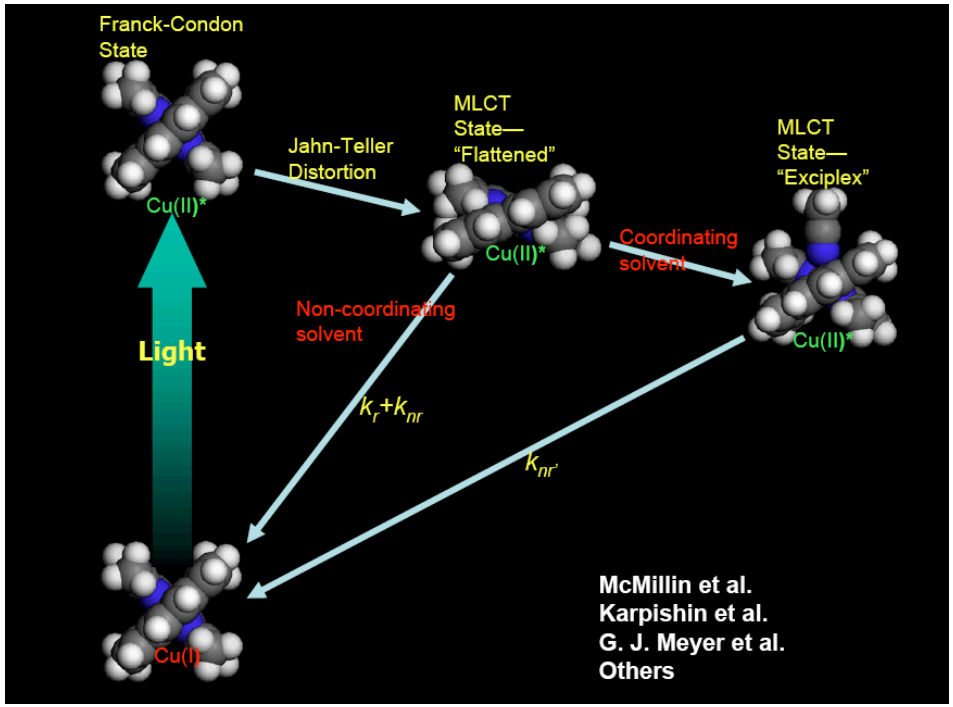




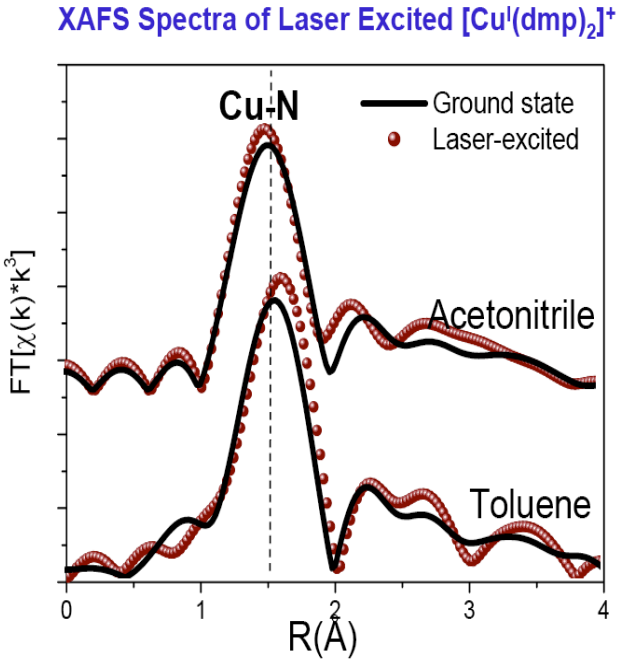
Orbital alignment dynamics in atoms in plasma - ns
Rotational alignment dynamics in molecules - ps

Chemistry: *Cu(DMP)$_2$ - a prototypical molecular machine*

Motivation: Understand excited-state dynamics on ultrafast timescale
structural origin of solvent dependence of MLCT state
structure-function relationship in MLCT state



Time-resolved XAS

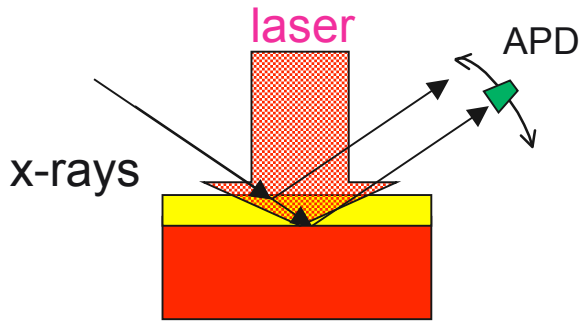


L. Chen et al., J. Am. Chem. Soc. (2002) **124**,*10861;* (2003) **125**,7022.

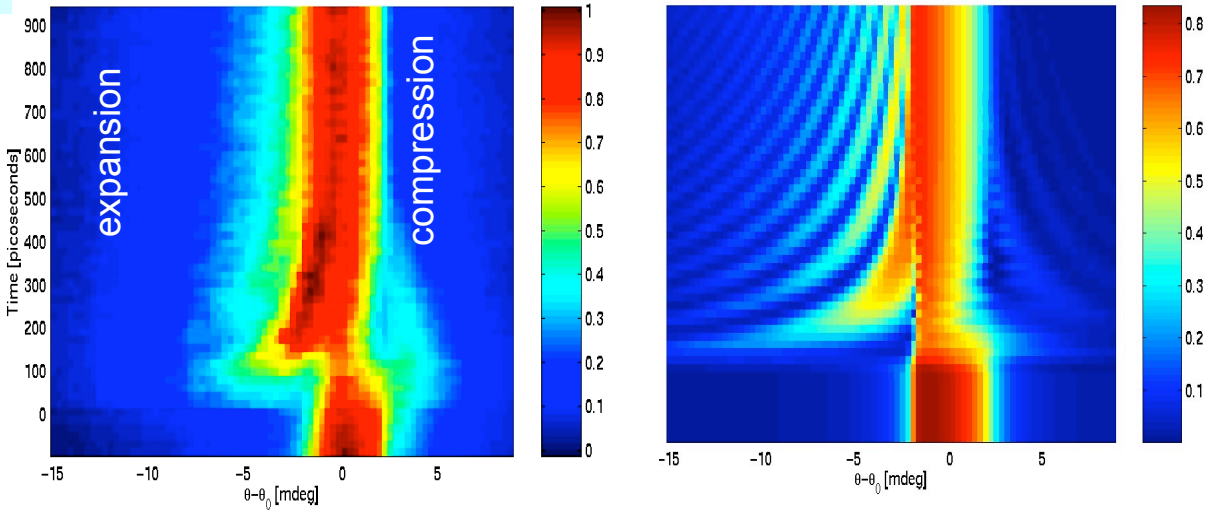
Condensed Matter: *Impulsive strain propagation in InSb*

Motivation: Understand response of condensed matter to impulsive optical excitation as fcn of fluence
quantitative measurements of atomic displacements
track energy flow from optical excitation into lattice modes

Time-resolved x-ray diffraction
proportional mode



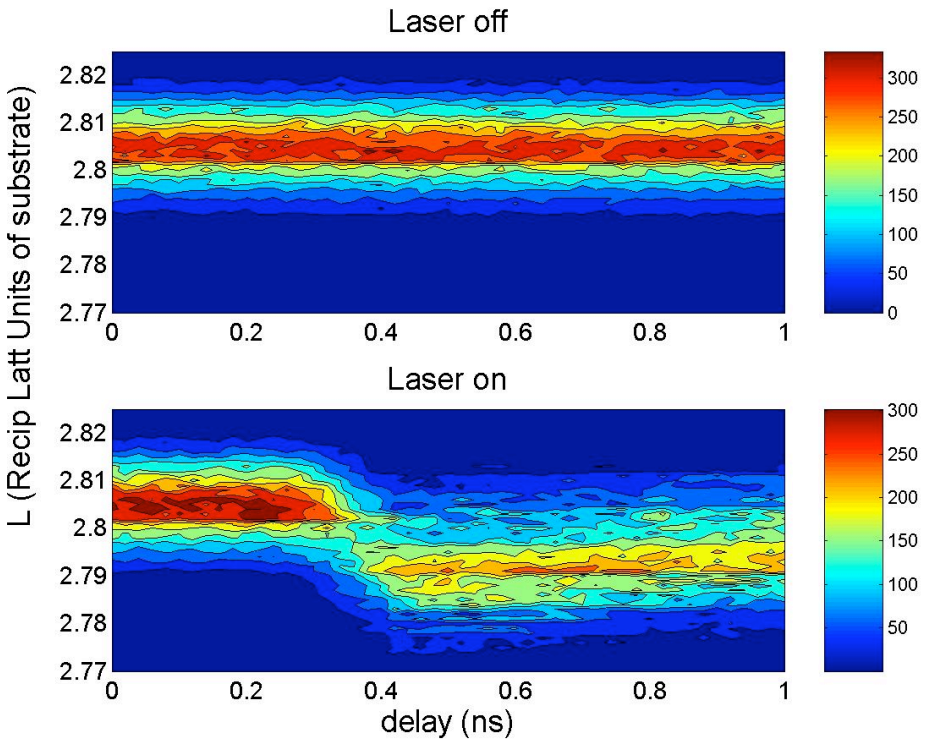
InSb diffraction vs laser/x-ray time delay



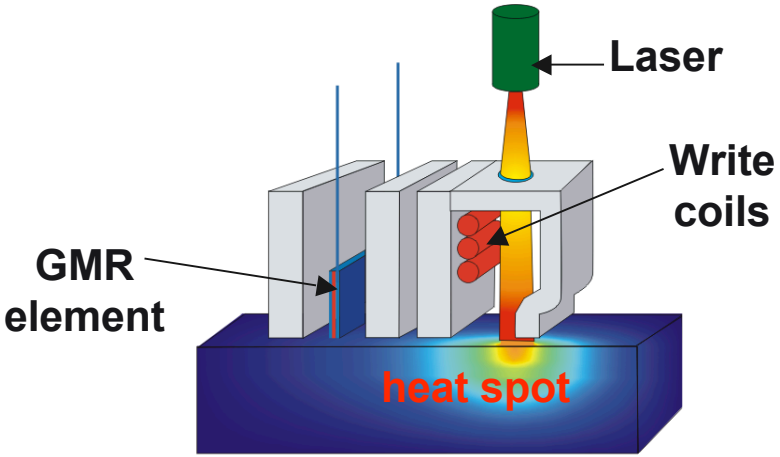
Materials Science: *FeRh lattice-expansion phase transition*

Motivation: Explore correlated magnetic and structural phase transitions in a material of technological interest

Time-resolved x-ray diffraction in counting mode

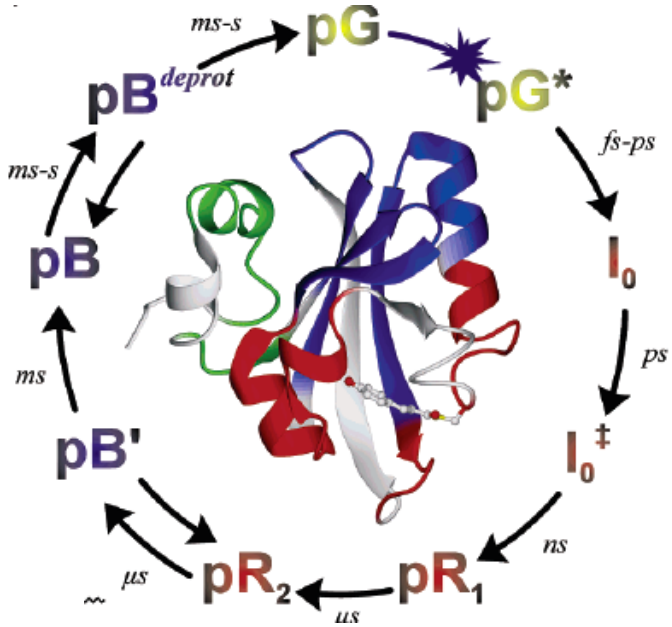


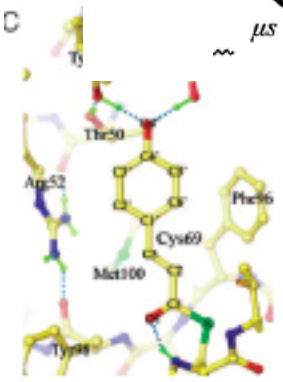
- Ultrafast laser heating induces magnetic phase transition (~500 fs) and anomalous lattice expansion
- Present measurements on homogeneous thin films
- Material may have applications in Thermally Assisted Recording:



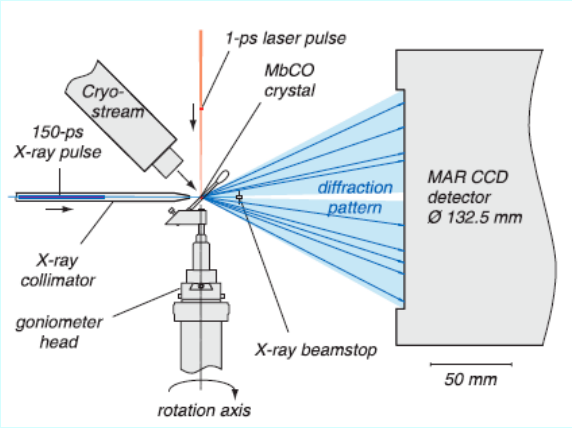
Biology: *Photoactive Yellow Protein - a reversible photosystem*

Motivation: track position of all atoms in space upon light activation in this blue-light photoreceptor exhibiting avoidance response.





Time-resolved Laue crystallography



Established four intermediate state structures from electron density maps from 1 ns to 1 s

Ihee et al PNAS 102,7145 (2005)
Rajagopal et al, Structure 13, 44 (2005)
Anderson et al Structure 12, 1039 (2004)
Rajagopal et al., Acta Cryst. D60, 860 (2004)

Missing info on primary cis-trans photoisomerization ~ ps timescale

X-ray parameters currently used for laser/x-ray pump probe expts at 100 ps

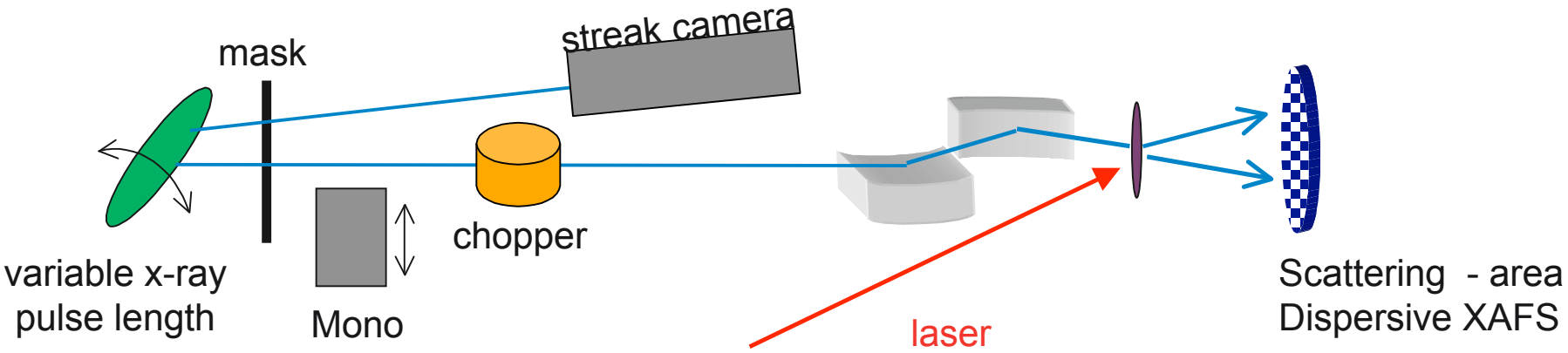
Experiment	Technique	Flux (ph/s)	Spot Size HxV (μm^2)	Bandwidth	Divergence	Rep rate (kHz) Current/max
AMO	Spectroscopy Scattering	10^9 10^{11}	10 x 10 10 x 10	10^{-4} 10^{-2}	~2 mrad ~2 mrad	1 / 3000
Chemistry (disordered)	Spectroscopy Scattering	3 x10^8 2.7 x10^7	500 x 100 1000 x 30	10^{-4} 10^{-4}	NA ~2 mrad	1 / 40 0.5 / 40
Condensed Matter	Diffraction Scattering	10^9 10^6 10^9	500 x 500 500 x 500 (10x10) 50 x 50	10^{-4} 10^{-2}	17 µrad ~2 mrad	1-5 / >100
Materials Science	Thin-film diffraction	10^8	100 x 100	10^{-4}	320 µrad	1-5 / >100
Biology (crystalline)	Laue diffraction	10^{10}/ pulse	100 x 150	1-3x10^{-2}	NA	1 -3 Hz / sample-limited

Phase 1 & Phase 2: short pulse x-rays

Phase 1: 120 Hz, 3 ps, slit source,10^6 x-rays/1% BW/pulse, 13x16 µrad^2

- Measure x-ray/laser jitter (shot-to-shot determination?)
- Study multiple slit option (x-ray diagnostics, x-ray pump/probe experiments)
- Prepare beamline optics, instrumentation, detectors for dispersive spectroscopies and scattering
- Possible science: time-resolved x-ray diffraction of terahertz phonons non-thermal melting, transient absorption

Phase 2: 1000 Hz, 1 ps, slit source, 3 x10^6 x-rays/1% BW/pulse, 13x16 µrad^2



Ultrafast x-ray science: a rapidly growing user community

Rapid growth timing proposals 2003-3 to 2006-3 @ Sector 7: 4 →14 UP 250%
Sector 11: 1 →7 UP 600%

APS growth: 2003 to 2006: number of proposals UP 40%

Jim Belak (Lawrence Livermore National Laboratory)
Phil Bucksbaum (Stanford University)
David Cahill (University of Illinois)
Roy Clarke (University of Michigan)
Phillip Coppens (State University of New York, Buffalo)
David Ederer (Tulane University)
Paul Evans (University of Wisconsin)
Eric Fullerton (Hitachi Global Storage Technologies)
Jacob Jones (University of Florida)
Jorgen Larsson (University of Lund)
Soo-Heyung Lee (KRISS, Korea)
Aaron Lindenberg (SLAC)
Hector Lorenza (Lawrence Livermore National Lab)
Roberto Merlin (University of Michigan)
Martin Nielsen (Copenhagen University)
Stanislas Pommeret (Commissariat à l'énergie atomique)
David Reis (University of Michigan)
Ed Stern (University of Washington)
Phil Anfinrud (NIH)
Keith Moffat (University of Chicago)
Vukica Srajer (University of Chicago)
William Royer (University of Massachusetts Medical School)
James Norris (University of Chicago)
Harry Ihee (KAIST, Daejeon, South Korea)
Marius Schmidt (Technische Universitaet Muenchen, Germany)

Uli Nienhaus (University of Ulm, Germany)
Maria Hrmova (The University of Adelaide, Australia)
Cynthia Stauffacher (Purdue University)
Douglas Ohlendorf (University of Minnesota)
Andrew Mesecar (University of Illinois at Chicago)
Nicola Armaroli (ISOF - CNR, Italy)
Felix Castellano (Bowling Green State U.)
Michael Hopkins (U. of Chicago)
Chuan He (U. of Chicago)
Michael Hopkins (U. of Chicago)
Joseph Hupp (Northwestern U.)
Himansu Jain (Lehigh)
Frederick D. Lewis (Northwestern U.)
Jonathan S. Lindsey (North Carolina State)
Braja K. Mandal (IIT)
Gerald J. Meyer (Johns Hopkins)
Martin Newcomb (U. Illinois Chicago)
Daniel Nocera (MIT)
Kirk S. Schanze (U. of Florida)
Russell Schmehl (Tulane)
Michael Schmittel (Universität Siegen, Germany)
Michael R. Wasielewski (Northwestern U.)
Glenn A Waychunas (LBNL)
Luping Yu (U. of Chicago)
Ling Zhang (Southern Illinois U.)
Jeffery Zink (UCLA)

Summary

- Exciting times for short pulse x-rays!
- 1 ps accesses *many* phenomena previously hidden at 100 ps
- Synchrotron advantage over laser plasma & LCLS : easily tunable
- Large & diverse user community growing rapidly.
- Working group is being formed to implement "fast track" Phases 1 & 2
  - -tunable, polarized, monochromatic, focused x-rays
  - -*variable* pulse length (1-100 ps)
  - -1 kHz, 10^9 x-rays/s, 1% bandwidth
- ERL would be a major advance for ultrafast time-resolved studies
  - 50 fs, 100 kHz, 10^9 x-rays/pulse, 0.1% bandwidth

(coexist short pulse + high coherence/high flux modes because short pulse at lower repetition rate doesn't require energy recovery - 2 guns)
I. Bazarov, ERL Workshop Cornell Jun 06

Acknowledgements

Klaus Attenkofer
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Tim Graber
Sarvjit Shastri
Vukica Srajer
Don Walko

60 + participants at the Aug 04 Lake Geneva Workshop

AMO: Bob Dunford, Conny Höhr, Elliot Kanter, Bertold Krässig, Emily Peterson, Steve Southworth