Challenges Far-from-Equilibrium Using Future Light Sources

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Recent References

CONDENSED-MATTER AND MATERIALS PHYSICS: THE SCIENCE OF THE WORLD AROUND US

Committee on CMMP 2010:

An Assessment of and Outlook for Condensed-Matter and Materials Physics

June 14, 2007

National Research Council *of*

The National Academies

Directing Matter and Energy: Five Challenges for Science and the Imagination

A Report from the

Basic Energy Sciences Advisory Committee

U.S. Department of Energy

December 20, 2007

What is Equilibrium State of Matter?

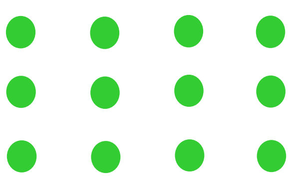
$$F = E - TS$$

Low T

Minimum energy, high symmetry, order

e.g. crystalline

E minimum

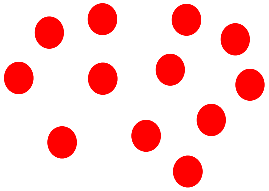


$F = E - TS$

High T

Maximum entropy, low symmetry, disorder

e.g. liquid

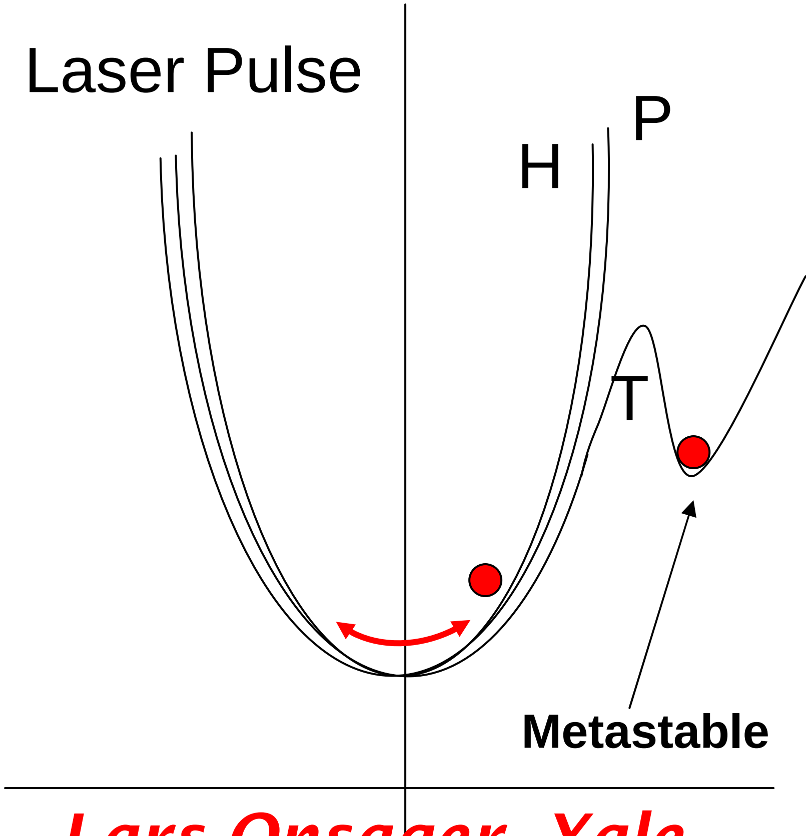


S maximum

$F = E - TS$

What do we generally mean by 'near- equilibrium studies' ?

- *Laws of thermodynamics and statistical mechanics apply*
- *Born-Oppenheimer approximation is valid*
- *Systems respond linearly when perturbed*
- *Often valid for large systems (10^{23} atoms or molecules)*

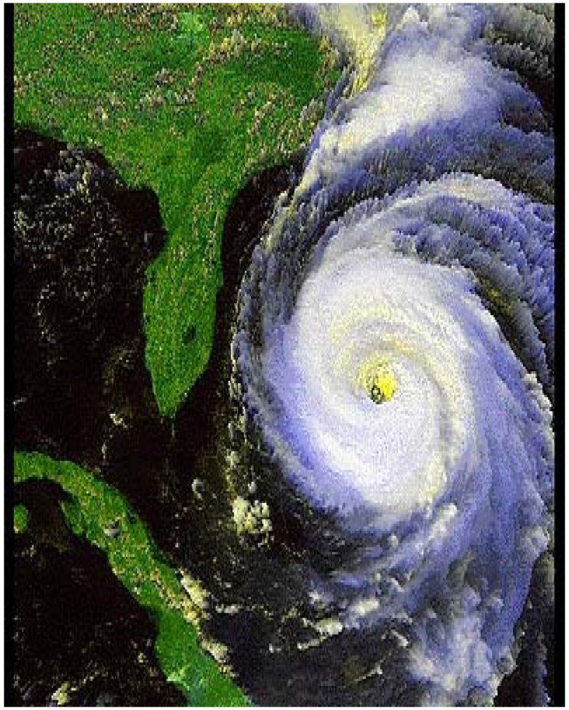


Lars Onsager, Yale
Nobel Prize, 1963

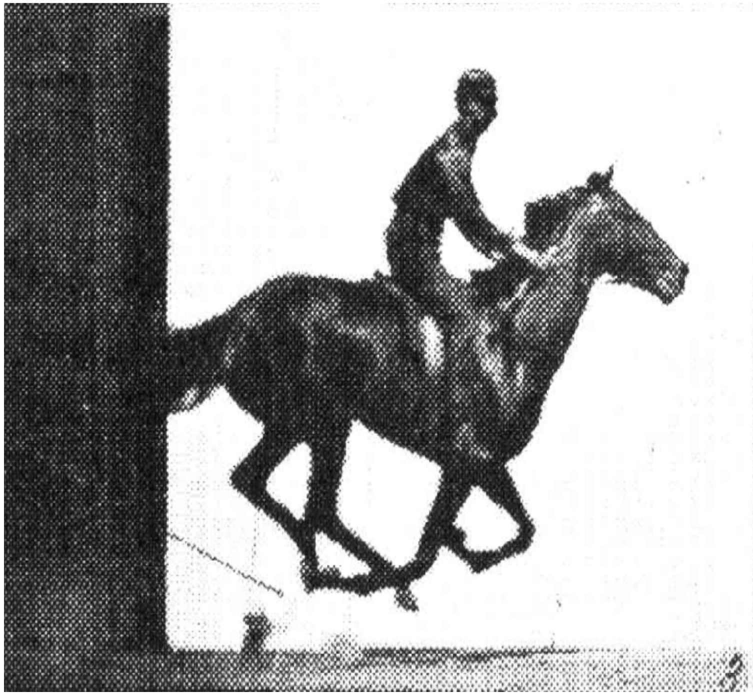
Why study systems that are driven far from equilibrium?

Far-from-equilibrium systems show non-linear response and are more common in nature

Tectonic dynamics

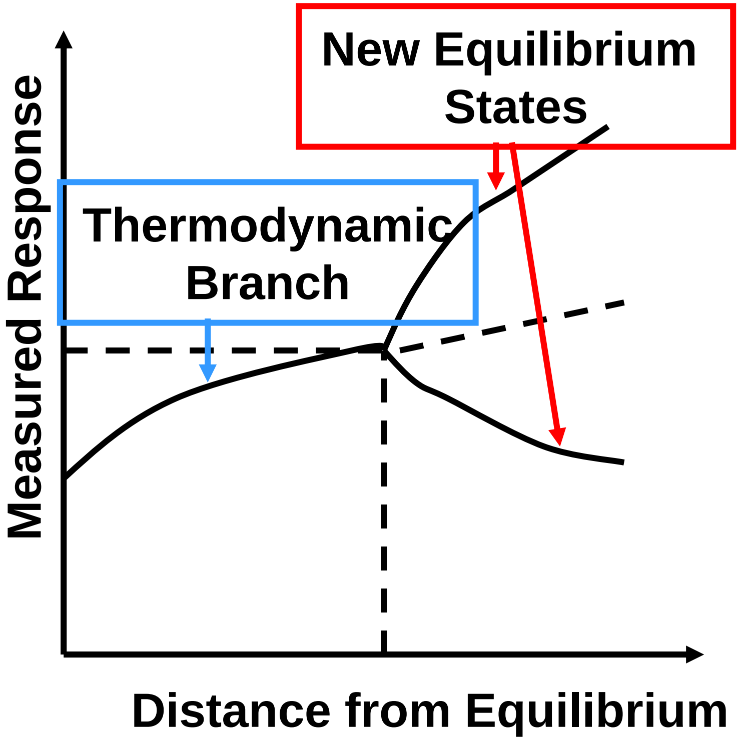


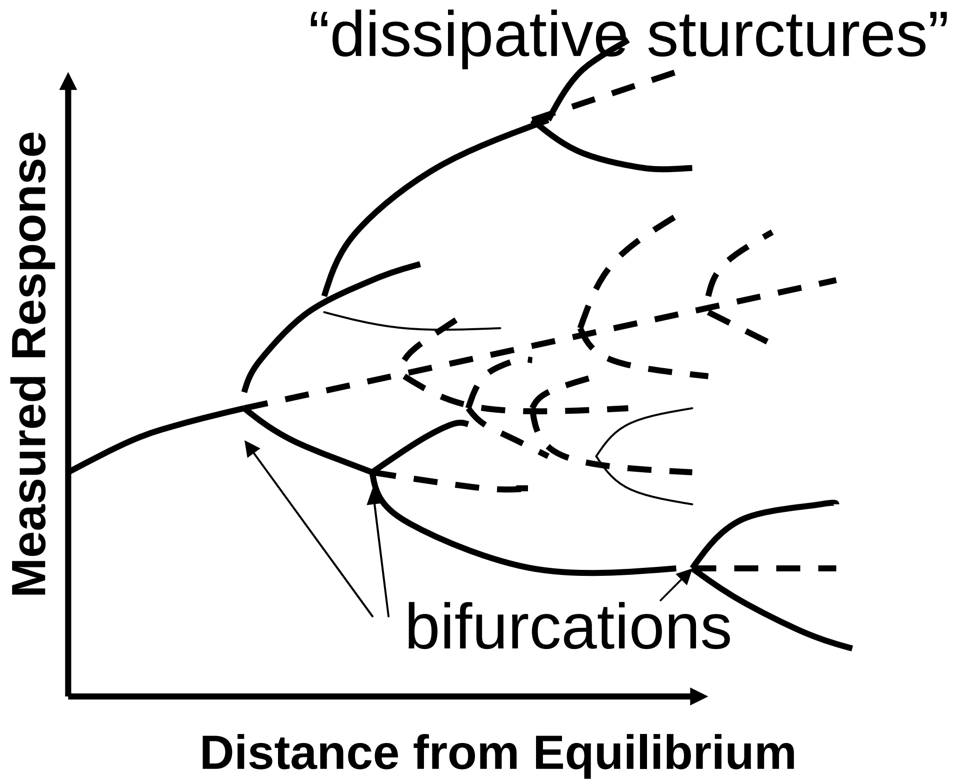
Hurricanes



Racing Horse

How to Describe the Systems Far From Equilibrium?





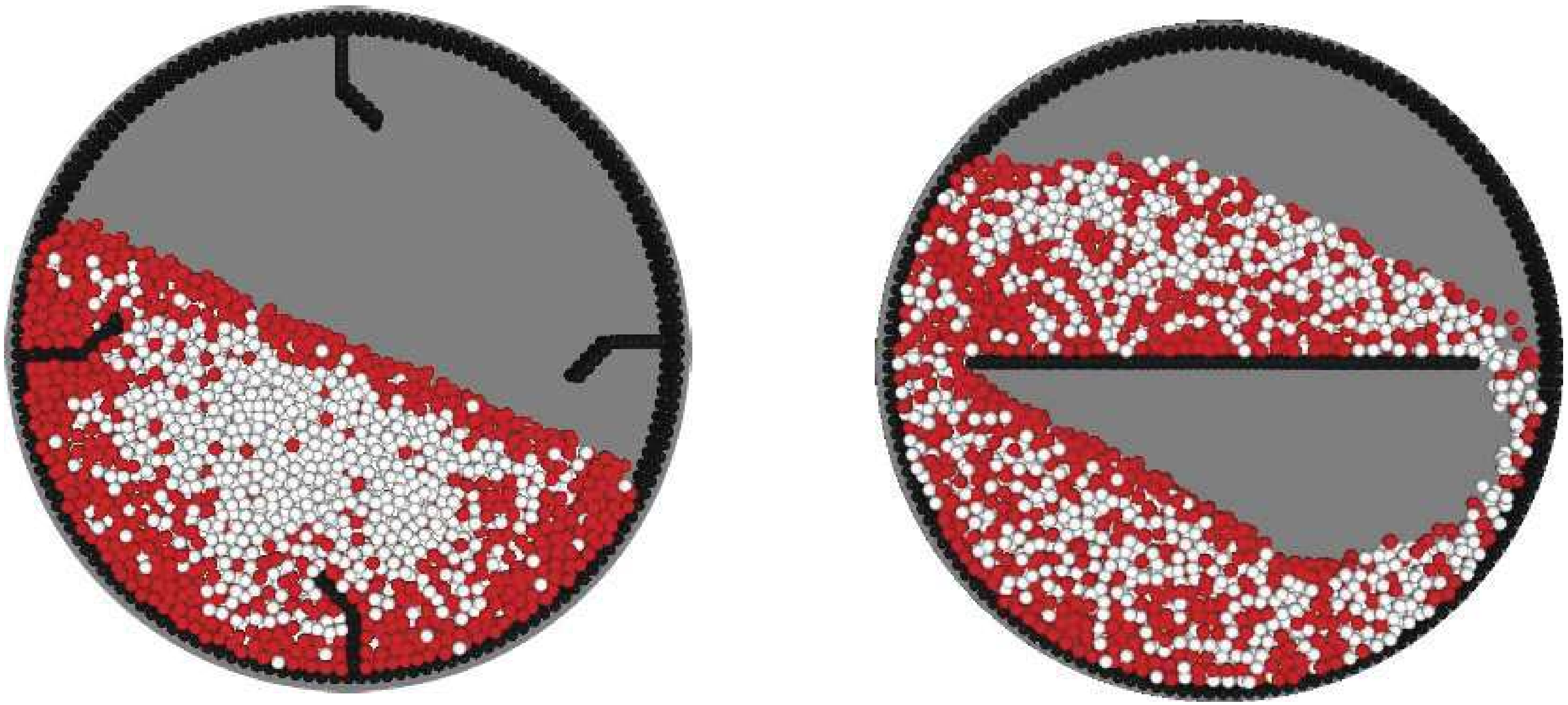
Evolution of Two New Equilibrium Organized States

Further Evolution of Multiple Equilibrium States Following Slight Perturbation

See: I. Prigogine, From Being to Becoming (W. H. Freeman, San Francisco, 1980)

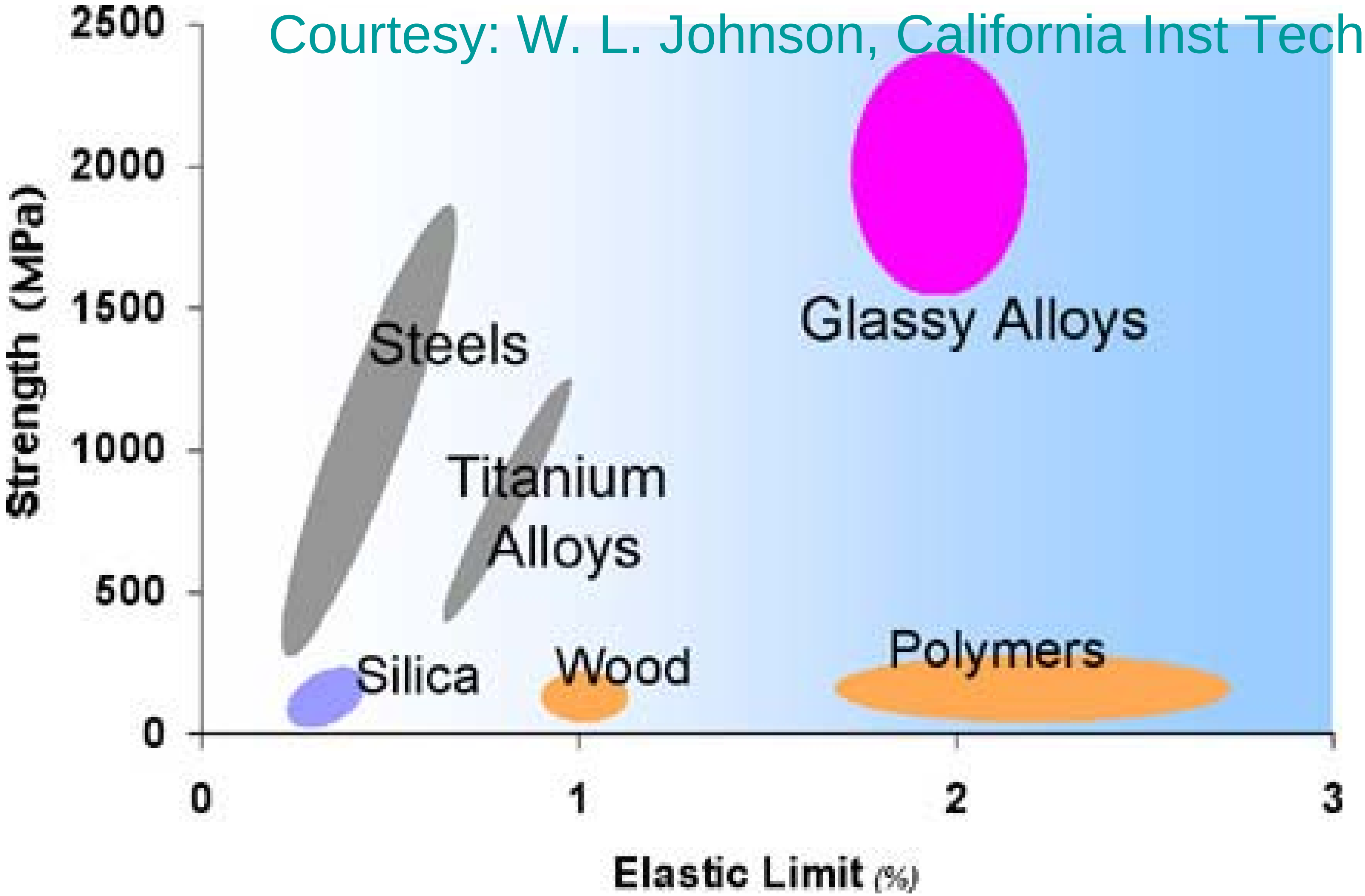
Mixing Granular Materials

Ottino et al: Science 319, 912, 2008



Rapid-Cooling Process of Glassy Alloys Far-from-Equilibrium Produces Highest Strength Materials

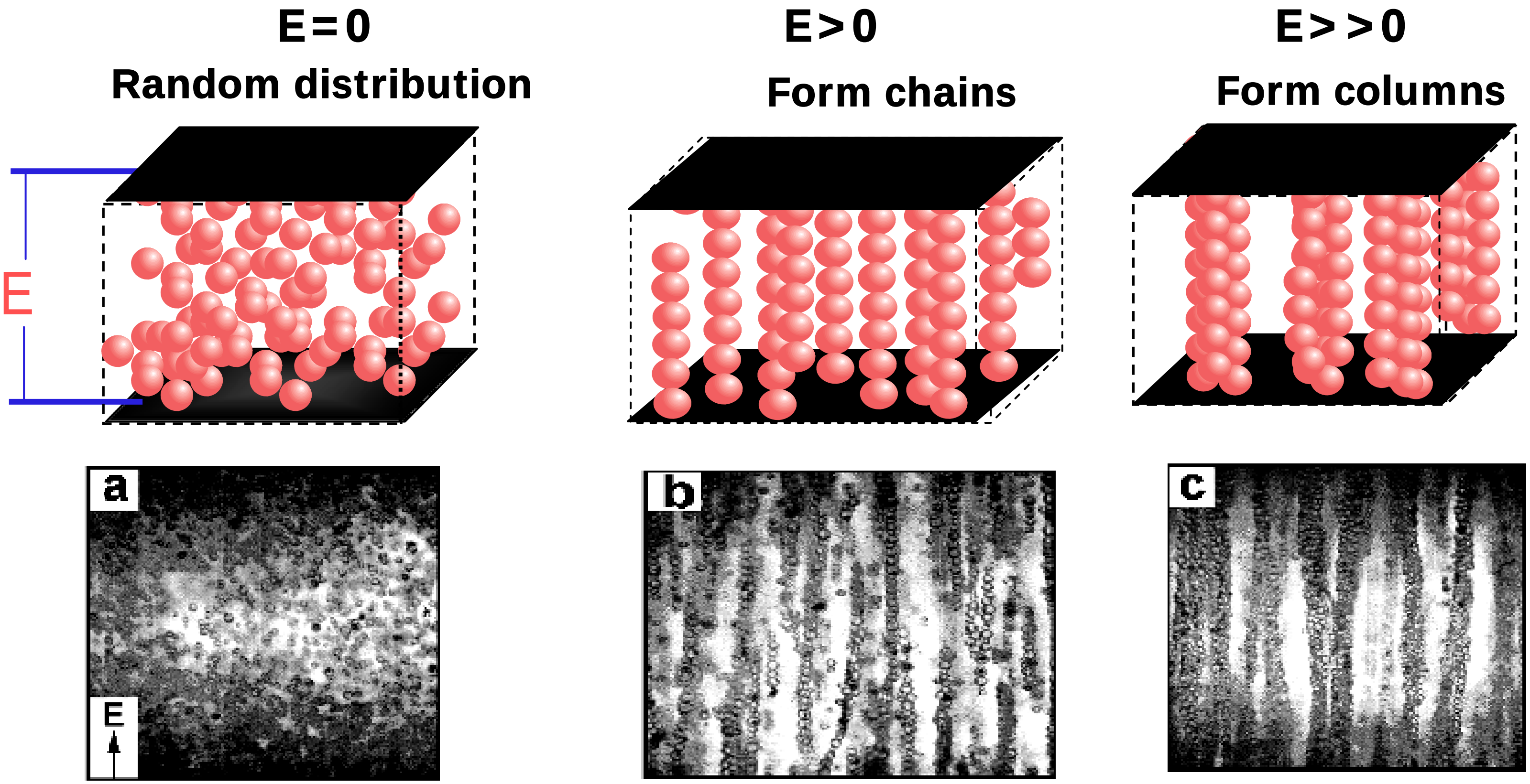
Courtesy: W. L. Johnson, California Inst Tech



What is Electro-Rheology (ER)?

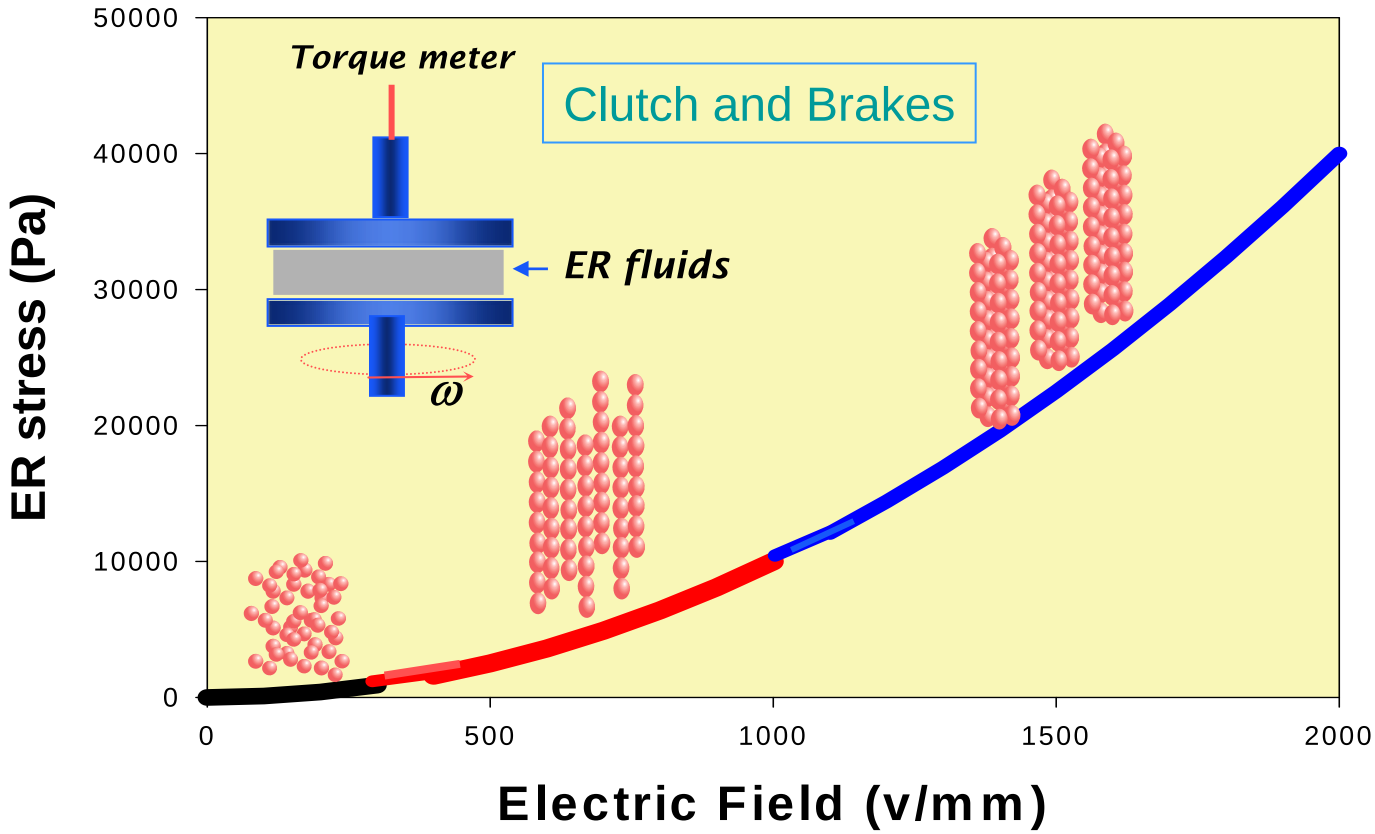
ER fluid consisting of dielectric nano-particles suspended in a nonconducting liquid can change from liquid state to solid state abruptly (1-10 ms)

Another Far-from-Equilibrium Phenomena.

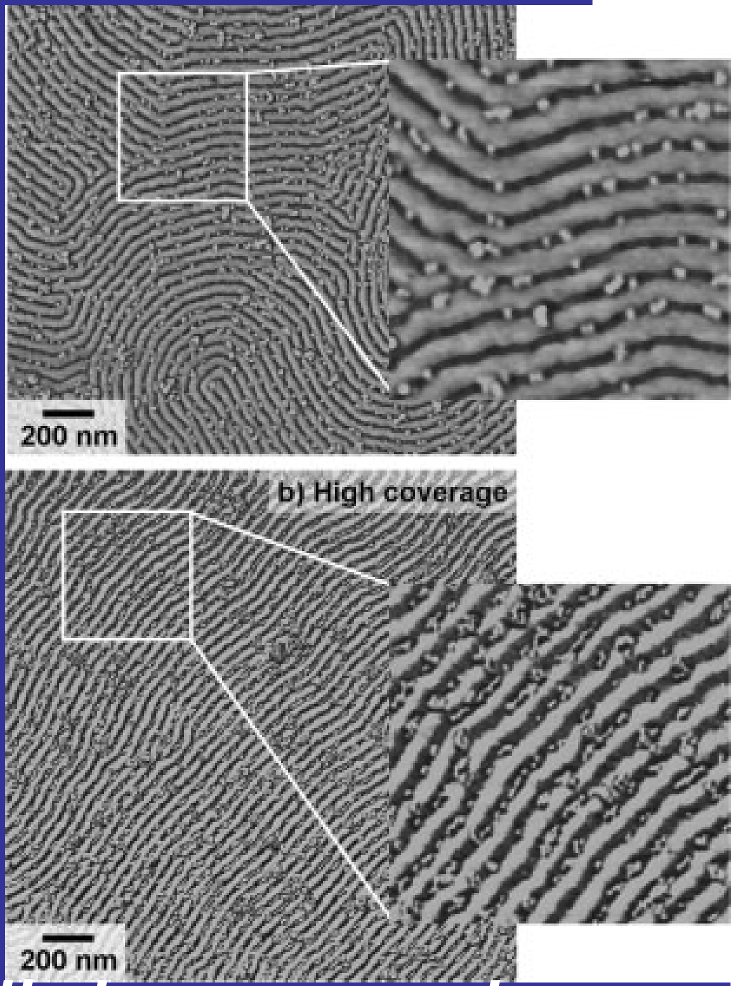


Giant Nonlinear ER fluids prepared with BaTiO3/Urea microspheres in silicone oil

X.X. Huang, S.H. Yang, K.Q. Lu and P. Sheng, Nature Materials 2, 727 (2003).



Surface Patterning and Nanoprocessing

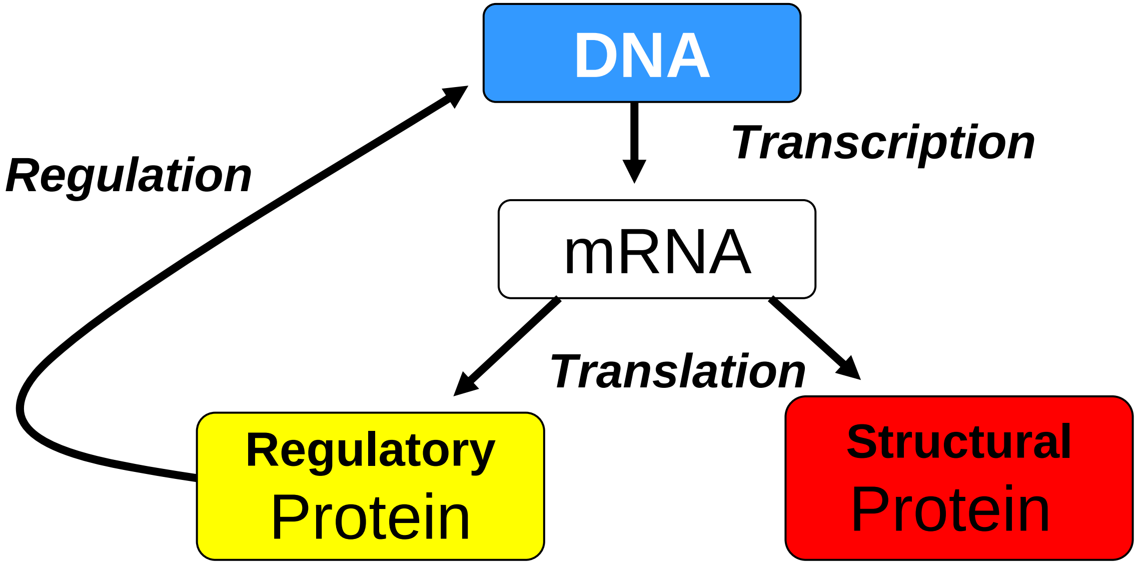


Selective adsorption of single FePt nanoparticles onto the photochemically modified polymer domains using VUV exposure of PMMA diblock copolymer films (dark stripes)

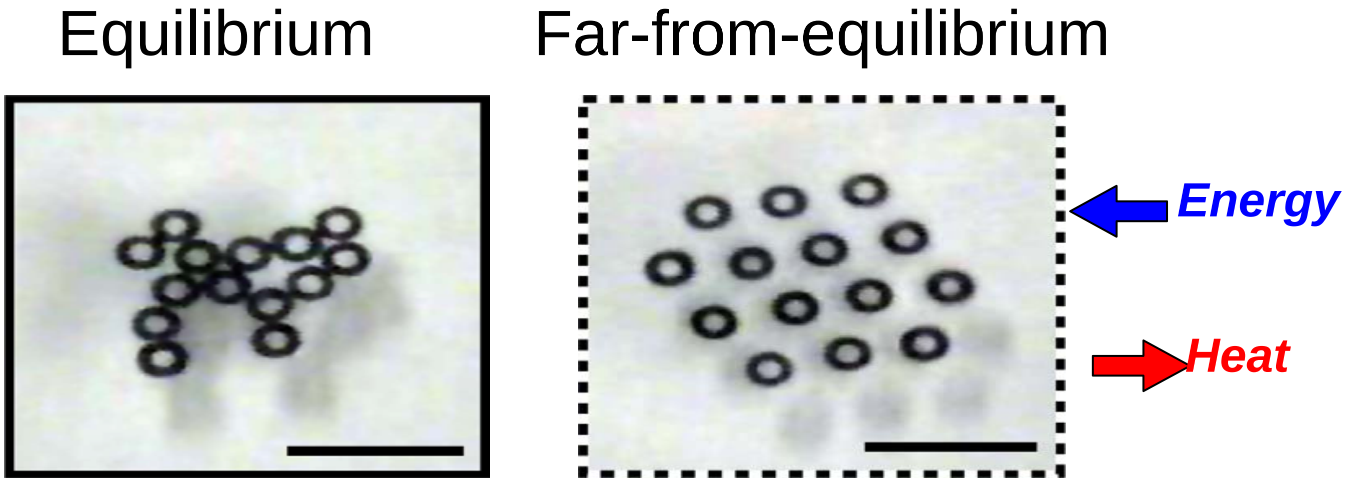
Seth B. Darling, et al., *Adv. Mater.* 17, 2446–2450 (2005).

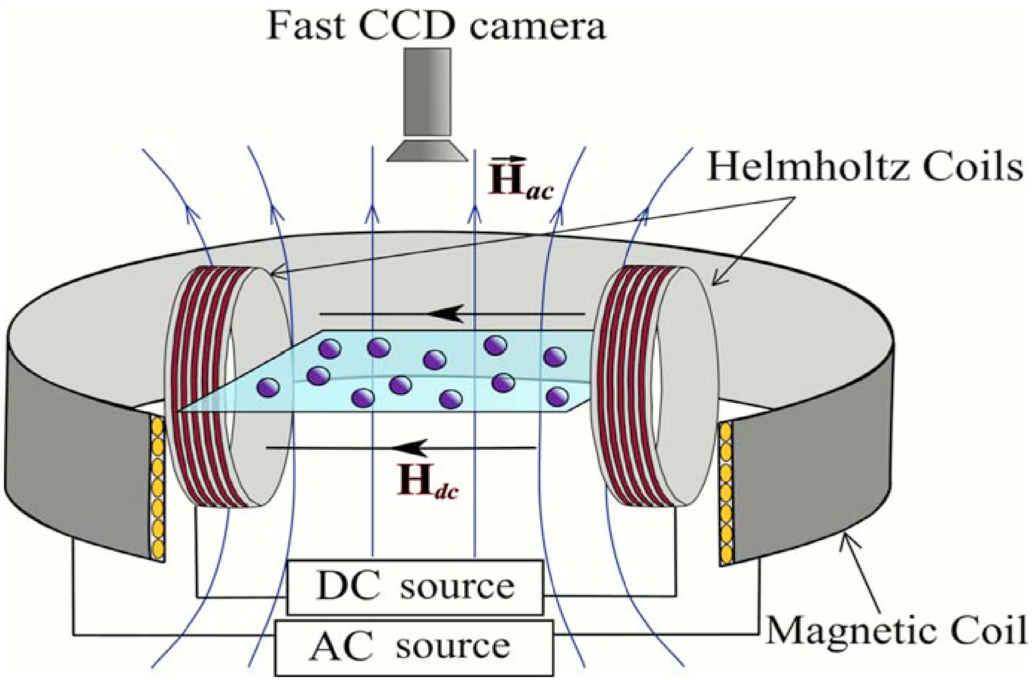
Ramifications of this far-from-equilibrium processing extend to potential future bit-patterned magnetic-storage media.

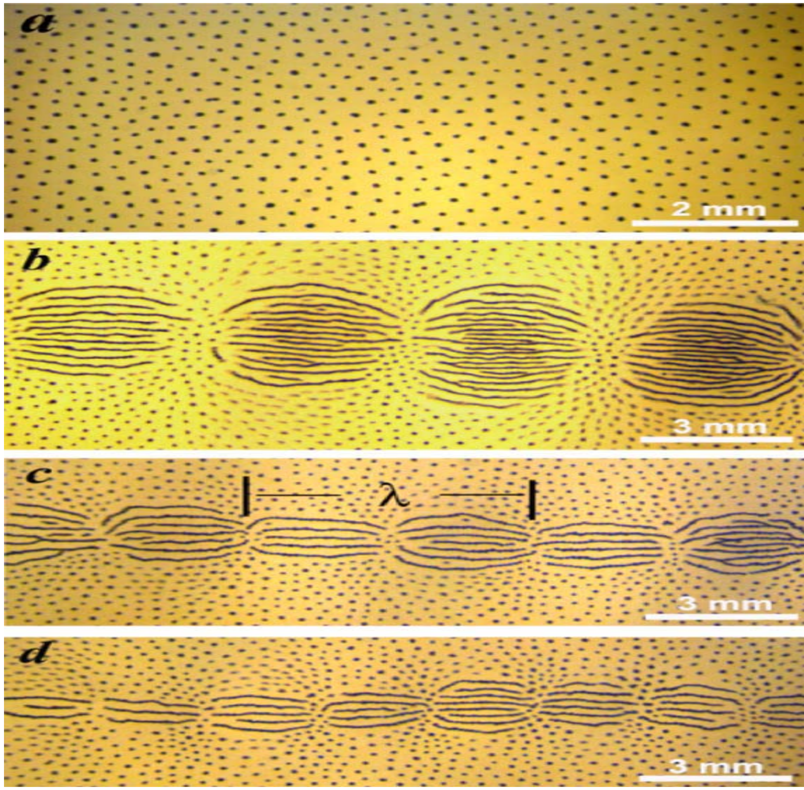
Belousov-Zhabotinsky (BZ) Cyclical Reaction: Extension to Living System



Living systems can reproduce themselves and can collect and exchange information







SNEZHKO et al Phys Rev E 73, 041306 2006

How to begin to understand far-from-equilibrium phenomena?

- ***Develop a quantitative understanding of far-from-equilibrium dynamics over the characteristic time of the system - from attoseconds to many hours***
- ***Determine the pathways, processes and configurations (from self-organized nano-assemblies to chaotic order)***
- ***Develop an understanding of the multi-scale behavior of materials and its relationship to emergent properties***

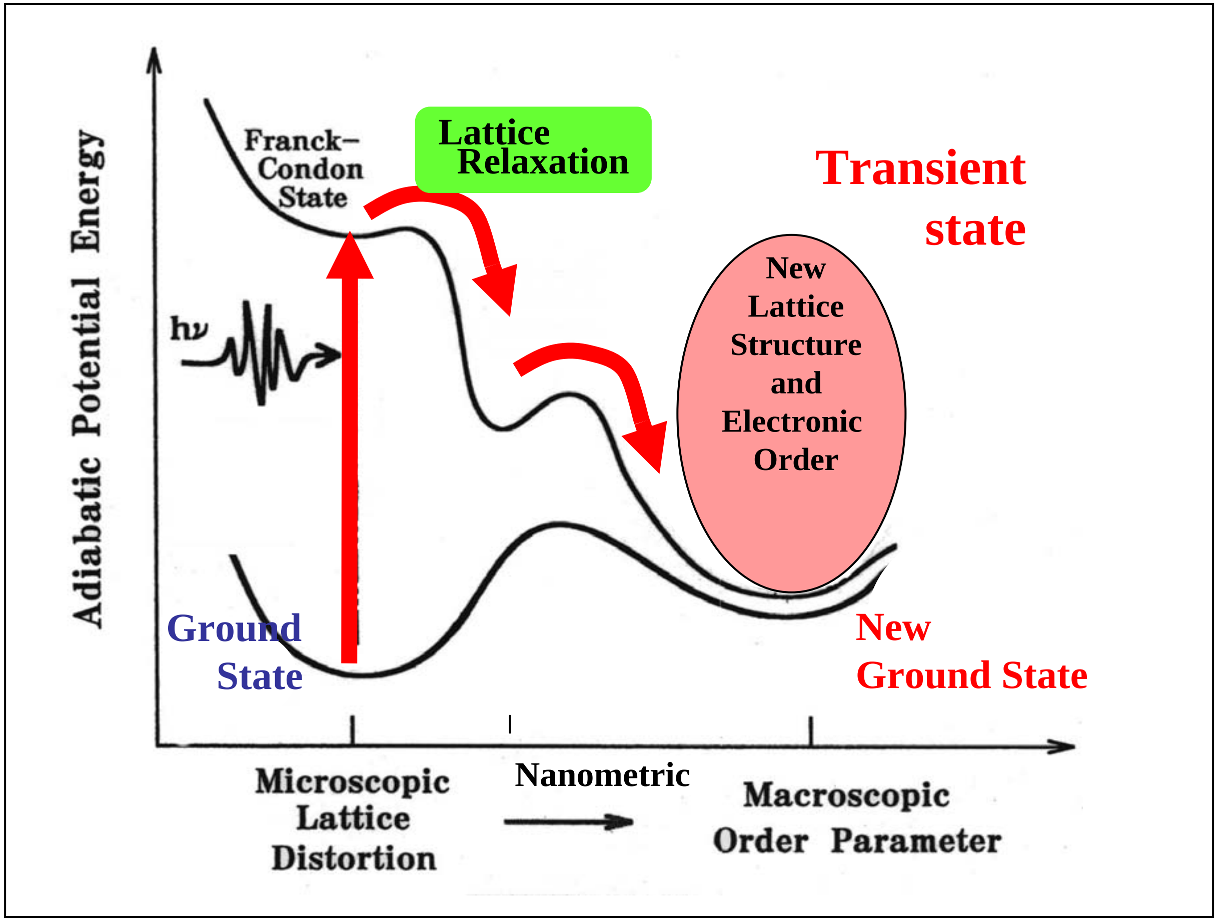
How do we prepare a system far from equilibrium?

- *Use of ultrashort laser pulses (fs, TW) to excite electronic excitation (≥1 ph/unit cell), photodoping, photochemical manipulation*
- *Use ultrsfast or rapid cooling to freeze the systems that are far from equilibrium*
- *Use of ultrashort high-amplitude strain pulses (shock waves) to develop dynamical "extreme conditions" and to coherently excite vibrational excitations*
- *Use of ultrashort duration several Tesla magnetic field pulses, for example, generated using ultra-relativistic electron bunches, to study the response of magnetic spins*

Etc. Etc. Etc………

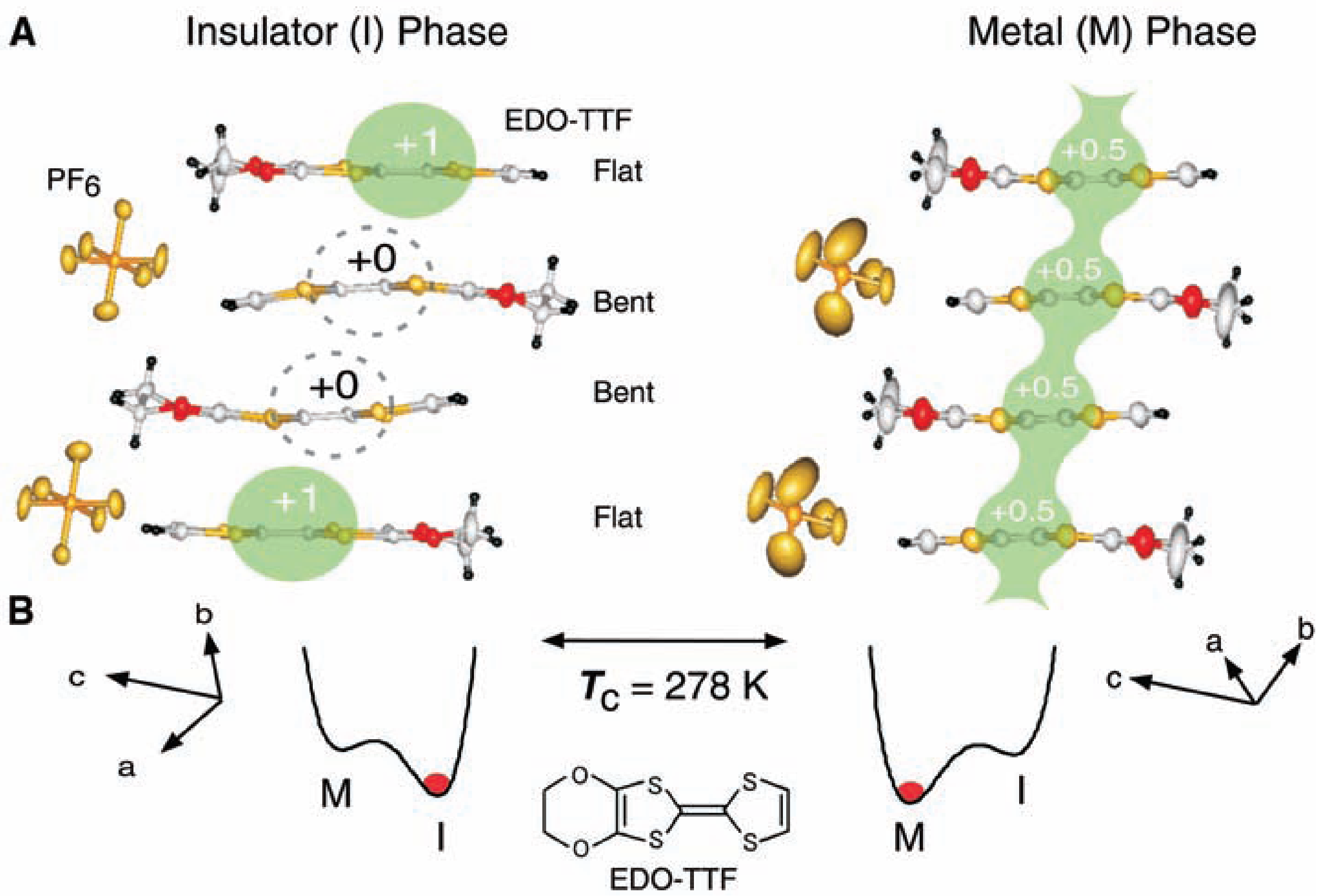
Photoinduced Solid State Phase Transitions

Far-from-Equilibrium and Multi-scale Processes in Solids



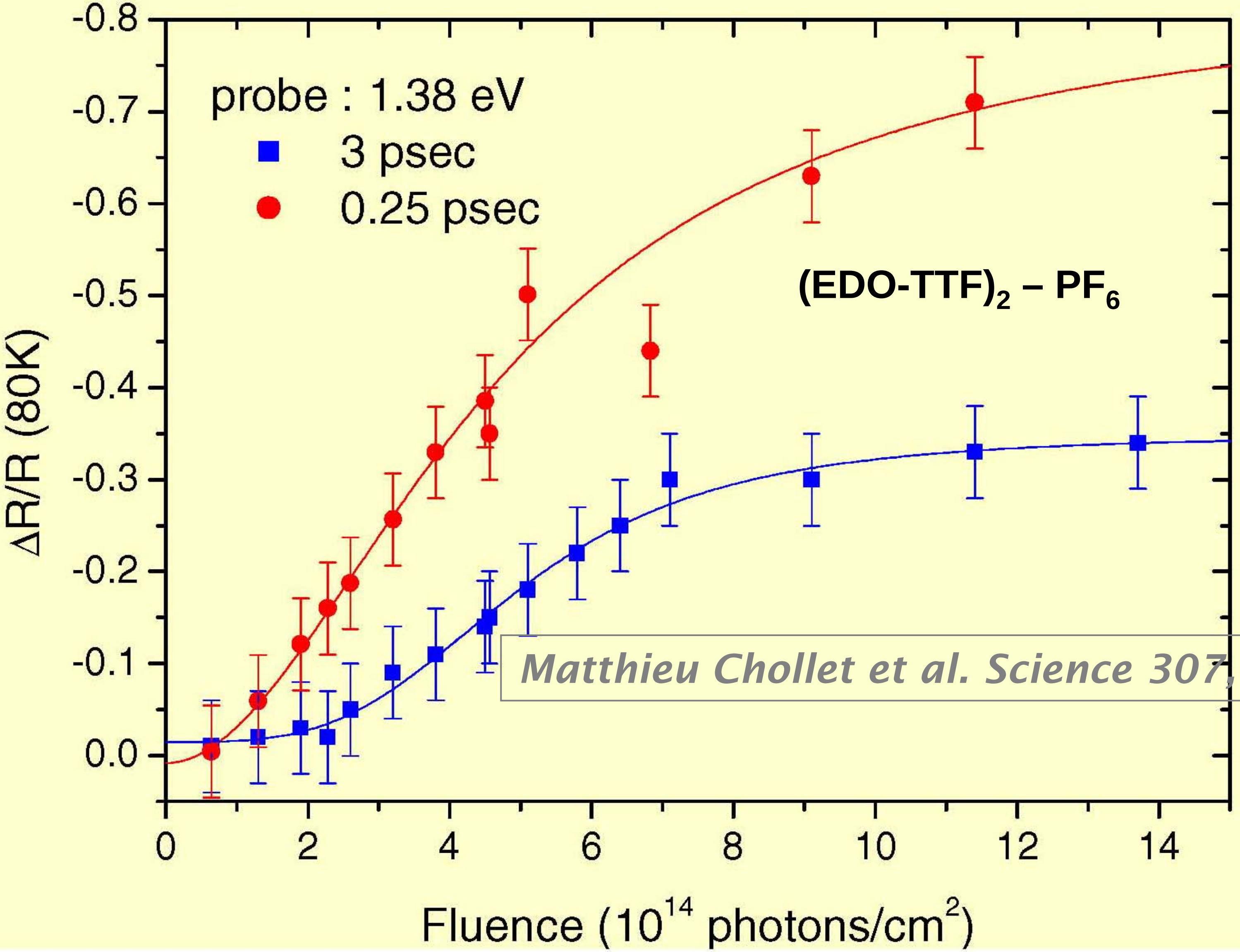
Energy Relaxation via Specialized Modes leading to Coherent State

Gigantic Photoresponse in Organic Salt $(EDO\text{-}TTF)_2PF_6$: Metal – Insulator Transition



Matthieu Chollet et al. Science 307, 86 (2005)

Nonlinear Response to Excitation Intensity

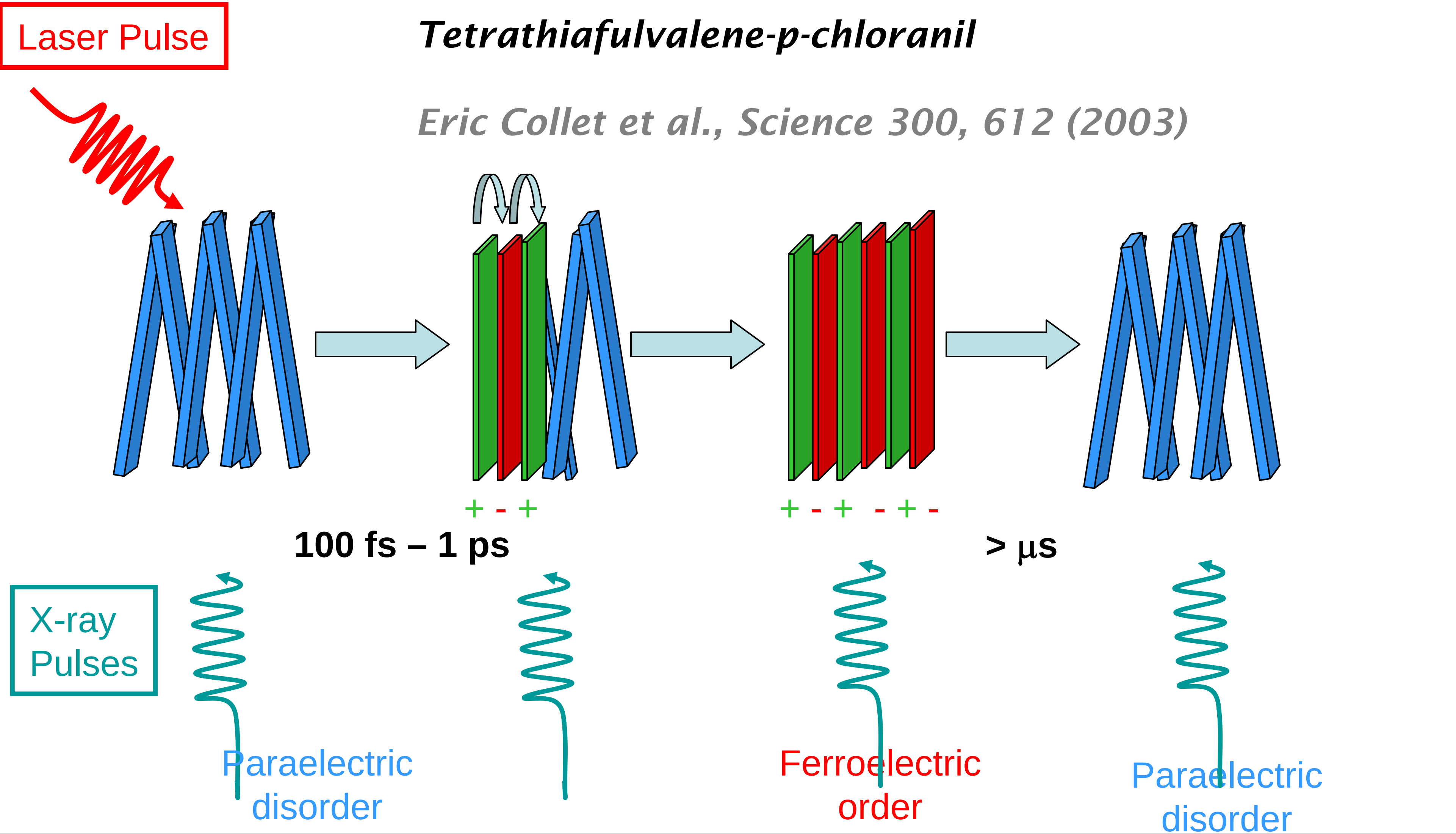


Matthieu Chollet et al. Science 307, 86 (2005)

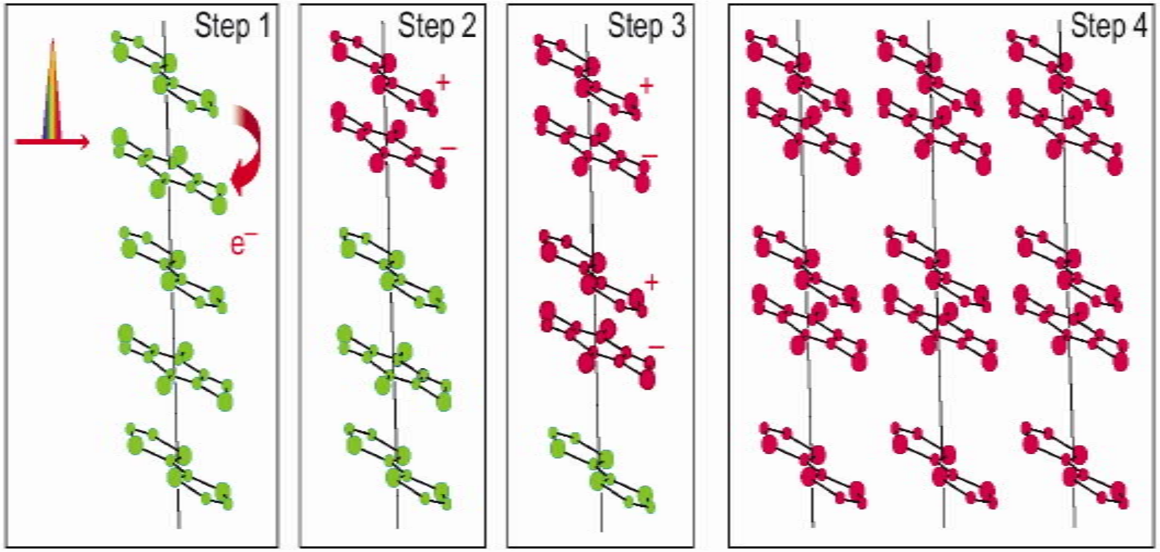
Laser-induced Ferroelectric Structural Order in an Organic Charge-Transfer Crystal

Tetrathiafulvalene-p-chloranil

Eric Collet et al., Science 300, 612 (2003)



Laser-induced Ferroelectric Structural Order in an Organic Charge-Transfer Crystal



100 fs – 1 ps   100 ps

Step 1. Laser pulse excites a molecule

Step 2. Intermolecular charge transfer occurs accompanied by a lattice relaxation, i.e. a dimerization process trapping the excitation

Step 3. Cooperative order take place with the self-multiplication of the excited molecule in the stack

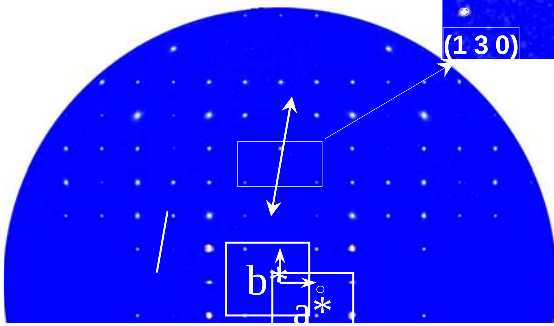
Step 4. Interstack interactions lead to the 3D ordering of the dimers, with a photon efficiency so high that one photon transforms a few hundred molecules

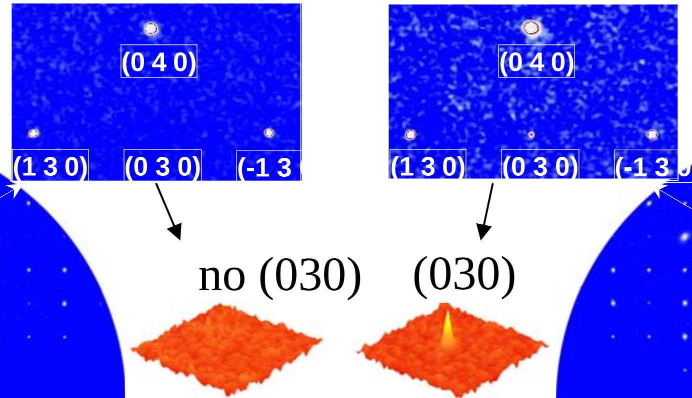
Eric Collet et al., Science 300, 612 (2003)

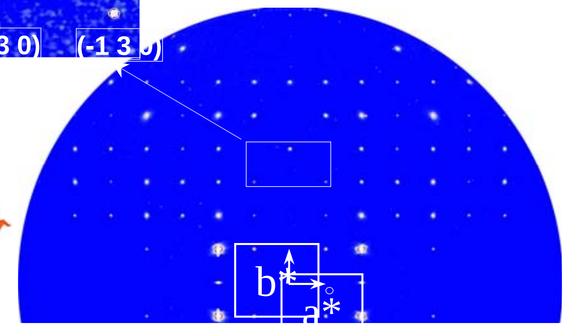
Laser-induced Ferroelectric Structural Order in an Organic Charge-Transfer Crystal

2 ns before laser irradiation

1 ns after laser irradiation







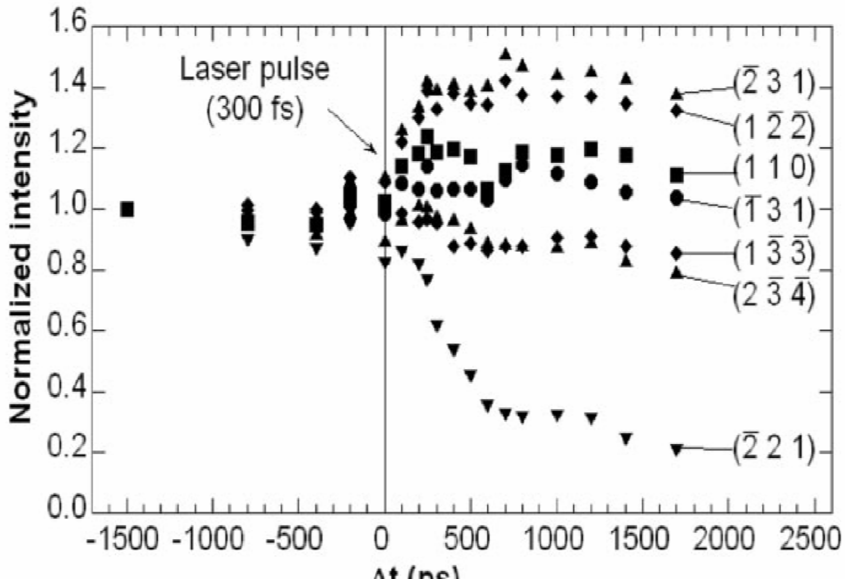
Paraelectric phase

Photo-induced ferroelectric order

Space group P2$_1$/n

(0 k 0) : k = 2n+1 absent

(h 0 l) : h+l = 2n+1 absent



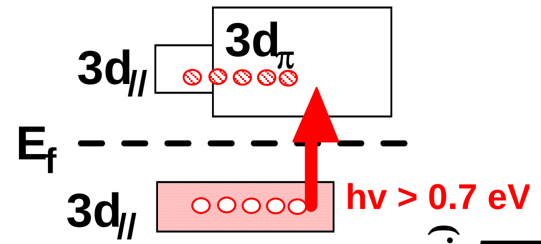
Space group Pn

(0 k 0) : k = 2n+1 present

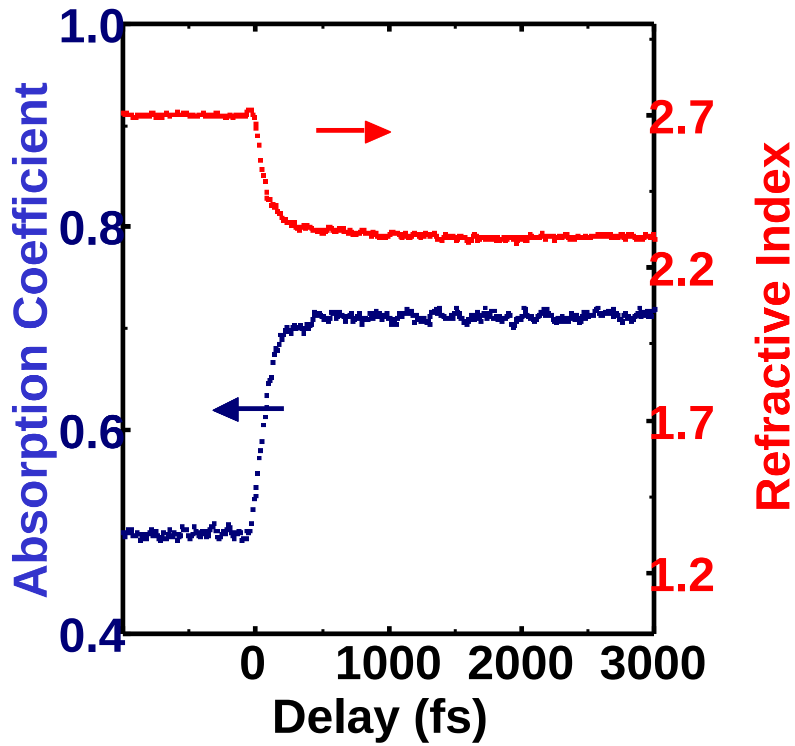
(h 0 l) : h+l = 2n+1 absent

E. Collet et al, Science 300 612 (2003

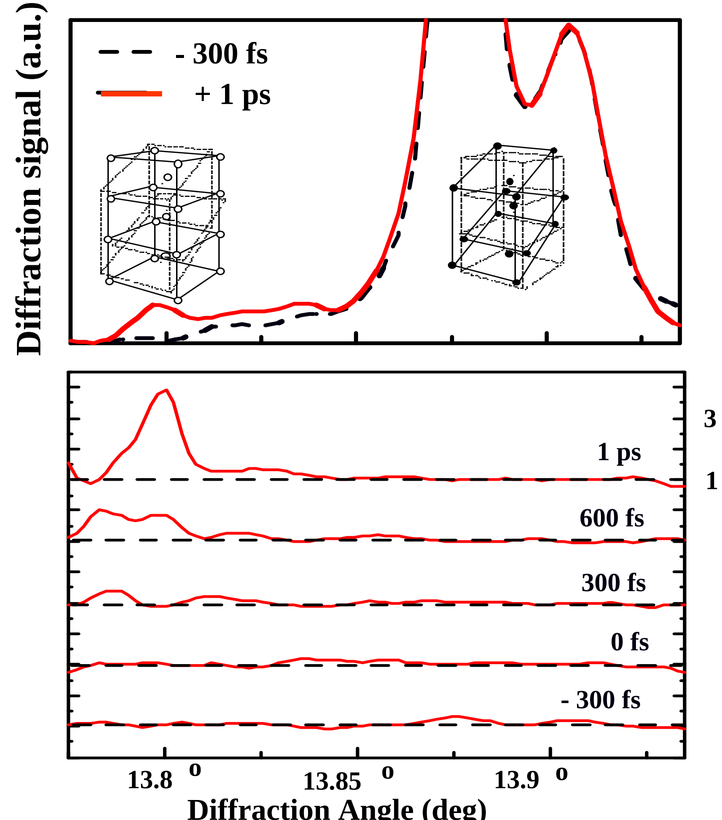
Structural & Electronic Transition in VO_2



Optical: 100 fs

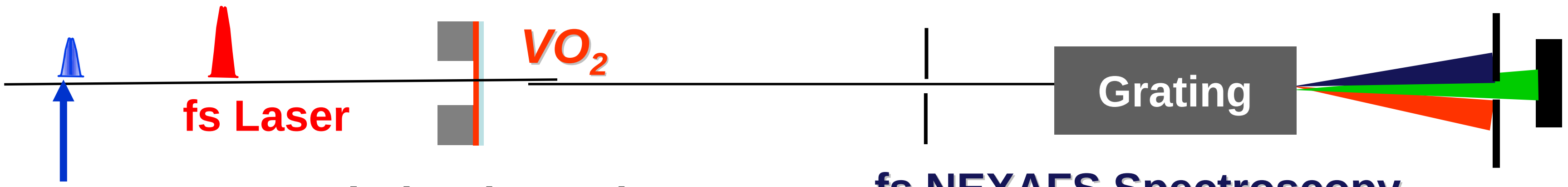


X-rays: 300 fs



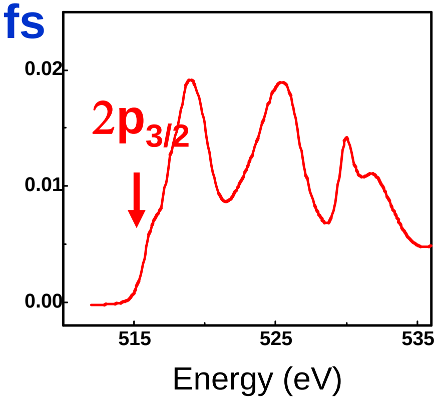
Cavalleri et al. *Phys. Rev. Lett.* 87, 237401 (2001)

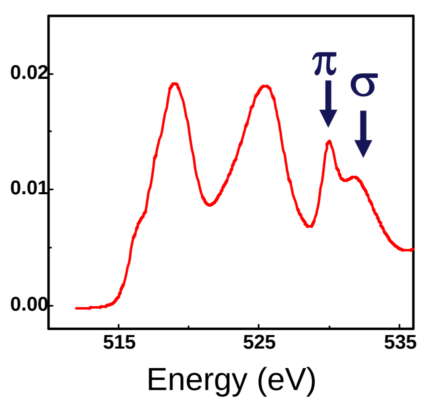
fs Oxygen Edge Spectroscopy: p bands



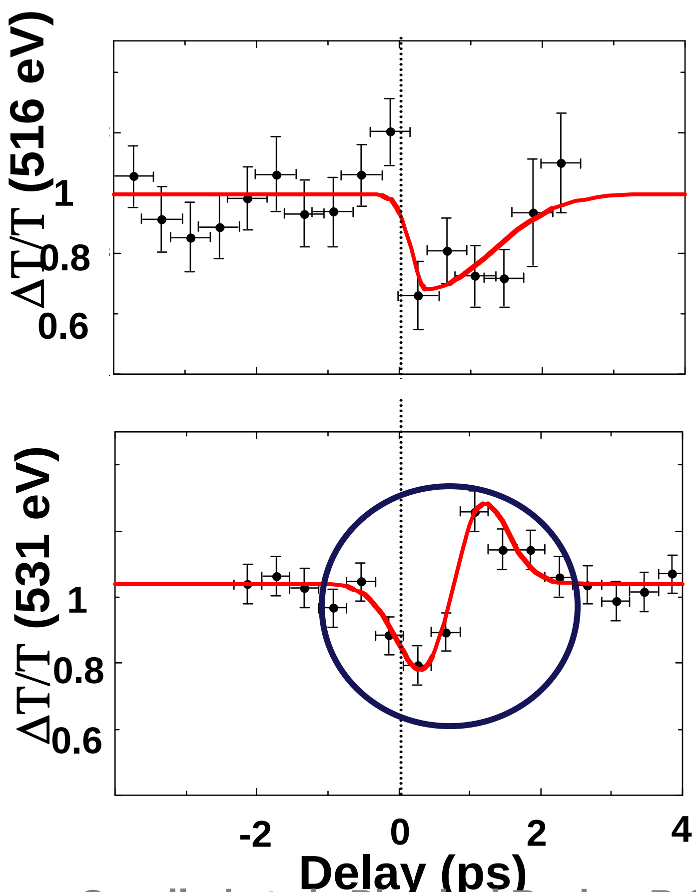
ALS Sliced Pulse: 100 fs

Resolution (0.1 eV)



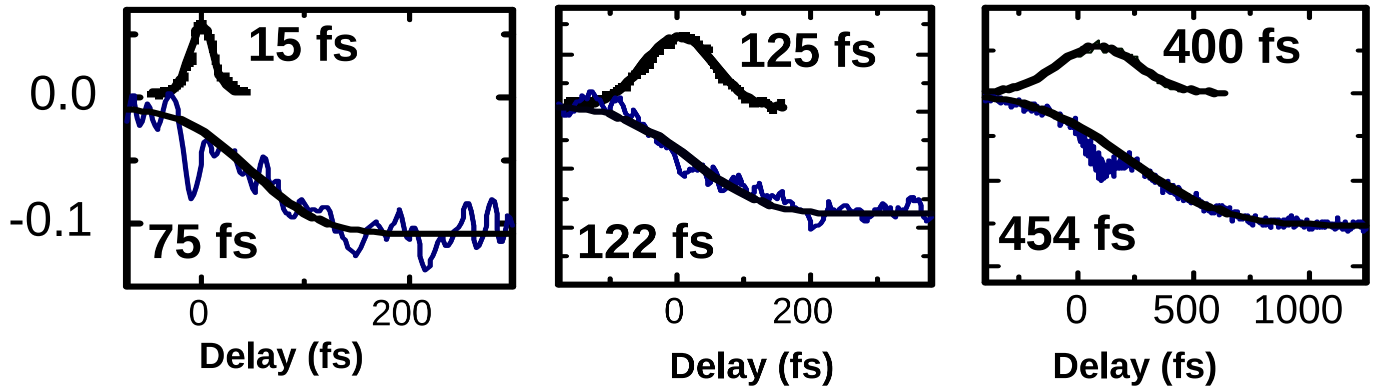


fs NEXAFS Spectroscopy

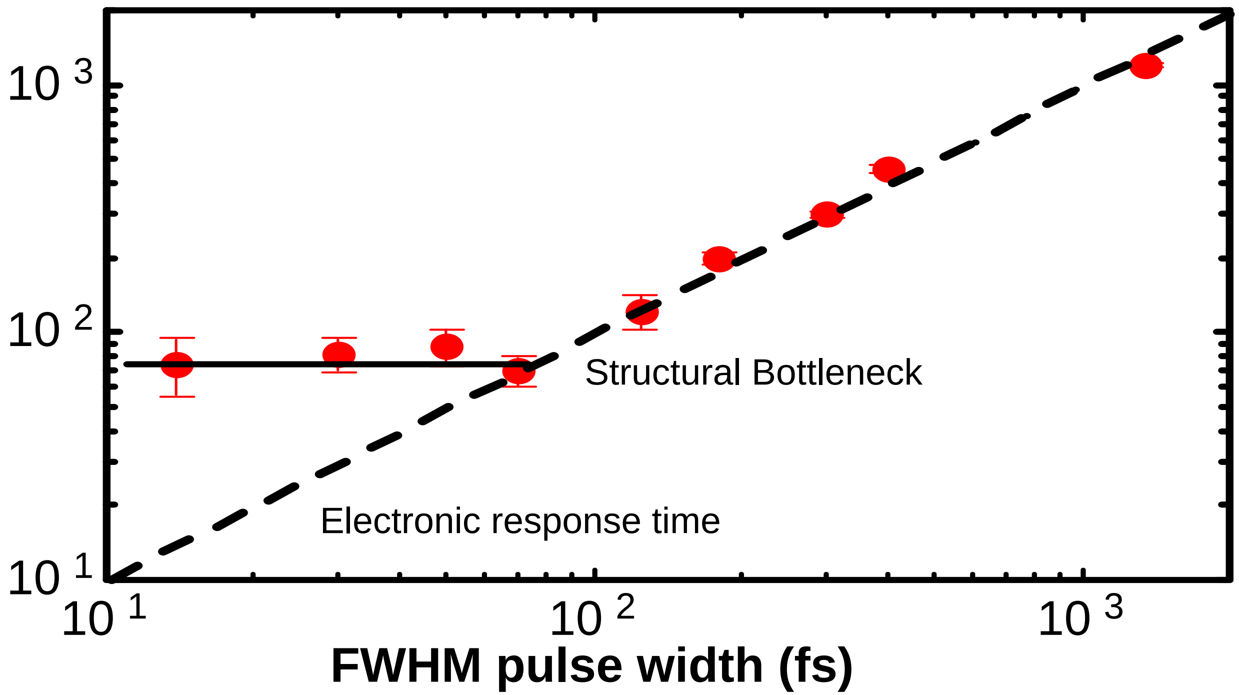


Cavalleri et al., Physical Review B 69, 153106 (2004)

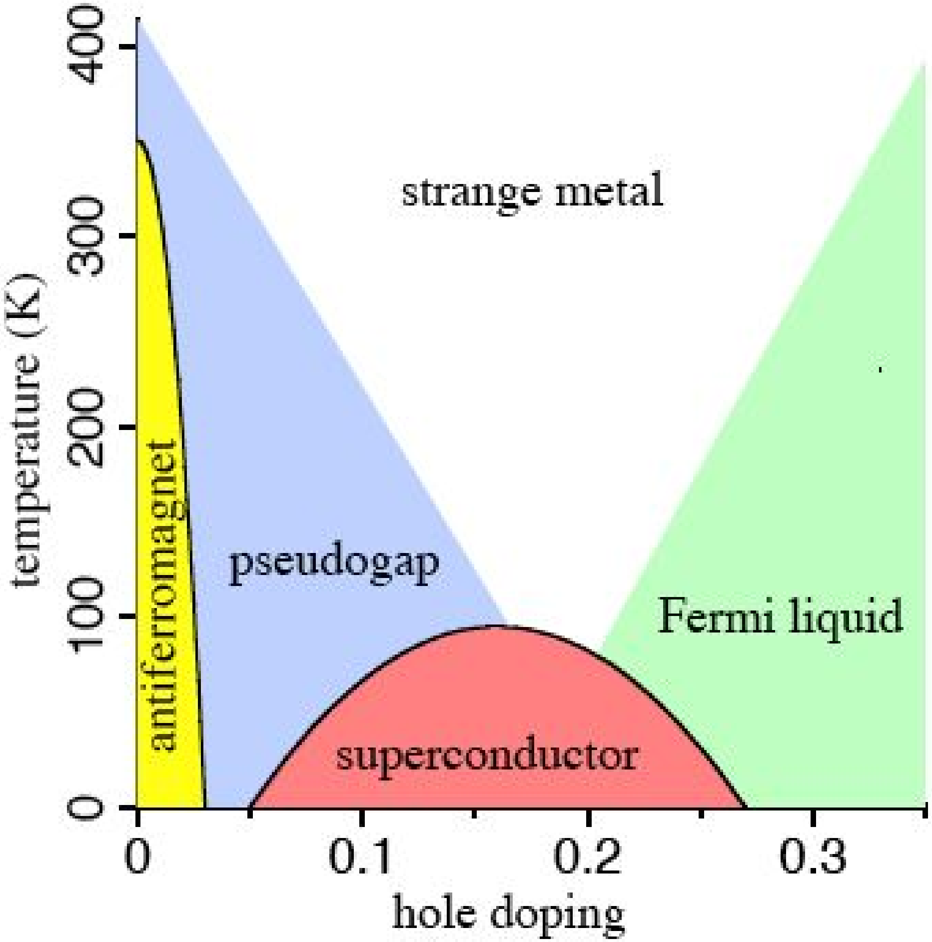
Phase Transition Time in VO_2: 75 fs



Cavalleri et al., Physical Review B 69, 153106 (2004)



Phase Diagram of the high Tc superconductors

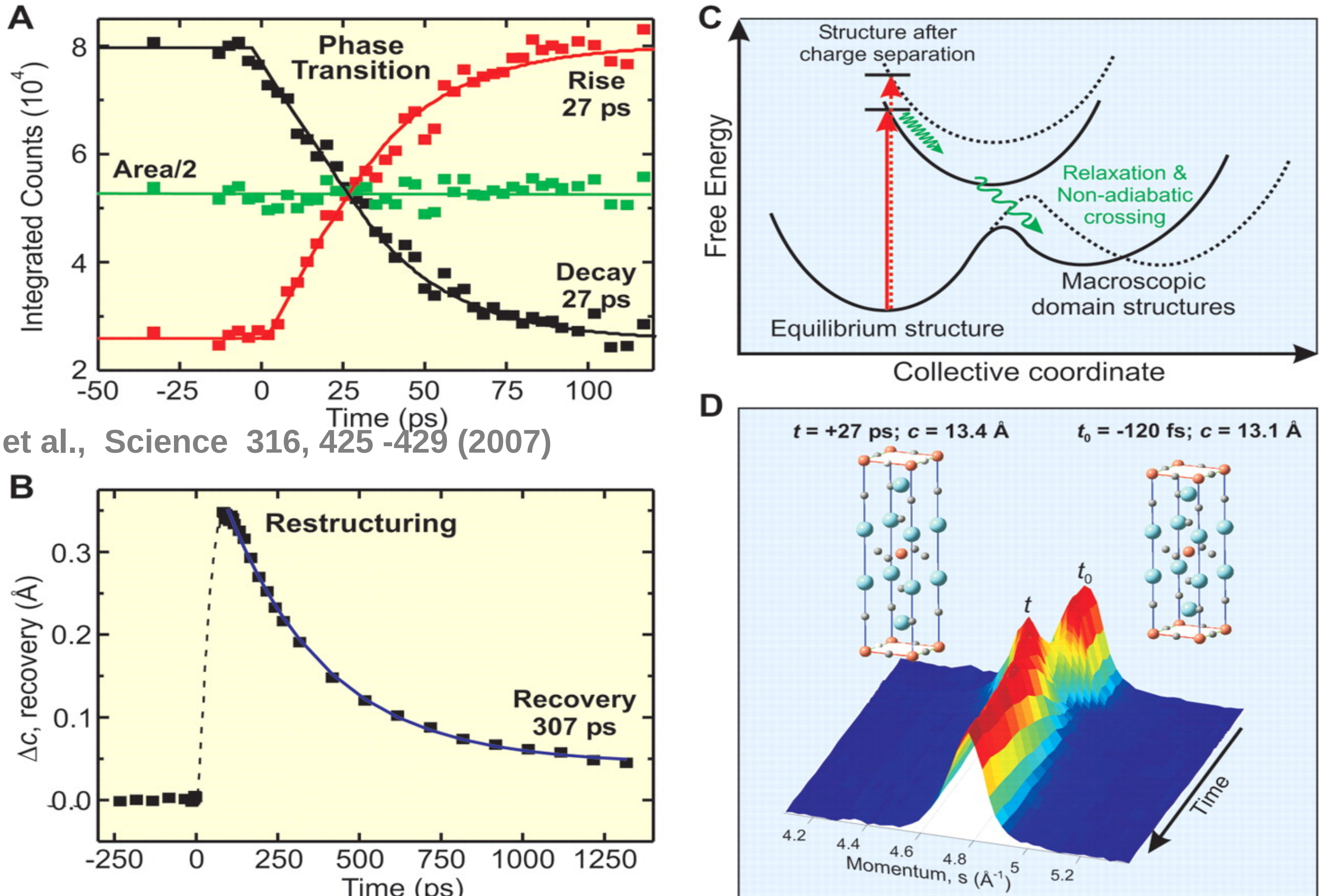


- *Coupled CuO_2 layers*
- *Doping the AFM insulator -> SC*
- *Nonmonotonic Tc versus doping*
- *Maximum Tc ~ 50-150 K*

Control ?

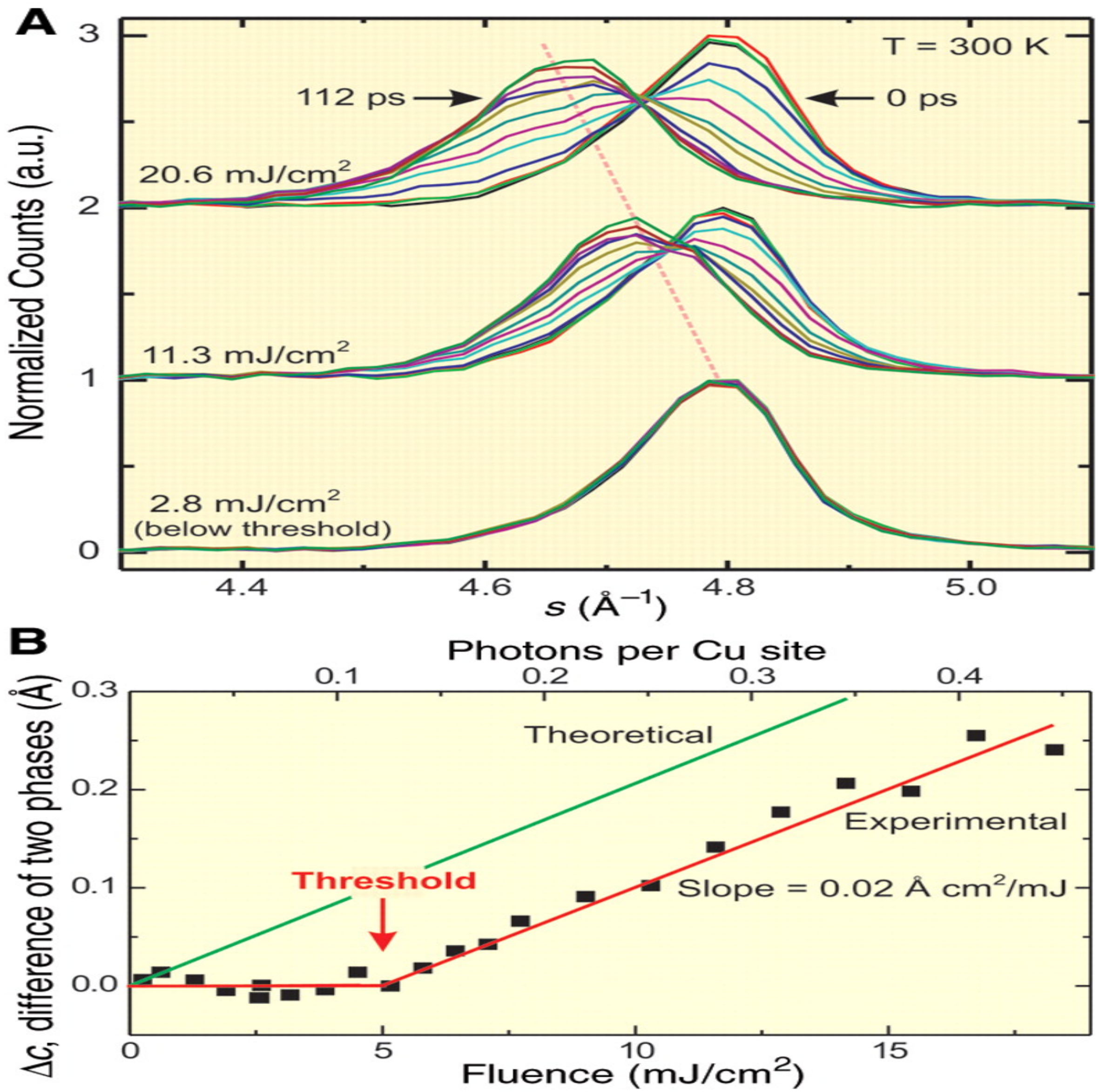
Can we replace 'chemical doping' with 'photodoping' to realize superconducting phase?

Far-From Equilibrium Phase Transitions in Cuprates Observed by Ultrafast Electron Crystallography
Time scales and trajectories of structural changes



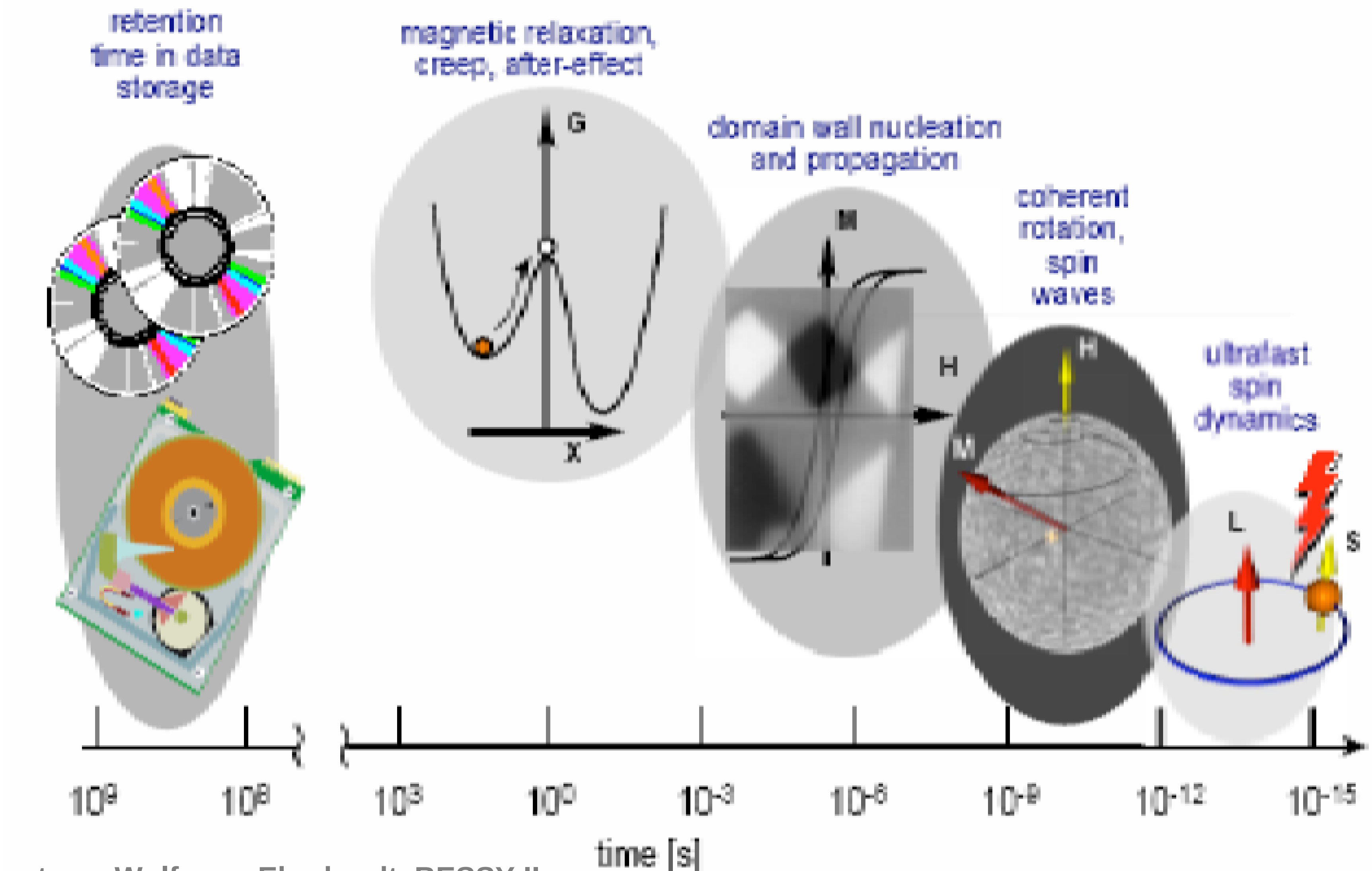
N. Gedik et al., Science 316, 425 -429 (2007)

Threshold behavior and fluence dependence



N. Gedik et al., Science 316, 425 -429 (2007)

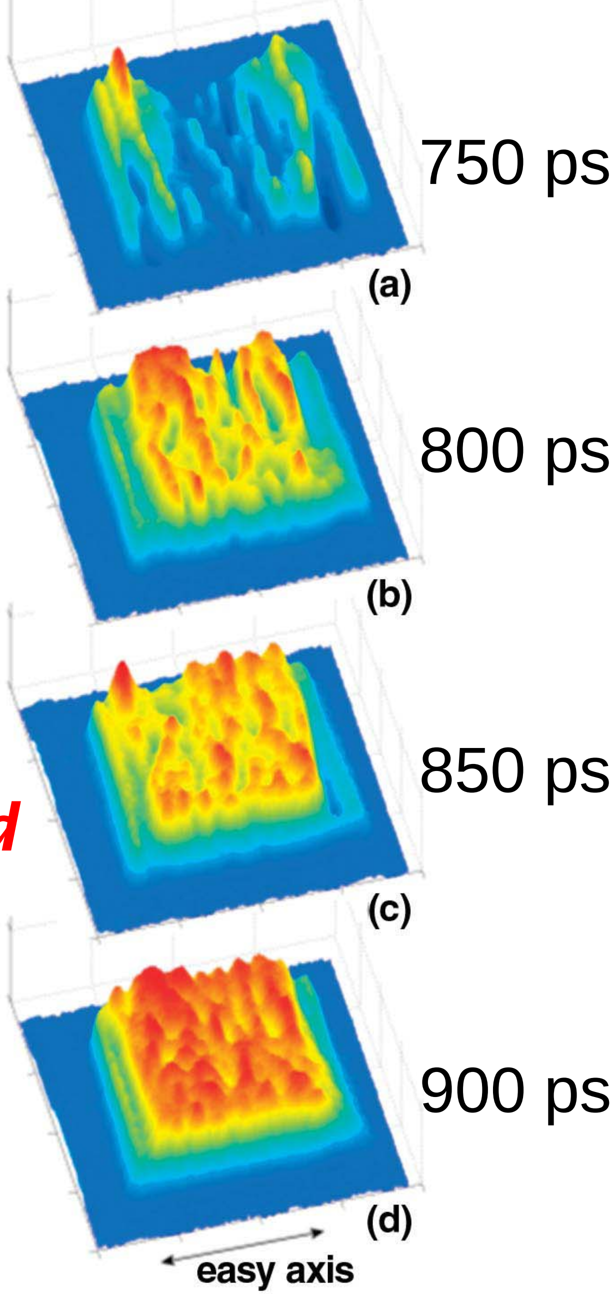
Magnetic Switching Timescales



Courtesy: Wolfgang Eberhardt, BESSY II

Far-from-equilibrium picosecond time-resolved evolution of a nonequilibrium magnetic domain pattern of mesoscopic $Ni_{80}Fe_{20}$ thin-film

Choi et al. PRL **95,** 237211 (2005)

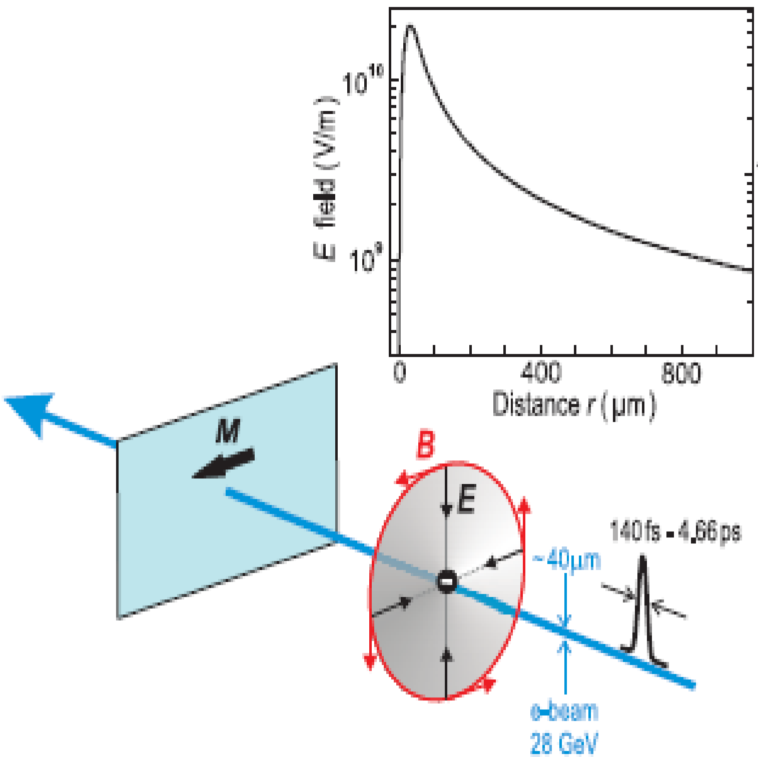


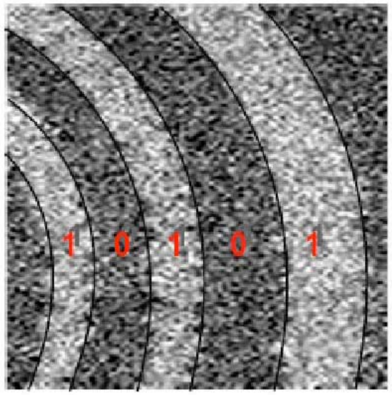
***Spatiotemporally** resolved domain images representing the easy-axis magnetization component at selected time points after the onset of a magnetic switching field of 0.24 ns rise time (10%–90%) and 24 kA/m.*

Pump: Relativistic GeV Electron Bunch (Generated Pulse E-M Field ~10^9 V/m)

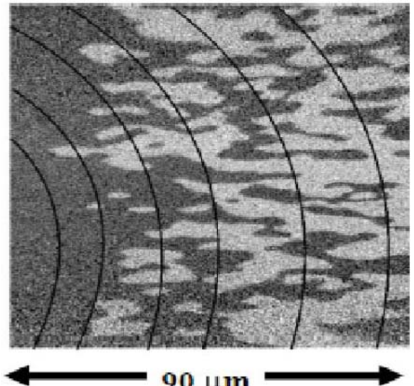
Courtesy: Jo Stohr, SLAC

Magnetization Pattern ferromagnetic $Fe_{30}Co_{79}$ film





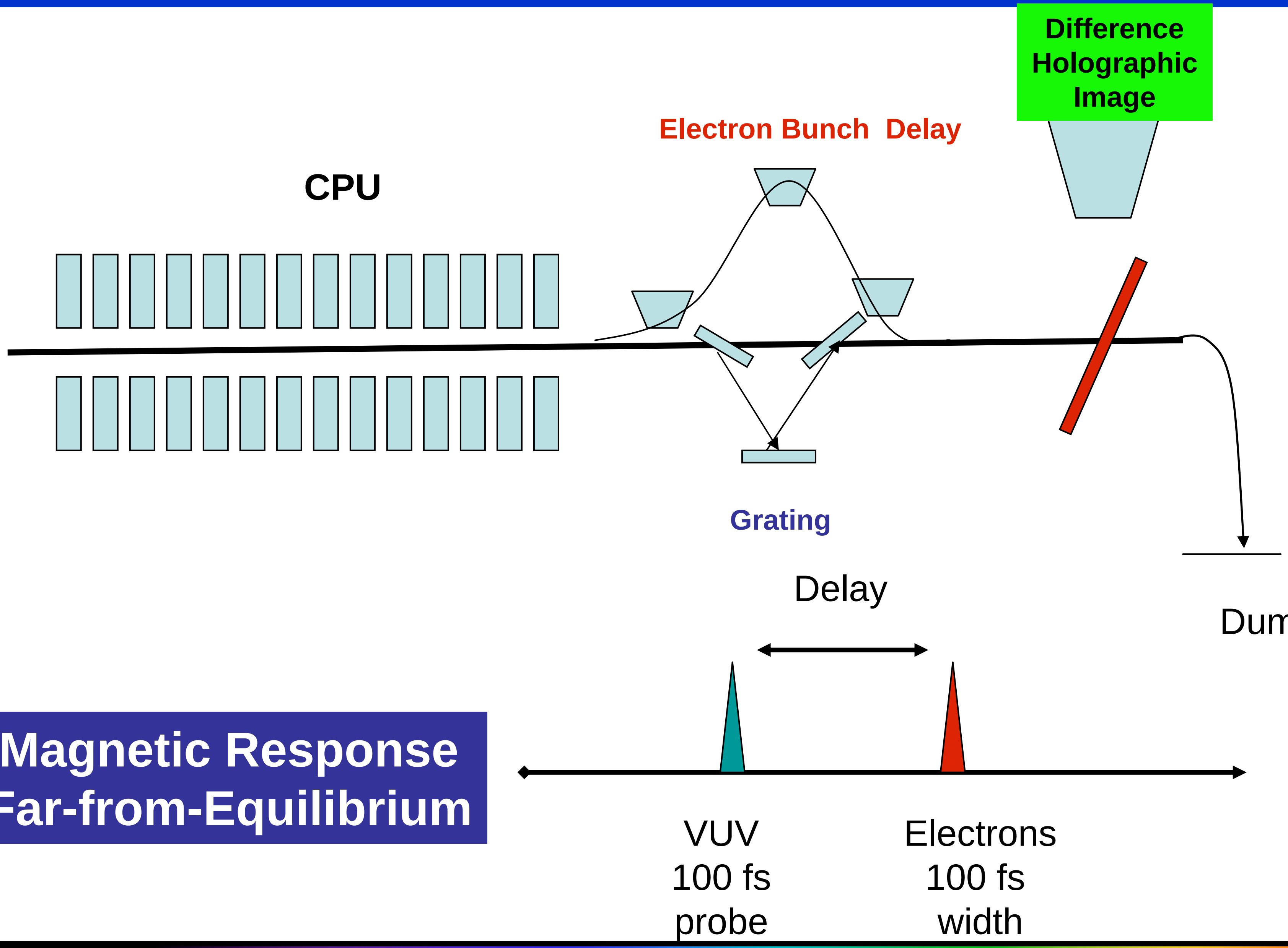
Pulse: 4ps



Pulse: 140 fs

C. Stamm, et al. Phys. Rev. Lett. 94, 197603 (2005)

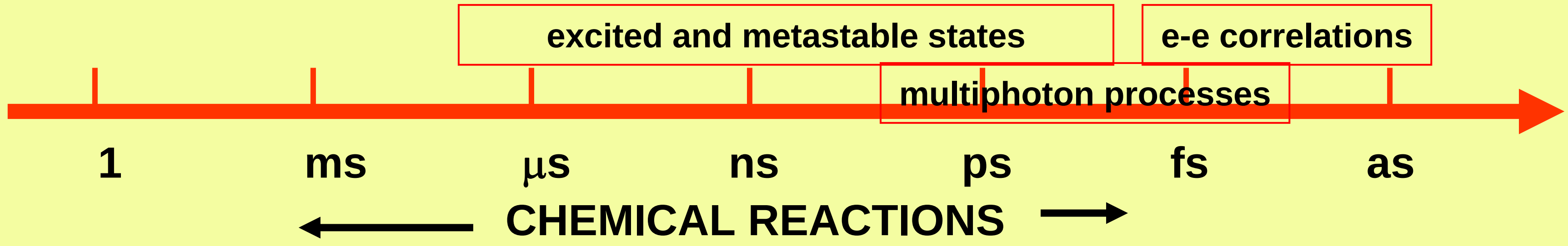
GeV Electron Pump – VUV Circularly-Polarized Probe



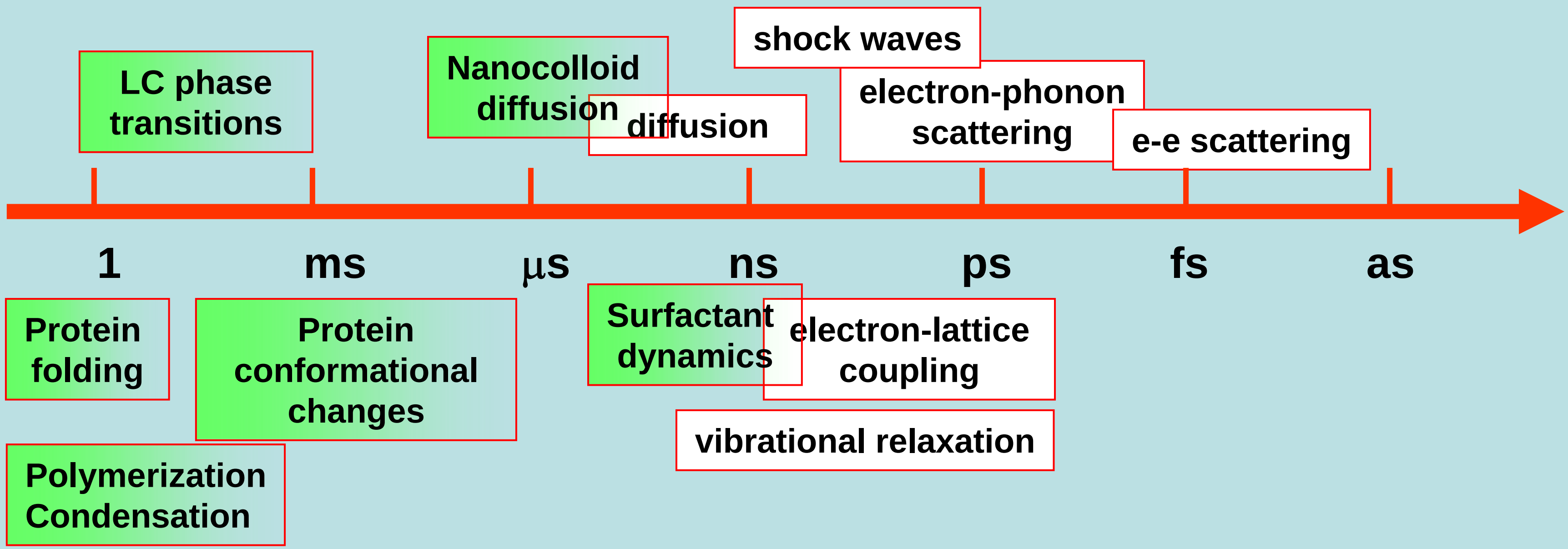
Magnetic Response Far-from-Equilibrium

Characteristic Times of Various Phenomena

Atomic, Molecular and Chemical Physics:



Hard and Soft Condensed Matter Physics:



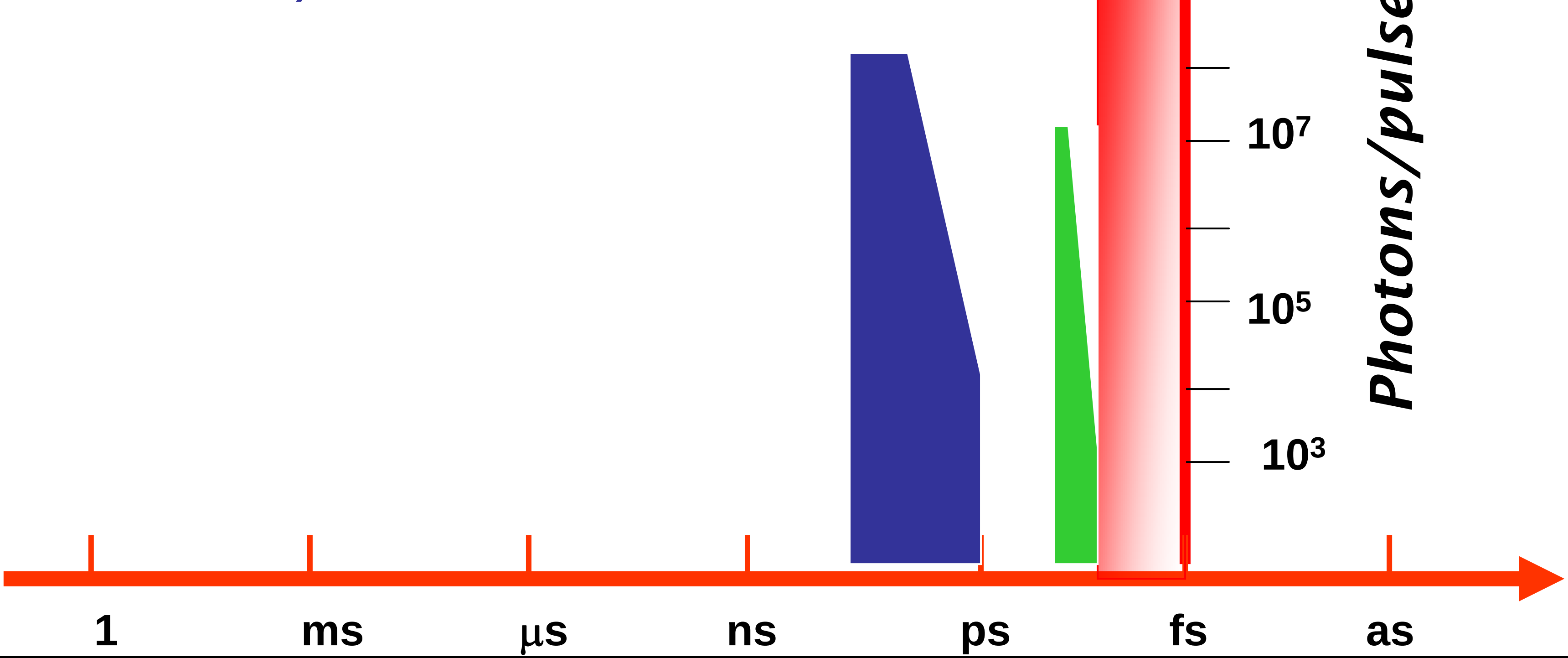
Typical Pulse Resolution and Rep Rate (Hz)

FEL $10^2 - 10^6$ Hz (~ ms – μs)

ERL 1.3 10^9 Hz (~ ns)

SR (0.3 – 100) X 10^6 Hz (~ 10 ns – 4 μs)

Sliced: laser, crab



Combining Many fs-ps Probes with nm spatial resolution

- *Optical reflectivity and absorption*
- *Impulsive Raman*
- *fs-ps ARPES*
- *fs-ps NEXAFS in the soft and hard x-rays*
- *fs-ps Diffraction*
- *fs-ps EXAFS*
- *fs-ps Coherent Imaging*
- *fs-ps Coherent Diffraction*
- *ps-ms SAXS*
- *Etc. Etc……….*

Summary: Studies Far-from-Equilibrium

- ***Light sources, present and future, will provide unprecedented opportunity to understand far-from- equilibrium processes in nature, i.e., bio-, chemical-, soft- and hard-matter and their interfaces.***

- ***Deterministic control of matter using extreme excitation pulses is not only feasible, but can be investigated using future and present light sources.***
  E.g.,quantum control with coherent light fields

Summary: Studies Far-from-Equilibrium

- ***Time-resolved* (fs-ps) *<u>along with</u> spatially-resolved* (nm) maps to trace the pathways, processes and *self-organization that follow the bifurcation are essential.***

. ***Support with modeling through solutions to non-linear differential equations to describe multi-scale dynamics can be accomplished using a combined molecular-dynamics and Monte Carlo approach to describe atom and carrier dynamics, respectively. These are peta-scale computational problems.***

Thank You All……