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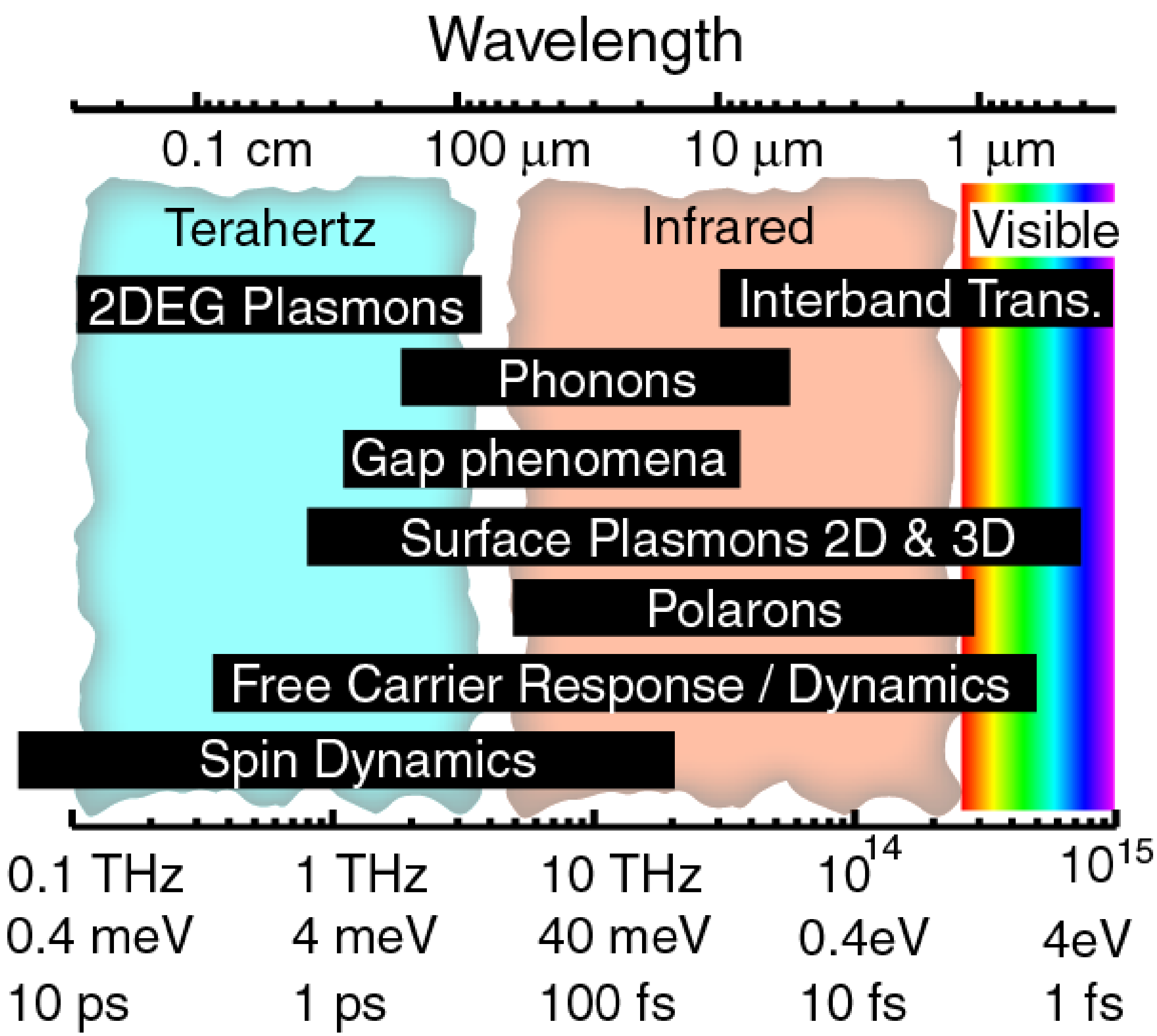
Scientific opportunities in solid-state/condensed matter physics using high-repetition rate, ultra-short pulse, hard x-ray sources

Joel D. Brock
Cornell University

Outline

- Scientific Drivers for SPX: Condensed Matter/Solid State
- Varieties of time-resolved x-ray studies
  - Spectroscopies
  - Diffuse X-Ray Scattering
  - X-Ray Reflectivity
  - X-Ray Diffraction
  - Coincidence Measurements
- Synergy and complimentary of SPX, ERL, and XFEL sources
- X-ray optics, detectors, and pump lasers: considerations

Why investigate dynamics from 0.001 to 5 eV?



(Toni Taylor - 2006)

Why use ultrafast optics to study complex materials?

~10-100 fs optical pulses are short enough to resolve processes at the fundamental timescales of electronic and nuclear motion allowing for the temporal discrimination of different dynamics.

Time

electron-electron (fs) electron-phonon (ps) spin-lattice (many ps)

Understanding the interplay between atomic and electronic structure

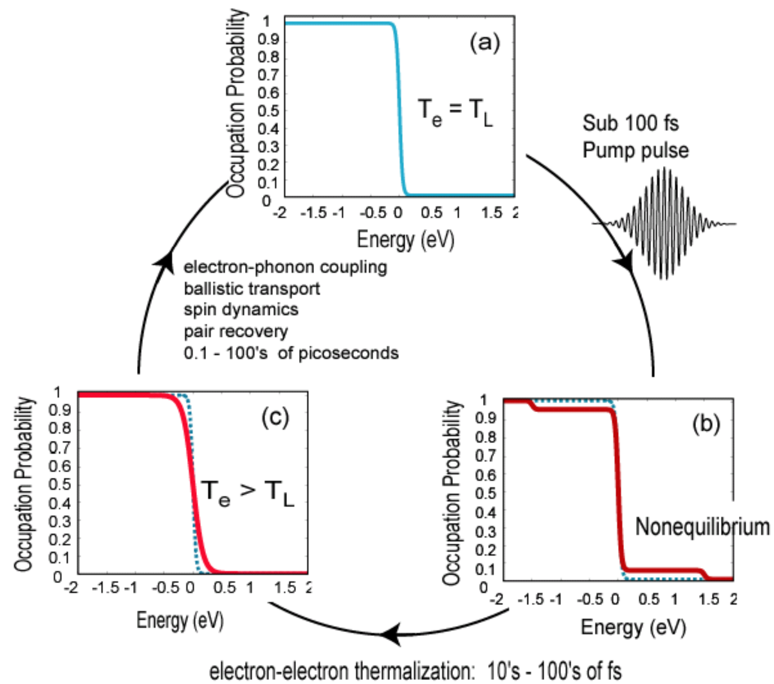
• Beyond single-electron band structure model: correlated systems (charge, spin, orbit, lattice)
• Beyond simple adiabatic potential energy surfaces

Understanding the nature of quasiparticles

• Formation dynamics, scattering processes, relaxation channels and dynamics

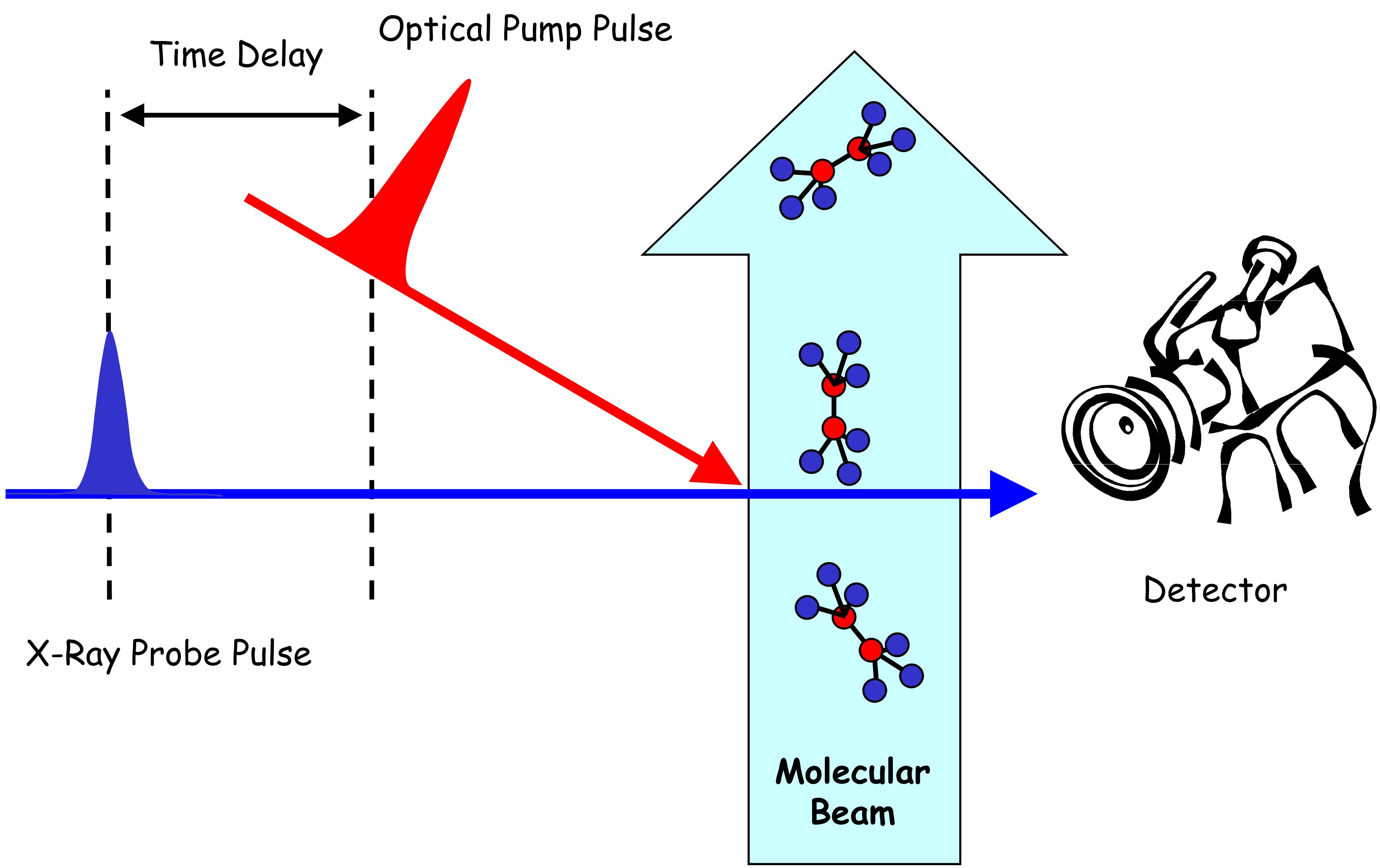
Creating new states of matter

Photoinduced phase transitions—fast switching, probing dynamics where the order parameter has been perturbed, creating nonthermally accessible phases.



(Toni Taylor - 2006)

Pump/Probe Measurements

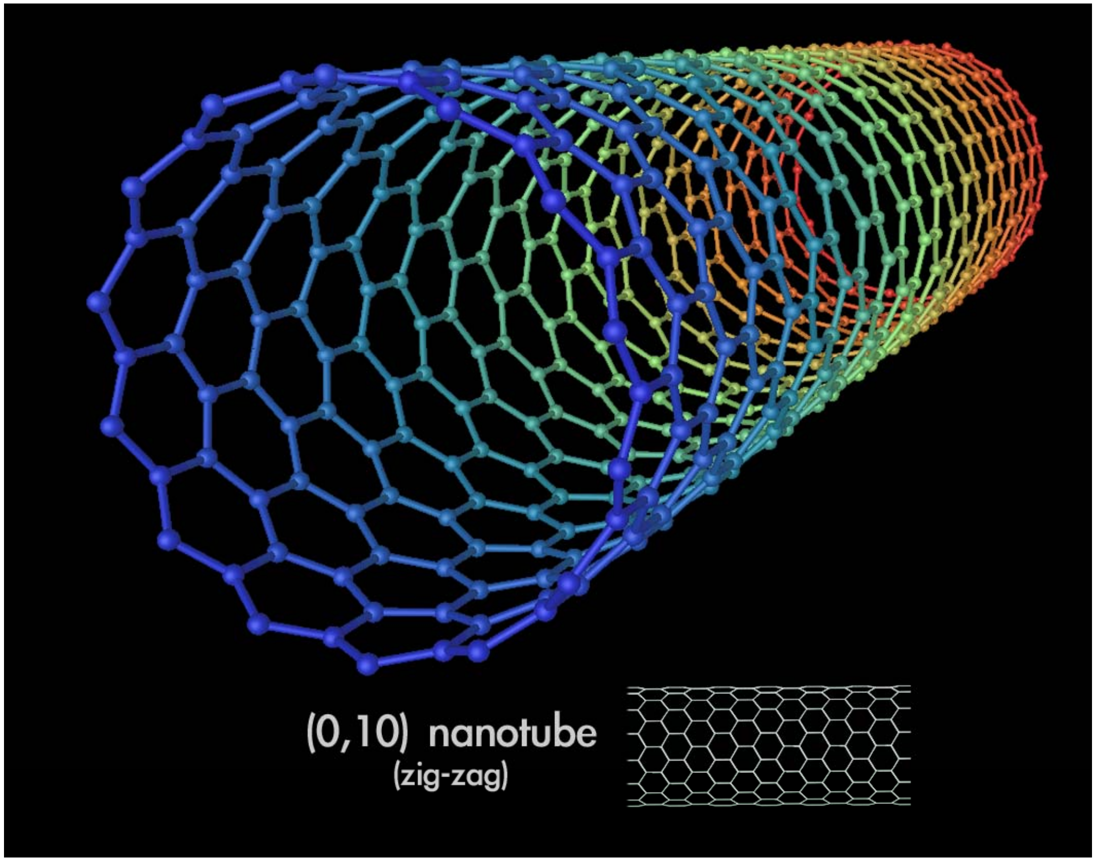


Pump/Probe – what are a high repetition rate source's advantages?

High repetition rate lends itself to studies of the linear response of systems to a perturbation.

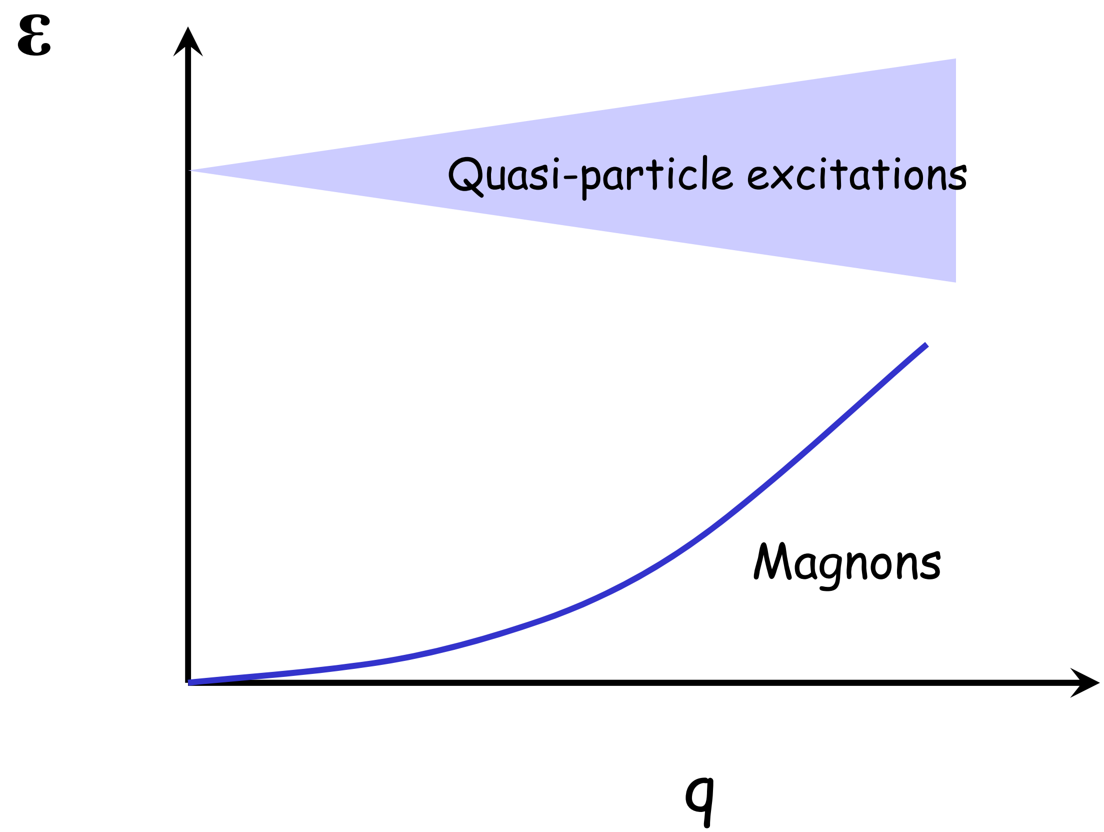
	Physics	
	Linear	Non-linear
Pump	weak, high rep rate	strong, slow rep rate
Probe	weak	weak

Goal is to deposit as little energy as possible so that the lifetime of the elementary excitation determines the repetition rate rather than thermal equilibration.



E.g.,. We want to study a <u>single</u> exciton in a single-walled carbon nanotube

Elementary excitations from ground state of correlated electron system.



Spectrum of elementary excitations of the ferromagnetic electron gas.

What do we need for pump?

- Want to excite only one quanta of the elementary excitation at time.
  - Each pump pulse excites one exciton
  - Pump pulses separated by ~10 decay times

Elementary excitations: relevant time-scales

- Collective modes
  - Low energy – typical energy gap ≤ 50 meV

$$\tau_{period} = \frac{4.135\times10^{-15} eV\cdot s}{0.05eV} \approx 100\ fs$$

  - Assuming 10xQ ~ 10^5 gives lifetime of excitation

$$\tau_{decay} \sim 10\ ns \rightarrow 100 Mhz$$

(Free running Ti:Al_2O_3 laser runs at about 80 MHz)

- Quasi-particles
  - Higher energy, faster

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VOLUME 83, NUMBER 4 PHYSICAL REVIEW LETTERS 26 JULY 1999

Single Particle and Collective Excitations in the One-Dimensional Charge Density Wave Solid $K_{0.3}MoO_3$ Probed in Real Time by Femtosecond Spectroscopy

J. Demsar,[1] K. Biljaković,[2] and D. Mihailovic[1]

[1]*Solid State Physics Department, "Jozef Stefan" Institute, Jamova 39, 1001 Ljubljana, Slovenia*

[2]*Institute for Physics, Bijenička 46, HR-10000 Zagreb, Croatia*

(Received 8 March 1999)

Ultrafast transient reflectivity changes caused by collective and single particle excitations in the quasi-one-dimensional charge-density wave (CDW) semiconductor $K_{0.3}MoO_3$ are investigated with optical pump-probe spectroscopy. The temperature dependence of nonequilibrium single particle excitations across the CDW gap and their recombination dynamics are reported for the first time. In addition, amplitude mode reflectivity oscillations are observed in real time. A T-dependent overdamped response is also observed which is attributed to relaxation of the phason mode.

PACS numbers: 71.45.Lr, 72.15.Nj, 78.47.+p

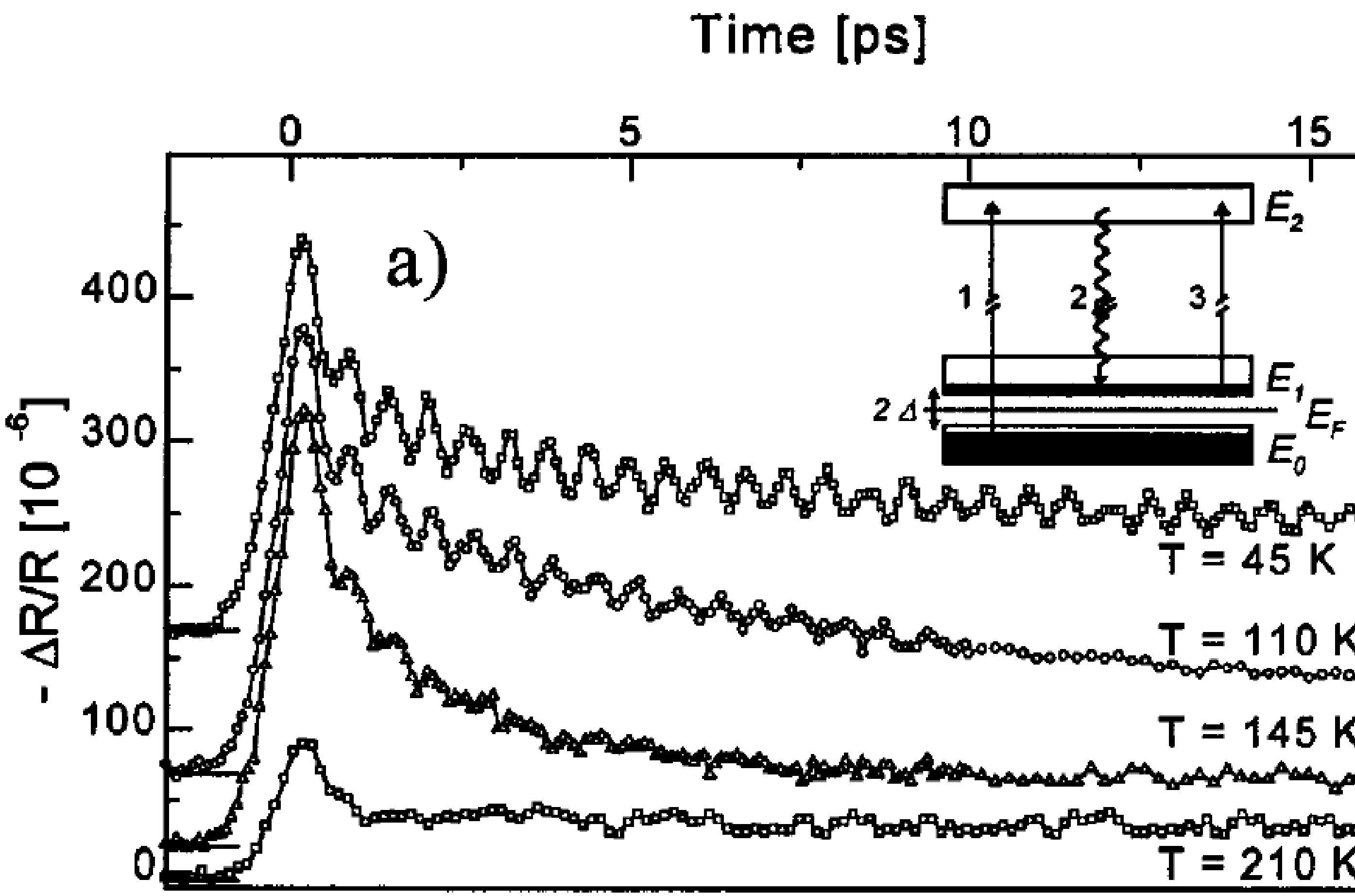
Relaxation time of photogenerated quasi-particles back to condensate: < 0.5 ps

Phason decay time ~10 ps

Frequency of amplitude mode ~1.7 THz

Optical pulse = 100 fs

Raw Data


Time [ps]
0
5
10
15
a)
400
300
200
100
0
- ΔR/R [10 -6]
E2
1
2
3
E1
2 Δ
EF
E0
T = 45 K
T = 110 K
T = 145 K
T = 210 K

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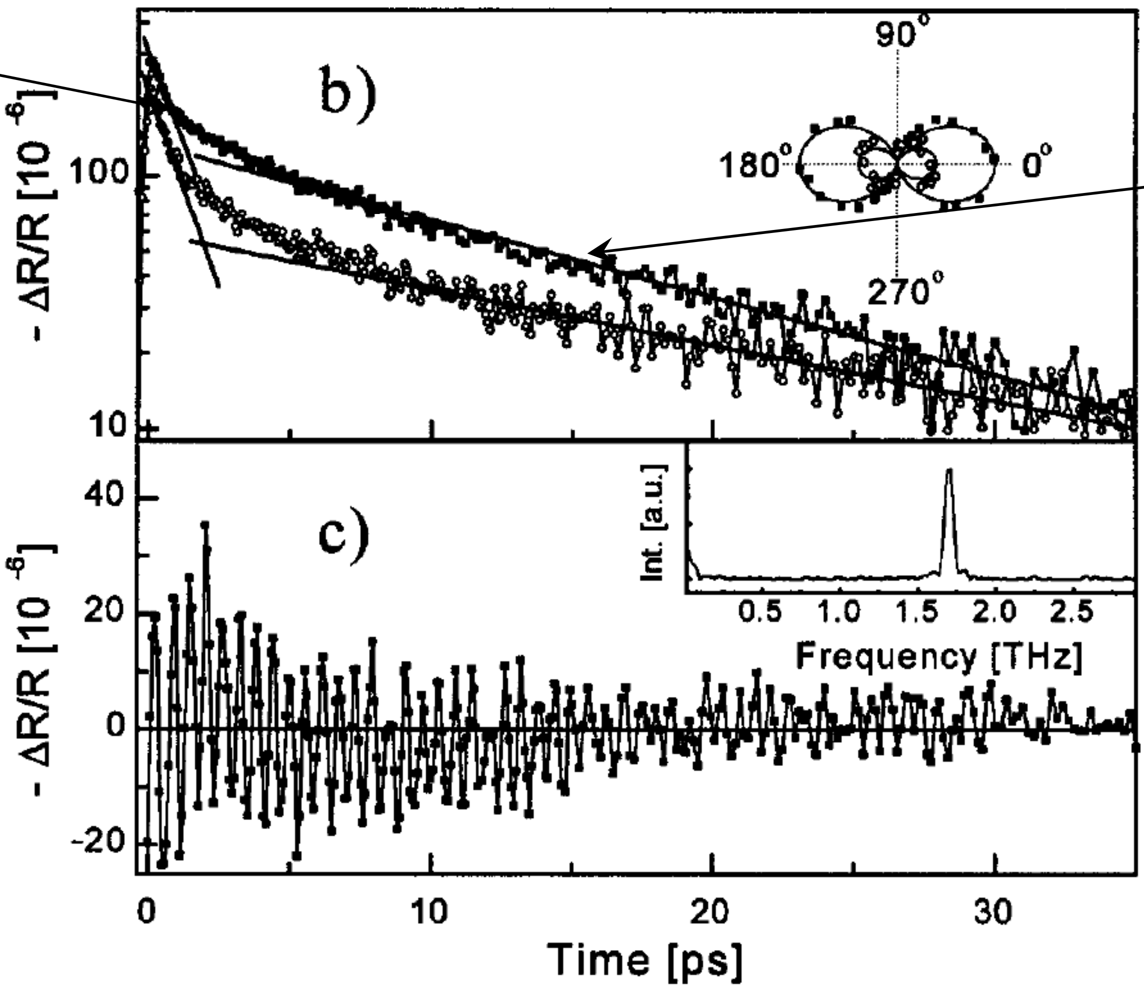
Reduced Data

$\tau_s \sim 0.5ps$

$\tau_p \sim 10ps$

$\nu_A \sim 1.7THz$

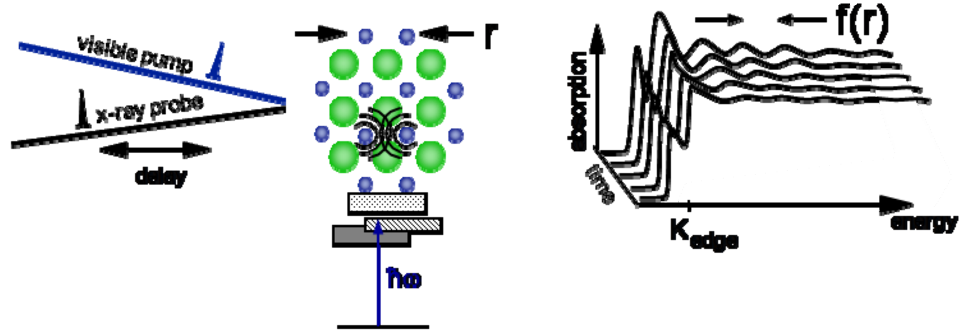


Ultrafast X-ray Science

Time-resolved x-ray spectroscopy

EXAFS (extended x-ray absorption fine structure) – local atomic structure and coordination

XANES (x-ray absorption near-edge structure) – local electronic structure and bonding geometry



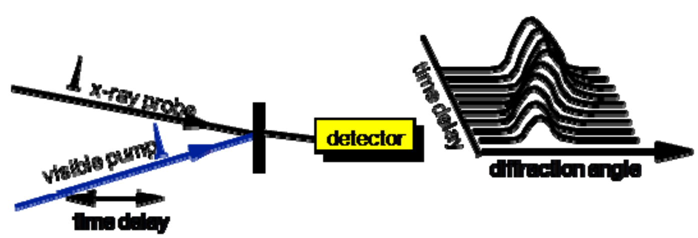
Time-resolved x-ray scattering

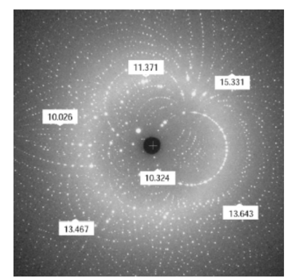
Bragg Diffraction: atomic structure in systems with long-range order/periodicity

Diffuse Scattering: clustering, nucleation, defects

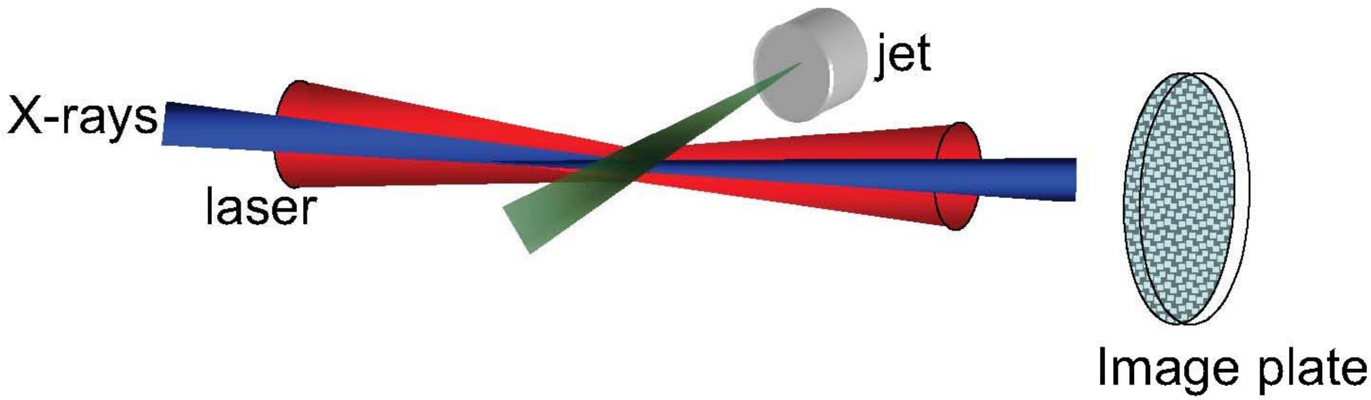
XRR (X-Ray Reflectivity): surface/interface structure

R.W. Schoenlein



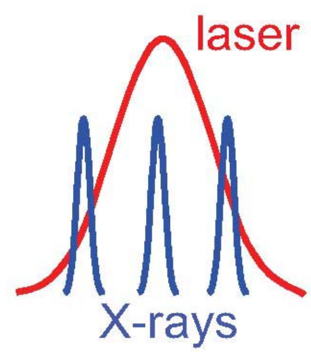


X-ray Studies of Laser Aligned Molecules



Aligning laser pulse at APS:

- 120 ps
- 10^{12} W/cm^2
- 30µm
- 1 KHz



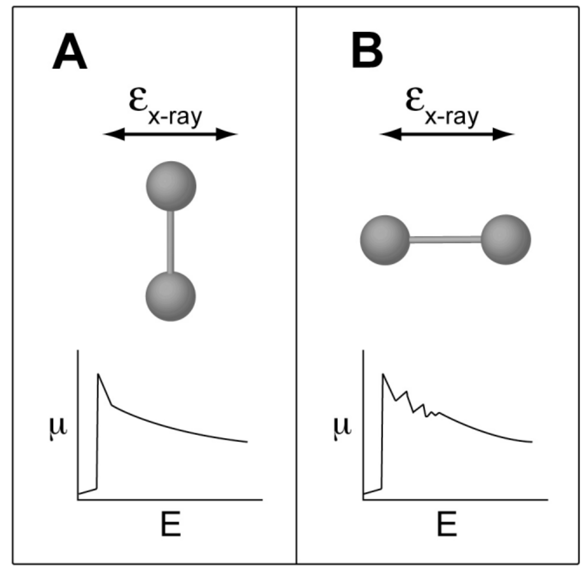
(Linda Young 2006)

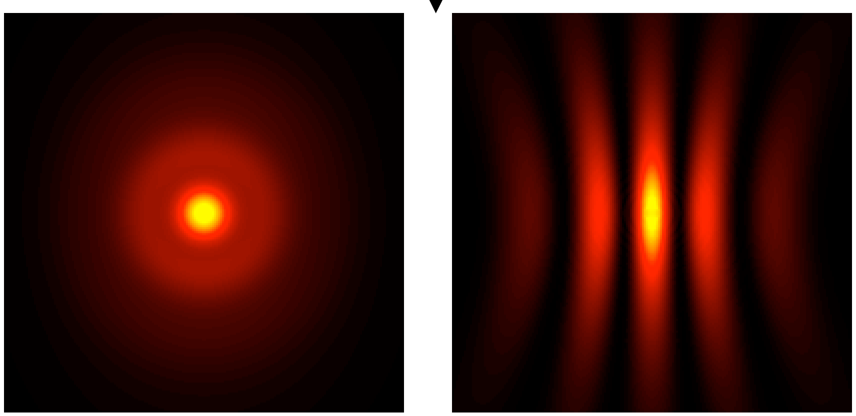
Laser Aligned Molecules

X-Ray Advantages:

- no dynamic alignment by probe pulse
- degree of alignment via near-edge structure
- change in structure via EXAFS
- atomic resolution structures via diffraction

Br_2 EXAFS

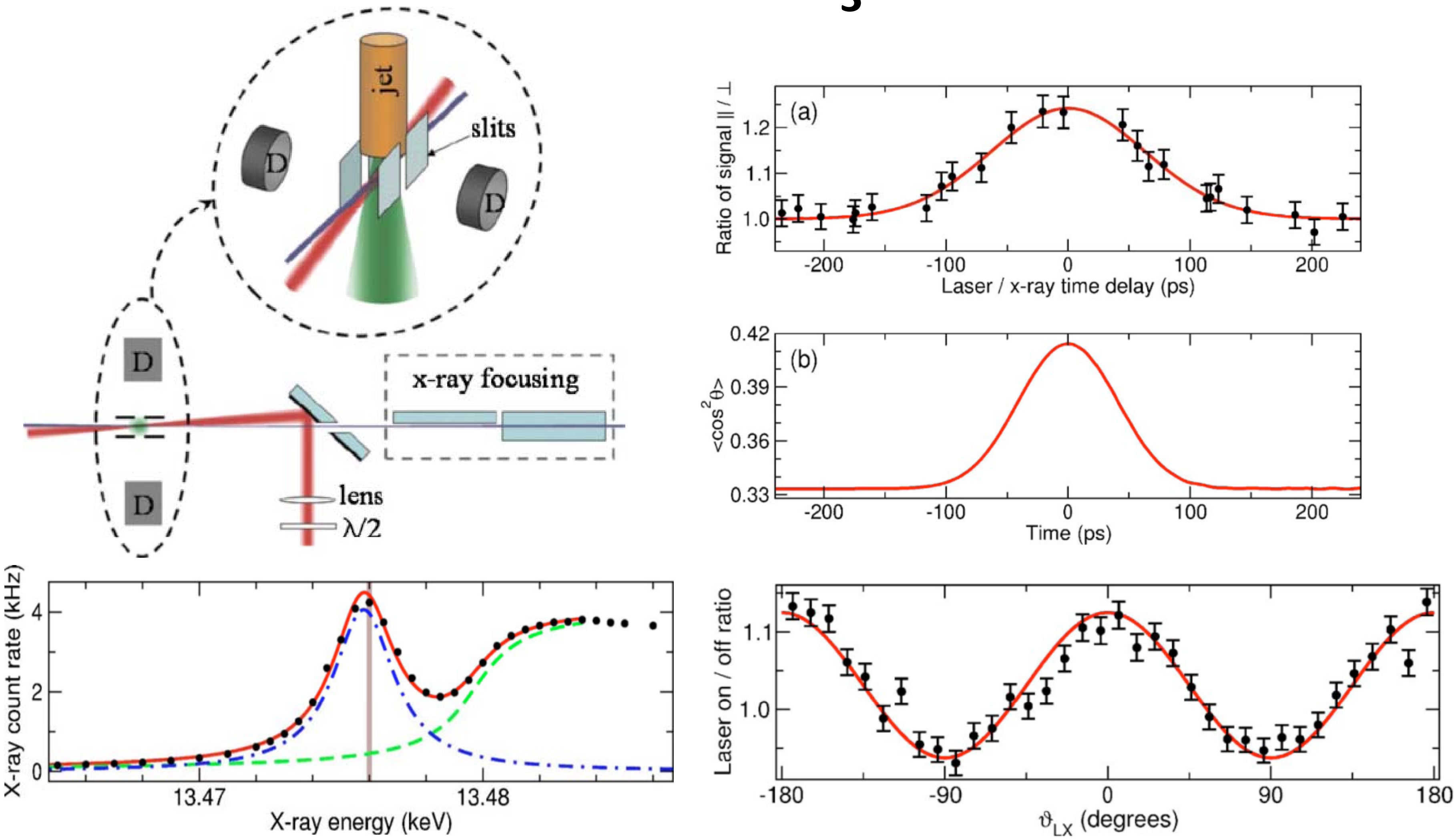




Isotropic Aligned

(Linda Young 2006)

It works! CF_3Br

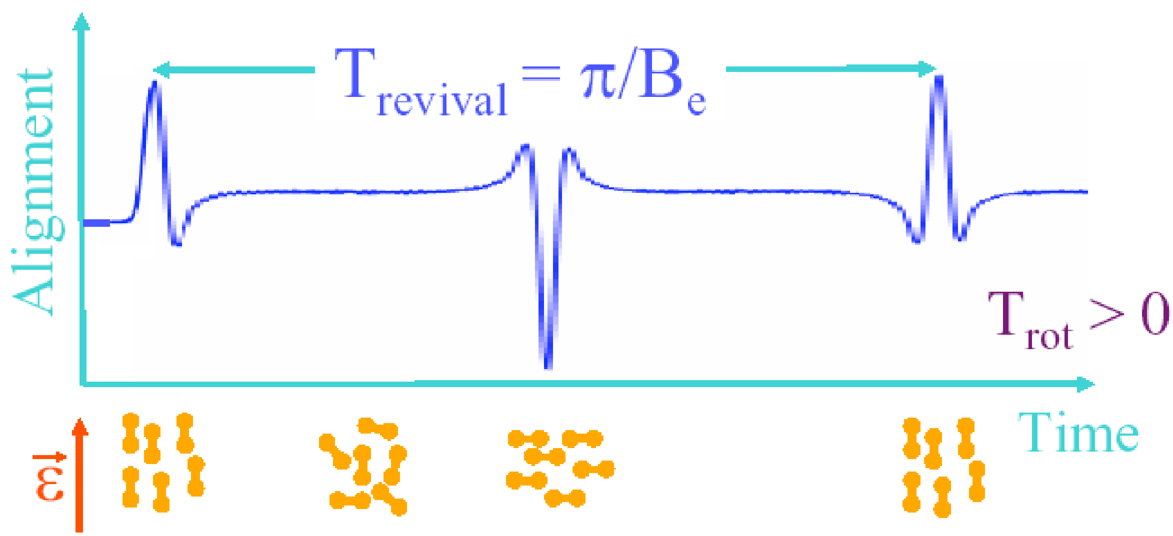


E. R. Peterson, et al., Journal of Applied Physics **92**, 094106 (2008).

Field Free Alignment

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

In general, a quantum mechanical superposition of states dephases. If the underlying dynamics is stable and periodic, the wave packetreconstructs after a time T_{rev} .



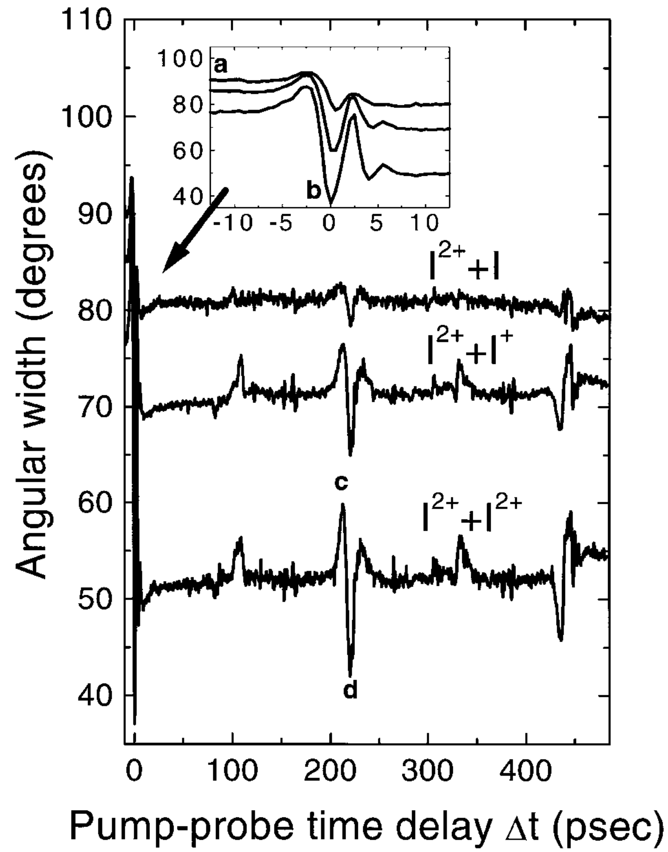
(Linda Young 2006)

T. Seideman, Phys. Rev. Lett. **83**, 4971 (1999).

Laser induced alignment of I_2

Pump laser:

- home made Ti:sapphire
- 800 nm, 0.7 x 80mJ
- 1-10ps, 50 Hz



F. Rosca-Pruna and M. J. J. Vrakking, Phys. Rev. Lett. **87**, 153902 (2001).

Time Resolved Diffuse Scattering

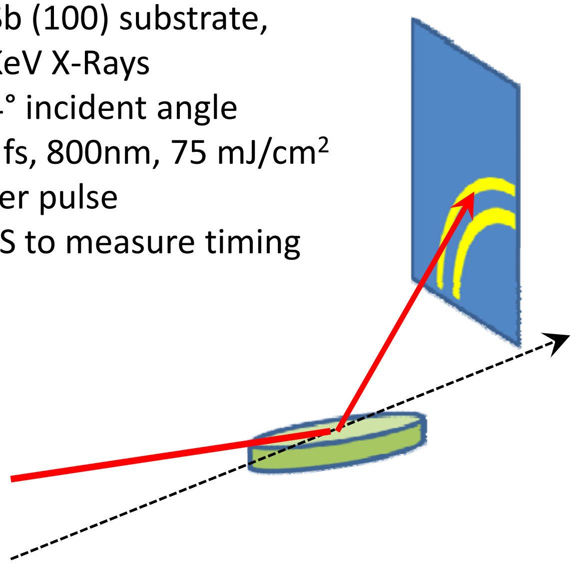
PRL **100,** 135502 (2008) PHYSICAL REVIEW LETTERS week ending 4 APRIL 2008

X-Ray Diffuse Scattering Measurements of Nucleation Dynamics at Femtosecond Resolution

A. M. Lindenberg,[1,2,3] S. Engemann,[1,3] K. J. Gaffney,[1,3] K. Sokolowski-Tinten,[4] J. Larsson,[5,1] P. B. Hillyard,[1,6] D. A. Reis,[7,1] D. M. Fritz,[1,7] J. Arthur,[3] R. A. Akre,[8] M. J. George,[3] A. Deb,[1,3] P. H. Bucksbaum,[1,3] J. Hajdu,[9,1] D. A. Meyer,[10,1] M. Nicoul,[4] C. Blome,[11] Th. Tschentscher,[11] A. L. Cavalieri,[7,12] R. W. Falcone,[13] S. H. Lee,[14] R. Pahl,[15] J. Rudati,[16] P. H. Fuoss,[17] A. J. Nelson,[18] P. Krejcik,[8] D. P. Siddons,[19] P. Lorazo,[20] and J. B. Hastings[3]

SPPS Experiment

InSb (100) substrate,
9 KeV X-Rays
0.4° incident angle
50 fs, 800nm, 75 mJ/cm^2
laser pulse
EOS to measure timing



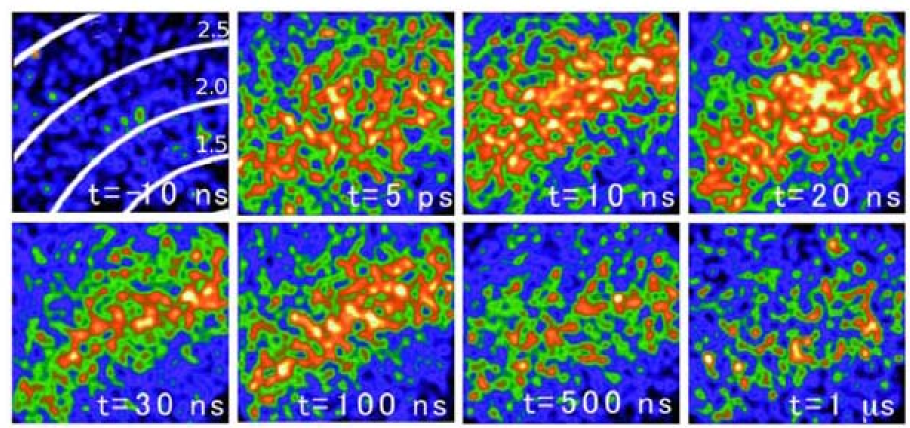
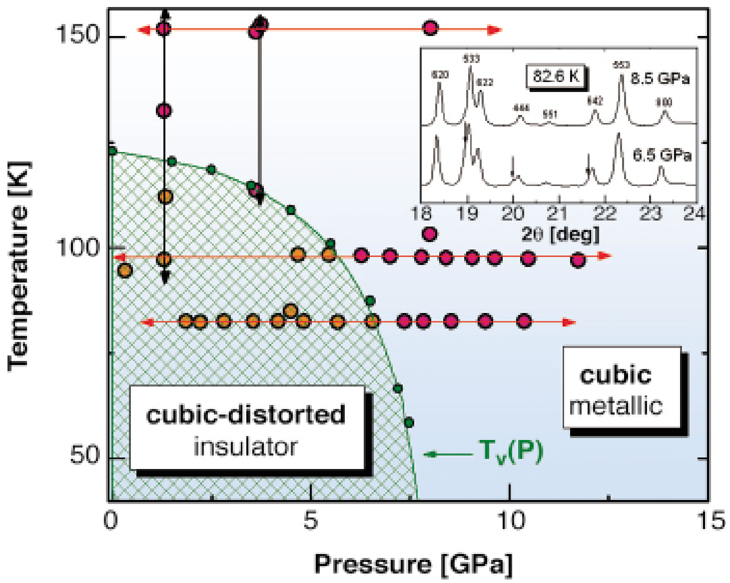


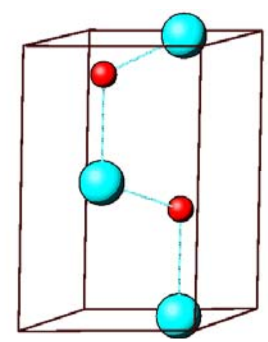
Fig. 1. X-ray diffuse scattering pattern measured on a CCD camera at various times before and after laser excitation of the semiconductor InSb. These images capture the dynamical evolution of the optically-induced disordered state on time-scales from picoseconds to microseconds. Top left image shows lines of constant Q, in units of Å^{-1}.



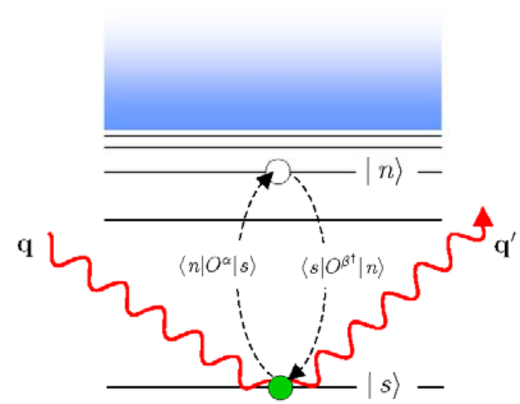
http://www.esrf.eu/UsersAndScience/Publications/Highlights/2006/MAT/MAT02

Selectively "tweek" atoms in a crystal and monitor the response of valence electrons.

ZnO Unit Cell



- •Coherent optical phonon changes overlap between valence electrons.
- •Excite coherent phonon with an ultrafast (50fs) optical pulse.
- •Access the resulting change in the wavefunction via the dependence of resonant scattering on the intermediate state.



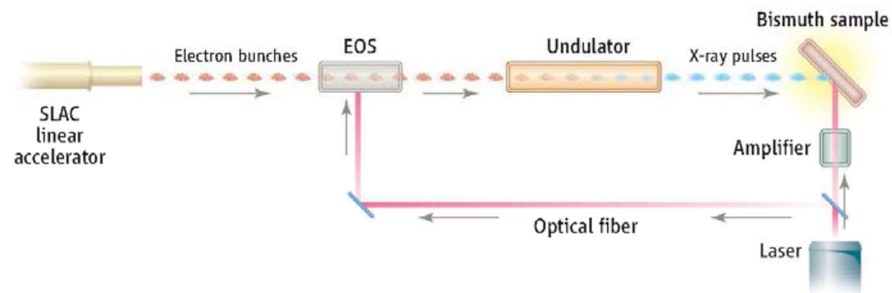
(Ken Finkelstein0

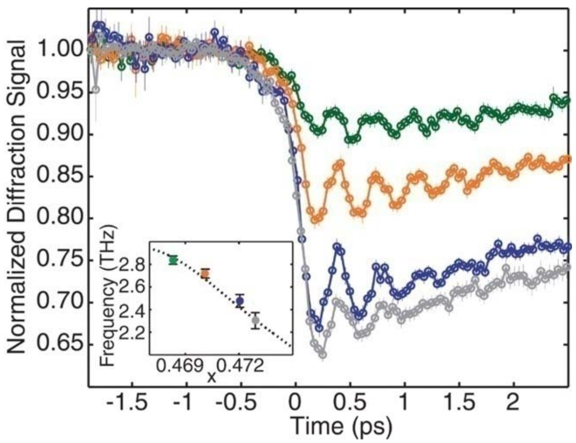
Time Resolved Diffraction

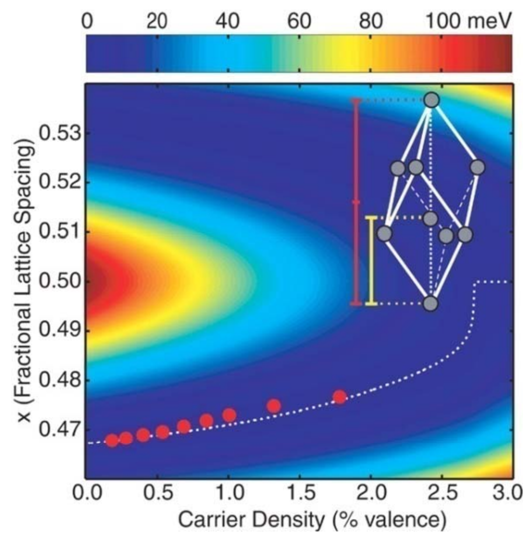
Ultrafast Bond Softening in Bismuth: Mapping a Solid's Interatomic Potential with X-rays

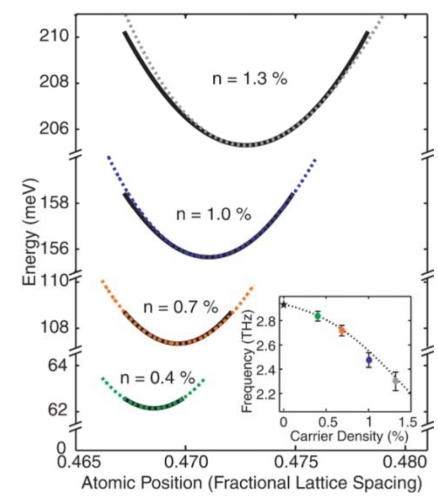
D. M. Fritz,[1,2*] D. A. Reis,[1,2] B. Adams,[3] R. A. Akre,[4] J. Arthur,[5] C. Blome,[6] P. H. Bucksbaum,[2,4,7] A. L. Cavalieri,[8] S. Engemann,[5] S. Fahy,[9] R. W. Falcone,[10] P. H. Fuoss,[11] K. J. Gaffney,[5] M. J. George,[5] J. Hajdu,[12] M. P. Hertlein,[13] P. B. Hillyard,[14] M. Horn-von Hoegen,[15] M. Kammler,[16] J. Kaspar,[14] R. Kienberger,[8] P. Krejcik,[4] S. H. Lee,[17] A. M. Lindenberg,[5] B. McFarland,[7] D. Meyer,[15] T. Montagne,[4] É. D. Murray,[9] A. J. Nelson,[18] M. Nicoul,[15] R. Pahl,[19] J. Rudati,[3] H. Schlarb,[6] D. P. Siddons,[20] K. Sokolowski-Tinten,[15] Th. Tschentscher,[6] D. von der Linde,[15] J. B. Hastings[5]

Science, **315**, 633 (2007)



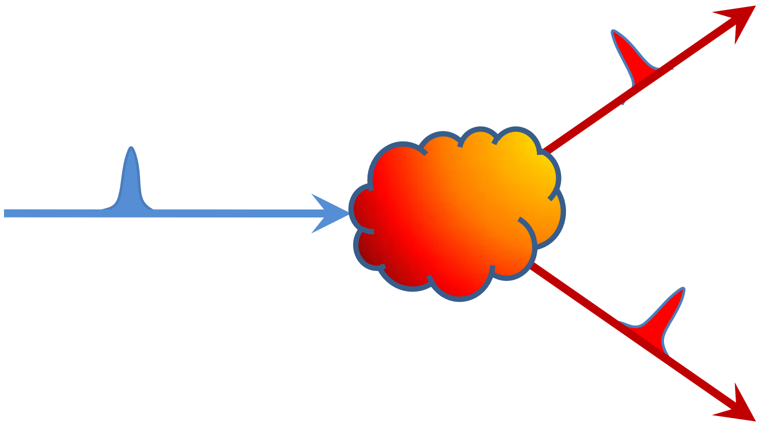






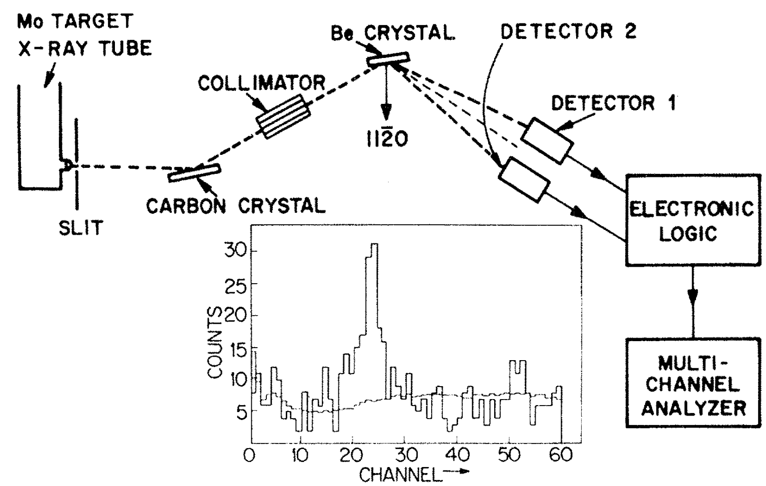
Coincidence Measurements

Example: parametric down conversion



$$\frac{T}{F} = \frac{\eta_t C f}{(\eta_1 C + N_1)(\eta_2 C + N_2)}$$

$\eta_{t,1,2}(\sigma,\theta,\varepsilon) \equiv$ detection coefficients
$N_{1,2} \equiv$ noise counts
$C \equiv$ average count rate
$f \equiv$ repetition rate



Eisenberger, P. and S.L. McCall, *Physical Review Letters, 1971.* **26**(12): p. 684-688.

Comparison of hard x-ray short pulse sources

	ALS	SPPS	APS SPX	ERL High Flux	ERL Short Pulse*	LCLS
Pulse Duration	100 fs	80 fs	1 - 3 ps	2 - 3 ps	100 fs	77 fs
Energy Range	2-10 keV		200 eV - 100 keV	4-30KeV	4-30KeV	
Repetition Rate	5 kHz	30Hz	6.5 MHz	1.3 GHz	100KHz	120Hz
Photons/Pulse (p/pulse/0.1%)	10^2	10^8	$7.3x10^7$ /10^2	$2.2x10^6$	$1.5x10^7$	10^{12}
Avg flux (p/s/0.1%)	10^6	10^9	$4.8x10^{14}$/10^2	$2.9x10^{15}$	$1.5x10^{12}$	10^{14}

*25m undulator, 2.5cm period, K=0.5, 5GeV. (Σ=80μm, Σ'=6.4μrad, σ_z=1.5×10^{-2}mm)

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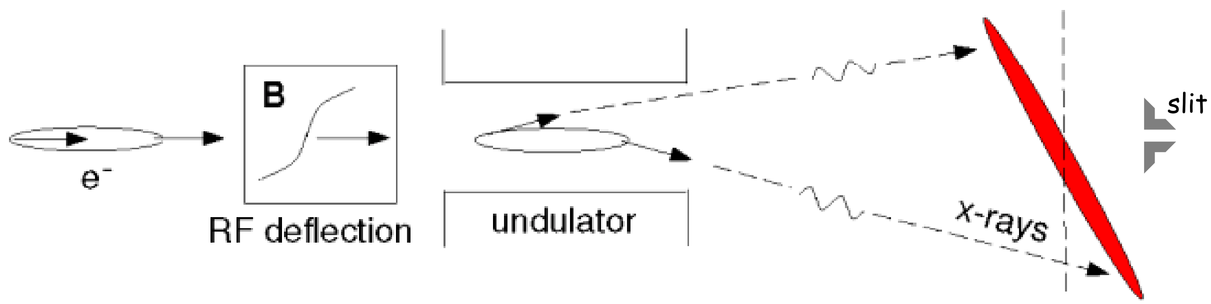
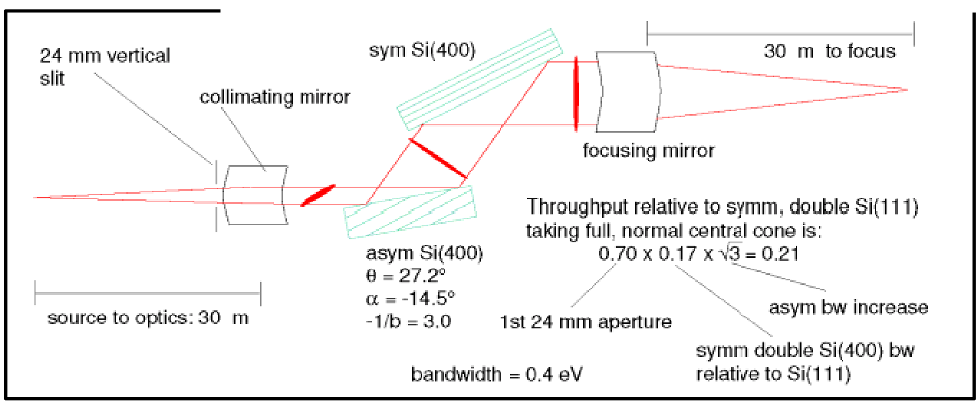
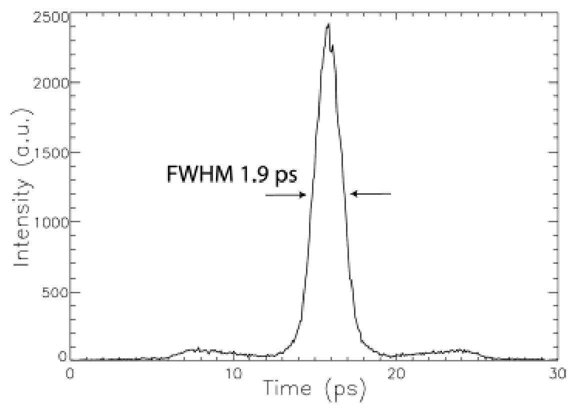
Proceedings of 2005 Particle Accelerator Conference, Knoxville, Tennessee

GENERATION OF SHORT X-RAY PULSES USING CRAB CAVITIES AT THE ADVANCED PHOTON SOURCE*

K. Harkay[#], M. Borland, Y.-C. Chae, G. Decker, R. Dejus, L. Emery, W. Guo, D. Horan, K.-J. Kim,
R. Kustom, D. Mills, S. Milton, A. Nassiri, G. Pile, V. Sajaev, S. Shastri, G. Waldschmidt, M. White,
B. Yang, ANL, Argonne, IL 60439, U.S.A.
A. Zholents, LBNL, Berkeley, CA U.S.A.







62% transmission (relative to ideal)

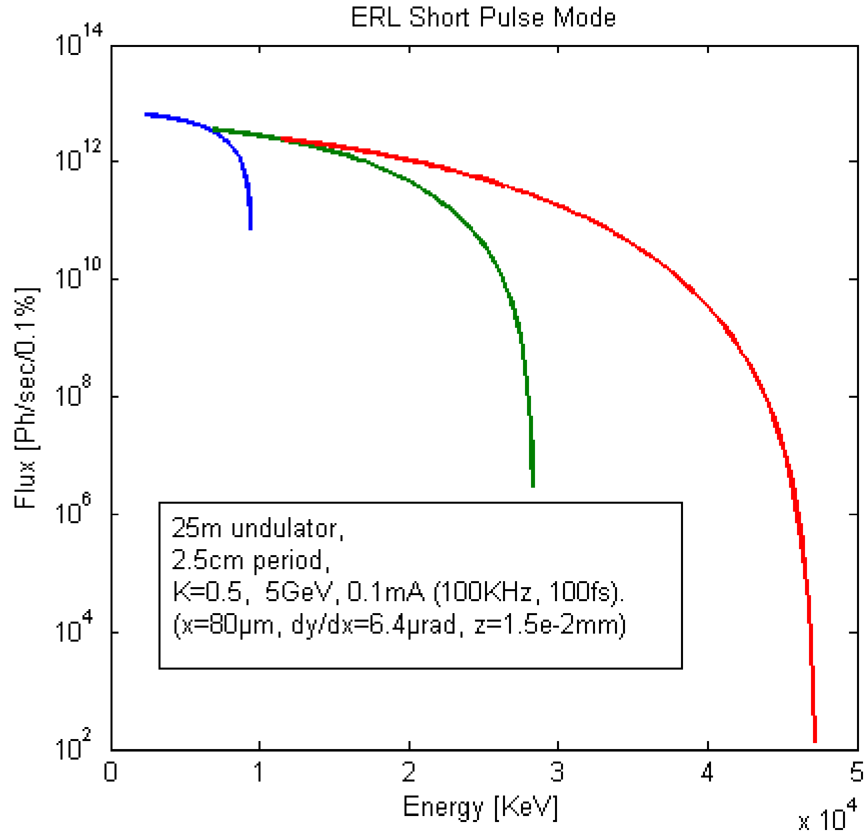
Characteristics of ERL:

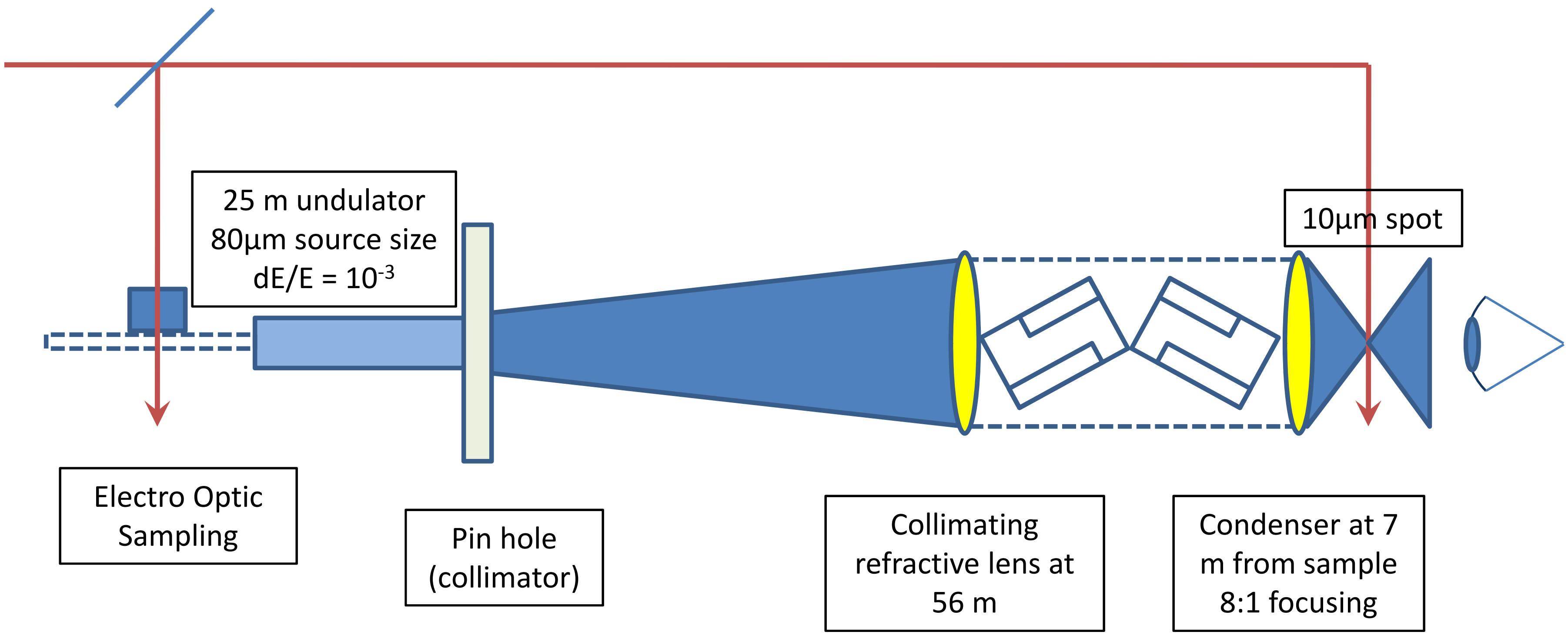
- 2-3 picoseconds,
- 10^6 photons/pulse,
- 1.3GHz

Issue:

- 1.3GHz is a challenge for conventional pump lasers and/or samples of interest

What can be done?
If willing to give up energy recovery, can utilize pulse compression to get short pulses.
Consider 0.1mA at 100KHz with nC bunches


ERL Short Pulse Mode
Flux [Ph/sec/0.1%]
Energy [KeV]
x 10^4
25m undulator,
2.5cm period,
K=0.5, 5GeV, 0.1mA (100KHz, 100fs).
(x=80μm, dy/dx=6.4μrad, z=1.5e-2mm)


25 m undulator
80µm source size
dE/E = 10-3
10µm spot
Electro Optic
Sampling
Pin hole
(collimator)
Collimating
refractive lens at
56 m
Condenser at 7
m from sample
8:1 focusing

To make these decisions about possible facilities, we need to begin calculating "luminosity," the # of useful events per second.

Bandwidth, repetition rate, spot size, and how often can we use the source (e.g., need for special ops) all matter for real experiments. Scattering, spectroscopy and spatial imaging require different specifications.

Short term need is the ability to perform ray optics calculations (e.g., 6D phase space calculations) of complete beamlines routinely. X-ray optics matter. Fortunately, both electron optics (accelerator physics) and laser optics communities have this capability. We should be able to leverage their tools.

Considerations:

1. Pump Laser:
   - does signal increase with laser power? If so, S/N improves with energy. This is where the XFEL is best suited.
   - Does laser bleach system? If so, high-repetition rate may give higher luminosity
2. Detectors:
   - Maximum count rate
   - Read out rate
   - Dynamic range
   - Energy resolution
3. Operations:
   - Simultaneous with other modes, or
   - Time-share on daily (e.g., 4/24 hours)

END