SPPS: New Science on the way to LCLS

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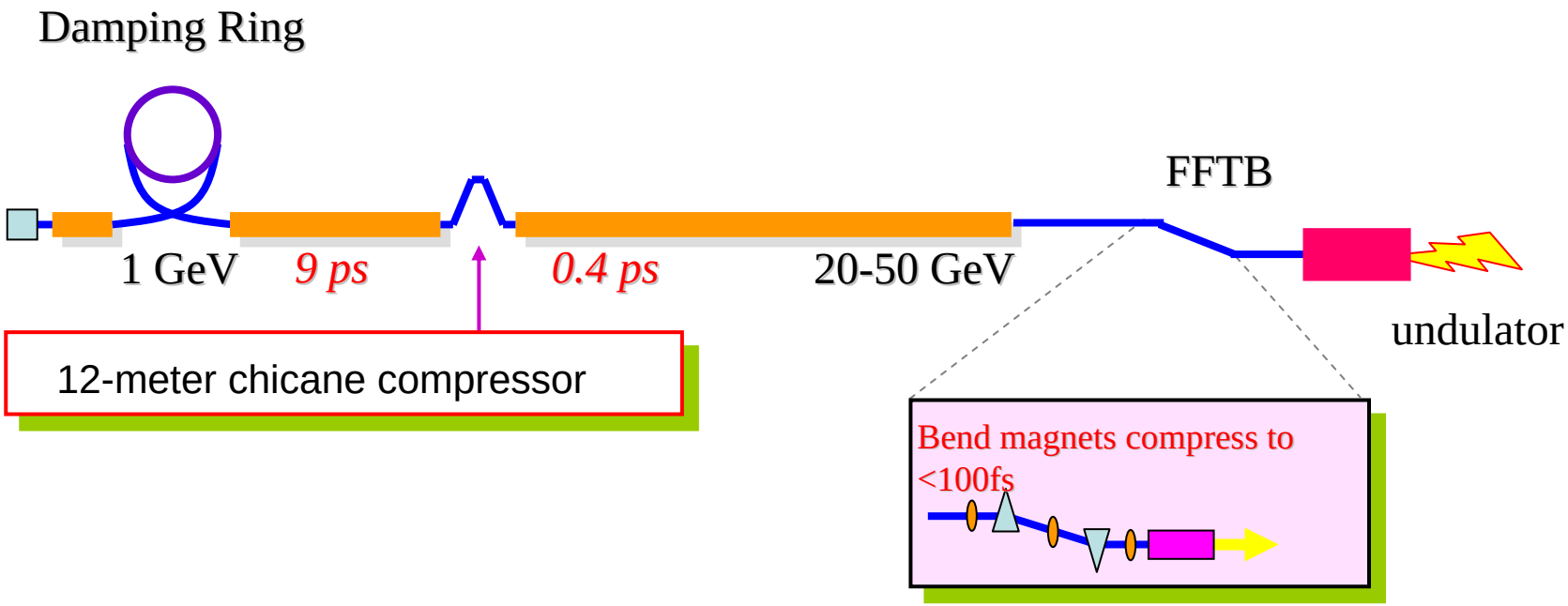
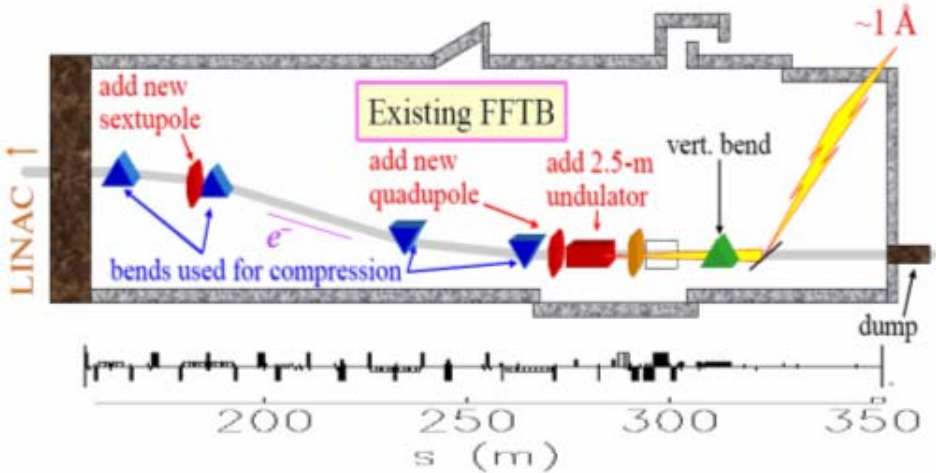
Outline

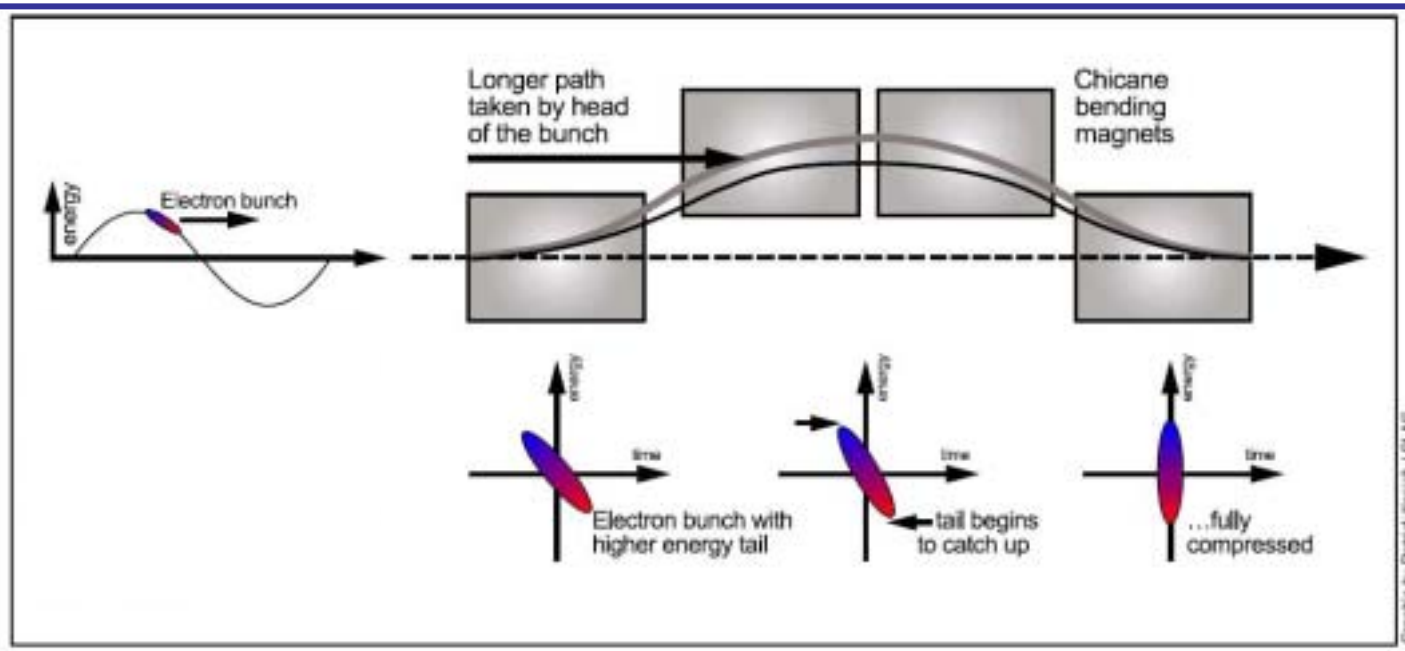
- SPPS compression scheme
- SPPS characteristics (photon numbers, pulse duration, spot size…)
- First experiments
- Future experiments

Femtosecond X-ray and Electron Source at SLAC Linac

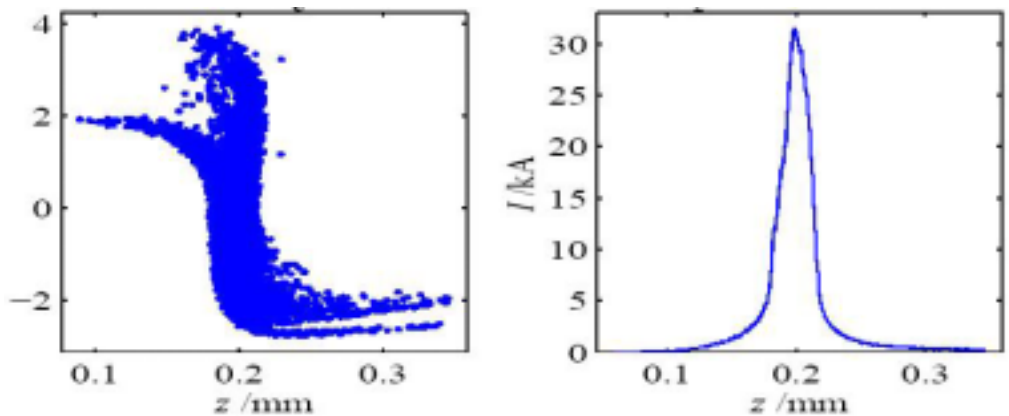




Electron Bunch Compression Scheme



Particle tracking codes

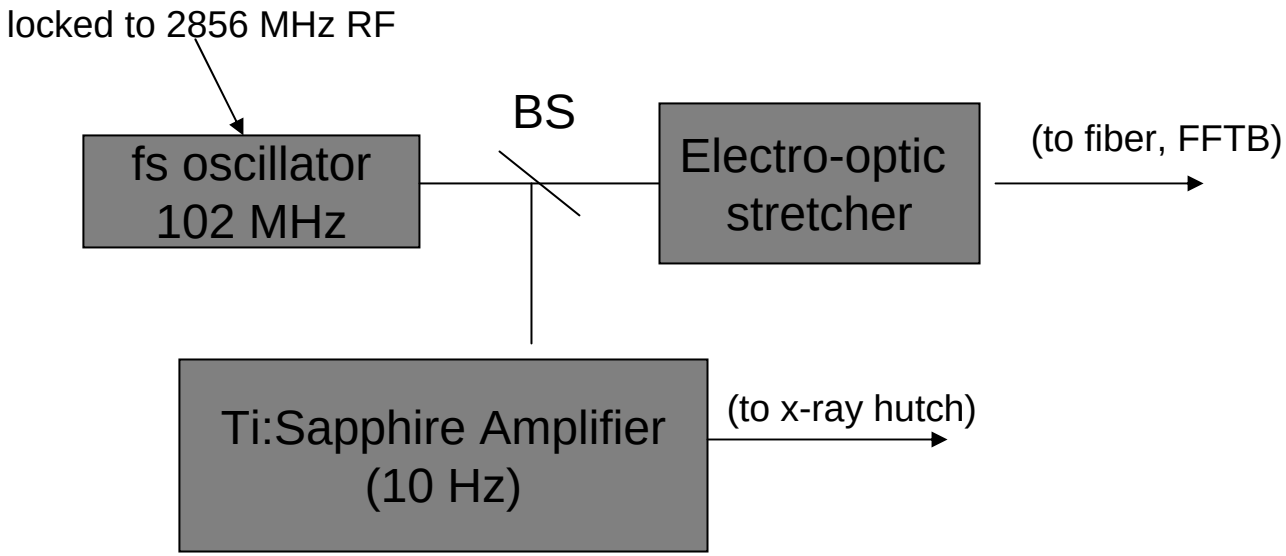


(P. Krejcik *et al*.)

SPPS Characteristics (x-rays)

- 2 x10^7 photons/pulse/1.5% BW at 10 Hz
- Pulse duration: ~ 80 fs
- Tunable over 8-10 keV
- Synchronization of laser/x-rays ~ 300 fs rms
- ~2 mm spot size (unfocused)
- ~250 μm (Be lens) (~3 x 10^6 ph/pulse)
- <1 μm (KB) (~5 x 10^5 ph/pulse)

SPPS Laser System and Timing

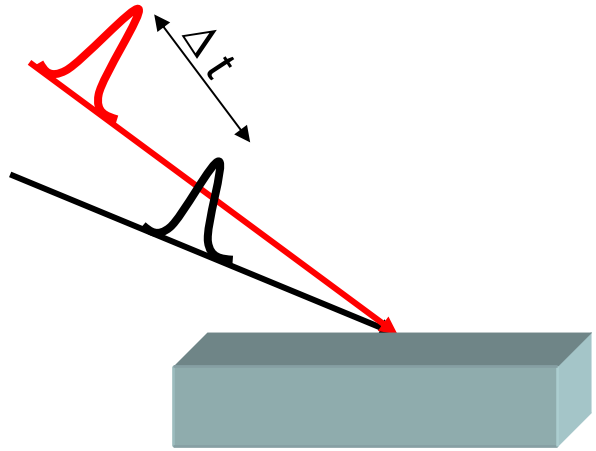


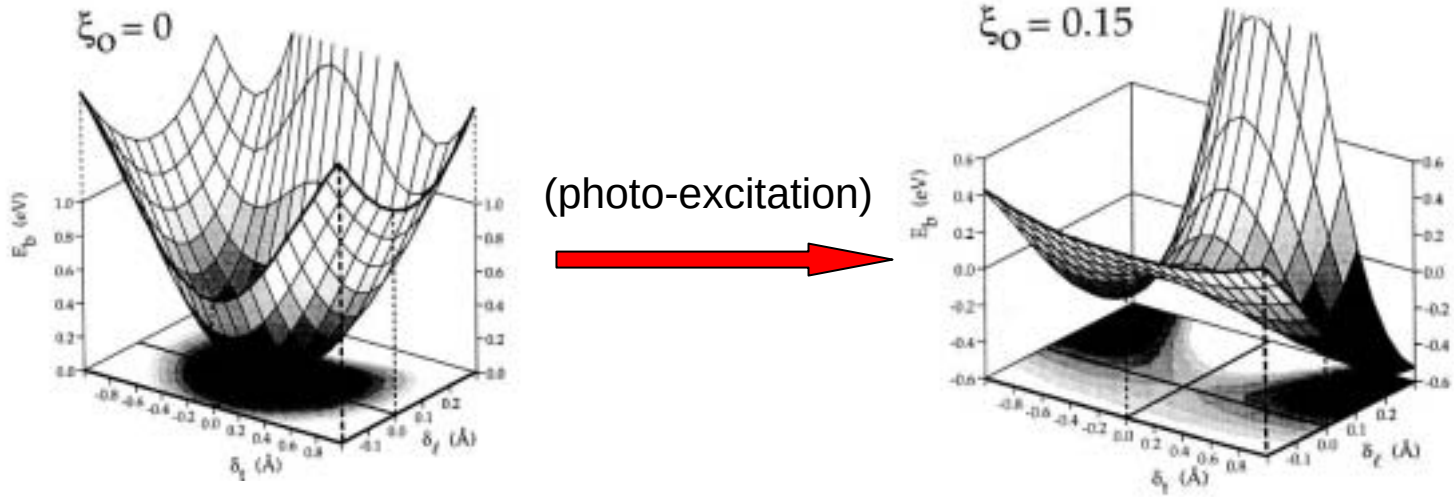
- Use EO timing diagnostics to post-process pump-probe data to overcome jitter. Permits integrating mode of operation.

Ultrafast Melting Experiments in Semiconductors

- Previous experiments:

Rousse *et al. Nature* **410**, 65 (2001)
Sokolowski-Tinten *et al*. *PRL* **87**, 225702 (2001)
Siders *et al*. *Science* **286**, 1349 (1999)
Chin *et al. PRL* **83**, 336 (1999)
A.M. Lindenberg *et al*. *PRL* **84**, 111 (2000)

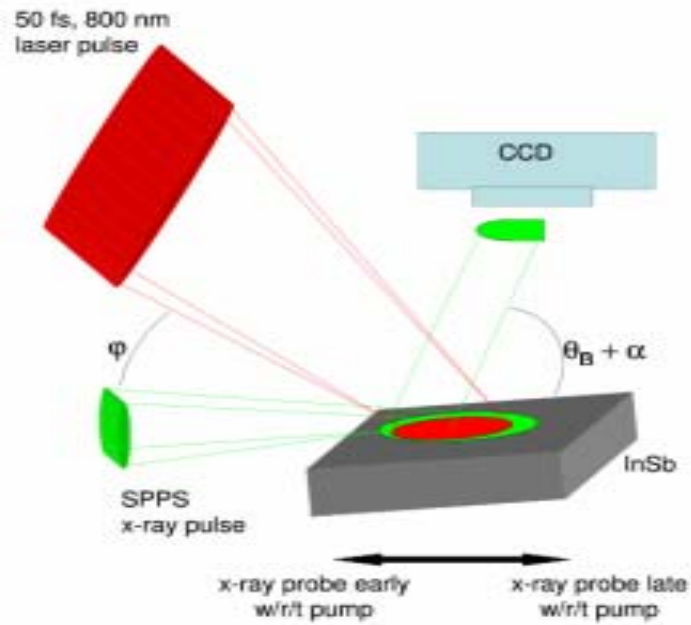




- But, melting is a difficult problem even under conditions of thermodynamic equilibrium…

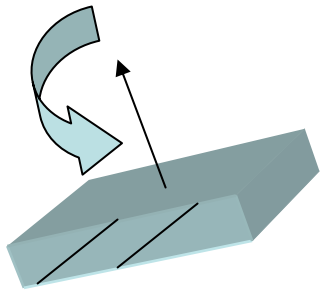
(Stampfli *et al.*)
PRB **49**, 7299 (1994)

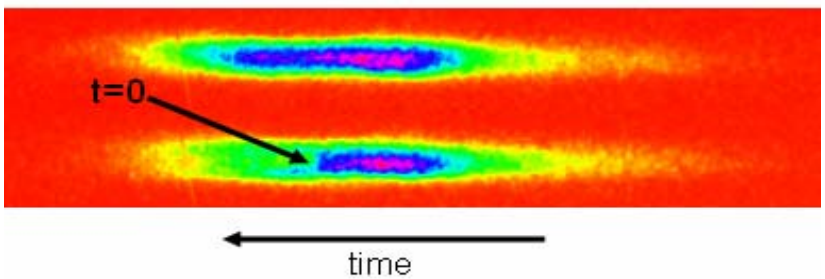
Experimental Setup: Crossed-beam topography



- Azimuthal rotation allows for continuous adjustment of crystal asymmetry angle (fixing x-ray penetration depth)

- X-ray incident angle set to 0.4 degrees for all measurements, gives x-ray penetration depth < 100 nm (set by photoabsorption).





- Crossed-beam technique transforms temporal information into spatial information.

- Measures complete time history around t=0 in single shot.

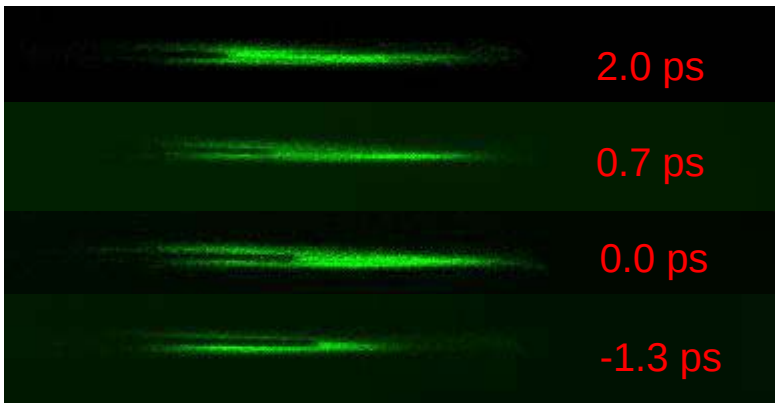
- Position of edge indicates x-ray/laser timing for that shot.

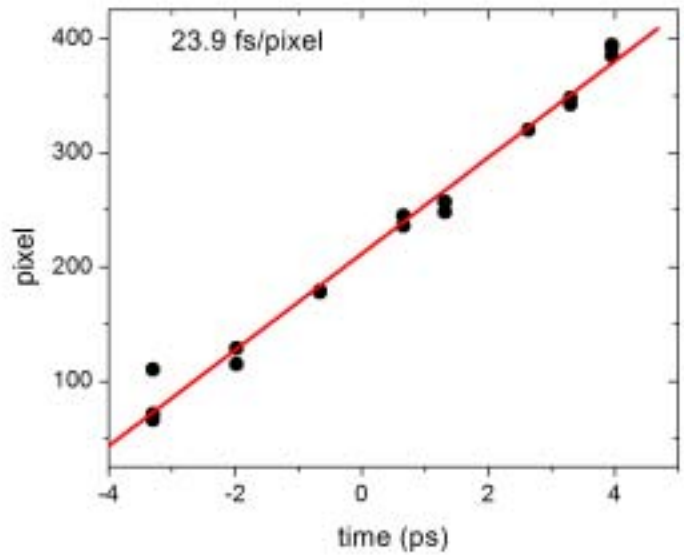
Calibration of Time Axis

•Scanning optical delay line changes x-ray/laser timing, moving the edge-position within the image

• Geometrically, one expects: $$fs/pixel = \frac{\delta(1-\cos\theta_L)}{\sin(\theta_B+\alpha)c}$$

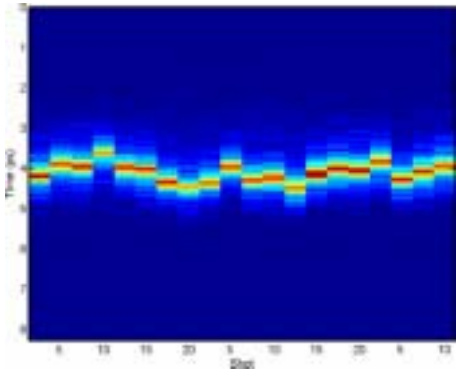
• Measurements are in good agreement with this geometric factor.



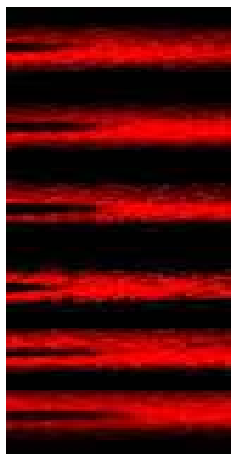


X-ray/Electron Bunch Timing

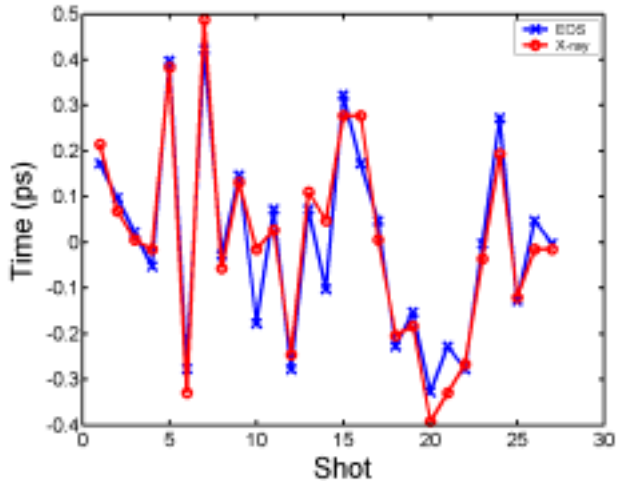
Electro-optic signal

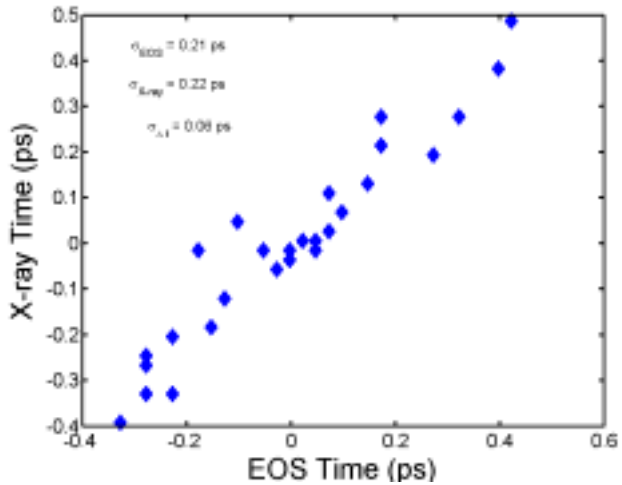


Melting signal



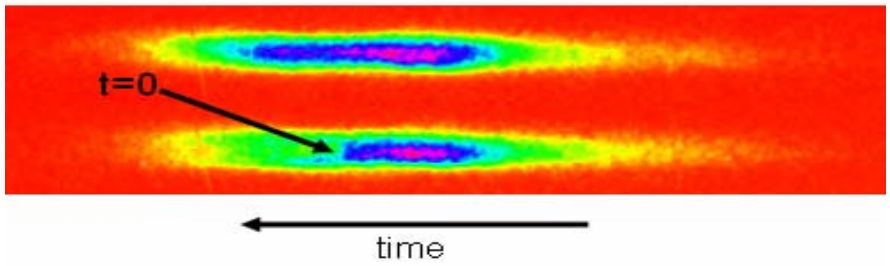
Correlation

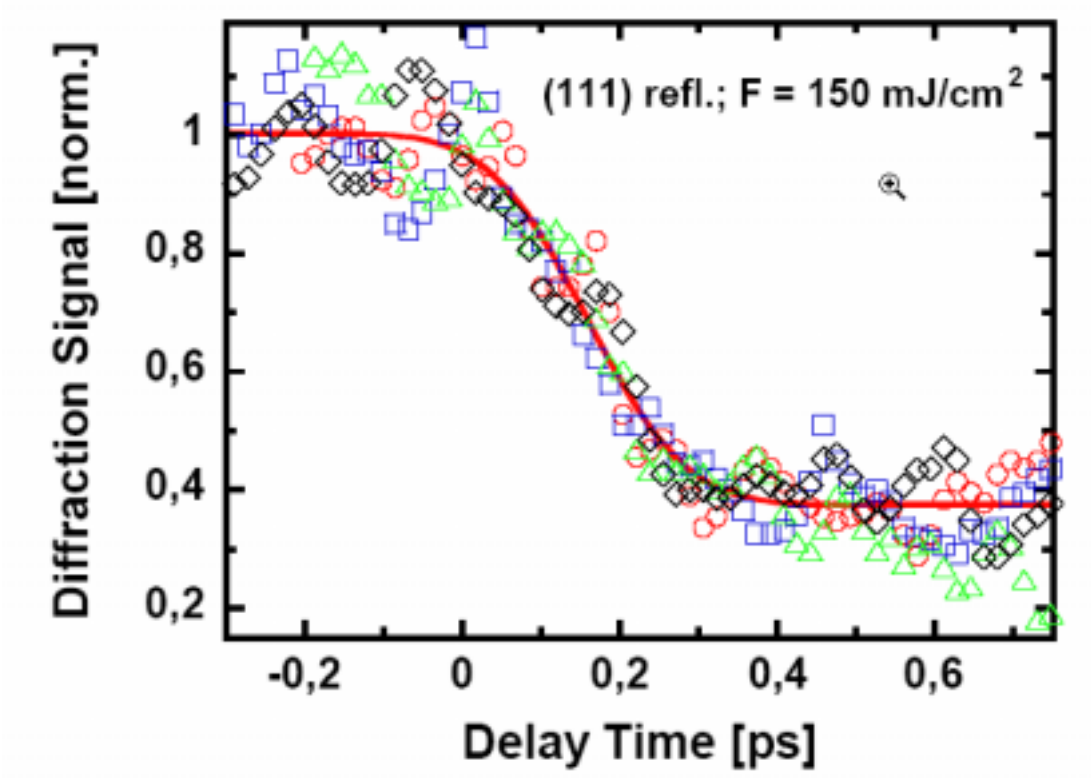




•Electro-optic sampling technique (sensitive to electron beam timing) agrees with location of edge in melting data to < 100 fs (60 fs rms)

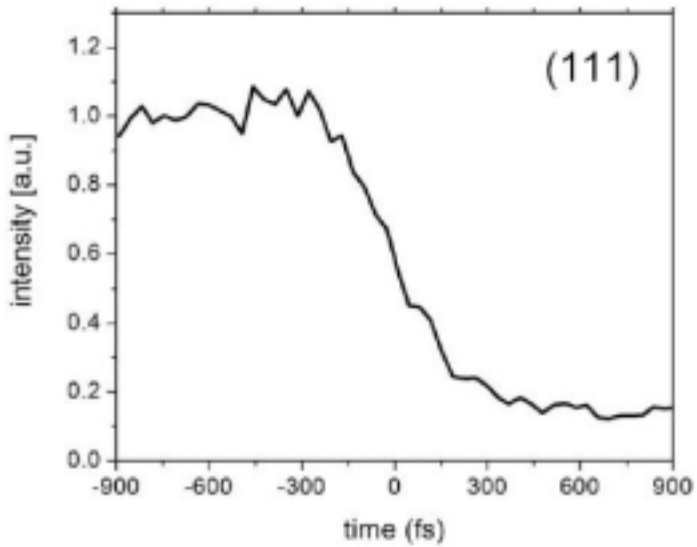
Raw data


t=0
time

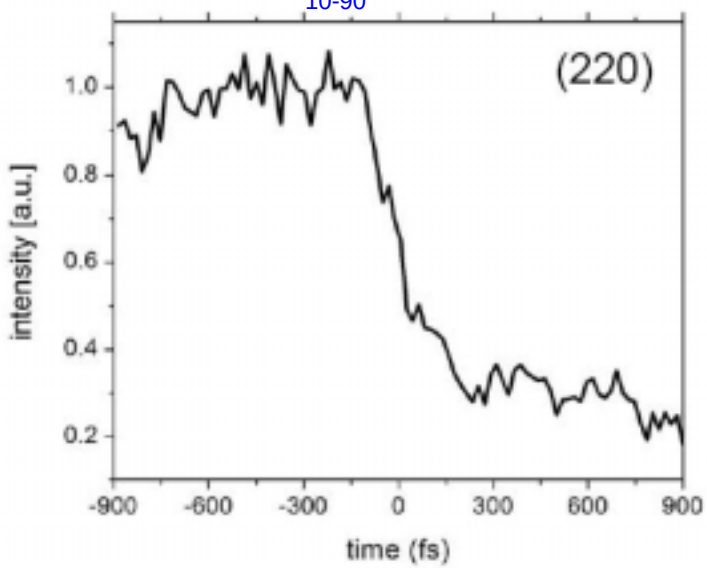

(111) refl.; F = 150 mJ/cm^2
Diffraction Signal [norm.]
1
0,8
0,6
0,4
0,2
-0,2
0
0,2
0,4
0,6
Delay Time [ps]

(111) vs. (220) Reflection

$\tau_{10\text{-}90}$=420 fs



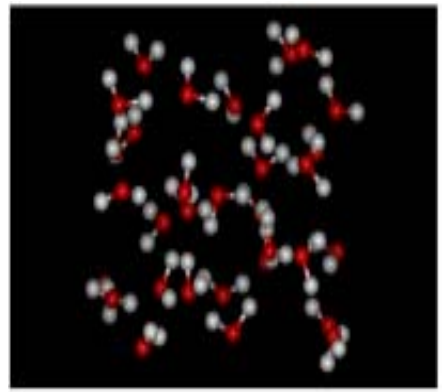
$\tau_{10\text{-}90}$=260 fs



- Two reflections measured under identical fluence conditions and identical pump and probe penetration depths.

- $$\frac{\tau_{(220)}}{\tau_{(111)}} = 1.62 \sim \sqrt{\frac{k_{(220)}}{k_{(111)}}} = \sqrt{\frac{8}{3}}$$

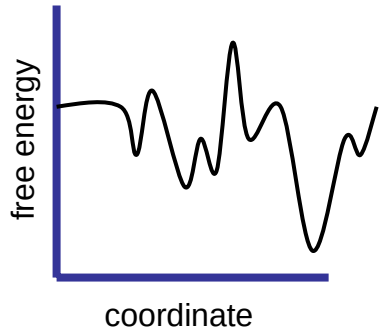
What is a liquid?



•Can be understood in terms of the topology of the instantaneous potential energy surface

•Instantaneous normal mode theory: Gives a representation of the potential energy surface at each instant in time, characterized by a set of frequencies, both real and imaginary (reflecting barriers and saddlepoints in potential surface.

•At short times, $q_\alpha(t) \sim v(0)t$ (inertial dynamics)



•Similar inertial dynamics observed in femtosecond optical measurements of solvation dynamics in liquids.

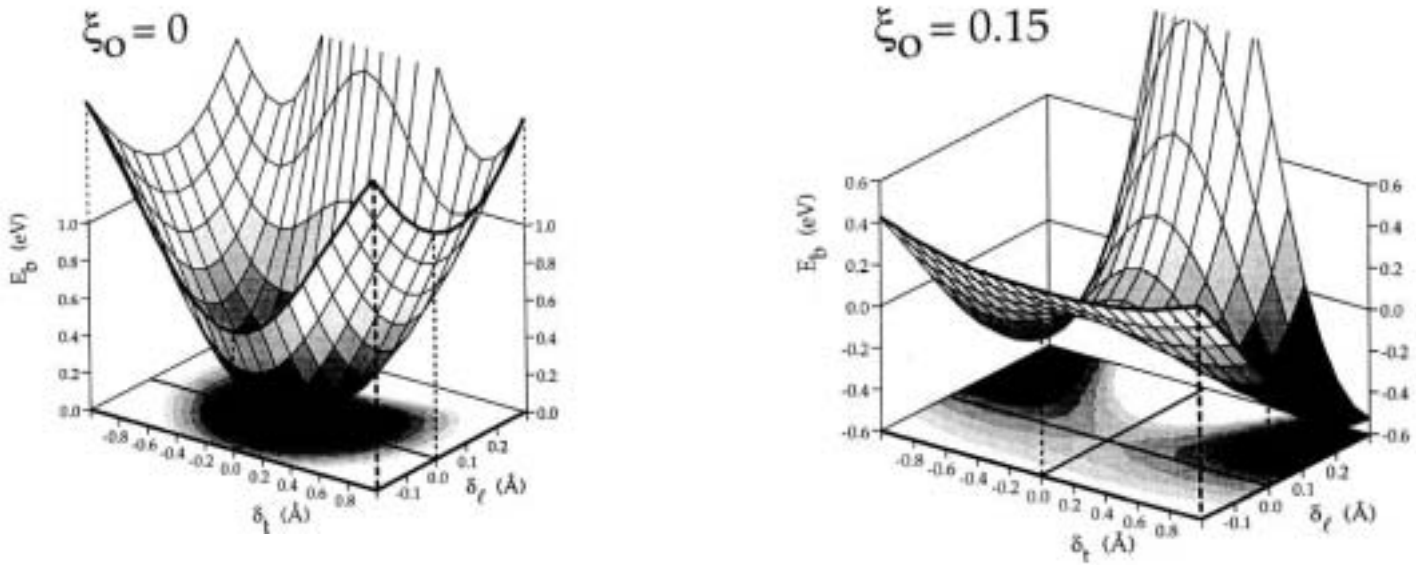
•Characterized by the spectral response function

$$S(t) = \frac{\nu(t) - \nu(\infty)}{\nu(0) - \nu(\infty)} \sim e^{-\frac{1}{2}\omega_{solv}^2 t^2}$$

(for short times)

•Represents ballistic motion of individual solvent molecules.

Visualization of Liquid State Dynamics



Debye-Waller Model:

$$I(t) \sim e^{-2W}$$

$$= e^{-\frac{16\pi^2}{3}(\frac{\sin\theta}{\lambda})^2 <u^2(t)>}$$

- Presumes that the disordering process corresponds to an ultrafast randomization of atomic positions (time-averaging is replaced by a spatial average over atomic positions.

- If dynamics are intertial for short times, predicts gaussian time-dependence (with u=vt).

Visualization of Liquid State Dynamics

Incoherent inelastic neutron scattering measures

$$S_s(k,w) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\omega t} F_s(k,t)dt$$

where $F_s(k,t)$ is the self-intermediate scattering function, defined as

$$F_s(k,t) =< e^{i\mathbf{k}\cdot[\mathbf{r(t)}-\mathbf{r(0)}]} >$$

The short time behavior of this function is gaussian and can be written as

$$F_s(k,t) = e^{-\frac{k^2 < r^2(t) >}{6}}$$

where for short times,

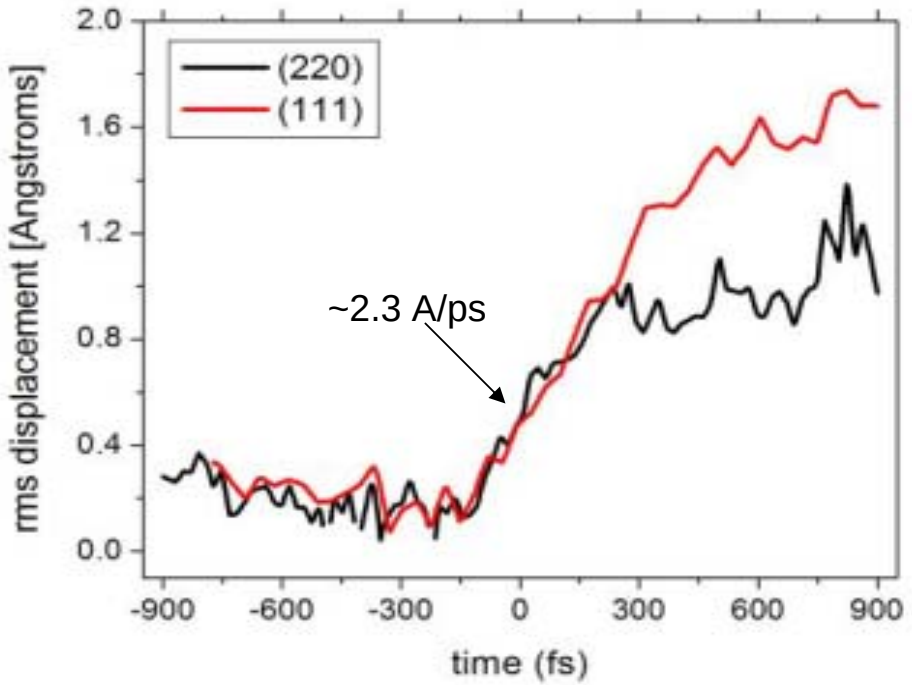
$$< r^2(t) >= \frac{3k_BT}{M}t^2 - \frac{k_BT}{4M}\Omega_0^2 t^2$$

inertial dynamics

influence of interatomic potential

Extraction of RMS Displacements

- On short time-scales, we observe gaussian (inertial) dynamics (Q-dependent time-constant)
- Short-time behavior for (111) and (220) reflections are the same
- Extracted velocities are in good agreement with equipartition.



collision frequency

Theoretical prediction: $$\delta r^2(t) \sim \frac{3k_bT}{M}t^2 - \frac{k_bT}{4M}\Omega_0^2 t^4 \qquad \left(\sqrt{\frac{3k_BT}{M}} \sim 2.5\text{Å}/ps\right)$$

Summary of Results and Open Questions

- Initial dynamics in ultrafast solid-liquid transition is governed by inertial dynamics, the ballistic motion of independent atoms on a softened potential surface.

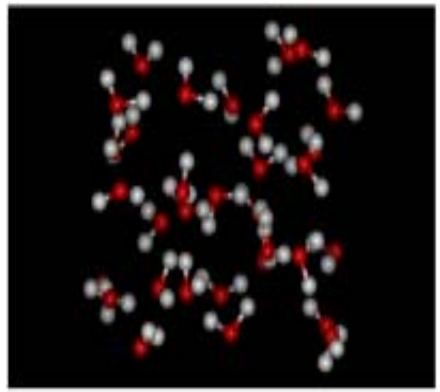
- Ultrafast melting phenomena may be thought of in analogy to the intrinsic structural changes which are continuously occurring in the liquid state.

- Time-resolved diffraction measurements give direct information on short-time dynamics previously only accessible through inelastic scattering techniques.

- For a wide range of potential landscapes, the dynamics are inertial to first order in t. Better signal to noise data should allow for more detailed measurements of the time-dependent interatomic potential.

Future Experiments

- Liquid State Experiments



- Measurements of liquid structure factor S(Q,t) gives information on pair correlation function and associated dynamics of both solute and solvent.

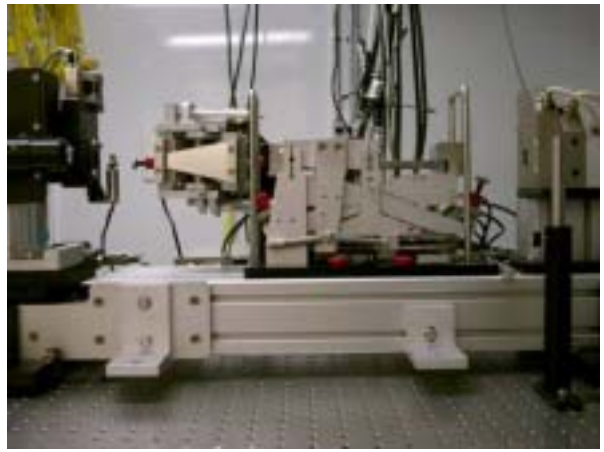
  -structure of H_2O
  -proton transfer dynamics
  -bond-breaking chemical reactions

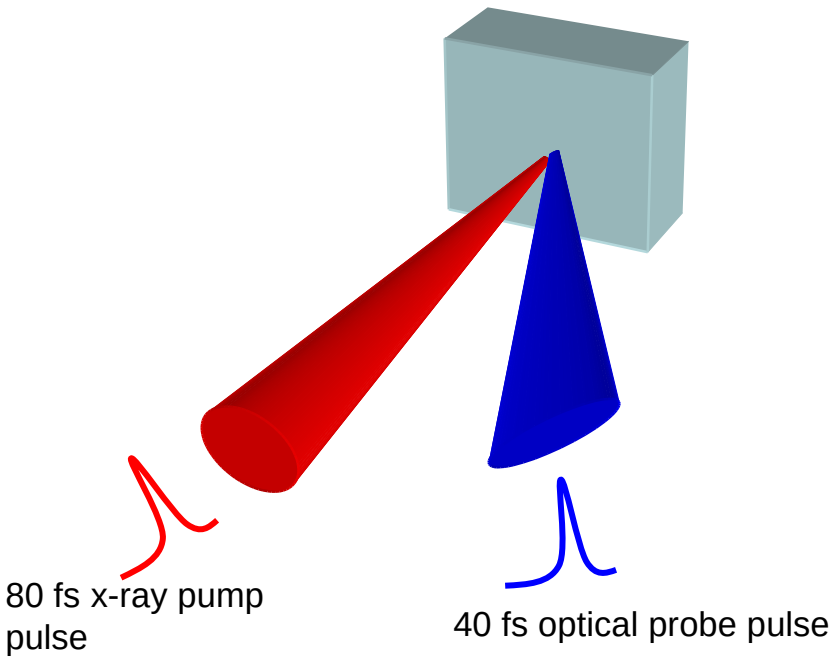
- Low-repetition rate of source allows for integrating CCD detectors

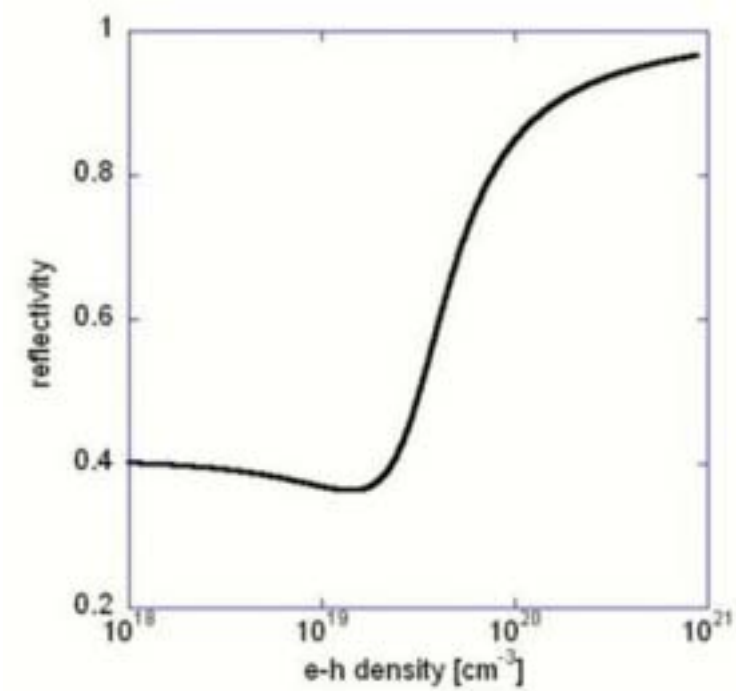
- Both diffraction and spectroscopy possible.

Future Experiments: X-ray Pump/Laser Probe

- Use of micro-focusing optics for creation of near LCLS conditions (~10^{13} W/cm^2)
- Measurement of secondary electron cascade dynamics
- Probing x-ray-induced damage mechanisms (limiting factor in structure determination for future XFELs)







Summary of Scientific Directions

•Solid-state dynamics:

-Vibrational excitations, structural phase transitions, strongly-correlated materials (See e.g. Sokolowski-Tintin *et al.* Nature, **422**, 237 (2003)

•Liquid-state dynamics

-Probing intrinsic fluctuations, bond-breaking, solvation dynamics

•Chemical reaction dynamics

-bond-breaking/formation, isomerizations, energy transfer, reaction intermediates (See e.g. A. Plech *et al*. PRL **92**, 125505)

•X-ray pump experiments, with goal to generate near-LCLS conditions.