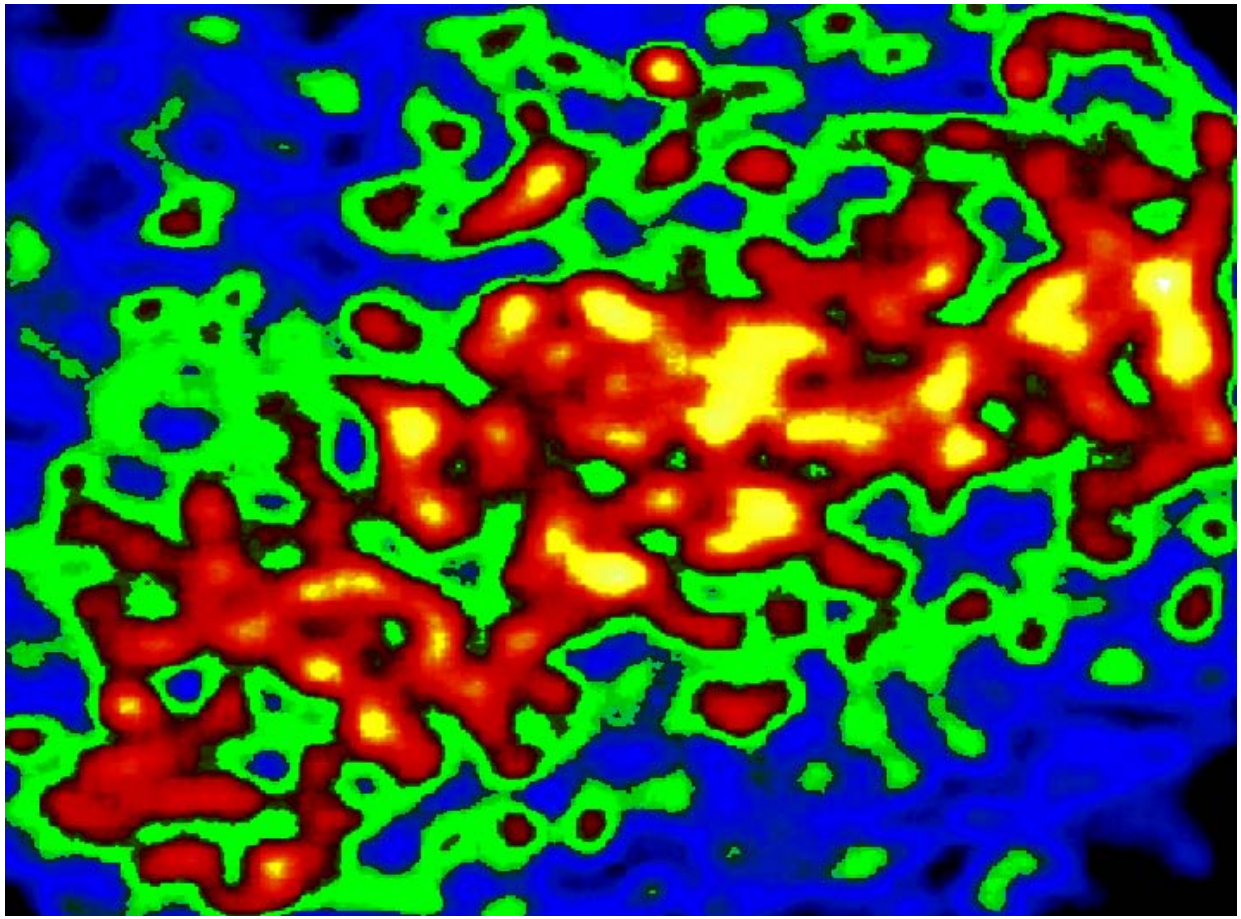
New Opportunities in Ultrafast X-ray Science



Aaron Lindenberg
Department of Materials Science and Engineering, Stanford University
PULSE Institute, Stanford Linear Accelerator Center

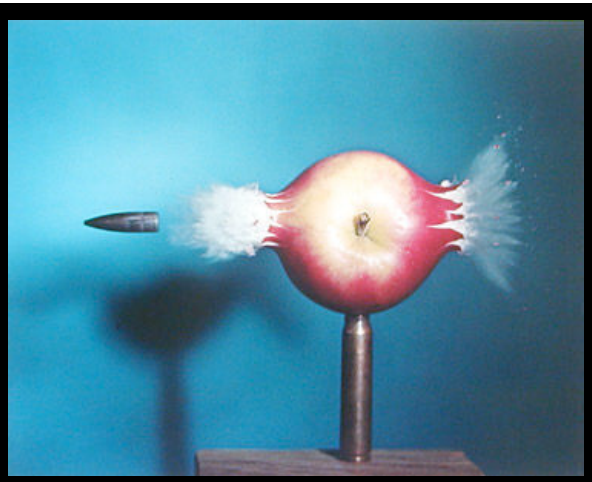
Outline

-A few words about existing sources

-Femtosecond x-ray measurements of phase transition dynamics,

-Future opportunities at free electron laser facilities

Taking Snapshots of Dynamic Phenomena



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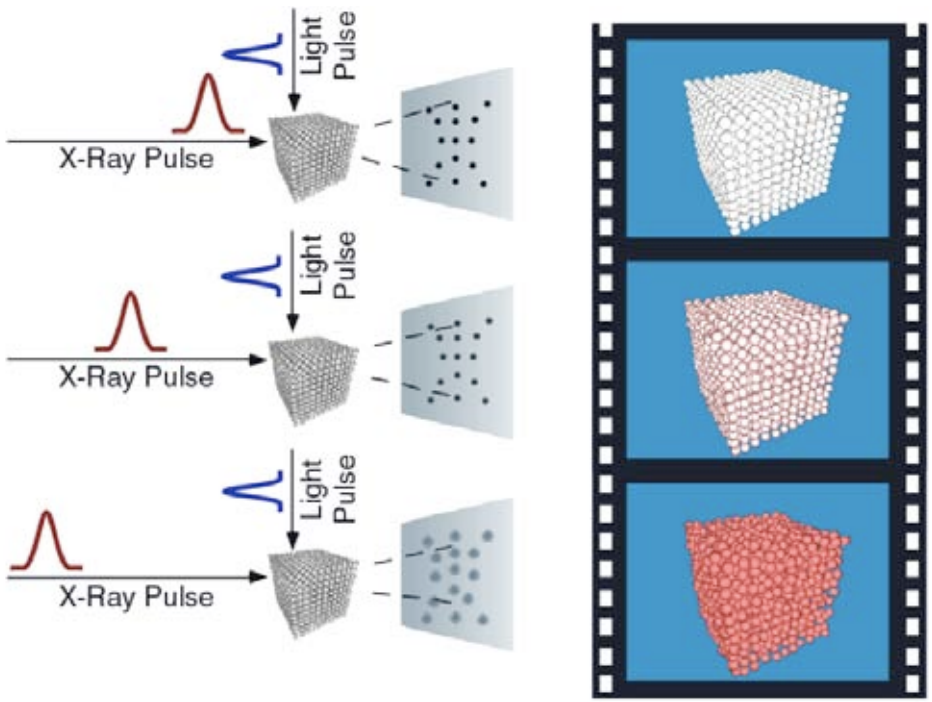

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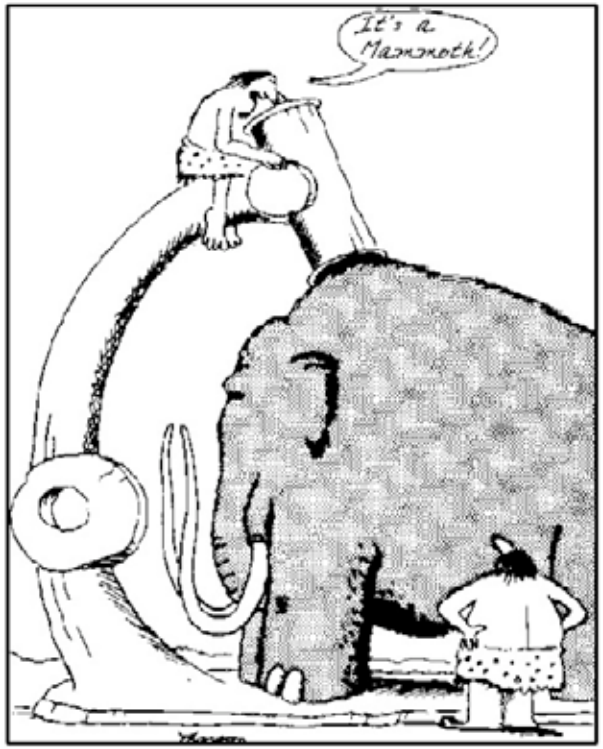
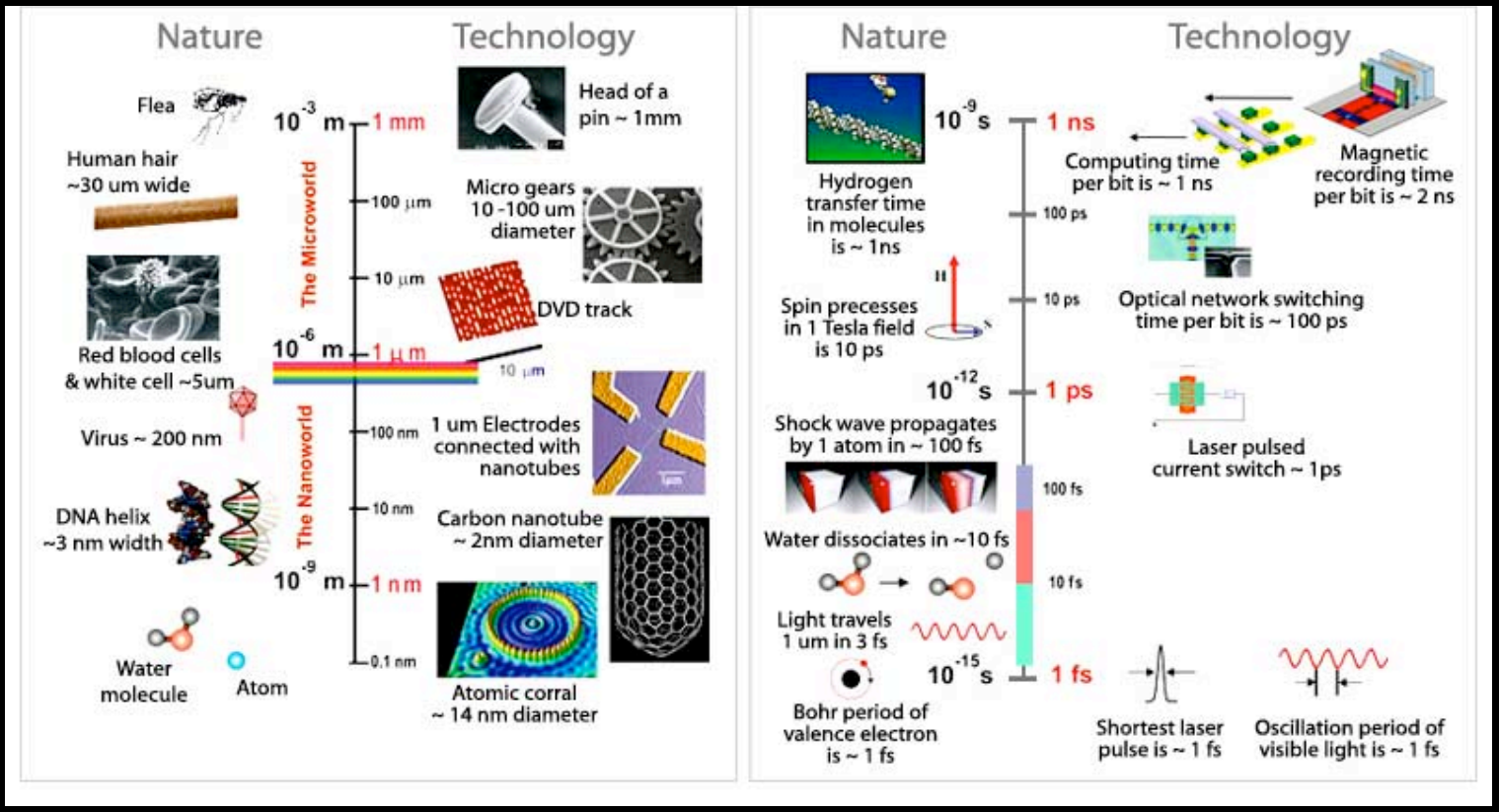


Early Microscope

Short length-scales implies ultrafast time-scales

Ultrasmall

Ultrafast



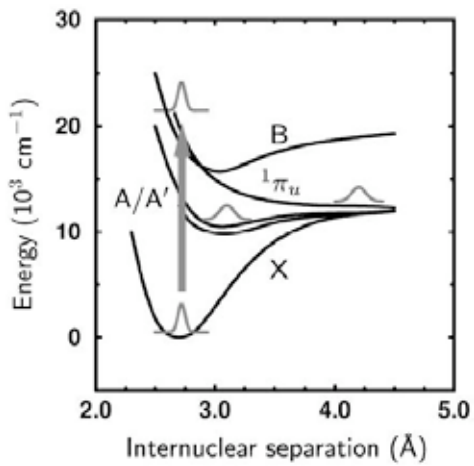
Short-pulse x-rays allow one to measure both technologically relevant length-scales and technologically relevant time-scales.

Goal is to elucidate the atomic and nanoscale dynamics underlying material transformations

-

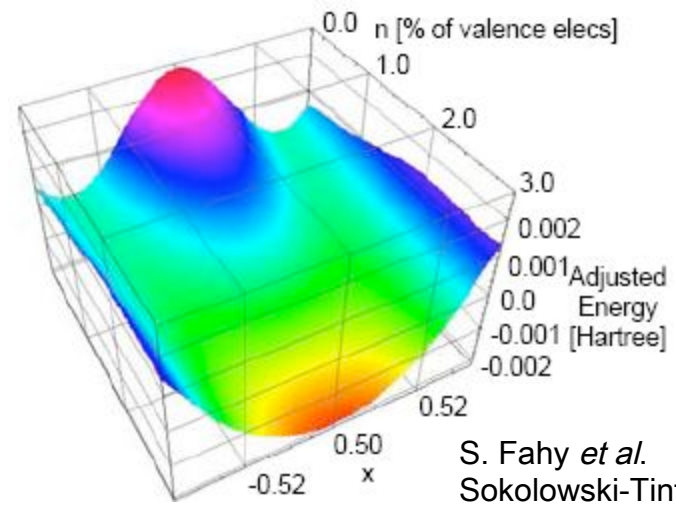
Mapping the forces on atoms

Molecular dissociation



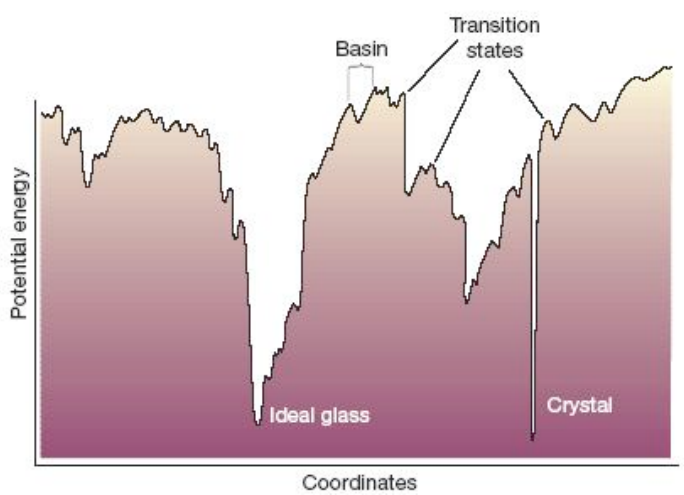
Neutze *et al. PRL*. **87** 195508 (2001)

Displacive excitation



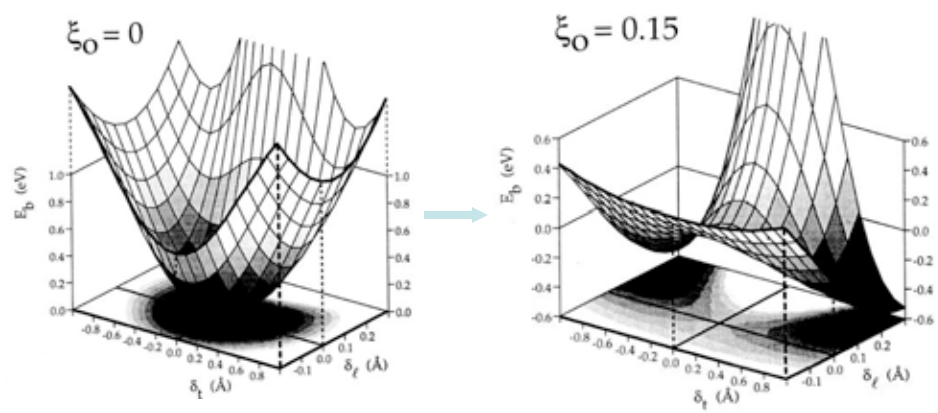
S. Fahy *et al*.
Sokolowski-Tinten *et al.*

Liquid/Glass dynamics



Debenedetti *et al*. Nature **410**, 259 (2001)

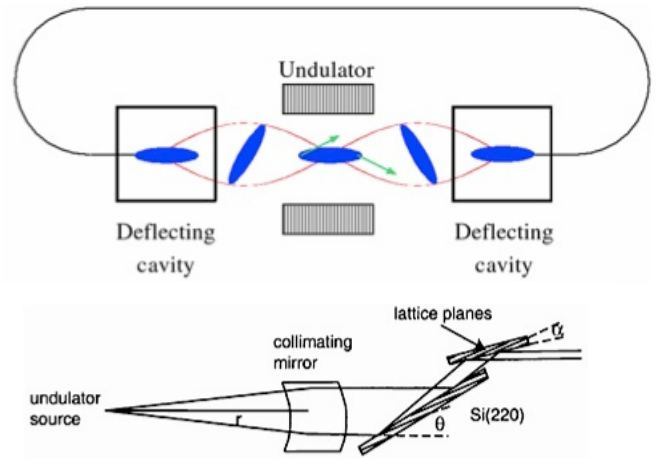
Melting transitions



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

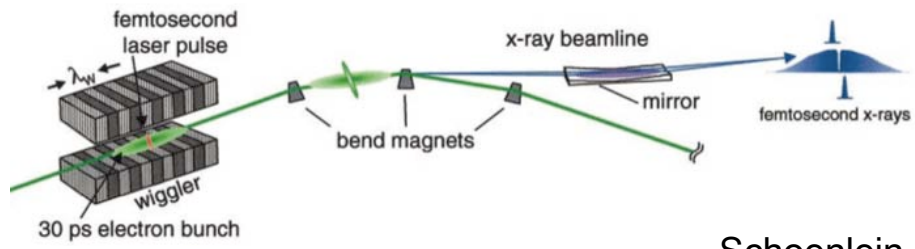
Going to shorter time-scales at synchrotrons

Crab cavities (APS) (5 ps, 100 Hz)



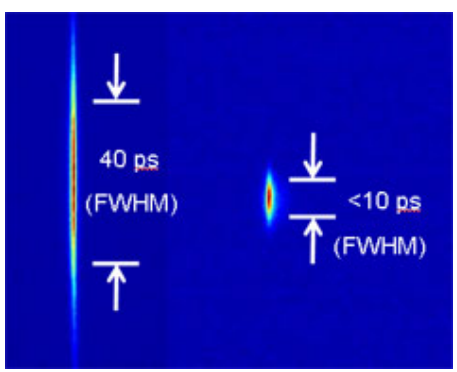
Zholents et al., NIMA (1999)

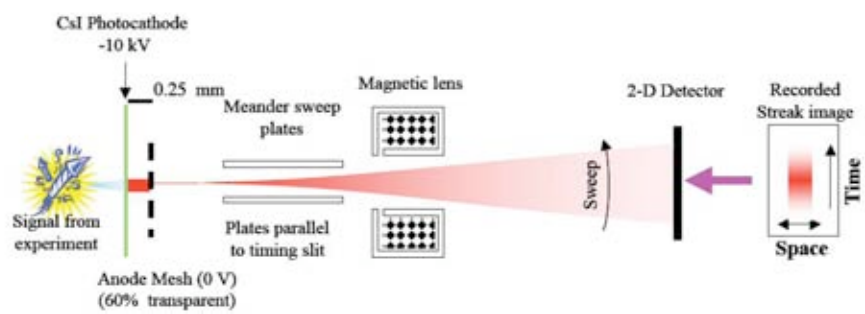
Slicing (ALS)
(100 fs, 20 kHz)



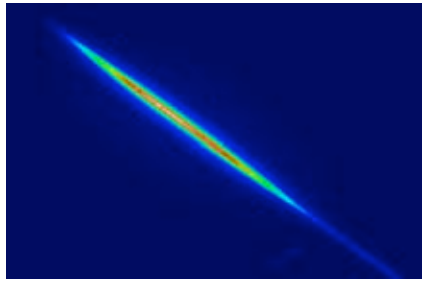
Schoenlein et al.
Science (2000)

Low alpha mode (SPEAR3)
pulse durations < 1 ps feasible
at full rep-rate

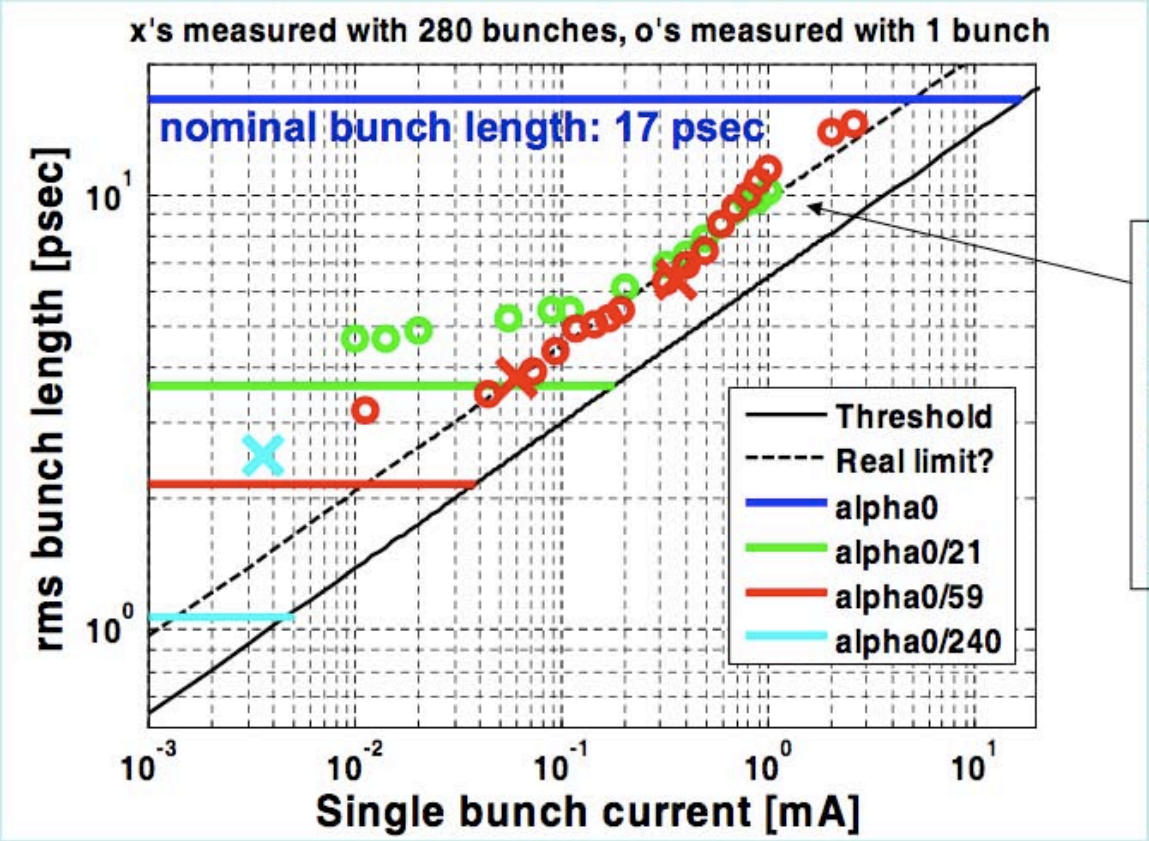




Streak cameras
(1 ps resolution
1-10 kHz)



SPEAR3 measured bunch length vs. current



CSR microbunch instability threshold defines bunch length for large bunch current.

Theory: Stupakov and Heifets, PRST-AB, May, 2002.

- Small-α, minimum bunch length:

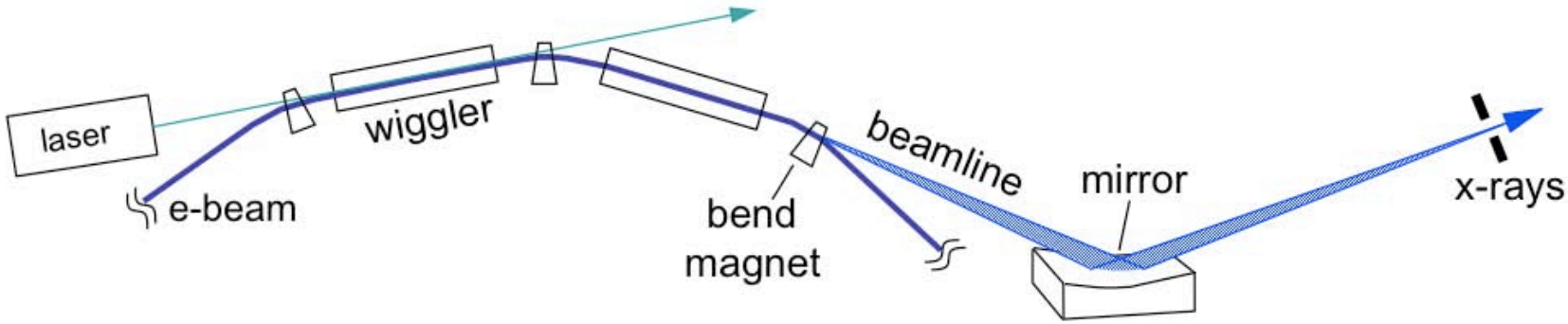
$$\sigma_{\min}[\text{psec}] \approx 9.7 * I_{bunch}^{1/3}[mA]$$

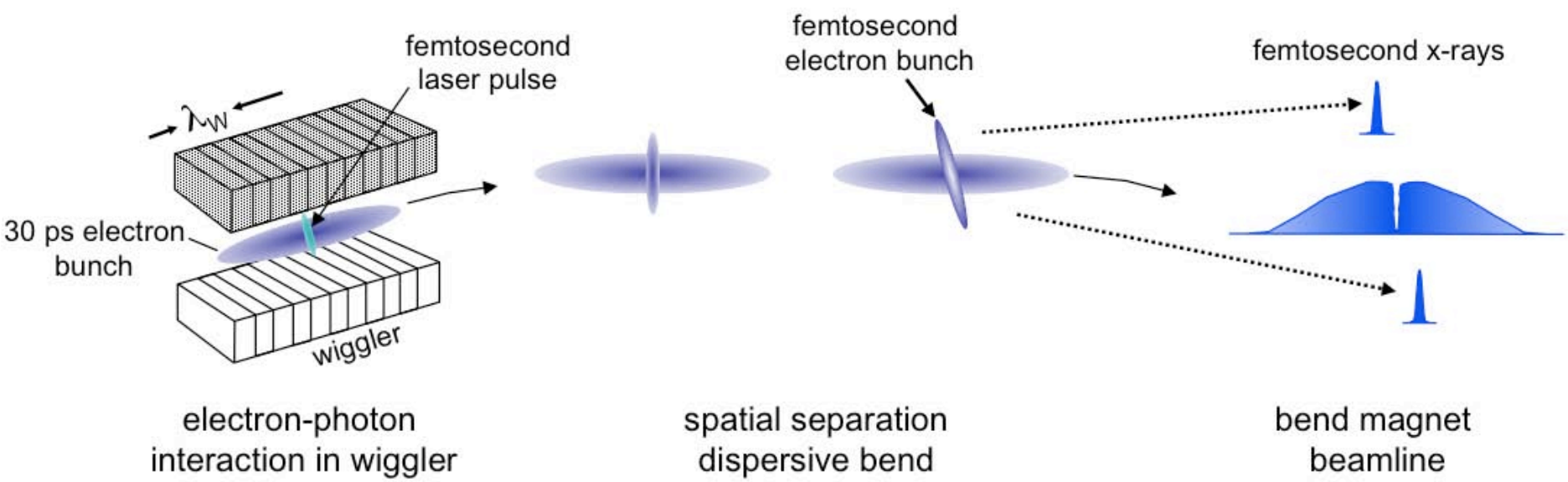
I_{total} [mA]	100	17	0.28	0.028
I_{bunch} [μA]	357	61	1	0.1
$\sigma_{\min}$ [psec]	6.9	3.8	1.0	0.45

$I_{total} = 280 * I_{bunch}$

From J. Safranek

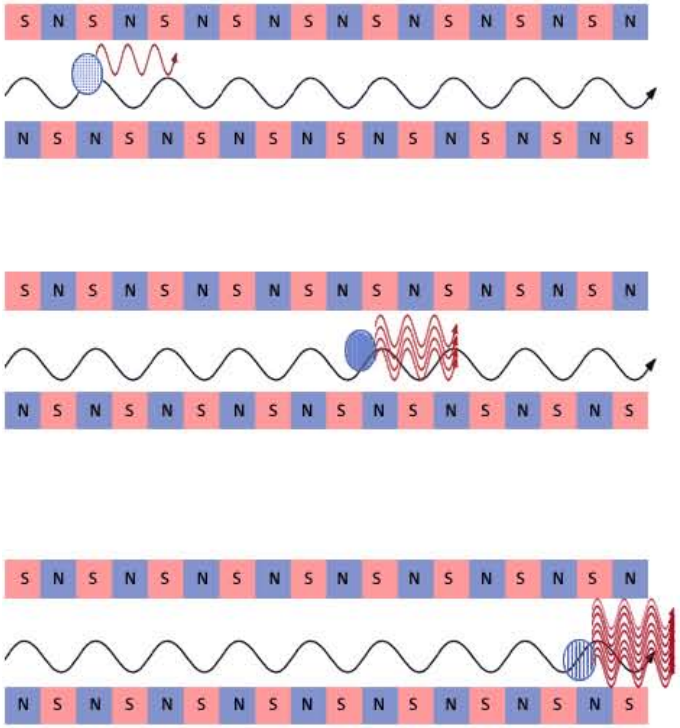
Slicing femtosecond pulses from long electron bunches

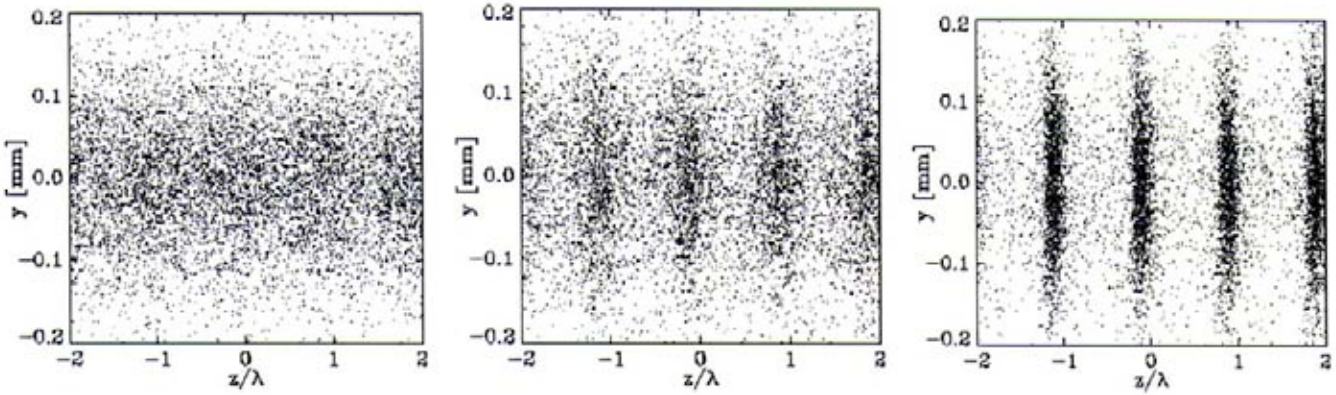




Zholents and Zolotorev, *Phys. Rev. Lett.*, **76**, 912,1996.

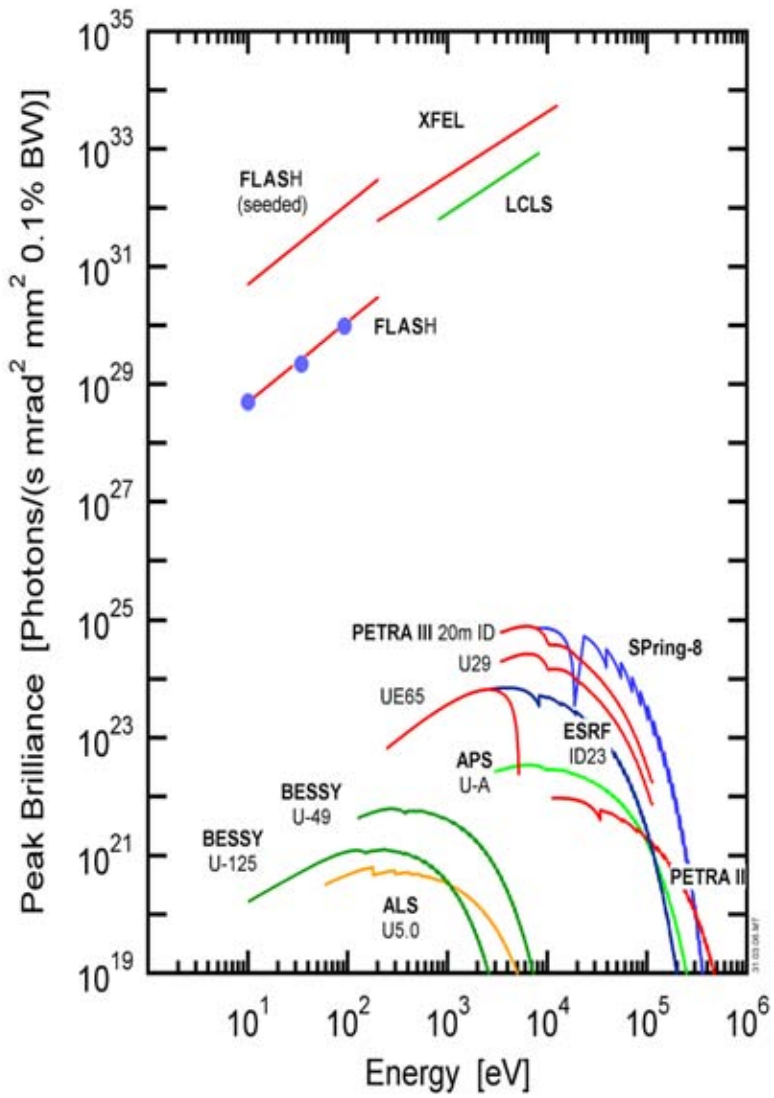
Free Electron Lasers and SASE Process



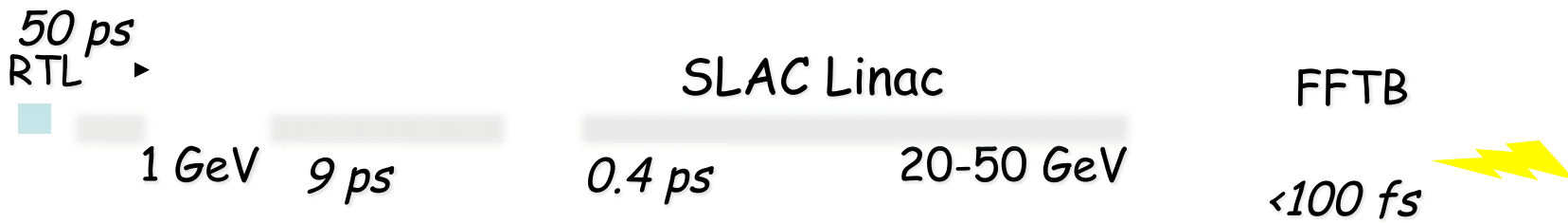


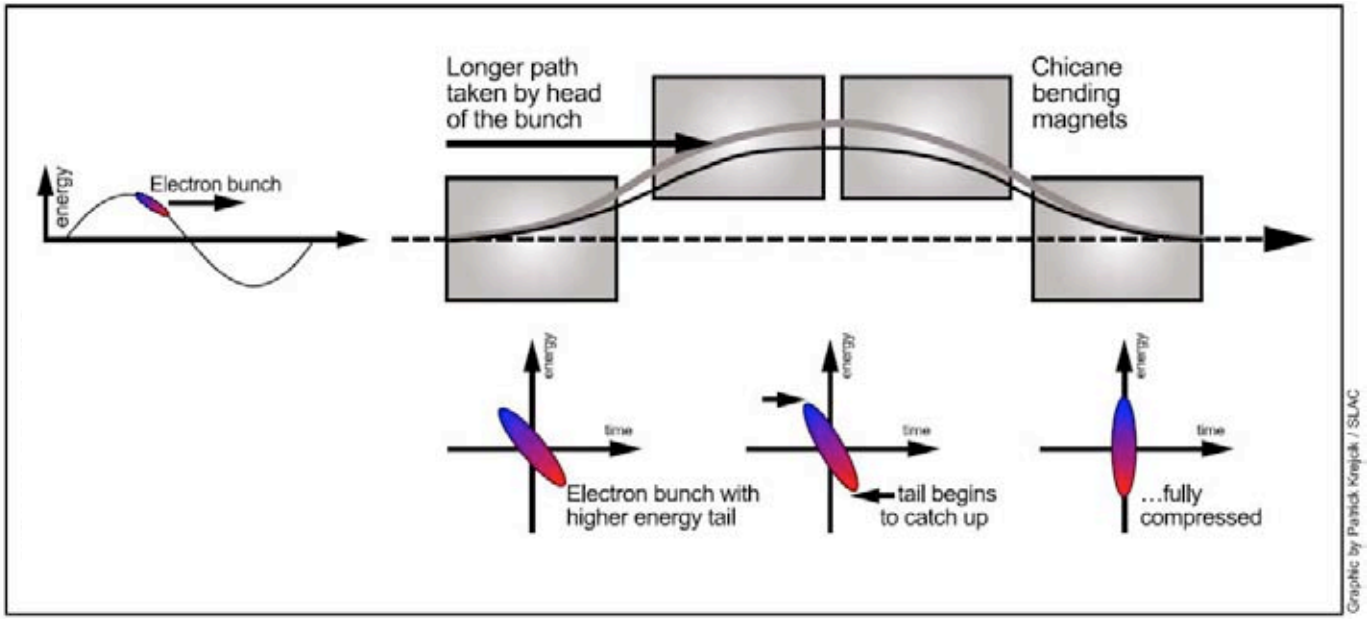
FLASH in Hamburg

Wavelength range (fundamental):
13-48 nm (90eV-25eV)
~ 6nm (200 eV in fall 2007)
FEL third harmonic:
4.6 nm (270 eV)
Pulse energy:
up to 65 µJ (average)
120 µJ (peak)
Peak power (fundamental):
> 4 GW
Average power (fundamental):
up to 30 mW
Pulse duration (FWHM):
7-50 fs
Spectral width (FWHM):
0.5-1 %
Peak brilliance:
up to 5×10^{29}

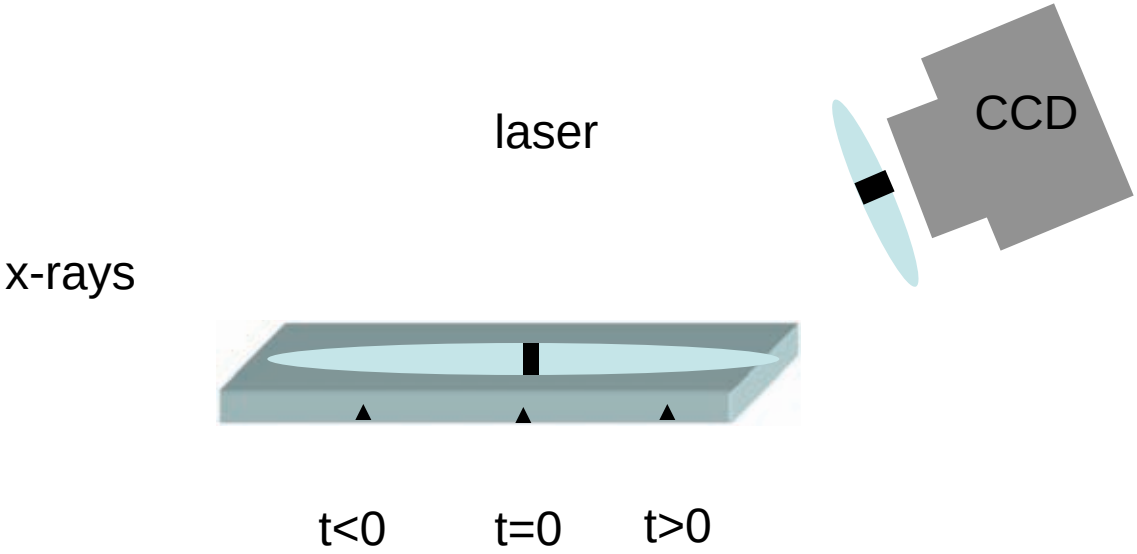


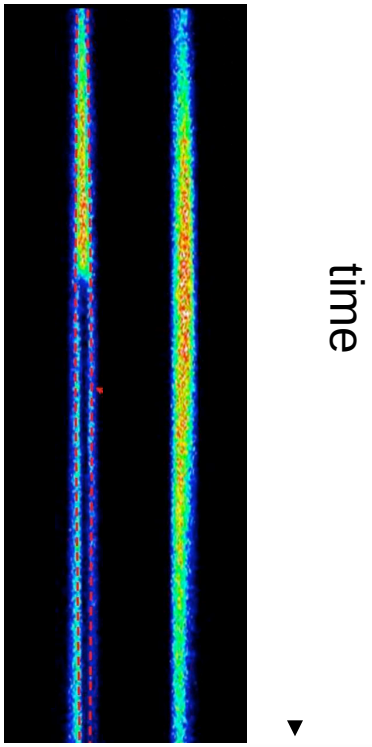
ecb□nSd□□FsTEbsT, a□SpEba□nTS.sb□nI I n□

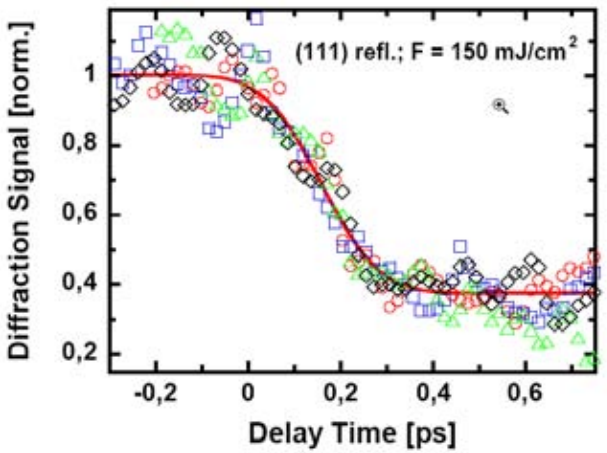




Example: Electrostatic interaction: How long does it take for a crystal to turn into a plasma? Where are the atoms going?

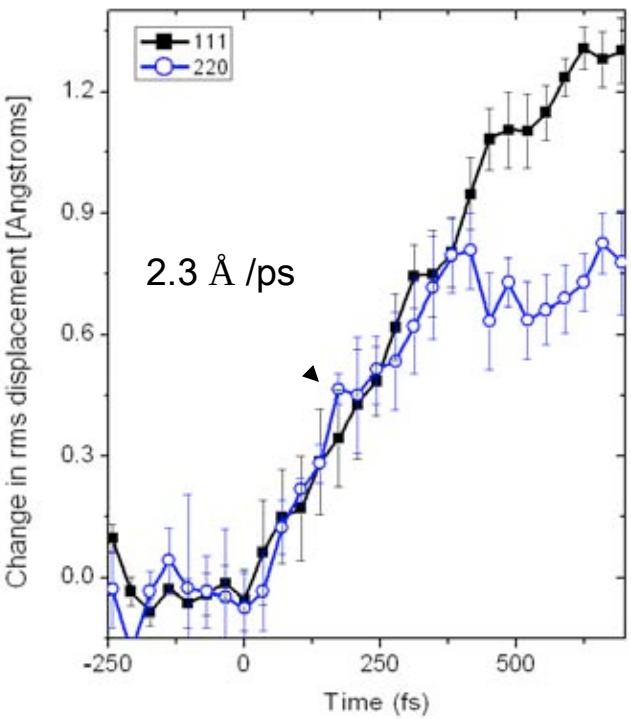


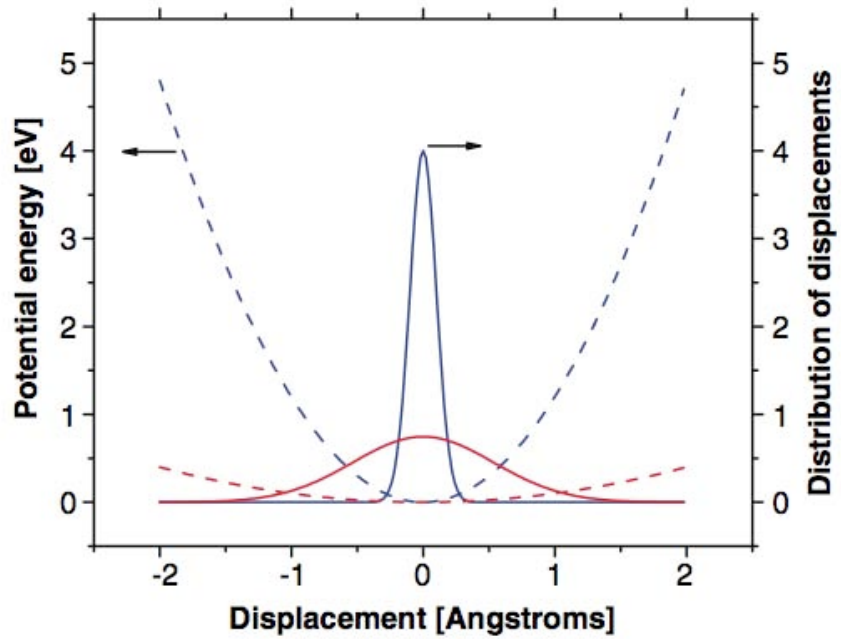




- [illegible]

Capturing the transition state: Ballistic motion of atoms on a softened potential energy surface

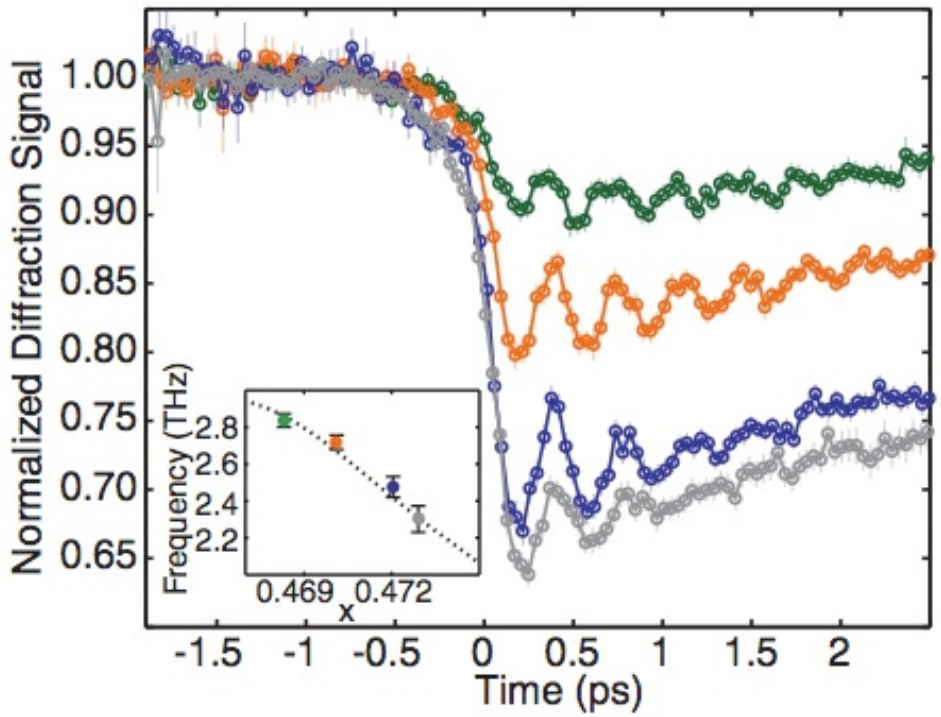


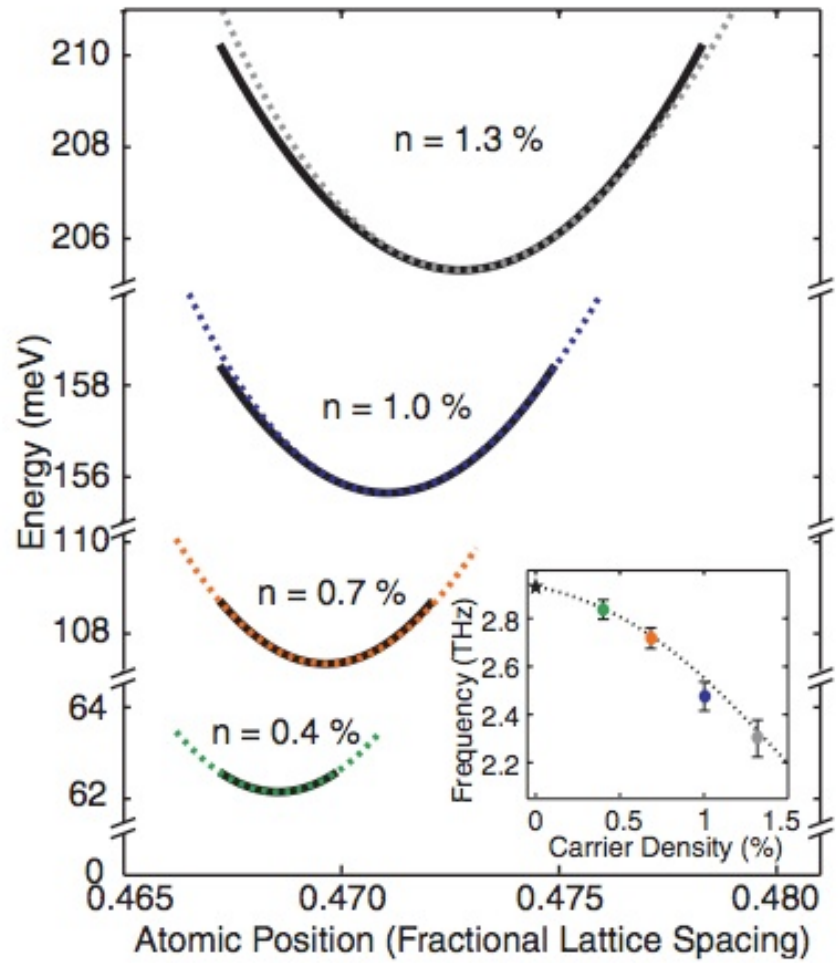


$$\sqrt{\frac{3k_BT}{M}} \sim 2.5\mathring{A}/ps$$

Lindenberg et al., Science **308,** 392 (2005)

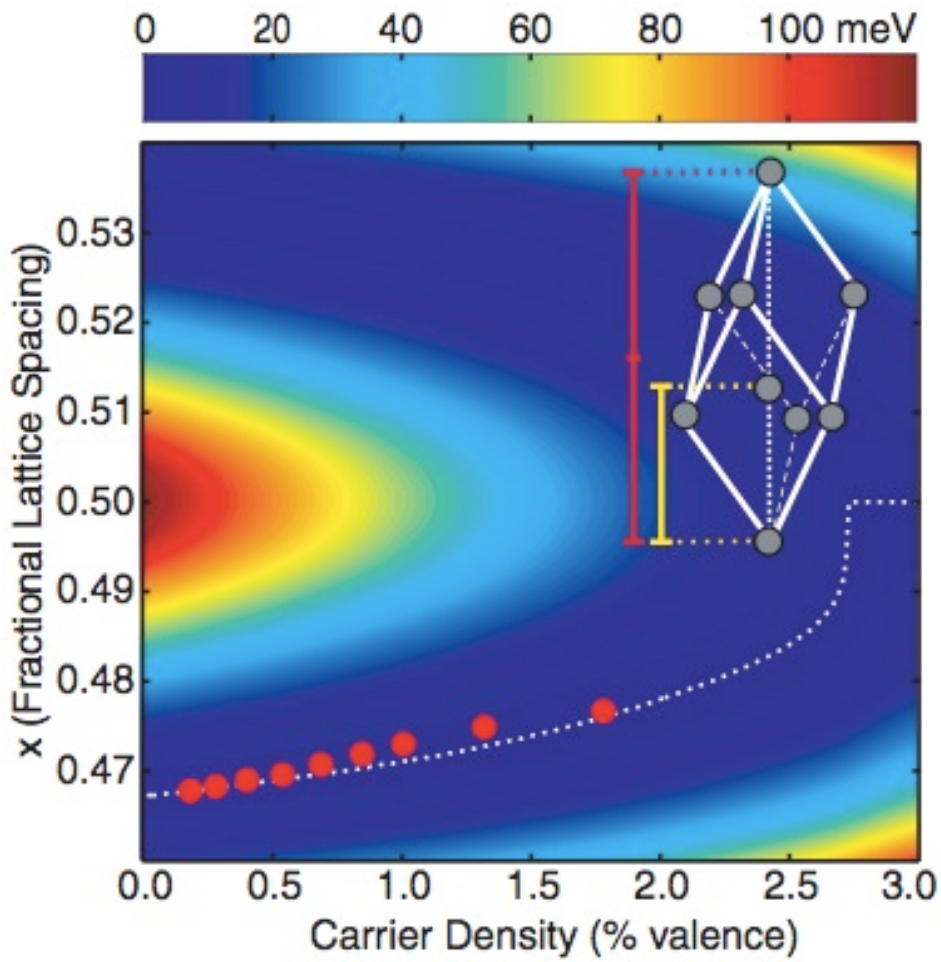
Example 2: Optical Phonon Dynamics in Bismuth



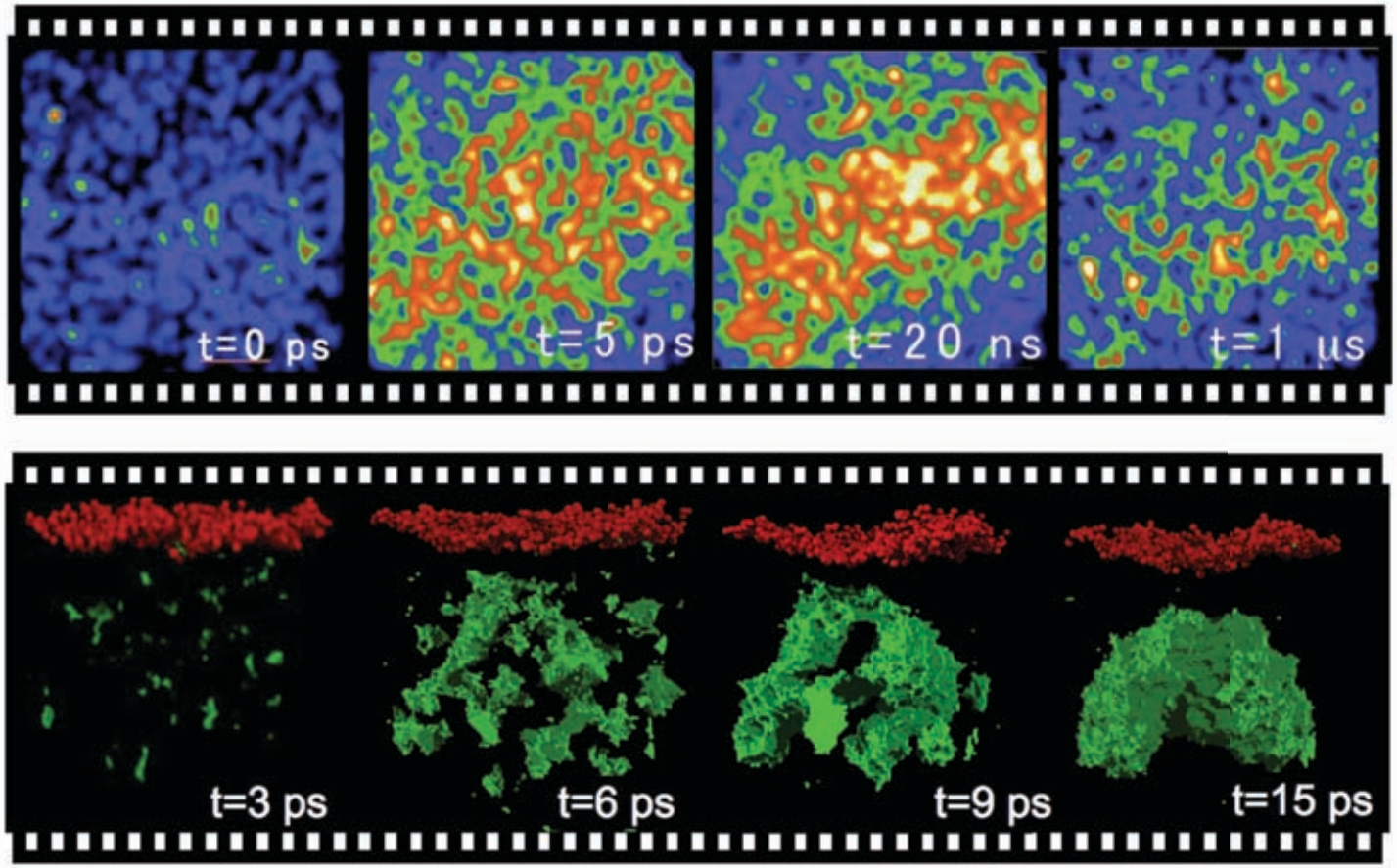


D.M. Fritz et al., Science **315**, 633 (2007)

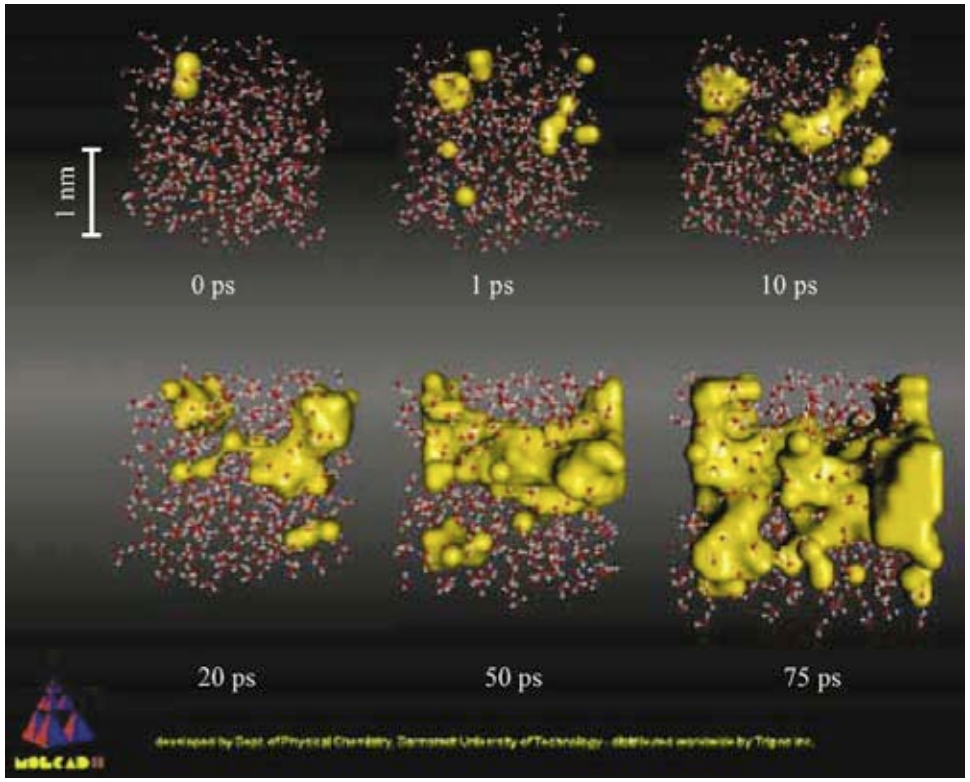
Comparison to excited state DFT calculations: Mapping excited state potential energy surfaces



Example 2: Imaging nanoscale, ultrafast, dynamics at the mesoscale

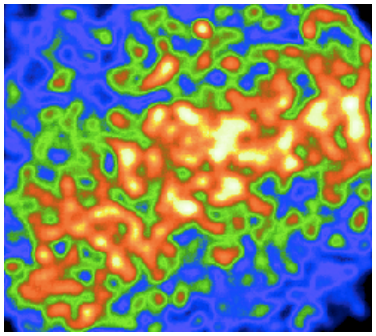


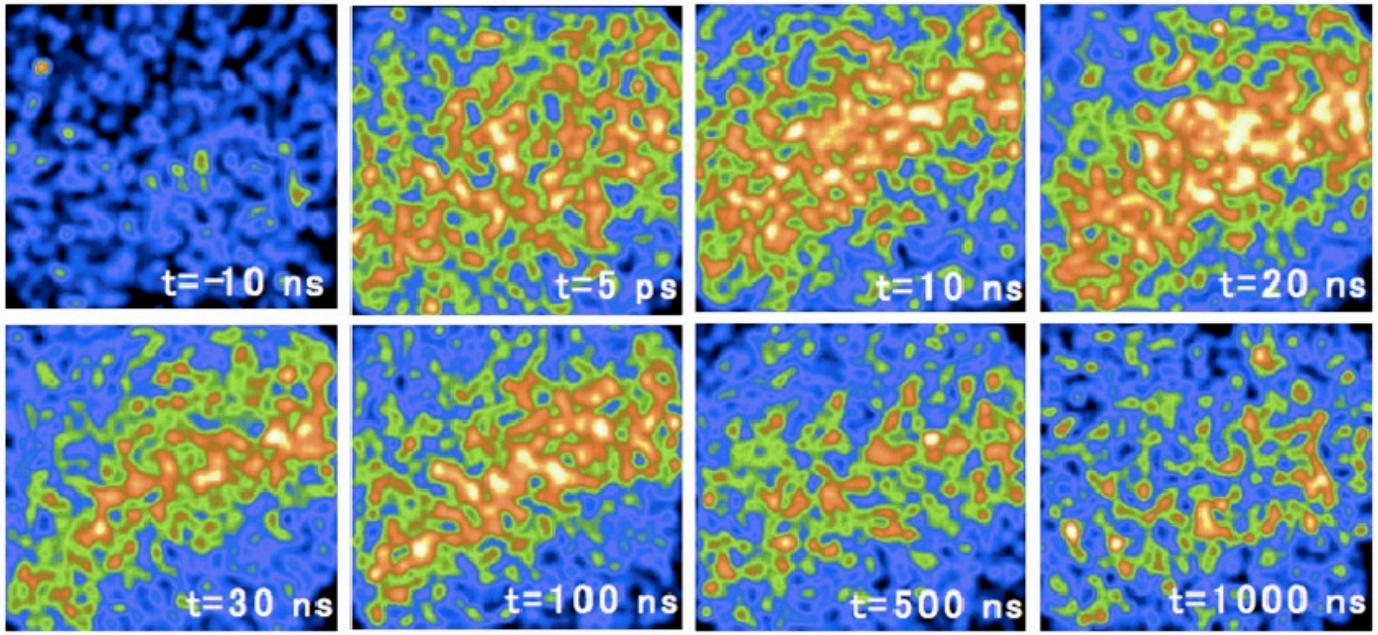
How does water boil?



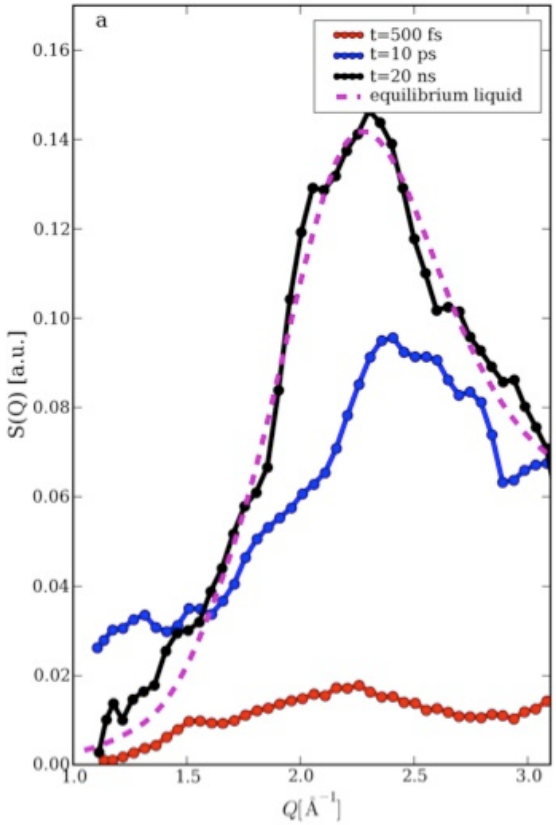
Zahn, PRL (2004)

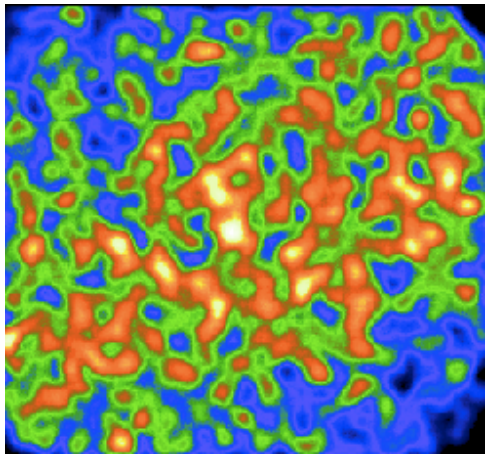
Snapshots of the growth and crystallization dynamics





Structure factor measurements of the optically-induced liquid state



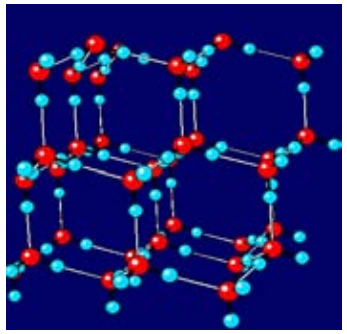


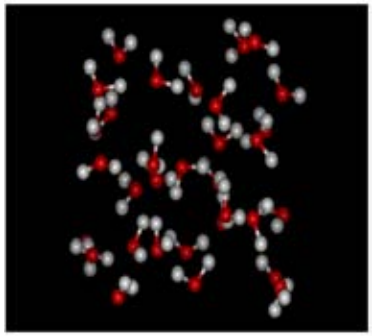
Observation of transient increase in small angle scattering

Simple model for short time observed behavior:

•At short times, the transition occurs at constant volume.

•A transient liquid with lower density than the equilibrium liquid will be formed





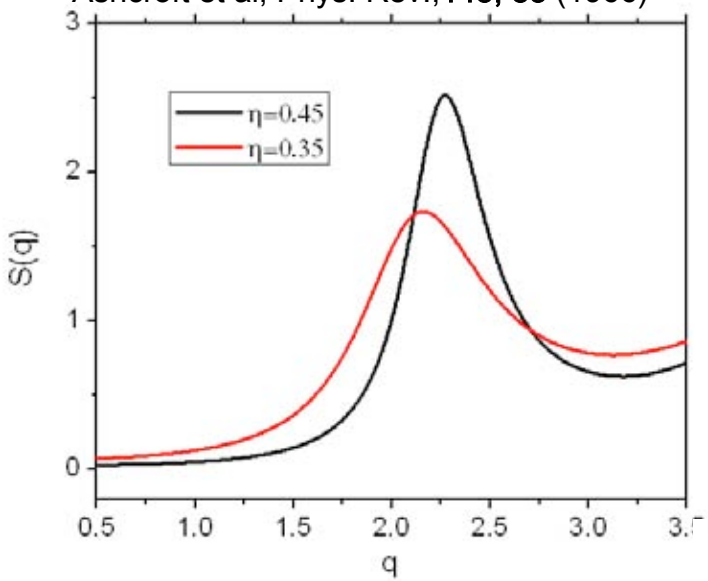
Increase in scattering at low q for liquid densities smaller than the equilibrium density ◂ ▸ Increase in compressibility, formation of softer material

$$\lim_{q \to 0} S(q) \sim \chi_T$$

•A supercooled low-density liquid

The structure factor as a function of packing density

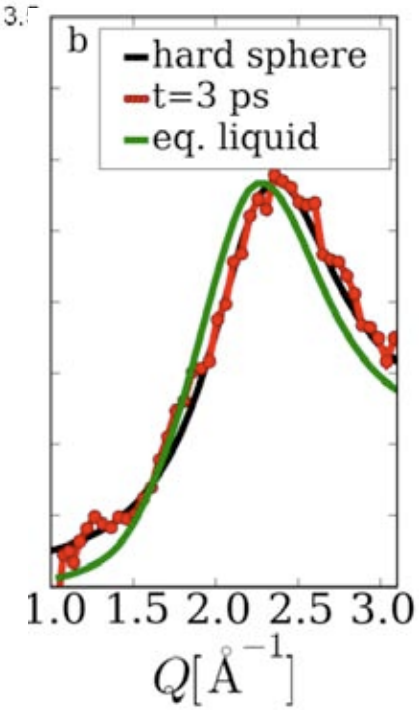
Ashcroft et al, Phys. Rev.,**145, 83** (1966)



$$\eta = \frac{\pi}{6} n \sigma^3$$

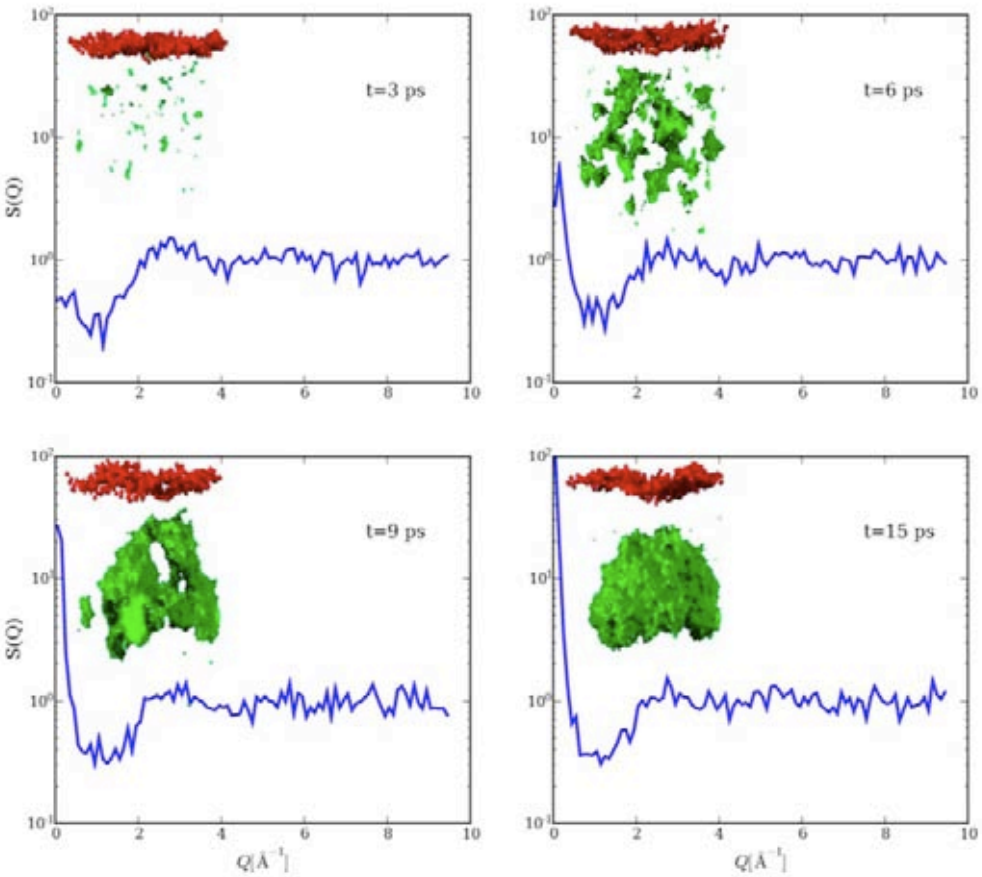
$$\eta_{diam} = \frac{\sqrt{3}\pi}{16}$$ =0.35 (short time packing density)

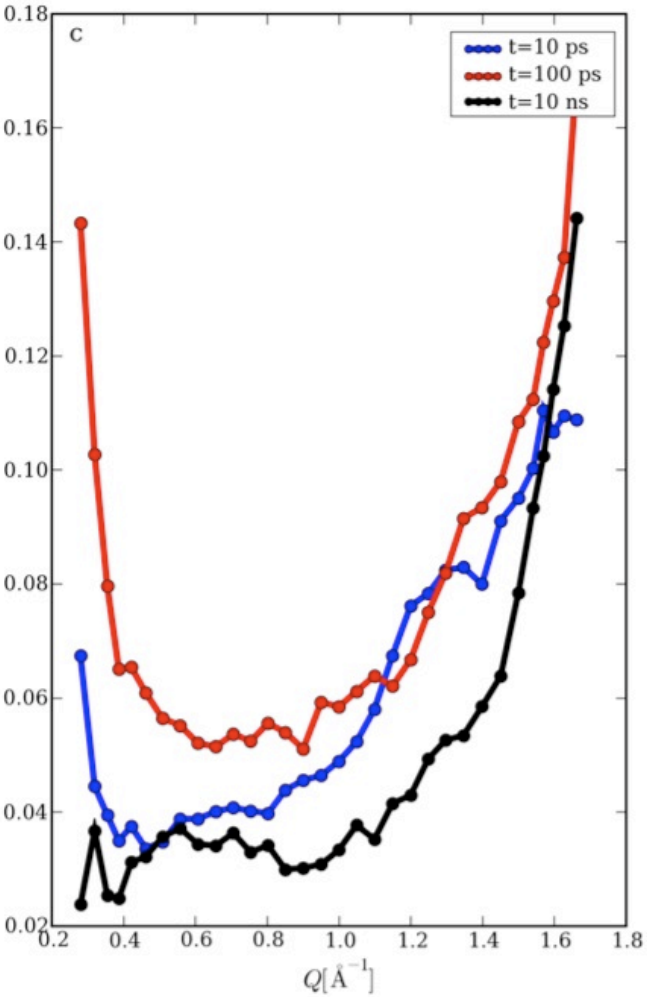
η(equilibrium liq) = 0.45 (long time packing density)



Observation of nanoscale nucleation phenomena and comparison to MD simulations of the ablation process

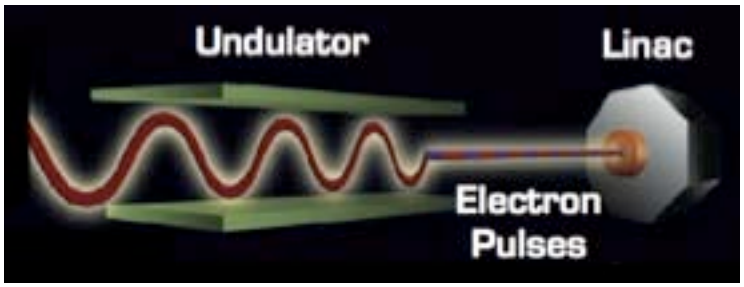
MD Simulations (Lorazo et al):

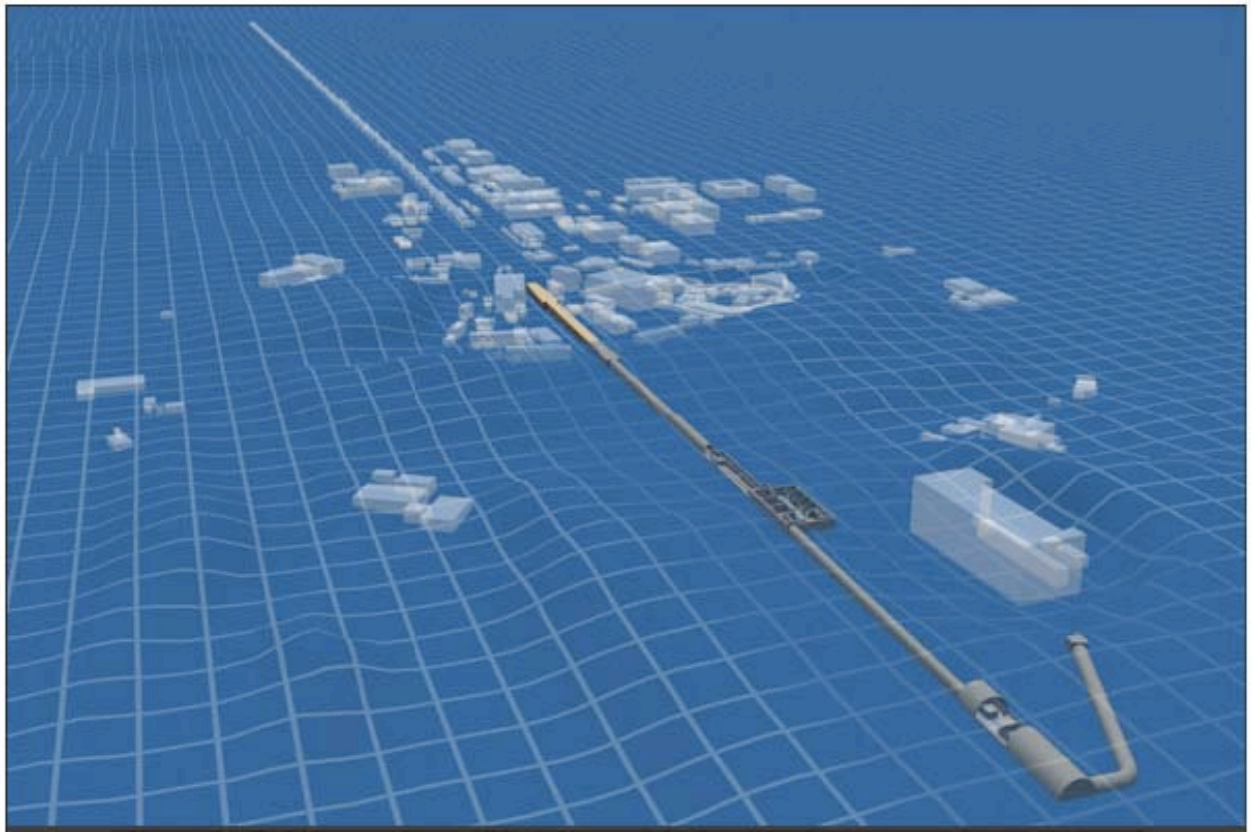




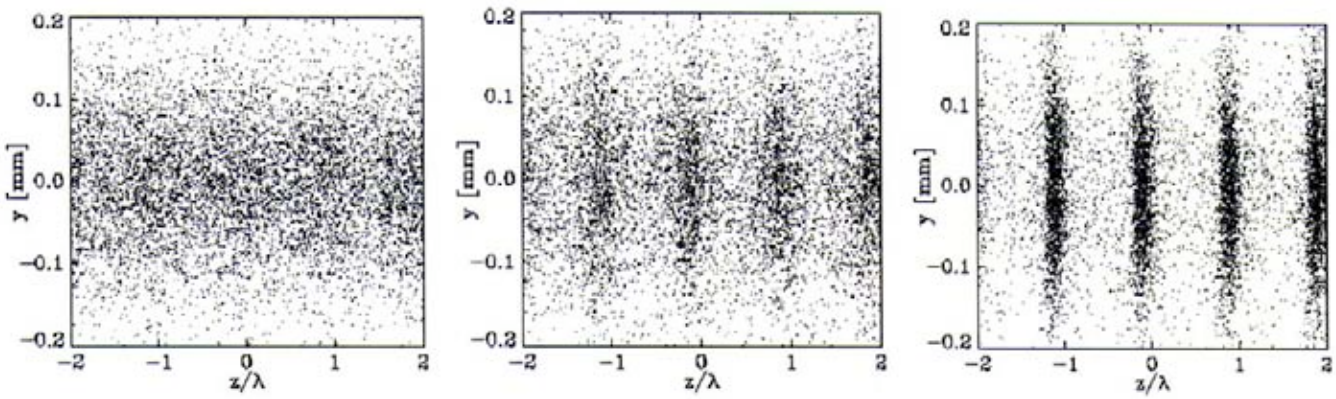
Phys. Rev. Lett. 100, 135502 (2008)

New Opportunties at the Linac Coherent Light Source





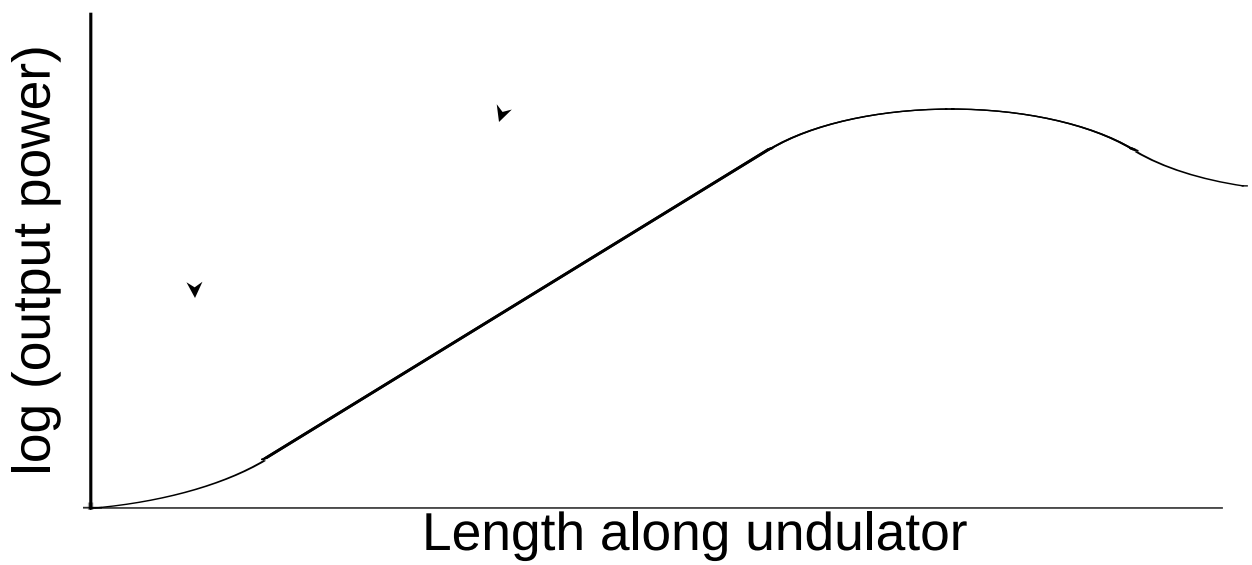
Self-Amplified Spontaneous Emission (SASE)



Undulator regime (unbunched)

Exponential gain regime (bunching)

Saturation regime



LCLS parameters:

10^{12} photons/pulse at 120 Hz rep-rate
800 eV – 8 keV photon energies
transversely coherent

1 mJ in 100 fs = 10^{10} W (total US energy consumption rate is about 10^{12} W)

focused to 1 um: 10^{18} W/cm2 (roughly the earth's solar flux focused to the head of a pin)

The Linac Coherent Light Source Construction Project

SLAC Research Yard

Near Hall Pump-Probe X-ray Hutch

LCLS undulator hall

LCLS Far Experimental Hall

First LCLS Call for Proposals

Experiments with soft X-rays at the AMO station

Closing date: September 1, 2008

The Stanford Linear Accelerator Center SLAC is currently building the world's first hard X-ray free-electron laser, the Linac Coherent Light Source (LCLS) (http://lcls.slac.stanford.edu), which will start operation in summer 2009. LCLS will produce intense, sub-picosecond pulses of spatially-coherent X-rays. In a flash of about 100 femtoseconds duration, LCLS will provide 10^{12} X-ray photons, roughly as many photons as obtained in one second at today's best storage-ring-based synchrotron radiation facilities. LCLS X-rays will enable investigation of systems at the atomic and nano-scale under conditions where the matter is far from equilibrium, and actually undergoing real-time reactions.

Four experimental stations are currently being constructed to serve the following scientific thrust areas:

1. Atomic, molecular and optical science (AMO)
2. Coherent imaging of non-periodic objects (CXI)
3. Coherent scattering of nano-scale fluctuations (XCS)
4. Diffraction studies of stimulated dynamics (XPP)

Two more stations are currently being planned to serve:

5. High energy density science (HED)
6. Investigation of materials with soft X-rays (SXR)

It is the goal of LCLS to attract a large number of users from diverse fields and to enable a broad set of important experiments that take advantage of the unique capabilities of LCLS.

The SPPS Collaboration

SLAC/SSRL
DESY
APS
UC Berkeley
University of Michigan
NSLS
Uppsala
ESRF
Copenhagen University
University of Lund
LLNL
Max Plank Institute
Oxford University
University Duisburg-Essen