Probing Complexity in Nonequilibrium Materials Processing

(Preliminary Version)

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Complexity in Nonequilibrium Materials Processing

- **Nonequilibrium processing is in a sense redundant, but emphasizes the often far from equilibrium conditions used, e.g. extreme conditions**

- **Developing materials with desired properties outside known envelopes has long been through the discovery of new and innovative processing methods guided by intuition and the accumulated understanding of physical principles**

- **Discovery is great when it happens, but it relies on serendipidy and becomes increasingly rare in complex and mature technologies**

- **The nature of this process is not likely to change dramatically; however, the level of understanding of the underlying physical principles required for continued technological advances increases inexorably**

Probing Complexity associated with Nonequilibrium Materials Processing

- Probing materials processing steps involving significant levels of complexity within ultra-short times, strong deformation, and strong time, pressure, or temperature gradients is likely to require high temporal resolution, high spatial resolution, or both.

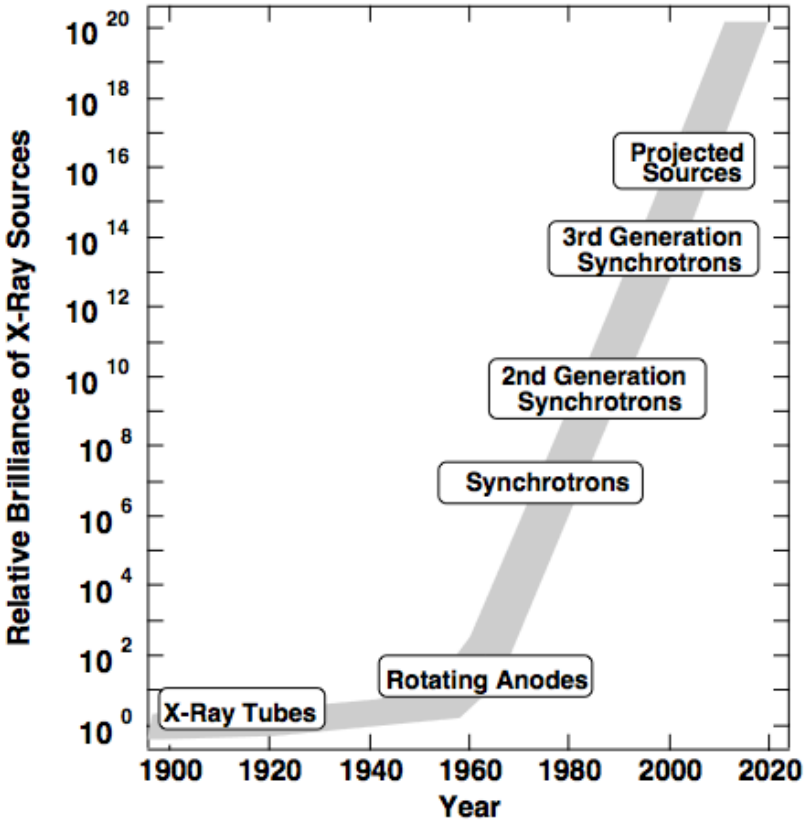
- Understanding complexity to the extent needed to predict or control desired materials processing results is likely to entail concommitant high fidelity, ultra-scale computational simulations and modeling as well as new and novel measurement capabilities

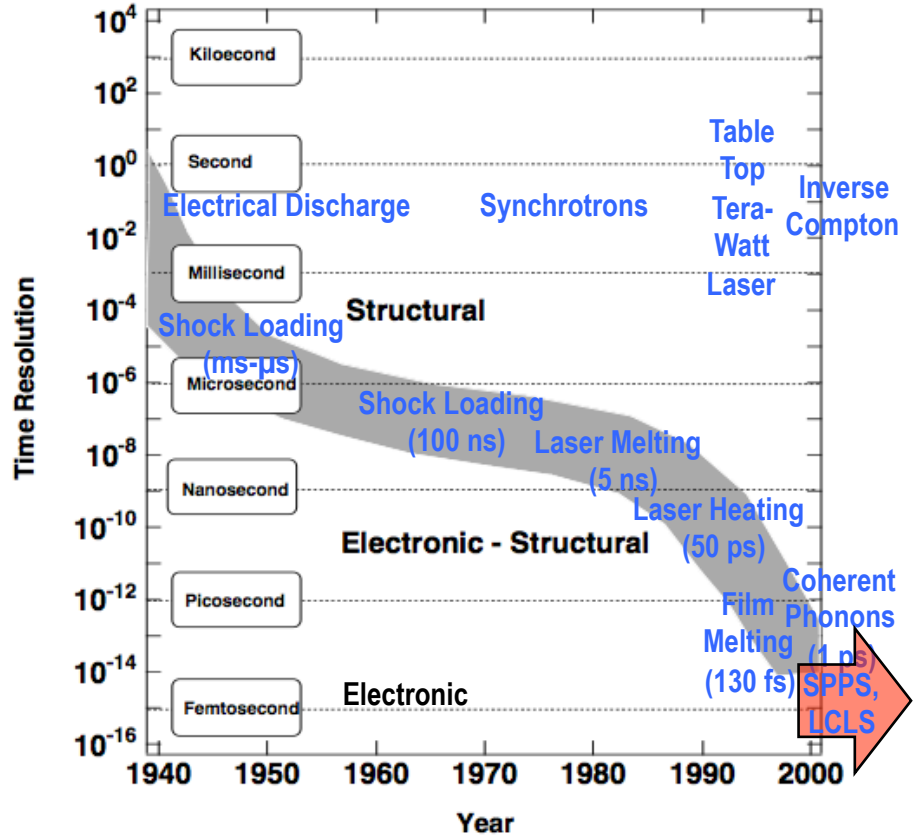
- This presentation will recall briefly x-ray source and time-resolution history

- The talk will consider nonequilibrium materials processing associated with:
  - Pulsed-laser processing/ablation
  - Pulsed-laser film deposition and growth
  - Plastic deformation and formation
  - Atomic dislacement cascade dynamics and evolution

Historical View of X-Ray Source and Time-Resolution Capabilities circa 2000

Structural Effects	Speed of Sound	~5 nm/ps
Electronic/Structural Effects	Electron-Phonon Interact.	<10 ps
Electronic/Atomic Effects	Lattice Vib., Bonding	~100 fs





Evolution & Control of Complexity

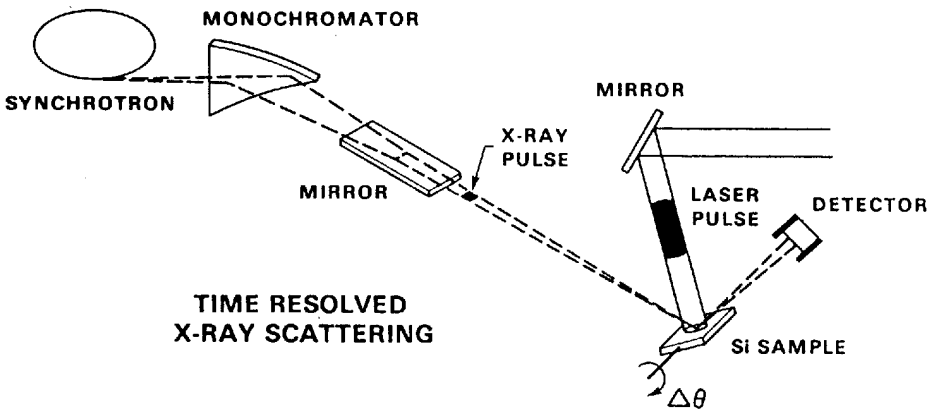
Pulsed Laser Processing:
Melting – Ablation – Deposition

- Pulsed laser heating and melting have been the subject of intense interest and remarkable progress on fundamental levels

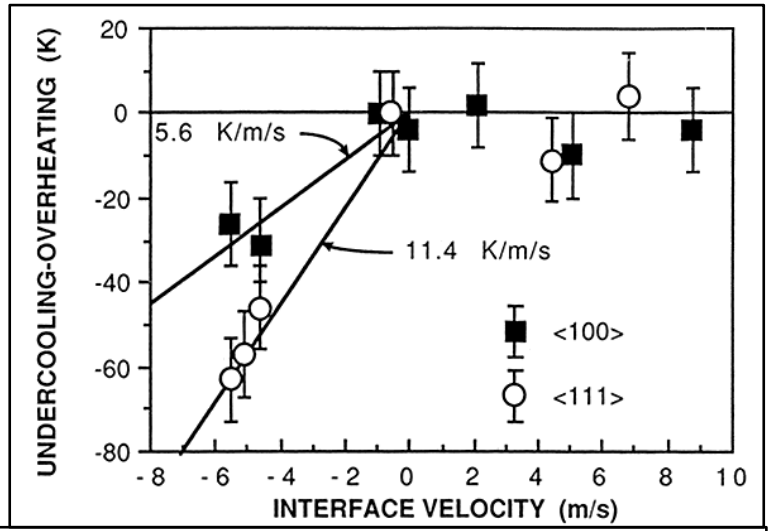
- The ablation process is both more complex and has received much less attention

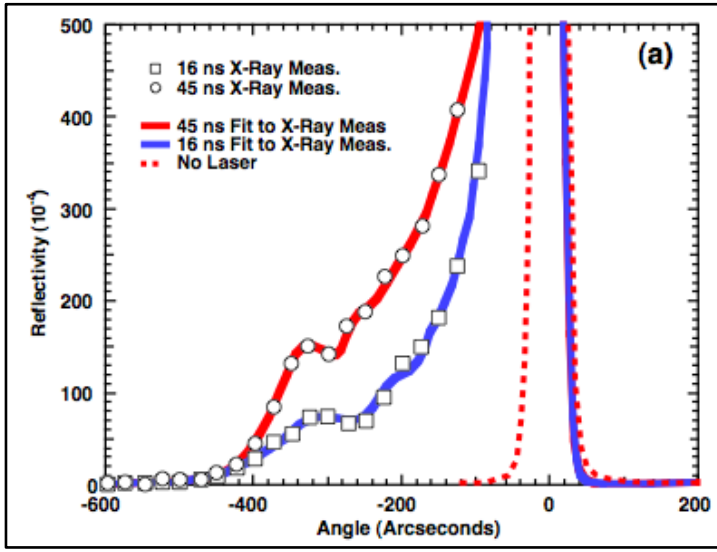
- Ablation is the basis of pulsed-laser deposition (PLD) film growth for many complex materials (e. g. oxides)

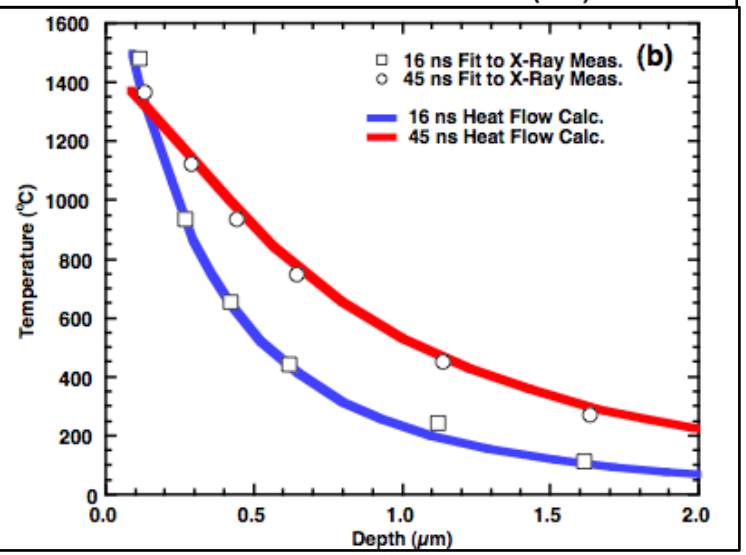
Time-Resolved (5 ns) Measurement of Interfacial Overheating and Undercooling During Pulsed-Laser Melting and Regrowth in Silicon



Larson et al., J. Mat. Res. 1, 144 (1986)

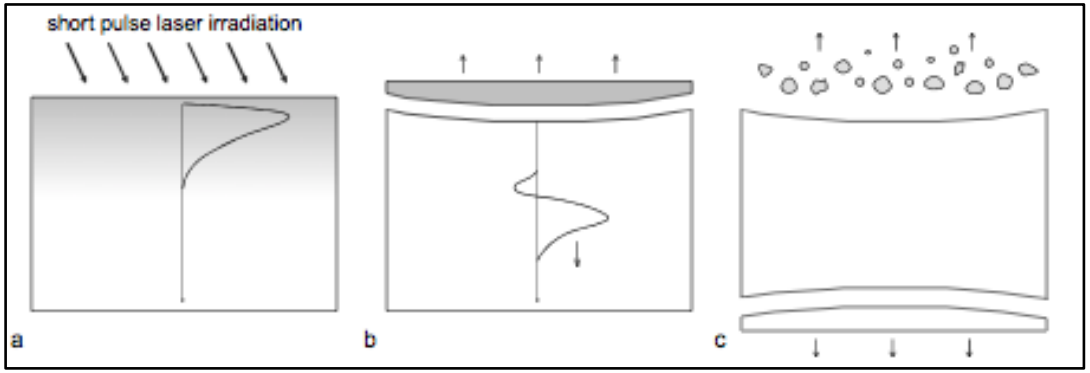






Rapid melting and refreezing with 1 J/cm^2, 25 ns laser pulses – with no ablation
Higher pulse energies and shorter pulse lengths leads to ablation

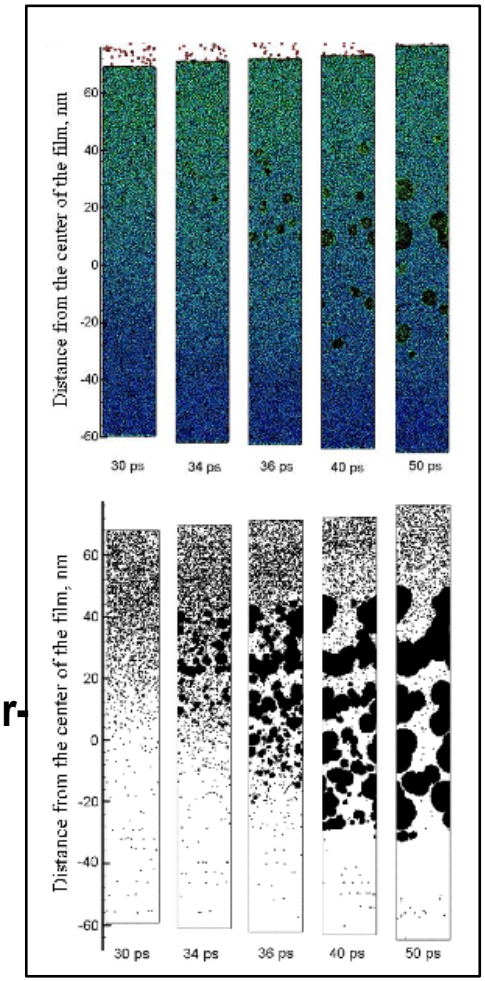
Modeling The Pulsed-Laser Ablation/Spallation Process



Schematic of an ultrashort laser induced pressure pulse penetrating into the surface of metal and ejecting a liquid surface layer.

The ablation/spallation process, the kinetic energy, the temperature, and clustering characteristics of the ejected material is critical for optimizing pulsed-laser deposition film growth

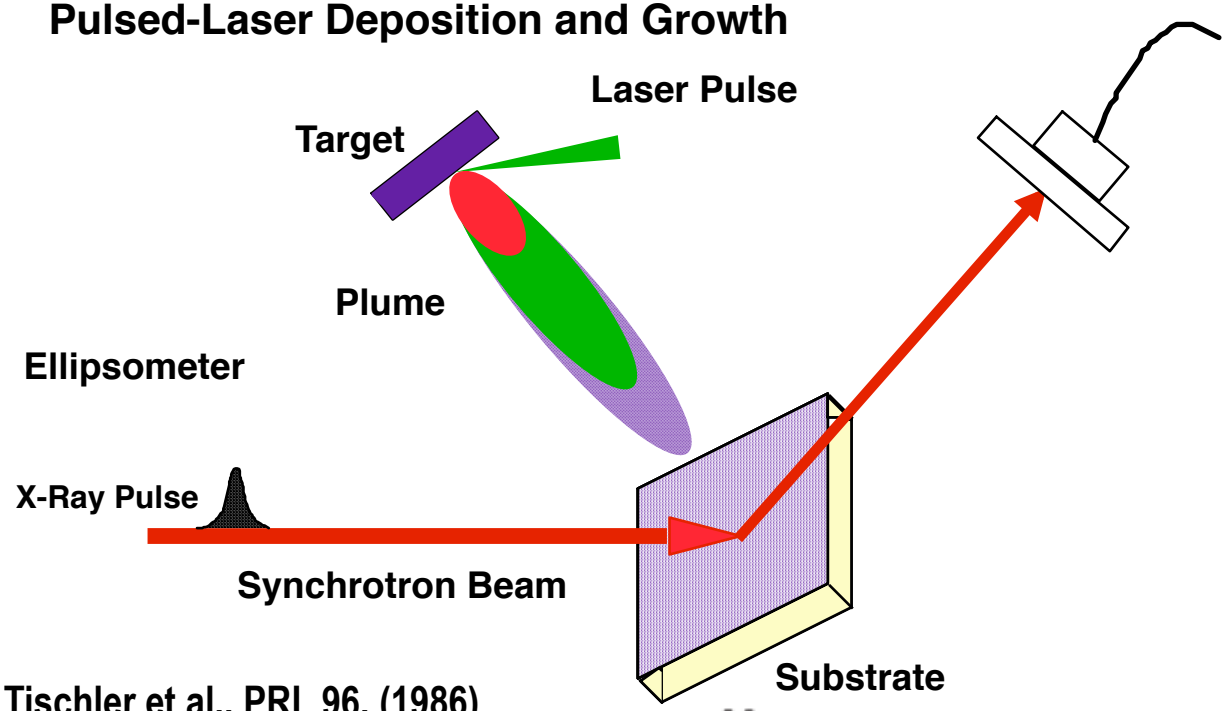
Molecular dynamics simulation of rapid laser-pulse heating and void nucleation and growth below the surface in Ni

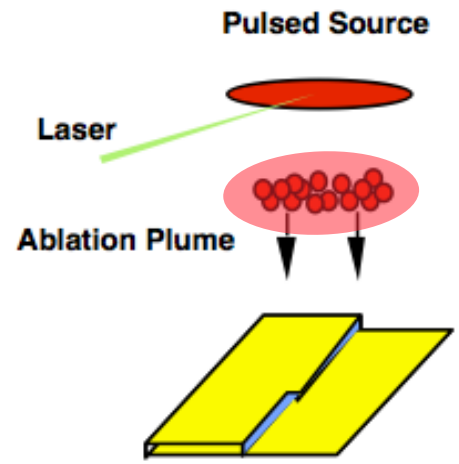


Zhigilei et al. Proc. SPIE 5448, 505 (2004)

Potential to Study Non-Equilibrium Pulsed-Deposition-Growth on Surfaces

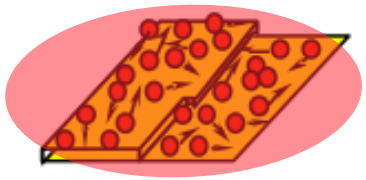
Pulsed-Laser Deposition and Growth



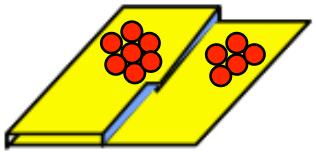


Growth Surface before Pulse

What happens during this few μs time duration as the plume arrives?

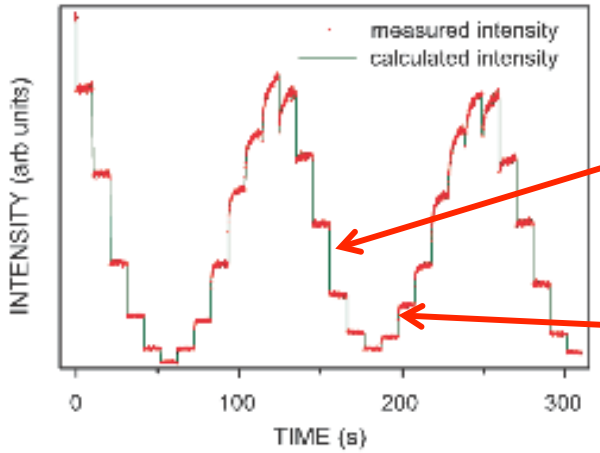


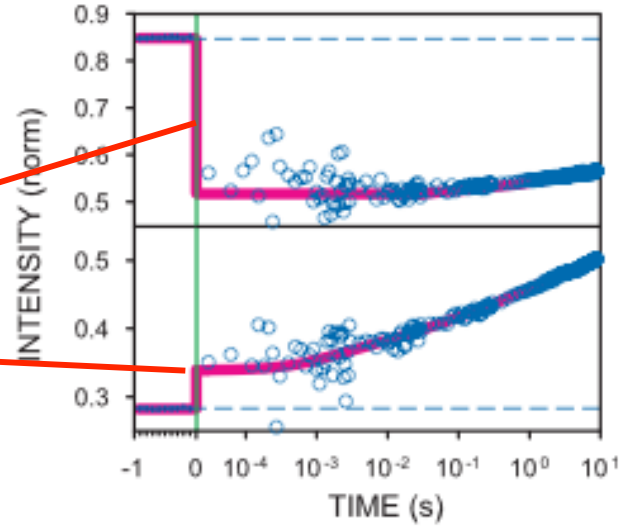
Growth surface as plume arrives



Growth surface after plume subsides

Tischler et al., PRL 96, (1986)



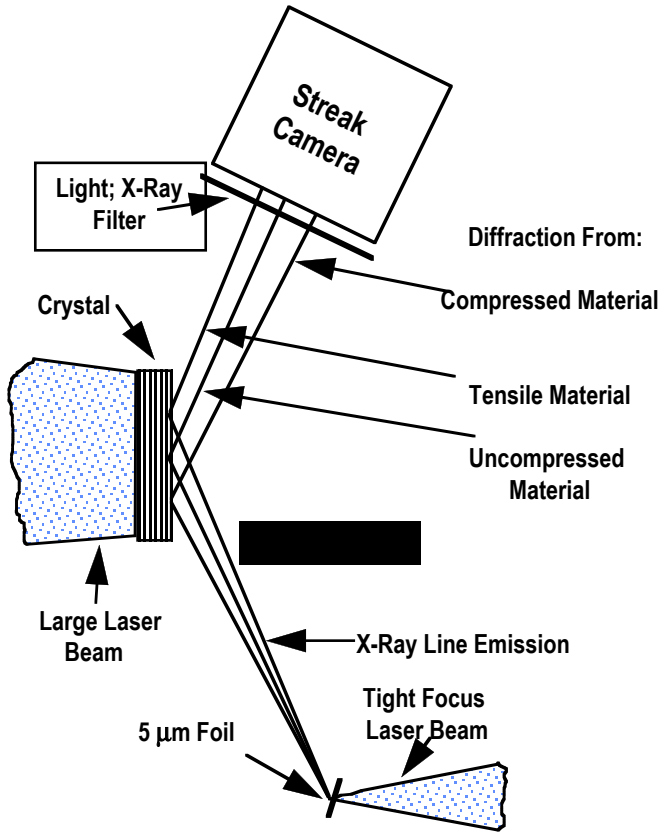


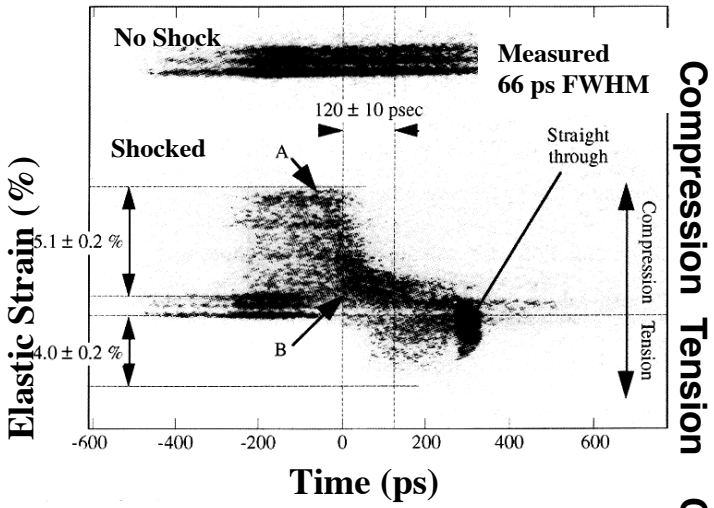
Deformation and Formation

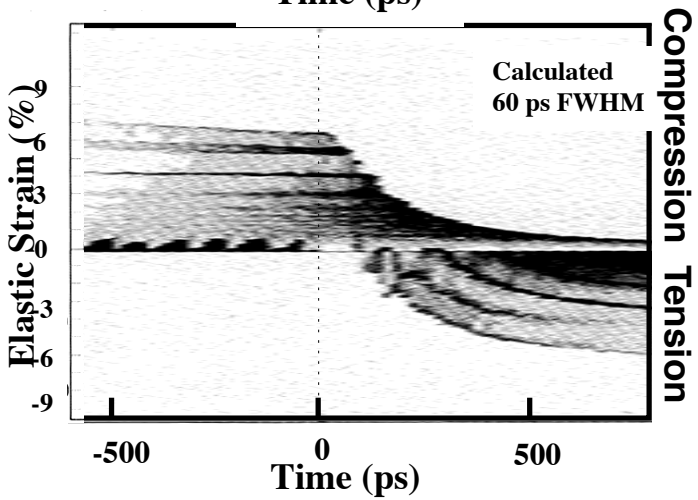
- Deformation cannot be predicted from first principles at low or high rates
- Shock deformation has long been of weapons related interest and was an early driver of time-resolved x-ray investigations
- Shock formation is of civilian materials processing interest as well

Shock Loading Investigations Have Driven Many X-Ray Time-Resolution Advances

Picosecond Resolution Shock Loading Strain Measurement



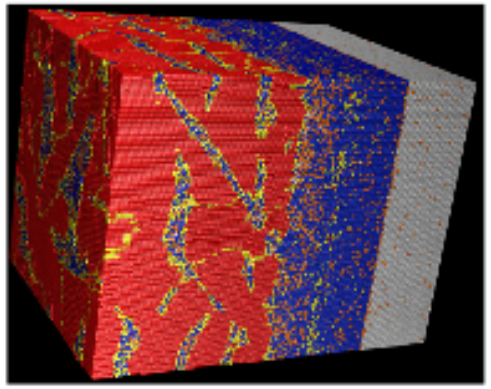




Picosecond resolution streak camera recording both compression (direct wave) and tension (reflected wave) ~120 ps after a 66 ps laser generated shock wave reaches the front surface at t = 0 ps.

After Whitlock and Wark, in *Time Resolved Diffraction,* ed. by J. R. Helliwell and P. M. Rentzepis (Clarenden Press, Oxford 1997) p. 106

First principles based (bottom-up) investigations of deformation Using molecular dynamics, discrete dislocation dynamics, and Statistical dislocation theory call for an entirely new level of experimental information



- Techniques for simultaneous temporal and spatial resolution are needed to address the type of detail provided in such simulations

- The irreversibility of deformation renders present ~100 Hz data sampling rates too confining

MD simulation of Fe shock loaded along [001] demonstrating phase change (red), dislocations (yellow), and unshocked region (grey) [40 x 40 x 50 nm^3]

Kimminau et al, J. Phys. Condens. Matter 20, 505203 (2008)

Structure and Dynamics of Atomic Displacement Cascade Dynamics

- Displacement cascades are the primary mechanism of fast neutron and ion irradiation

- The time scales of cascades range from sub-picosecond generation to picosecond - nanosecond annealing, evolution on microsecond – millisecond time scales, and accumulation to giga-seconds (40 years) in reactor service

- Because of the sub-picosecond time scales involved in the generation of cascades, the dynamics has remained the domain of molecular dynamics for more than 50 years

Dynamics of Irradiation Induced Defects

Point defect generation,
Close pair annihilation

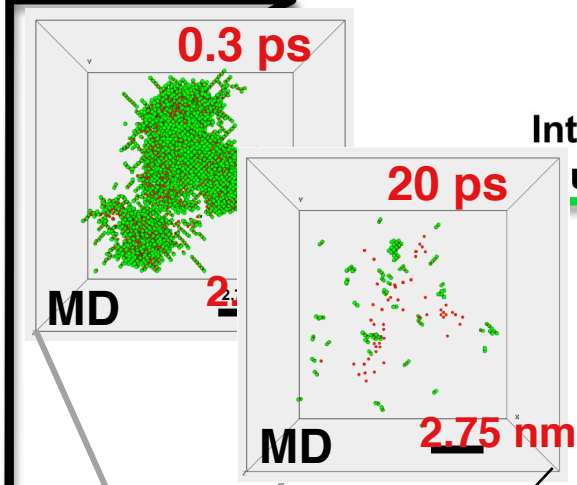
Point defect diffusion
and aggregation

Damage accumulation,
Radiation enhanced/induced
microstructure evolution

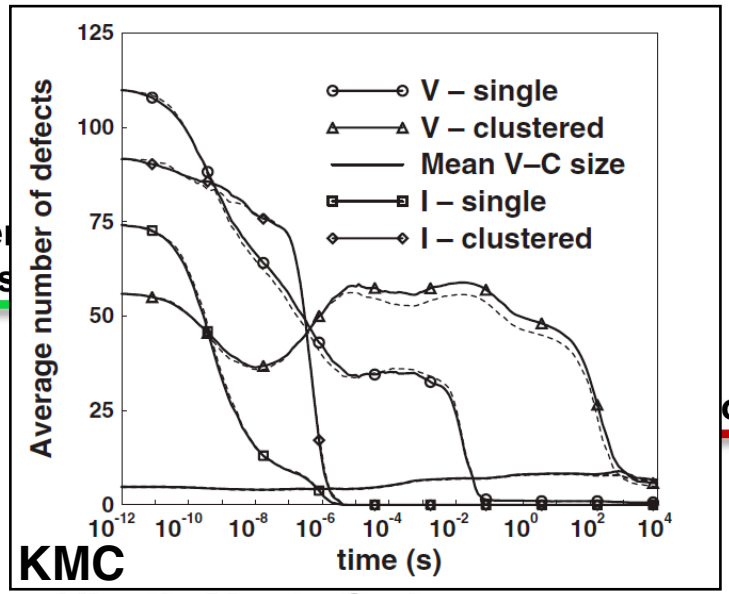
Cascade Dynamics/Evolution Measurements Missing

Defect Character

Interstitial, vacancy creation,
Cascade formation



Molecular Dynamics, DFT



Kinetic Monte Carlo,
Lattice phase field

on, cluster diffusion

Adapted from
Brian Wirth

fs ps ns μs ms s ks

CDP-EFRC ORNL

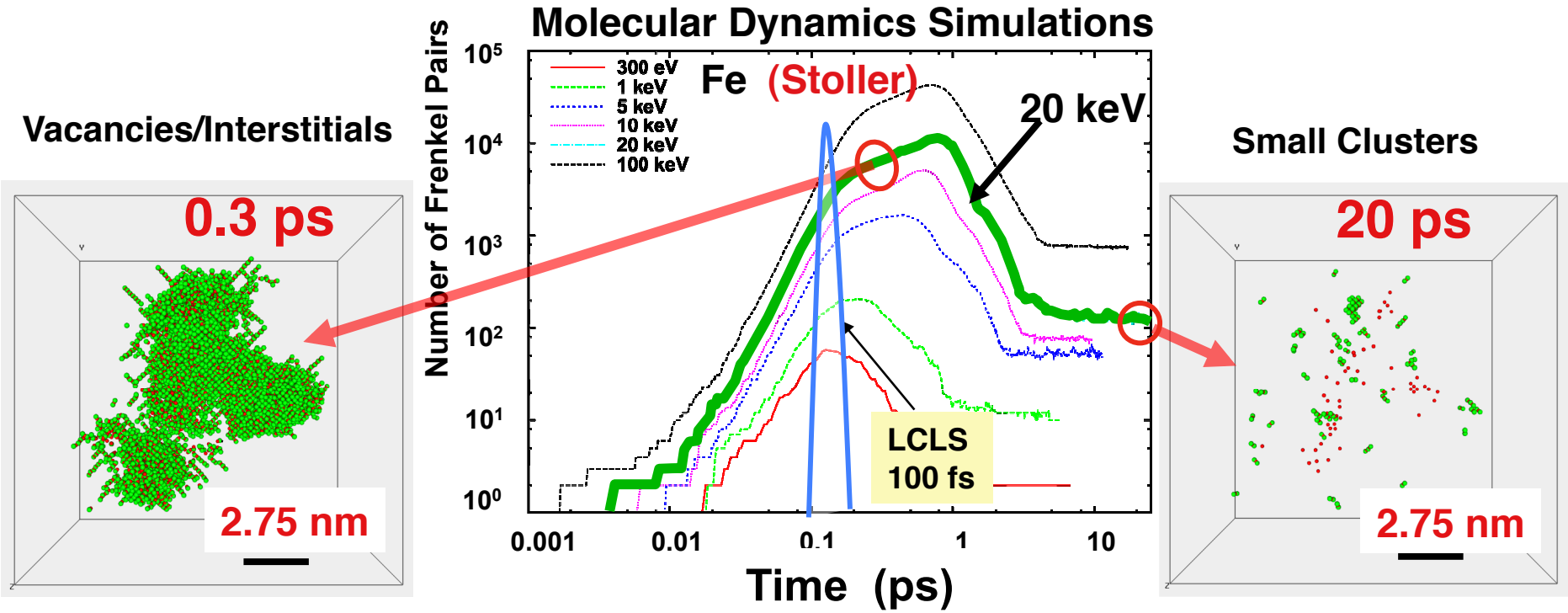
Time Scales

Managed by UT-Battelle
for the U.S. Department of Energy

Evolution & Control of Complexity

OAK RIDGE National Laboratory

Molecular Dynamics Simulations of Displacement Cascade Evolution Have Spanned 50 Years



Experimental Measurements of Cascade Dynamics and Evolution Will Be Possible for The First Time Using Sub-Picosecond X-Ray Pulses At The LCLS and Diffuse Scattering from The Transient Strain Fields

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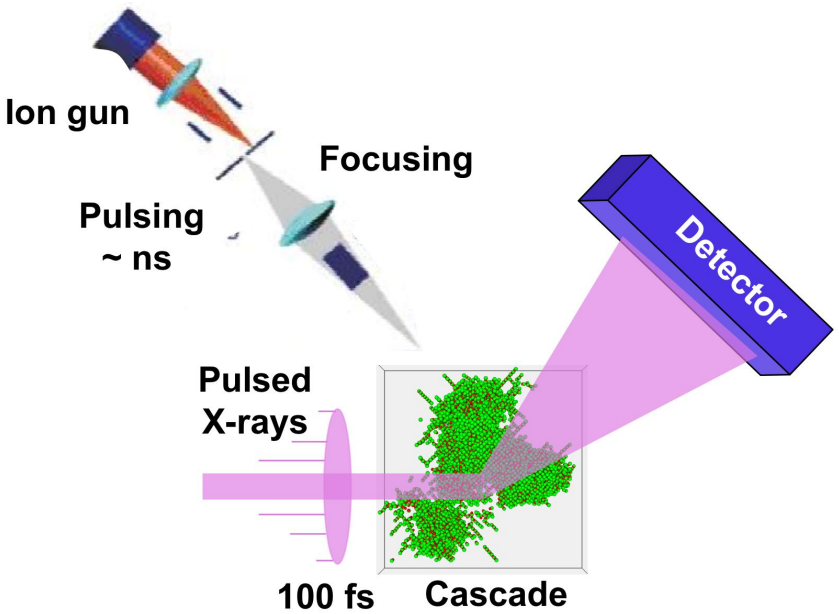
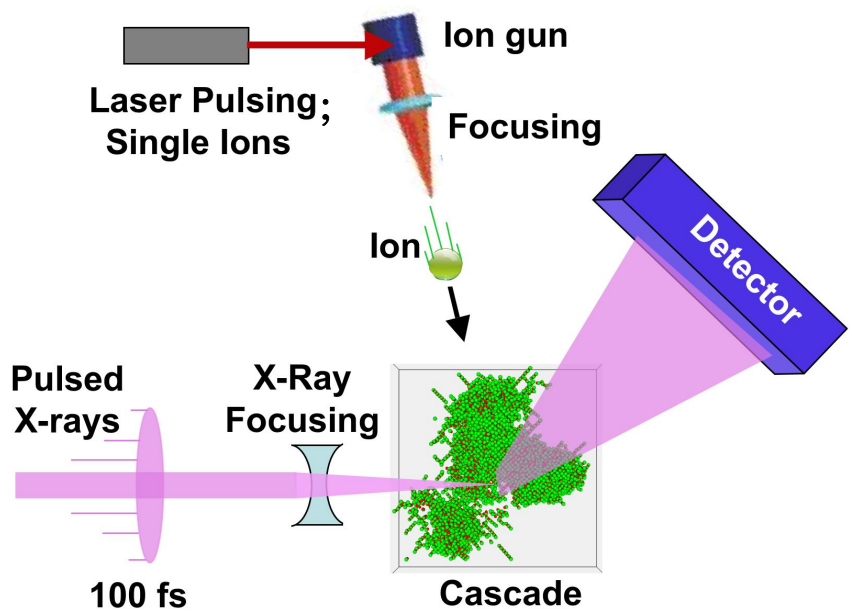
Time-Resolution from Subpicoseconds to Milliseconds



Nanosecond to millisecond ion pulses by sweeping focused ion beam (APS/LCLS)



Subpicosecond to nanosecond resolution by ion time of arrival signal (LCLS)

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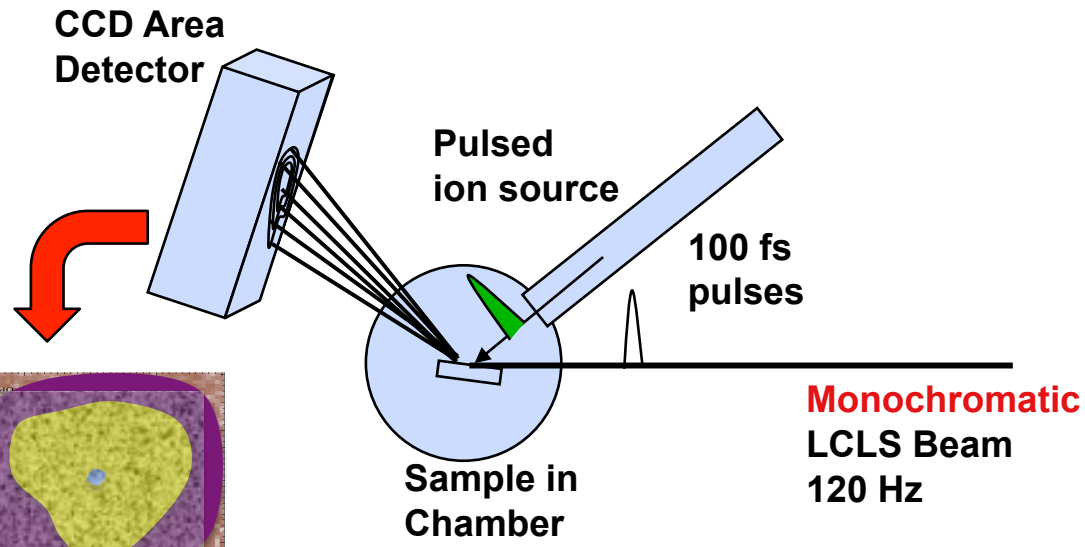
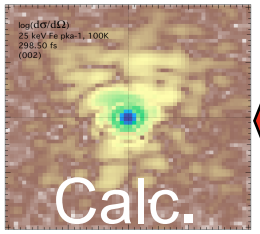
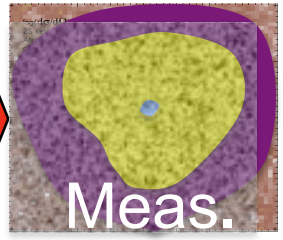
Diffuse Scattering Analysis of Cascade Dynamics-Evolution

Synergistic Exp.-Theory
Reverse Monte Carlo
Data analysis

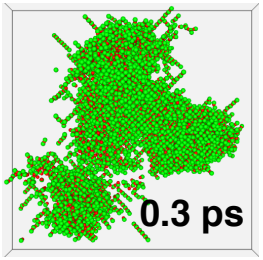


Diffuse
Scattering
Patterns



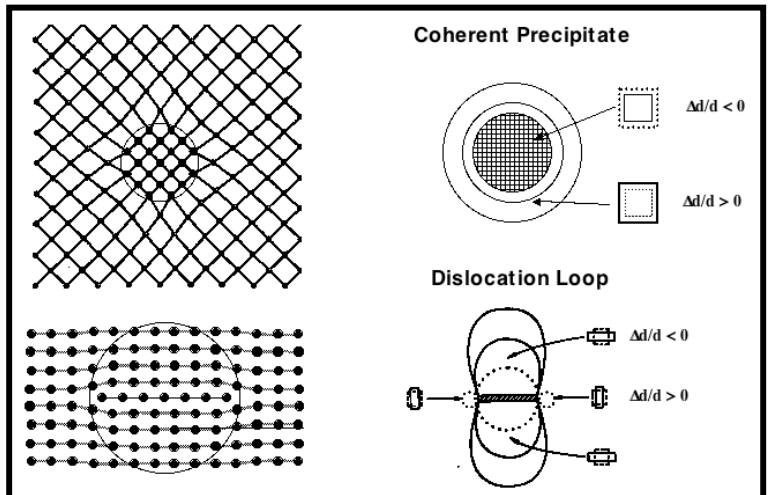


(Schematic)



Cascade Dynamics
Simulation

- Each cascade is different
- Time structure will be statistically equvalent
- Micro-x-ray pulses would provide time history of individual cascades



Local Bragg scattering origin of diffuse scattering from defects

B. C. Larson, in *Diffuse Scattering and the Fundamental Properties of Materials*, (Momentum Press, New York, NY 2009).

CDP-EFRC ORNL

Concluding Comments

- Toward the goal of transforming materials processing into a predictive rather than a discovery issue:
  - Increasingly detailed temporal, spatial, and energy resolved structural information will be required in close connection with first principles based theory and modeling
  - Increasingly close coordinations between facilities and scientific users will be required
  - A hierarchy of user facilities will be required to match sources with experimental requirements