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Simulation of Laser-Material Interactions for Dynamic Transmission Electron Microscopy Experiments

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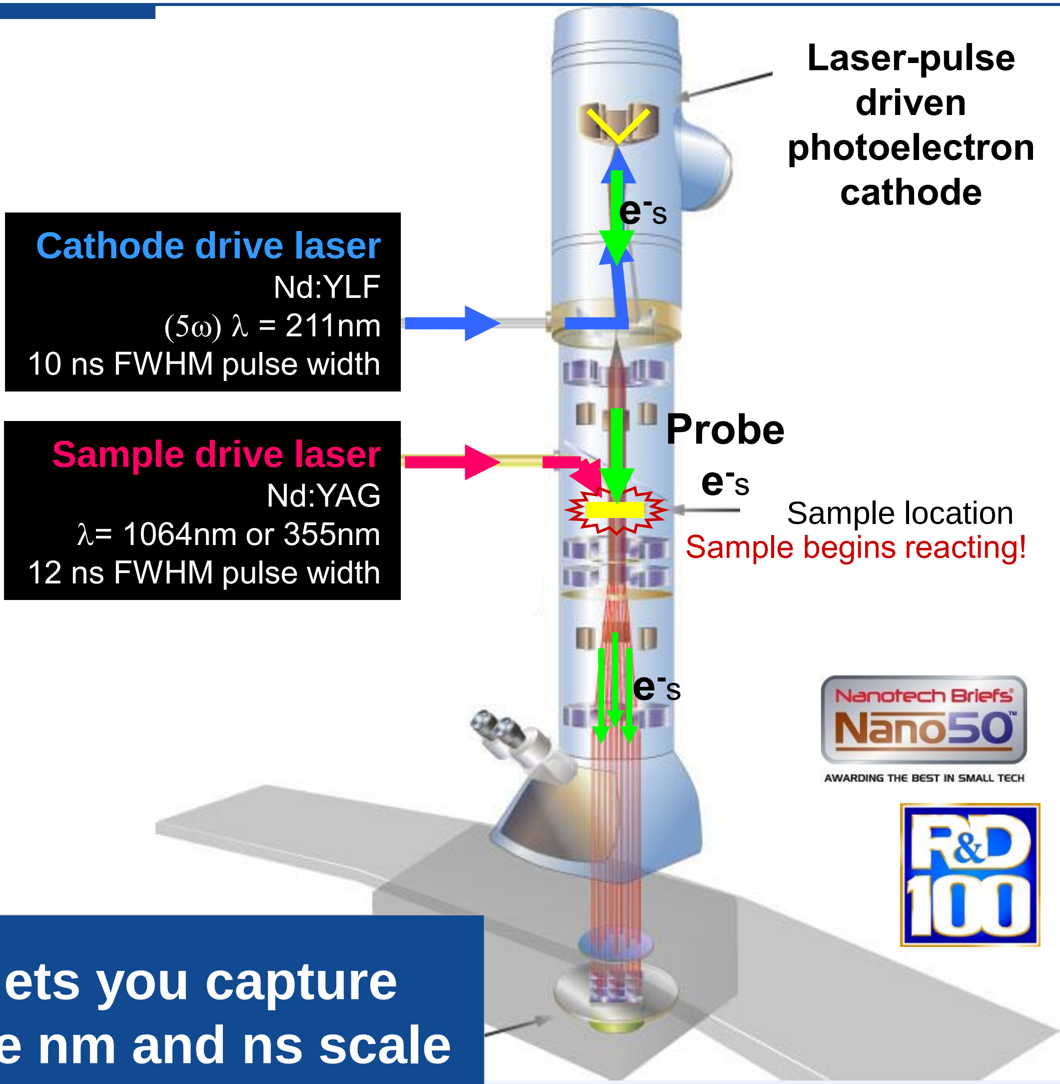
Overview: The LLNL DTEM is a nanosecond-scale *in situ* TEM with single-shot capability

DTEM adds two lasers to a conventional TEM to enable:

- Driving sample events with extreme spatiotemporal temperature gradients
- Real-space imaging and diffraction with ~15 ns exposures
- Enough signal in one exposure to form a complete image (up to 2×10^9 electrons)



Cathode drive laser
Nd:YLF
(5ω) λ = 211nm
10 ns FWHM pulse width

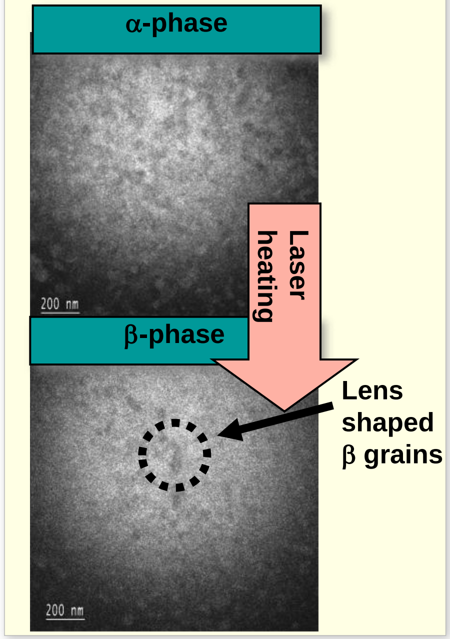
Sample drive laser
Nd:YAG
λ= 1064nm or 355nm
12 ns FWHM pulse width

DTEM's single-shot approach lets you capture *unique, irreversible* events on the nm and ns scale

Scientific Context: DTEM enables applications in physics, materials science, chemistry, and biology

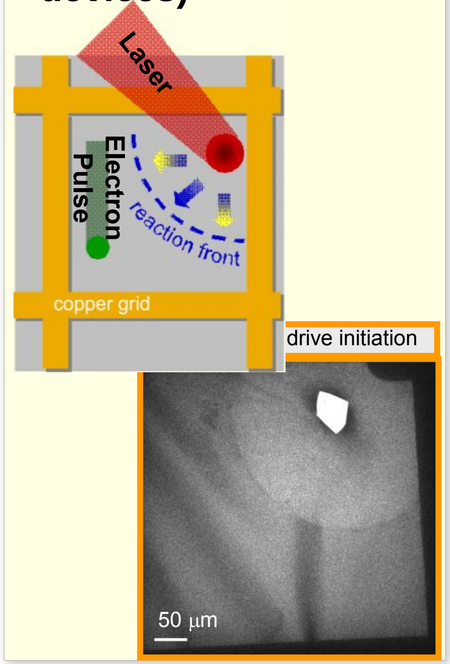
Structural Materials

- **Diffusionless phase transformations (martensites)**
- **Dislocation dynamics nucleation/ interactions**



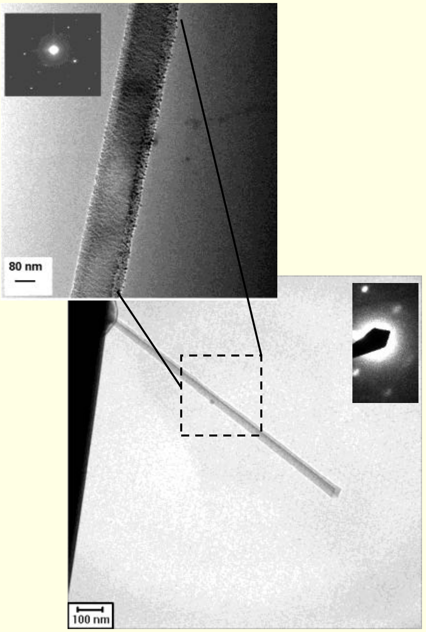
Solid State Reactions

- **Reactive Multilayer Foils (RMLF)**
- **Small scale diffusional transformations in thin films (electrical devices)**



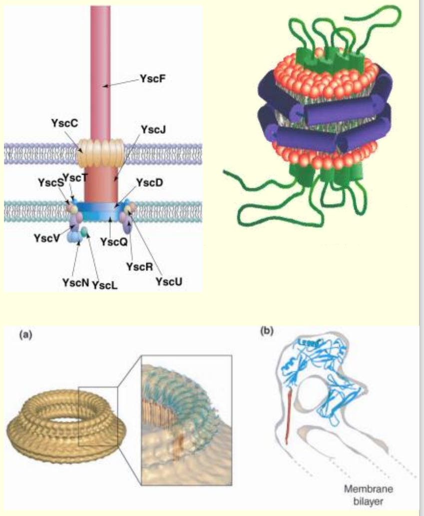
Catalytic Reactions

- **Nanowire and nanoparticle growth**
- **Catalyst/substrate interactions in gaseous and liquid environments**



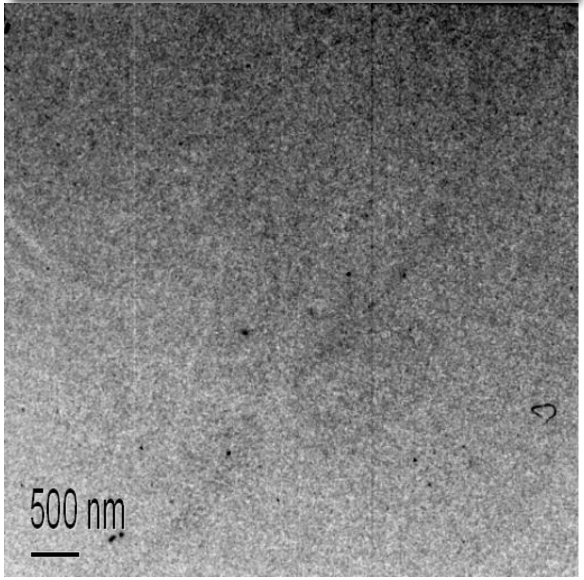
Biological Processes

- **Dynamics of cellular modification in the presence of toxins**
- **Pathogen identification**
- **Radiation damage in organic molecules**

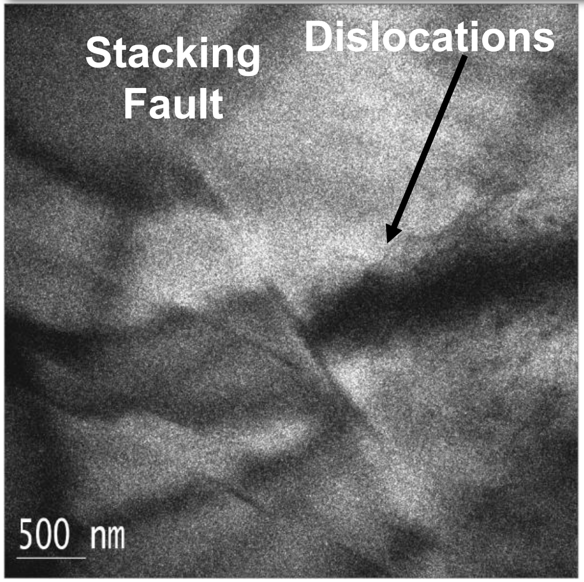


Current DTEM performance enables 15 ns diffraction contrast imaging.

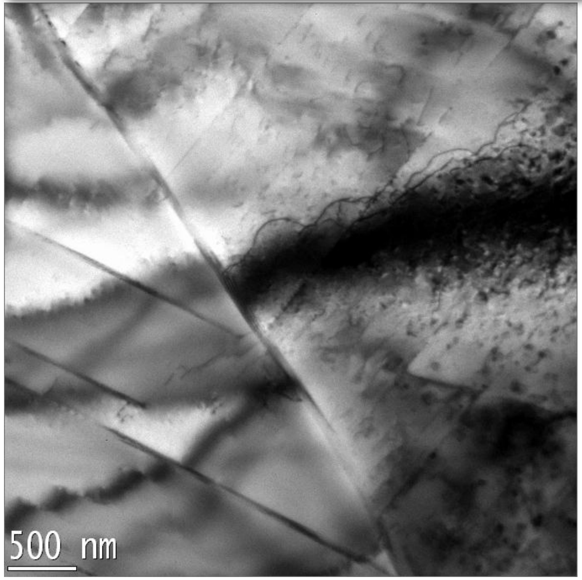
Pulsed Image 2005:
1×10^{6} e^{-}/pulse



Pulsed image 2008:
15 ns, 1×10^{9} e^{-}/pulse



Conventional TEM image:
1 s exposure



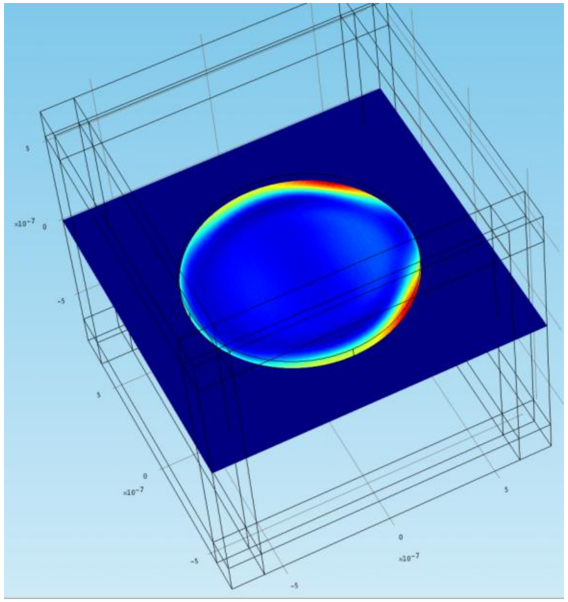
The latest upgrades enable images of dislocations, stacking faults, and other microstructural features in a single 15 ns exposure.

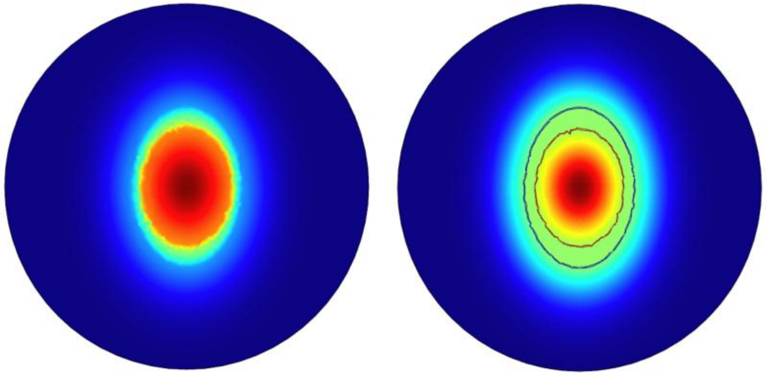
Previously, these features could have only been seen by accumulating a large number of pulses.

Quantitative interpretation of DTEM experiments requires an understanding of laser-material interaction

Two aspects:

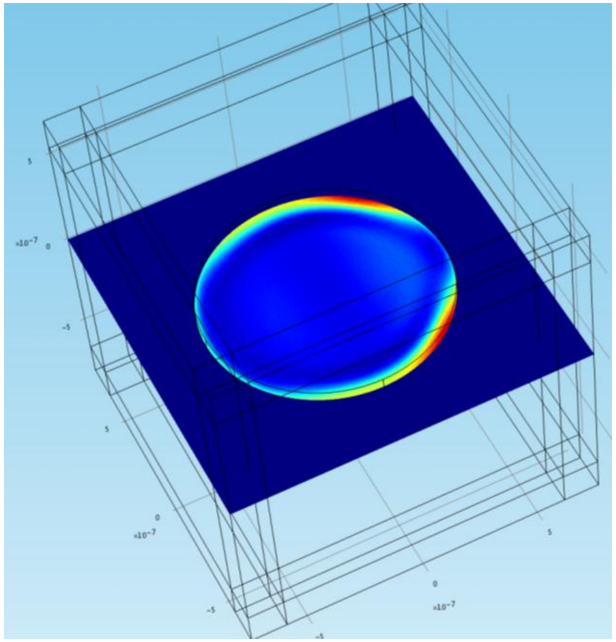
- Laser absorption
  - Polarized light incident at an angle onto nanostructured materials
  - Spatial distribution of absorption is important and complicated
- Heat diffusion
  - Normal direction (~100 nm) is a fast (few ns) 1D problem
  - Transverse direction (~50 µm) is a slow (many µs) 2D problem
  - Transformations and reactions are a nonlinear heat source/sink

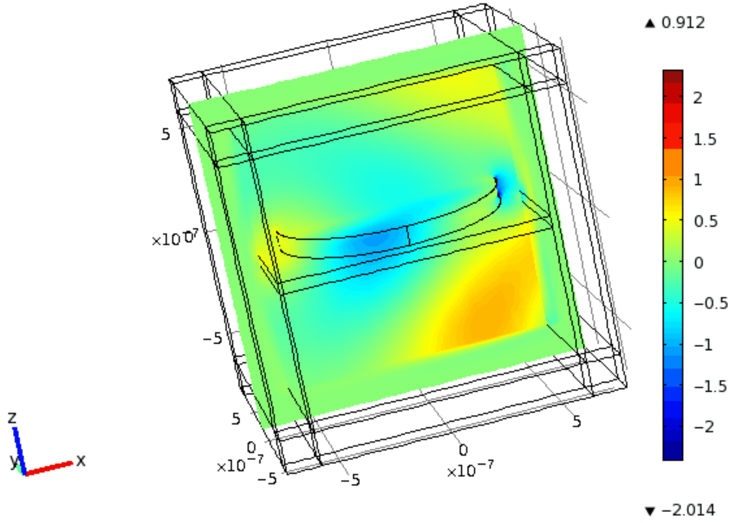




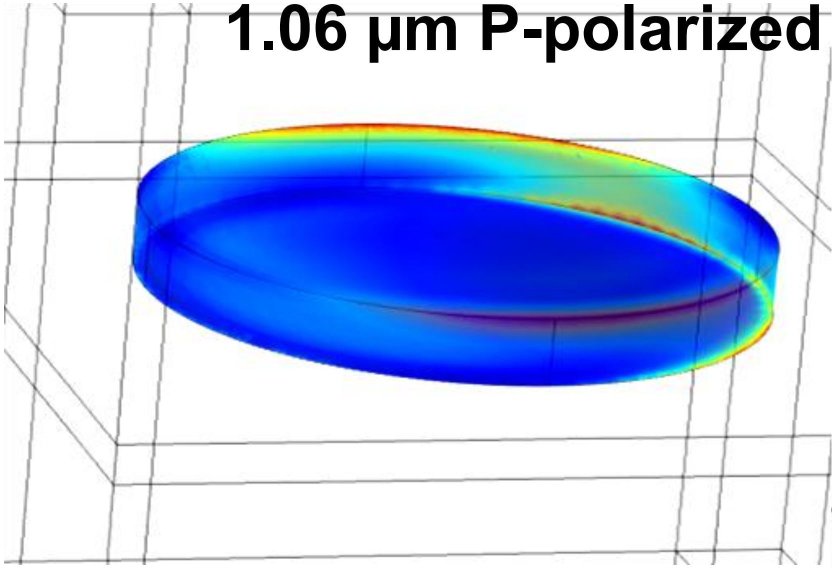
Laser absorption is calculated in a 3D scattered-wave formalism

- User specifies wavelength, complex vector polarization, incident angle, geometry, and complex $\varepsilon(\omega)$ for each material
- This example is 1 µm diameter, 85 nm thick $Ge_2Sb_2Te_5$ on a 50 nm Si_3N_4 membrane hit with 1.06 µm p-polarized light at 42.5°
- Standard single-frequency scattered-wave formalism with perfectly matched layers and scattering boundary conditions
- Direct PARDISO solver is fast, stable, memory-hungry
- Validated against analytical solutions for planar thin-film stacks
- Volumetric absorption can couple directly into subsequent heat diffusion simulations

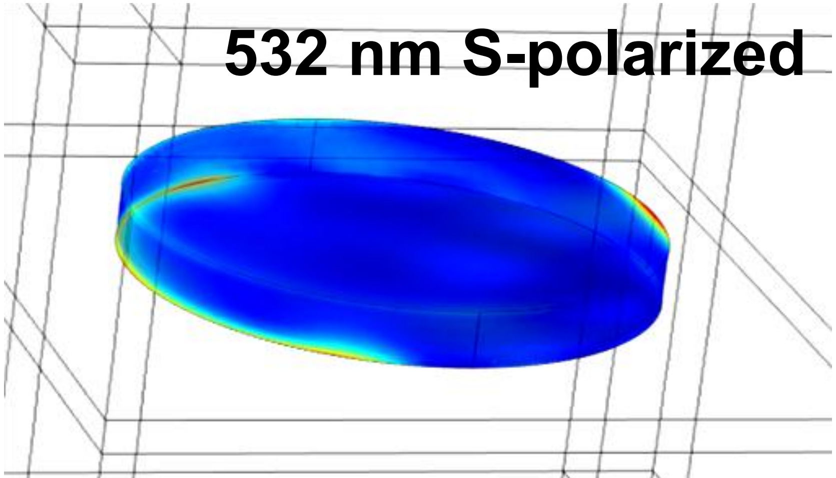




Laser absorption shows interesting three-dimensional polarization/wavelength dependence



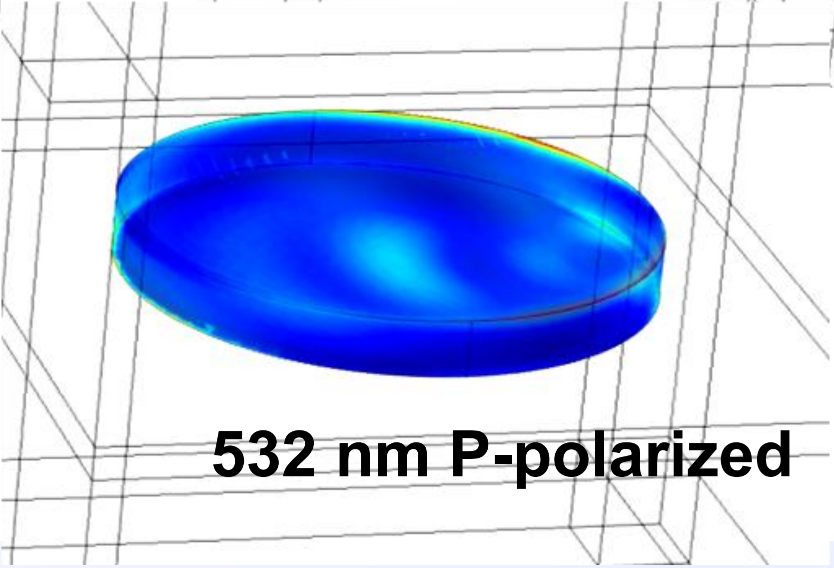
1.06 µm P-polarized



532 nm S-polarized

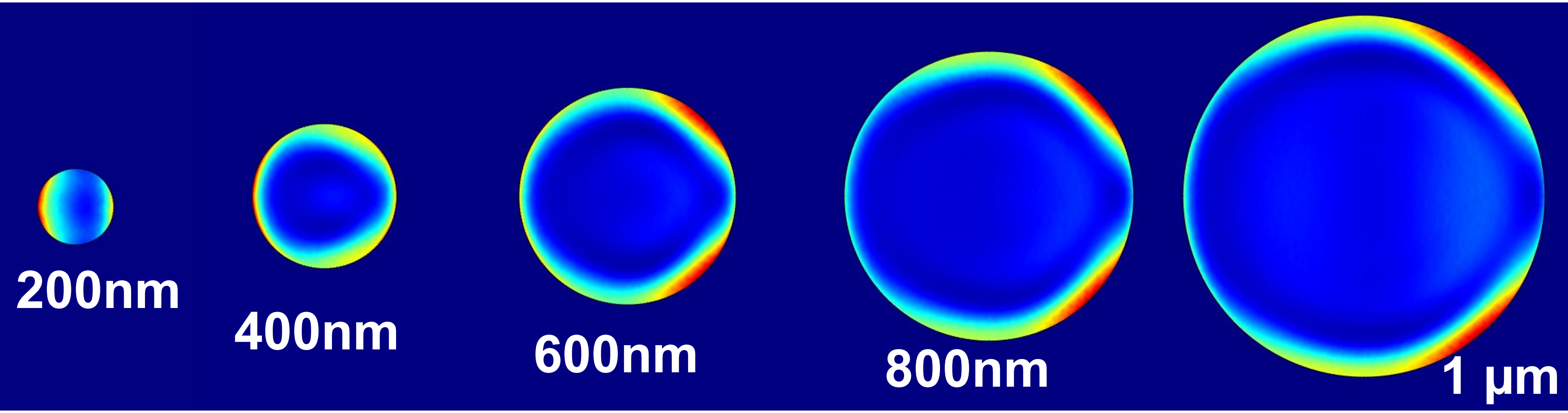
Example is a 0.8 µm disk

Plots show absorbed power density



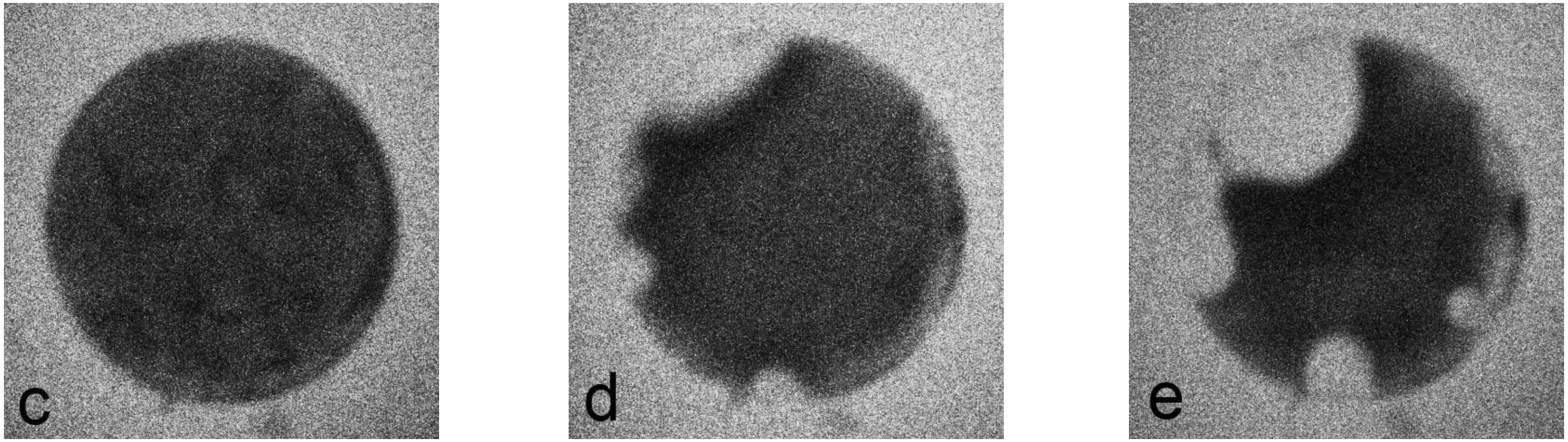
532 nm P-polarized

There is also a strong size dependence for diameters much less than λ



Absorption profile halfway through the thickness of the disk for 1.06 µm light

Experiments show absorption to be very inhomogeneous, and this affects phase transformations and morphology evolution

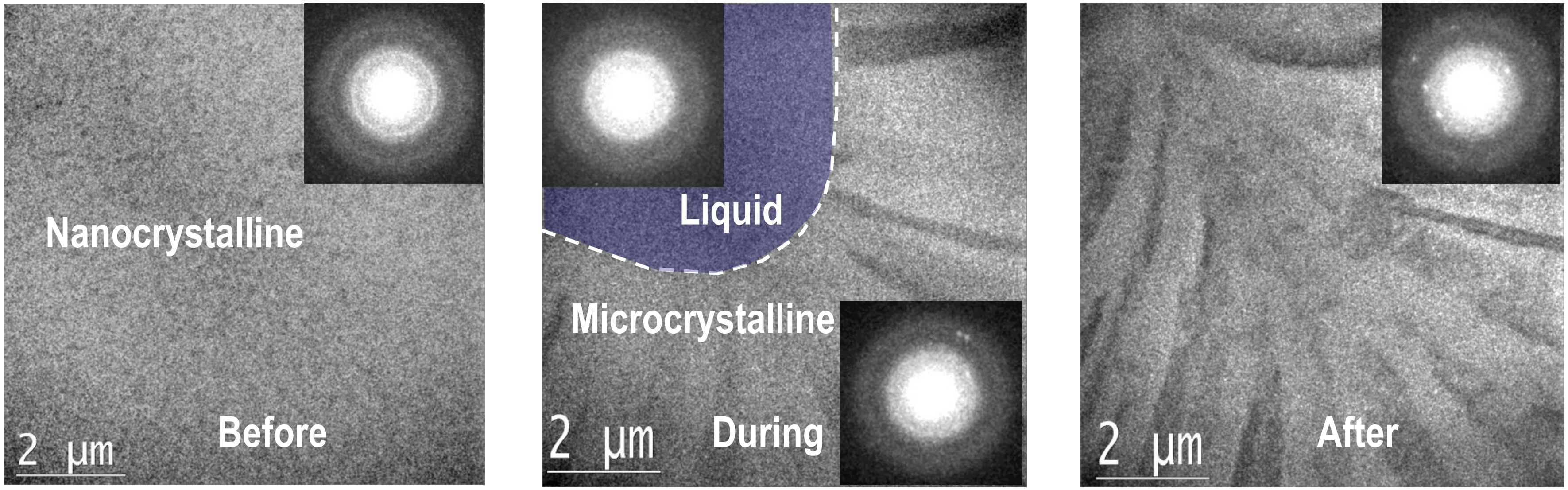


Before **During (t = 4 ns)** **After**

- Experiments show certain spots around the edges consistently melt long before the rest of the material gets hot
- Once laser shuts off (at t ~ 12 ns), the heat can diffuse and equalize—but the damage is already done

Collaboration with S. Meister and Y. Cui, Stanford

DTEM can also track solid-liquid phase transformation fronts

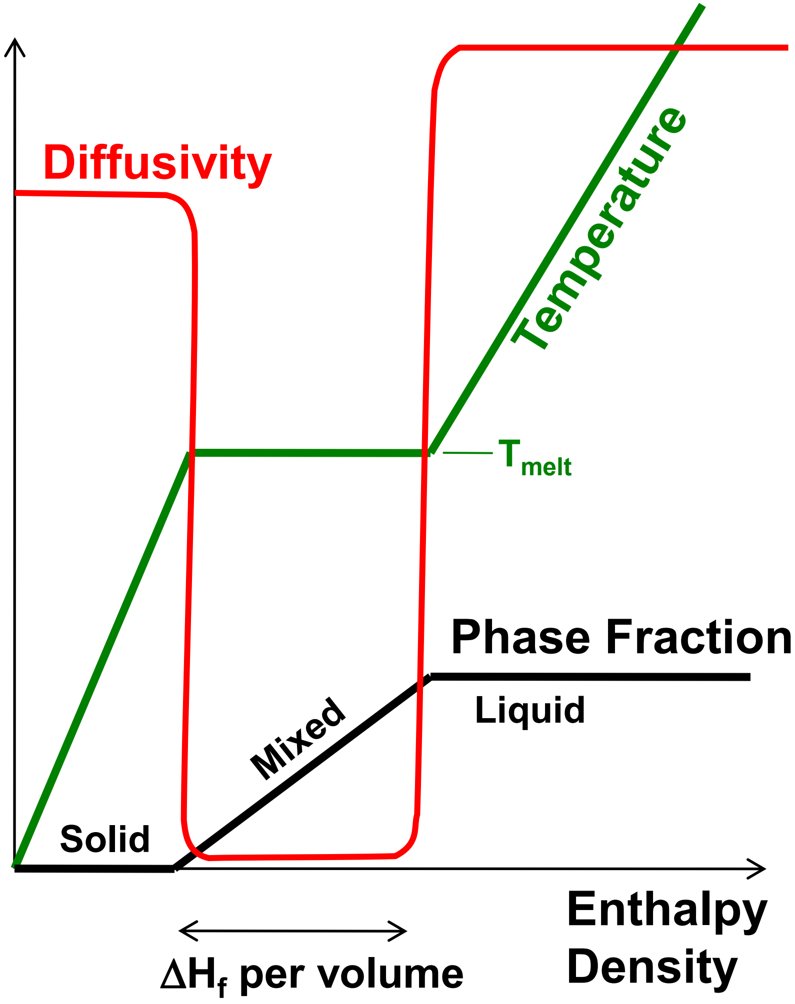


- DTEM captures rapid lateral solidification front moving at ~3.5 m/s near edge of an elliptical laser spot
- Microstructural evolution is of interest and depends on nonlinear nonequilibrium dynamics at the front

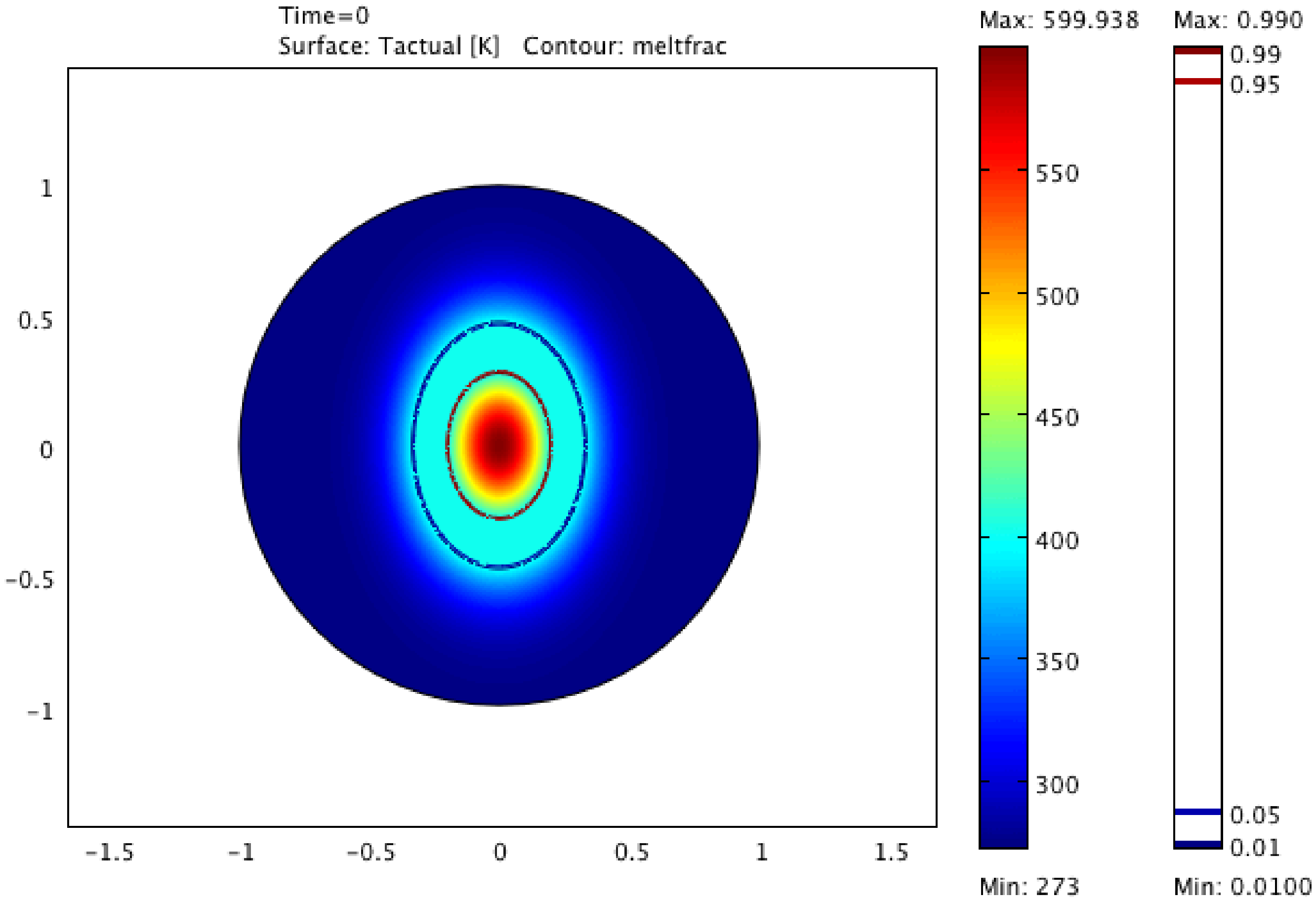
Collaboration with A. Kulovits and J. Wiezorek, U. Pitt.

Heat of transformation creates nonlinearity that can be handled within an enthalpy formalism

- Computer solves directly for enthalpy density, not temperature
- Defined functions calculate the actual temperature and phase fractions in post-processing
- Essence of the method is in an appropriate nonlinear enthalpy-dependent diffusivity
- Smoothed corners and artificial diffusivity in mixed-phase regions stabilize the solution
- Fifth-order finite elements provide high precision while keeping reasonable computational costs
- A practical compromise: Simpler than phase field, but neglects kinetics



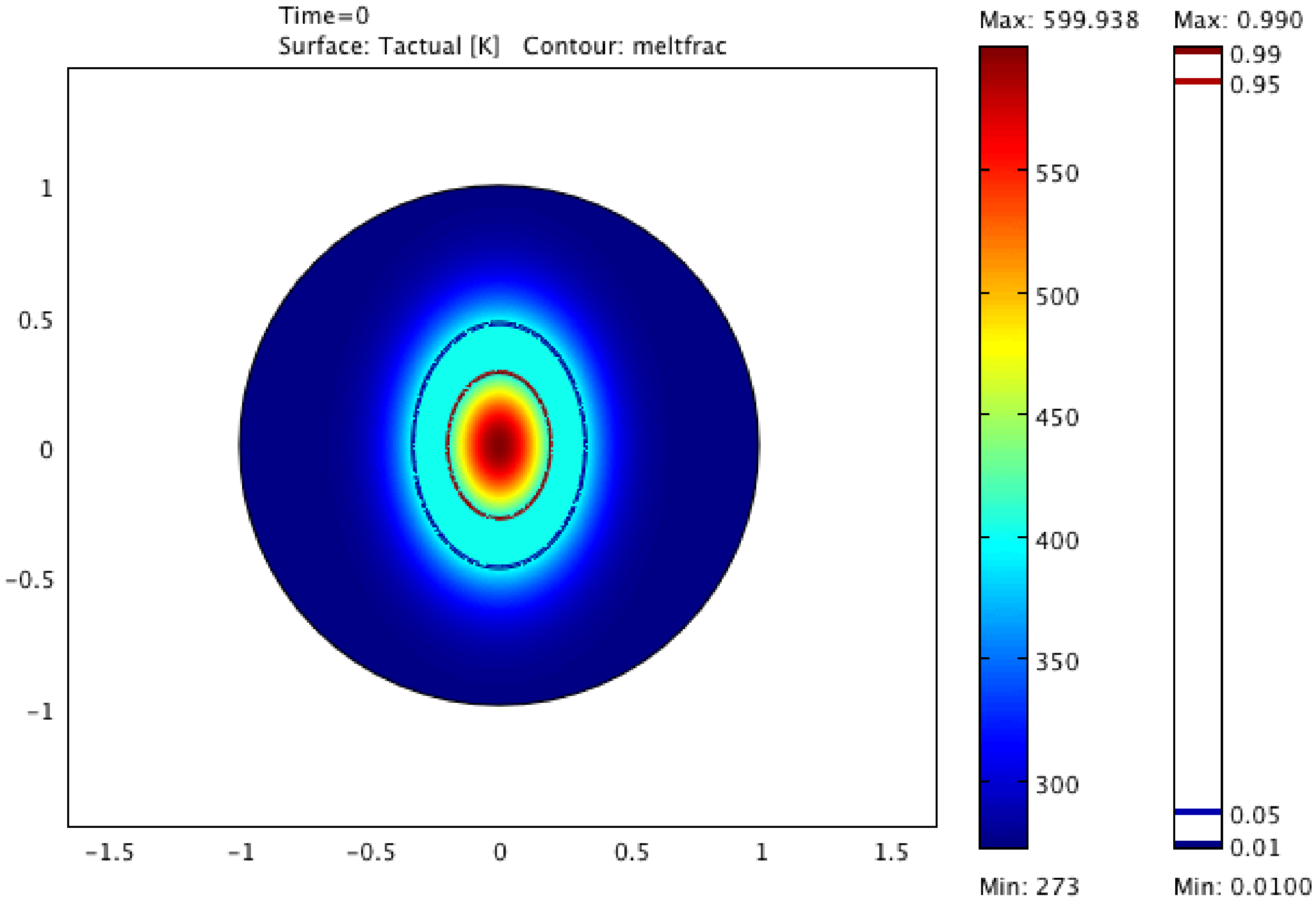
Simulation quantitatively predicts anisotropic collapse of mixed-phase region followed by slow resolidification



Color scale = Temperature

Contours = Boundaries of mixed-phase region

Simulation quantitatively predicts anisotropic collapse of mixed-phase region followed by slow resolidification



Color scale = Temperature

Contours = Boundaries of mixed-phase region

Summary

- We have a TEM that can perform single-shot *in situ* experiments on the scale of nanometers and nanoseconds
    - Example applications include chemical reactions and phase transformations
    - Reveals transient material structures that couldn’t be seen any other way
- Understanding experimental results depends on understanding laser-material interactions
- Simulations provide handle on two important aspects of this
    - Geometrical effects in laser absorption
    - Nonlinear heat flow coupled with transformations