Simulation of Laser-Material Interactions for Dynamic Transmission Electron Microscopy Experiments
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 $2 \times 10^9$ electrons)

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

<table>
<thead>
<tr>
<th>Structural Materials</th>
<th>Solid State Reactions</th>
<th>Catalytic Reactions</th>
<th>Biological Processes</th>
</tr>
</thead>
<tbody>
<tr>
<td>• Diffusionless phase transformations (martensites)</td>
<td>• Reactive Multilayer Foils (RMLF)</td>
<td>• Nanowire and nanoparticle growth</td>
<td>• Dynamics of cellular modification in the presence of toxins</td>
</tr>
<tr>
<td>• Dislocation dynamics nucleation/interactions</td>
<td>• Small scale diffusional transformations in thin films (electrical devices)</td>
<td>• Catalyst/substrate interactions in gaseous and liquid environments</td>
<td>• Pathogen identification</td>
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<td>• Radiation damage in organic molecules</td>
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</tbody>
</table>

α-phase

β-phase

Lens shaped β grains

Laser heating

β-grains

Electron Pulse

reaction front

drive initiation

Lawrence Livermore National Laboratory
LLNL-PRES-458677
Current DTEM performance enables 15 ns diffraction contrast imaging.

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 $\text{Ge}_2\text{Sb}_2\text{Te}_5$ on a 50 nm $\text{Si}_3\text{N}_4$ 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 $\lambda$.

Absorption profile halfway through the thickness of the disk for 1.06 $\mu$m light.
Experiments show absorption to be very inhomogeneous, and this affects phase transformations and morphology evolution.

- Experiments show certain spots around the edges consistently melt long before the rest of the material gets hot.
- Once laser shuts off (at $t \sim 12$ 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

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.
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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