Ultrafast Laser Diagnostics for Energetic-Material Ignition Mechanisms: Tools for Physics Based Model Development (12-0515)

SAND2011-6976P

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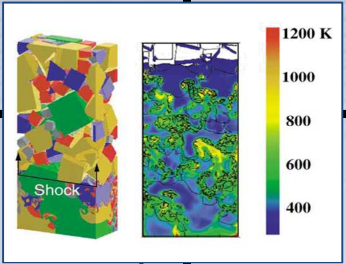
Project Purpose

Develop a set of experimental tools to provide a fundamental description of initiation. Deliver the first-ever mesoscale-resolved data to enable development of a predictive non-empirical treatment of initiation

R&D Goals & Milestones

- Develop cutting-edge experimental tools for “extreme-scale” probing of shock, chemistry and thermal response in dynamically loaded heterogeneous materials
- Provide mesoscale data for code support: discovery and validation



R&D Approach

Exploit rapid development of femtosecond laser technology for:

- Controlled high-turnover shock-drive
- Extreme-resolution laser diagnostics
- Quantitative imaging of heterogeneous samples under strong dynamic loads

Significance of Results

This capability represents a major scientific breakthrough toward non-emprical prediction of initiation, which is critical for design, QMU, and stewardship of the stockpile and for credible assessment of threats from adversaries

LDRD
LABORATORY DIRECTED RESEARCH & DEVELOPMENT

Sandia National Laboratories

The Problem: Initiation of Heterogeneous Explosives

• Despite its importance, a fundamental description of initiation eludes researchers.

• Initiation physics are complex:

• Tight coupling of mechanical (shock), thermal, and chemical phenomena

• “Extreme” time (ps) and length (μm) scales

• Uncertain chemical pathways

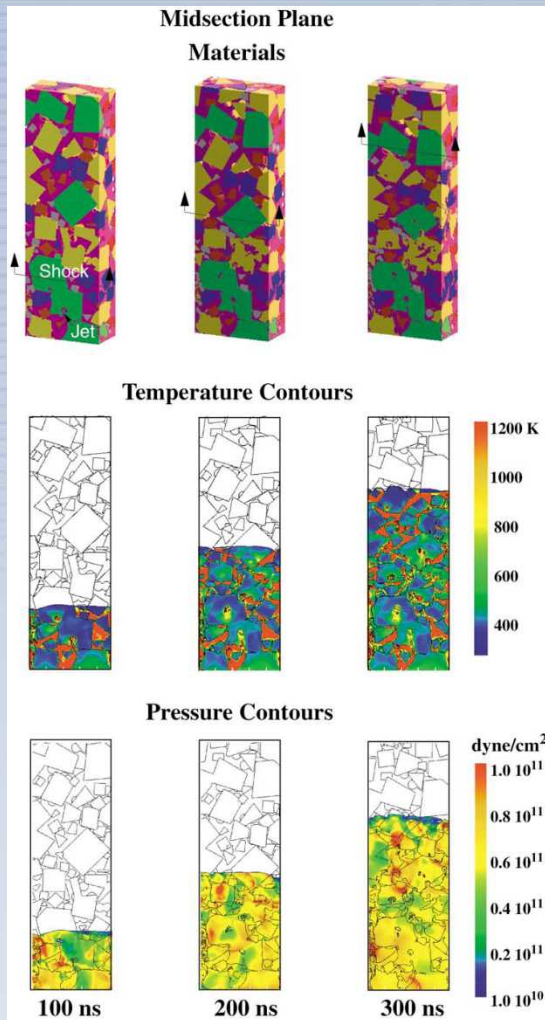
• Poorly understood mechanical to vibrational energy transfer

• Complex microstructure can dominate the overall response

• Void collapse, hot-spot formation, coalescence

• Ignition at boundaries? Across grains?

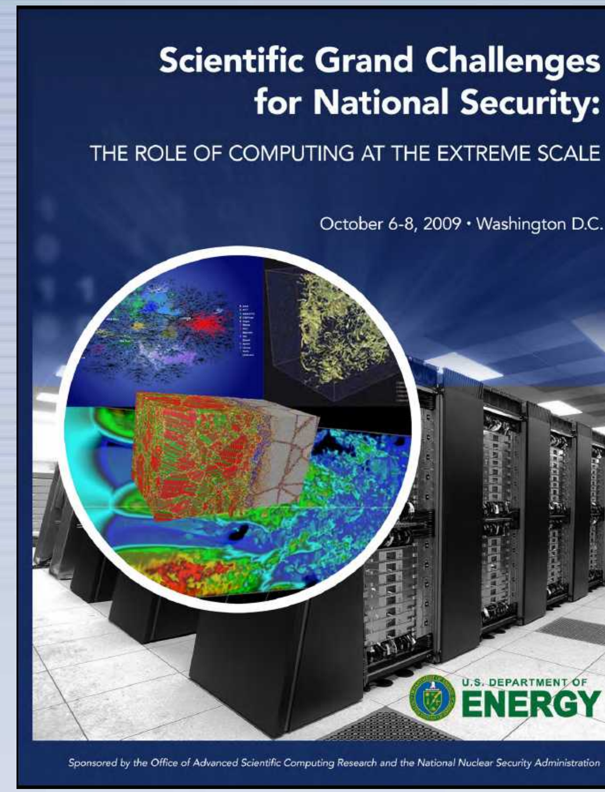
• Localized, complex ignition response



M.R. Baer, Thermochimica Acta **384**, 351-367 (2002).

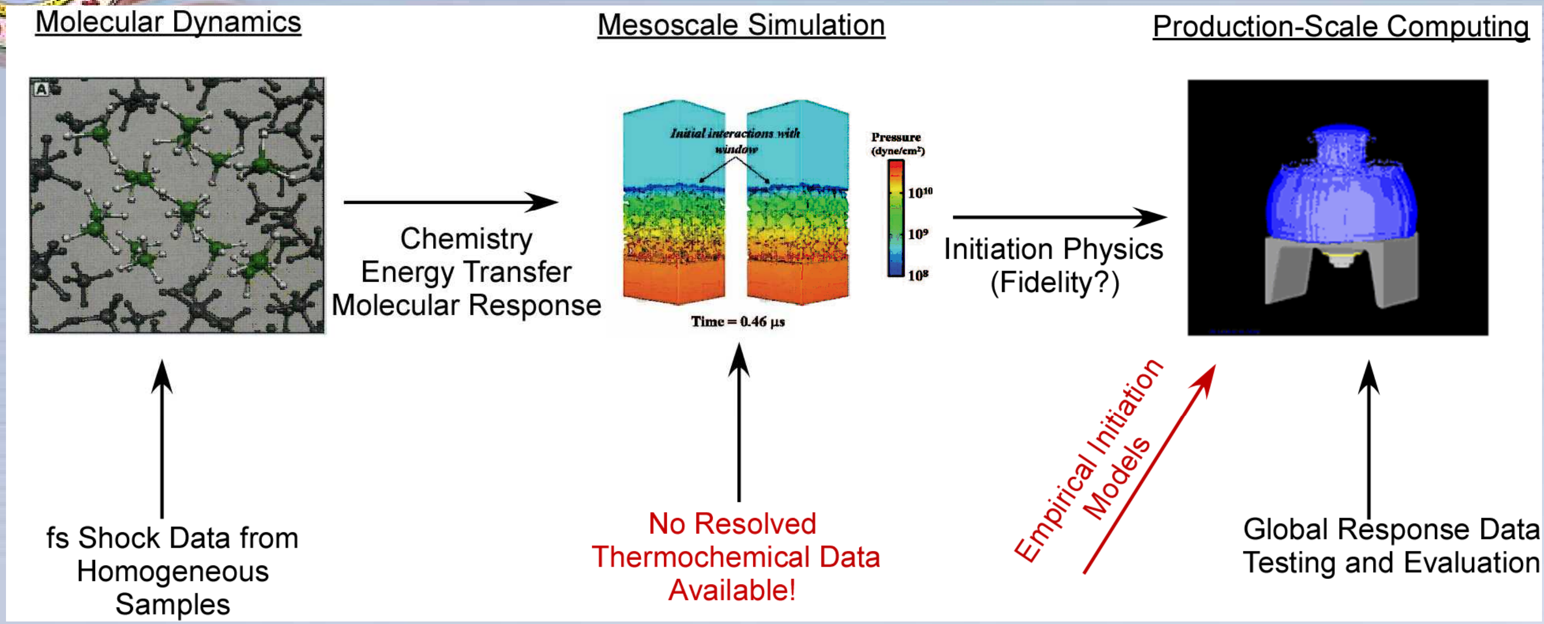
Why is this difficult problem important to solve?

- Design, QMU and stewardship of explosive components is critical to the evolving stockpile
- Nonideal explosives developed by adversaries are additionally critical to simulate and understand
- Initiation is presently handled using problem-specific empirical models tuned to experiment/testing observation of global response
    - Slow-turnaround, high-cost of testing
    - Models cannot be extrapolated outside the conditions of test data
    - Slow response to new customers
    - Decreased fidelity of computational solutions
- A first-principles understanding is sorely needed to improve model fidelity and agility



"…there is a significant common need from NNSA, the U.S. Department of Defense (DoD), and the U.S. Department of Homeland Security (DHS) agencies to dramatically improve current understanding of high explosive formulations, especially regarding safety and performance."

How will these new experimental capabilities enable predictive simulation?

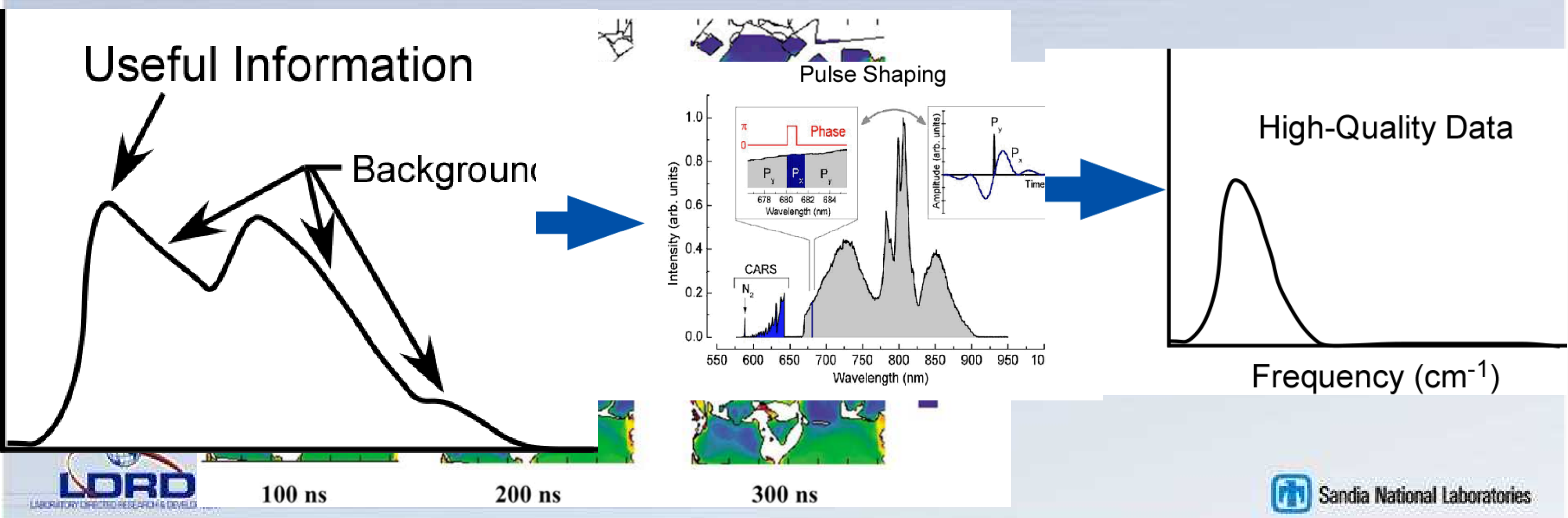


- Thermochemical access to the mesoscale has, until recently, been restricted to simulations
- Femtosecond lasers are enabling a new class of experiments to access thermochemical response on extreme scales
- Data (LANL, Illinois) are emerging on homogeneous systems to support MD codes
- Mesoscale-resolved experiments on heterogeneous samples are required

The LDRD Challenge

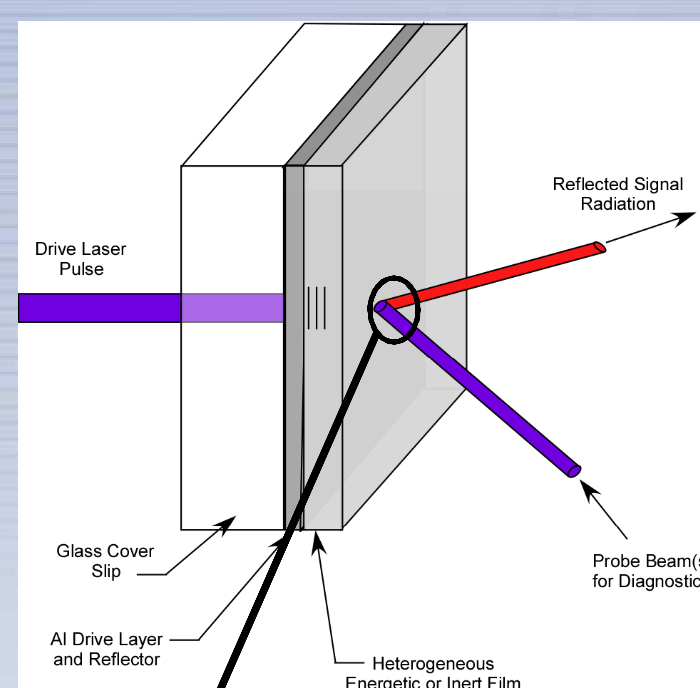
• ***Develop state-of-the-art diagnostics for extreme mesoscale-resolved probing of mechanical, thermodynamic, and chemical parameters in complex heterogeneous materials under intense dynamic loading***

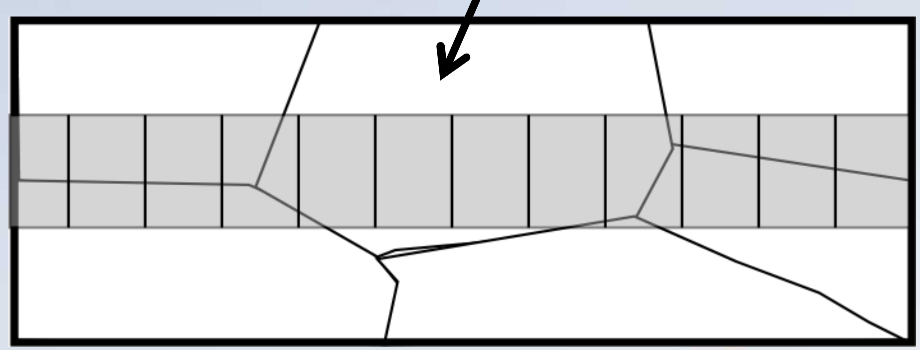
• Major scientific advances will be required:
  - Extension to realistic heterogeneous microstructures (optical effects?)
  - Spatially correlated imaging (subgrain resolution and realistic grain size)
  - Spectroscopy in complex chemical and topological environments
  - Quantitative data: temperature measurements are still a “holy grail”!



Our solution: Use femtosecond laser pulses for ***mesoscale-resolved*** experiments on ***heterogeneous*** materials

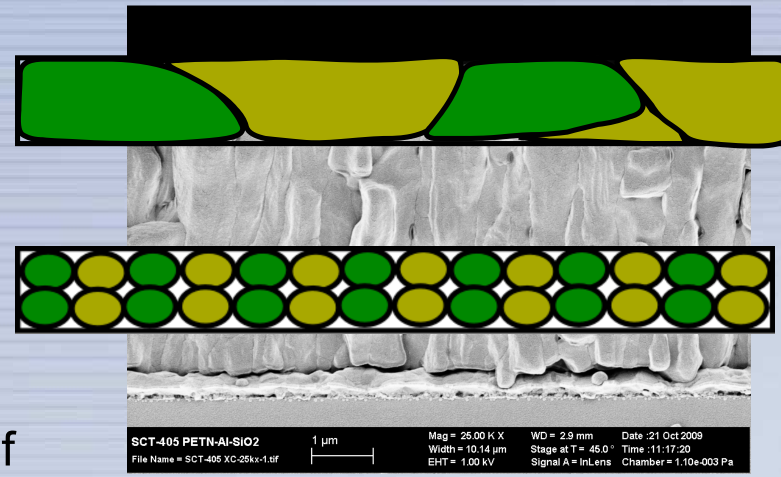
- Reliable femtosecond (fs) pulsed laser sources have become commercially available within the last ~10 years
- Ultrashort 10^{-14}–10^{-13} sec pulses permit access to ps-scale events
- Well-controlled laser drive has been demonstrated in homogeneous films (Moore group, LANL)
- Broadband yet low-noise laser bandwidth enables multiple spatially resolved *imaging* spectroscopies
- Spatially resolved measurements on *heterogeneous* samples at realistic grain sizes





The heterogeneous samples themselves are a key part of the solution

- Grain growth occurs in 3-D so large grains may dictate thick samples
  - More difficult laser drive
  - Tall/thin microstructures
  - “Pancake” grains
  - Engineered (ordered) samples?
- Sandia (Alex Tappan, 2554) has developed facilities for fabrication and characterization of explosive and inert films
  - Preparation of homogeneous samples for LANL experiments
  - Cost leveraging with multiple ongoing projects
- Candidate energetic materials:
  - HNAB, HNS
  - PETN, HMX
  - Polymers (Sylgard)
- Inert films will also be utilized



Thin-grain PETN film

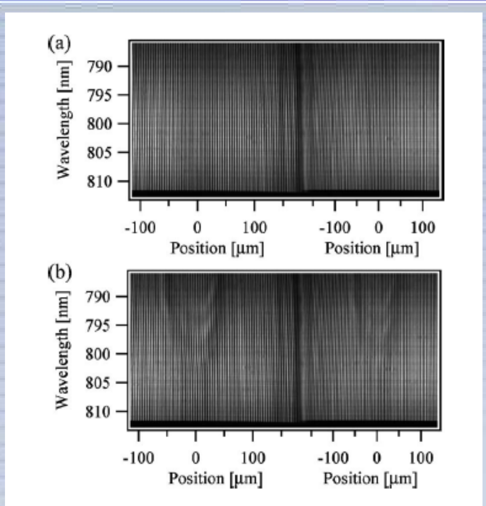
PETN / Al on 22 mm glass coverslips

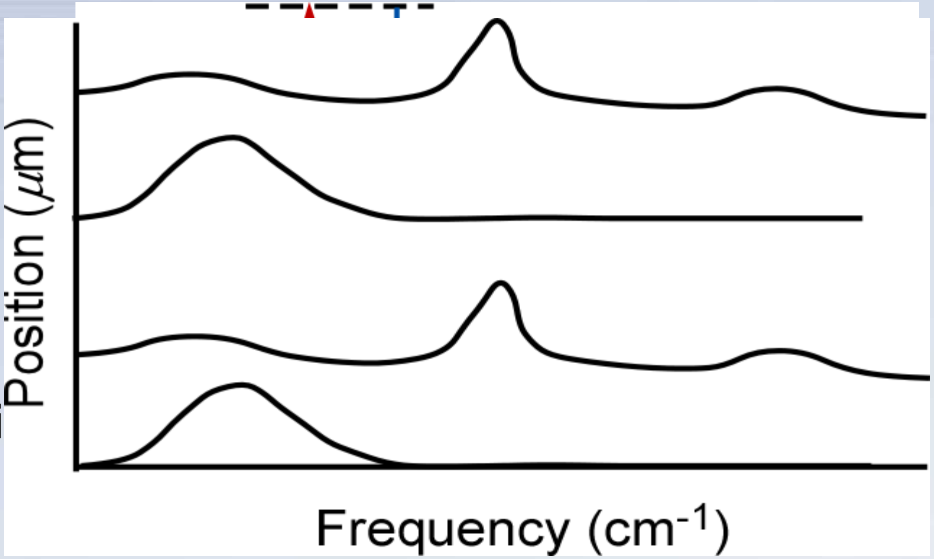
Measurement Approaches

- <u>Mechanical Characterization (Shock Propagation): u_s, u_p, *Pressure*</u>
- Ultrafast Dynamic Ellipsometry (UDE) develop by Moore Group at LANL

- <u>Temperature, Vibrational Energy, Species, Chemistry</u>
    - Nonlinear Raman spectroscopy
    - Coherent anti-Stokes Raman Scattering (CARS)
    - Stimulated Raman Gain/Loss Spectroscopy (SRS)

- All Measurements performed in imagi mode for identification of complex mesoscale spatial features





Project Team and Research Strategy

- A "parallel processing" approach to diagnostic development
- Three separate femtosecond laser lab resources
  - Building 865 (1500): IR/UV Diagnostics and Laser Amp Development
  - Building 962 (5400): Raman Diagnostics Development
  - Building 905 (2500): UDE Diagnostics Development
- Three distinct areas of expertise drawn from three unique Sandia centers
  - Center 1500: laser diagnostics, thermal science, mesoscale modeling
  - Center 5400: optical physics, ultrafast laser applications
  - Center 2500: energetic materials, chemistry, laser diagnostics
- Key Team Members
  - Sean Kearney (1512), Jeremy Lechman (1516), Mel Baer (1500/contract)
  - Junji Urayama (5444)
  - Brook Jilek (2554), Alex Tappan (2554), Mike Kaneshige (2554)
- Collaboration with Moore group at LANL

The Role of Predictive Simulation in this Project

- While the goal here is to develop state-of-the-art experimental tools, the measurements must add value for model/code development.
- The resulting experiments are cultivated in the context of *mesoscale simulations.*
- The selected thin-film/laser drive configuration and film characterization provides well-controlled conditions needed for model-development support.
- Mesoscale simulations will be conducted (Baer, Lechman) for the films under consideration to help guide the experiments and provide early feedback to the code.
- The Ultimate Milestone for this project will be a direct comparison of mesoscale-resolved shock and thermochemical data to a mesoscale simulation result.

Schedule/Goals/Milestones

Fiscal Year 2012

Design and Fabrication of Material Samples

Homogeneous sample fabrication 12/31/2011

Fiscal Year 2014

Evaluate Heterogeneous Sample and Diagnostic Performance and Redesign

Construction of final heterogeneous samples for code-comparison data 12/31/2013

Down selection and modification of thermochemical state diagnostics 03/31/2014

Conduct Final "Capstone" Code-Comparison Experiments

Fabrication of Final Heterogeneous Samples 03/31/2014

Temperature/Pressure/Species/Shock u_s and u_p measurements 08/01/2014

Fundamental energy transfer and hot-spot observations 08/01/2014

Final Documentation/SAND Report 09/30/2014

Raman (temperature. & vibration state) imaging on stationary targets 09/30/2012

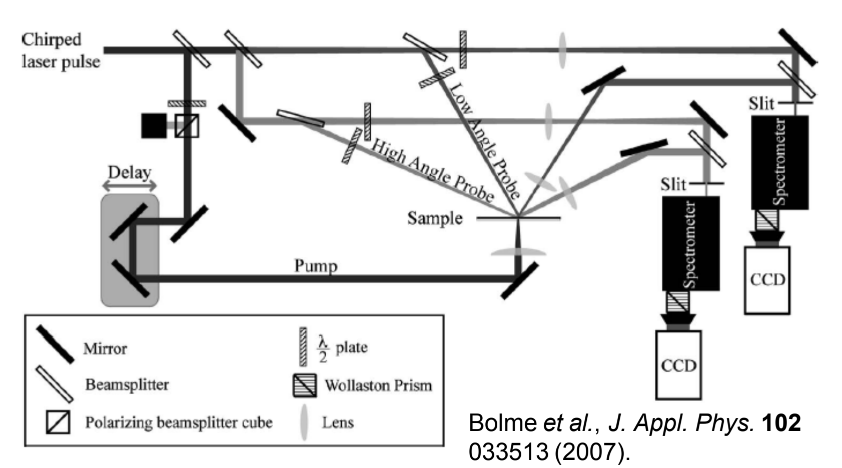
IR/UV/LIF imaging (species, temperature, energy) on stationary targets 09/30/2012

Evaluate performance for temperature, pressure, species, and state data 09/20/2012

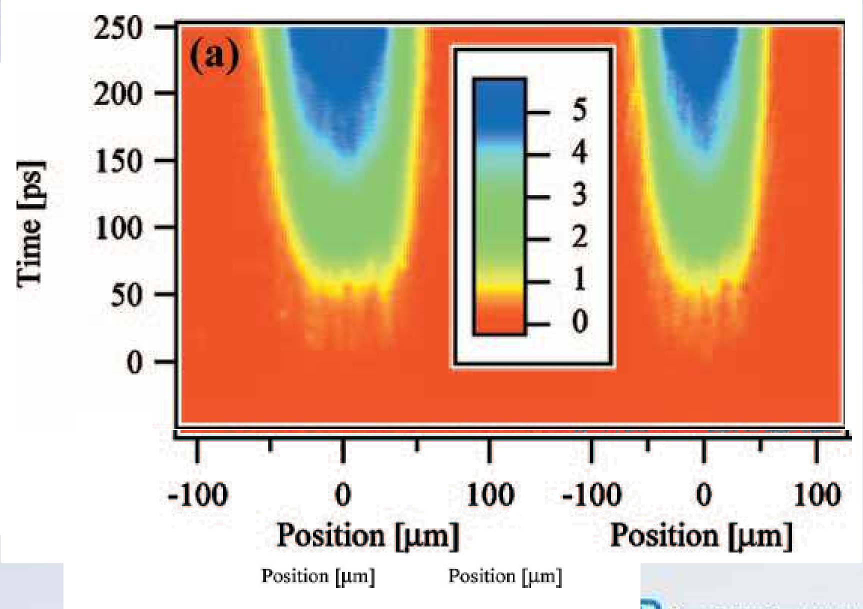
BACKUP SLIDES

What will we measure and how will we measure it?

- Mechanical Characterization (Shock Propagation): u_s, u_p, *Pressure*
  - Ultrafast Dynamic Ellipsometry (UDE)
  - Developed at LANL for studies of homogeneous materials
  - Adaptable for spatially resolved imaging
  - Chirped probe beam encodes time-domain information into frequency domain
  - Interferograms are spectrally (time) resolved and imaged along a 1-D spatial line.
  - Phase shift information yields displacement vs. time information.
  - u_s, u_p, *Pressure* obtained from Hugoniot and unshocked density

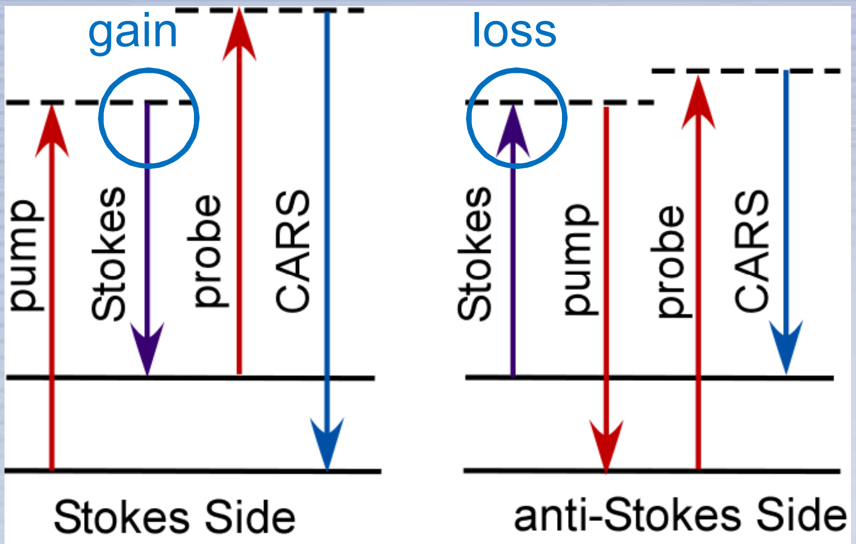


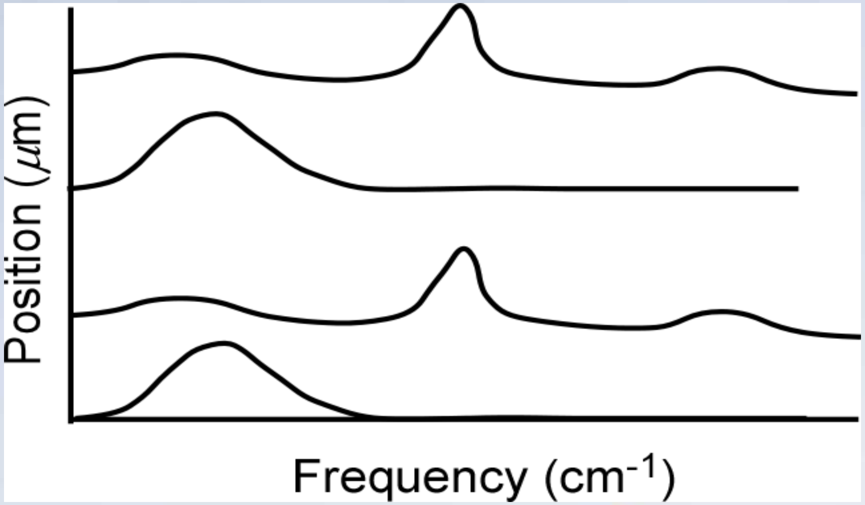
Bolme *et al.*, *J. Appl. Phys.* **102** 033513 (2007).



What will we measure and how will we measure it?

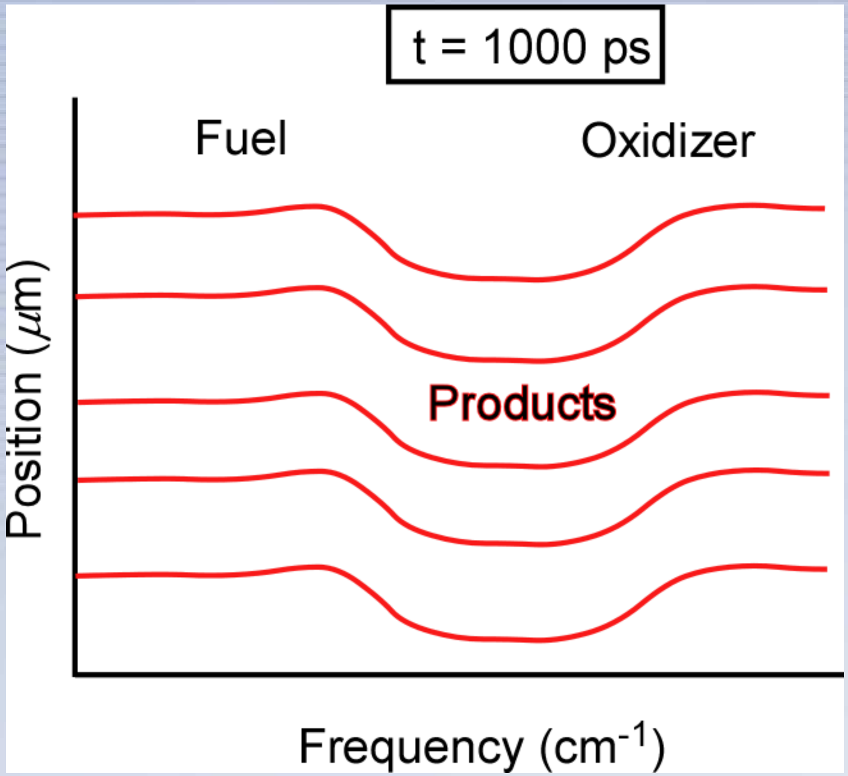
- <u>Temperature, Vibrational Energy, Species</u>
    - Nonlinear Raman spectroscopy
    - Coherent anti-Stokes Raman Scattering (CARS)
    - Stimulated Raman Gain/Loss Spectroscopy (SRS)
- Vibrational state population is reflected in CARS/SRS spectra
- Raman frequencies are species (or bond) specific
- Quantitative temperatures may be possible with SRS Stokes/anti-Stokes ratio
- “Hot-Spot” detection with spatially correlated imaging spectroscopy





What will we measure and how will we measure it?

- Chemistry/Species/Vibrational Energy
  - Linear IR/UV Absorption Spectroscopy
  - Observe temporal evolution of spatially correlated spectra
  - Broadband fs pulses may provide access to multiple species simultaneously
  - More straightforward to implement than nonlinear Raman
  - Can readily performed in 2-D imaging mode



Risks and Mitigation

1. *Laser drive experiments in heterogeneous films will be difficult. Sample thickness and shot-to-shot repeatability may be problematic.*
- Construct Laser Amplifiers for higher energy drive
- Sapphire substrates may improve impedance match
- Stochastic description is enabled by experiment design

2. *Complex state-of-the-art experiments are high risk and non-trivial. Schedule is very aggressive.*
- Staff has extensive experience with fs lasers and diagnostics
- Parallel work approach in multiple femtosecond laser facilities
- Close collaboration with Moore group at LANL
- Qualitative information may still yield enormous value in many cases

3. *Budget needed for significant diagnostic development, sample fabrication and mesoscale simulations*
- Cost leveraging with sample fab projects
- Mesoscale code is sufficiently developed to run simulations