Lawrence Livermore National Laboratory

Utilization of Versatile Experimental Diagnostics to Characterize Materials Response

October 12, 2015

Joseph (Joe) Zaug
Physical Chemist
Experimental Group Leader - PLS/MSD

R&D Support by WCI HE Science Campaign – II and the Joint Munitions Program, TCG-III

WCI WEAPONS AND COMPLEX INTEGRATION

JOINT DoD/DOE MUNITIONS PROGRAM 1985 MOU

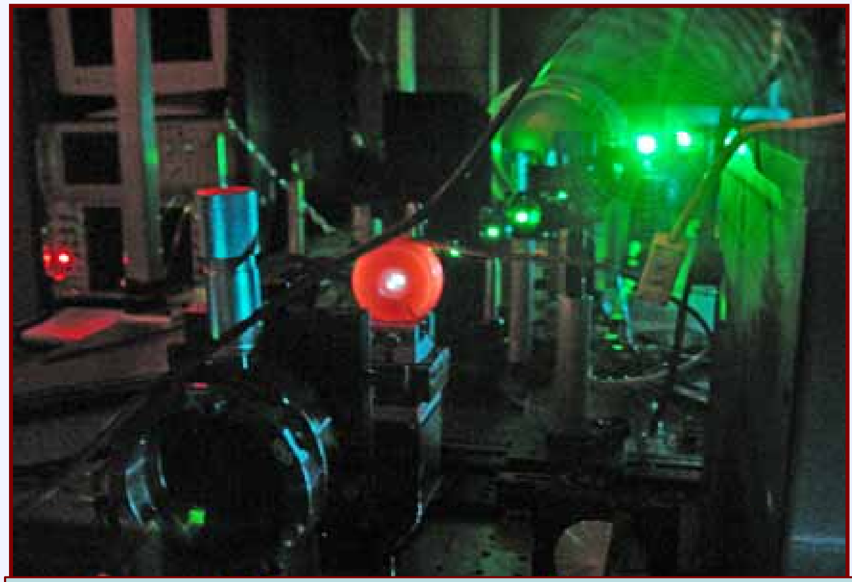
Lawrence Livermore National Laboratory, P. O. Box 808, Livermore, CA 94551
This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

EMC ENERGETIC MATERIALS CENTER

"The Accuracy of Computational Predictions, Rests on the Quality of Relevant & Available Extreme Condition Materials Experimental Data"

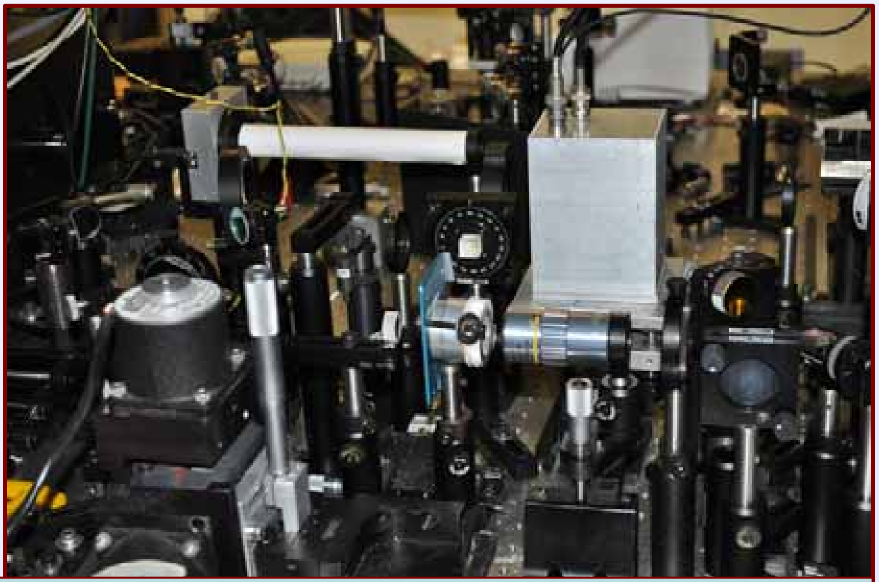
***I**mpulsive **S**timulated **L**ight **S**cattering*
***P**hotoacoustic **L**ight **S**cattering*

Ultrasonics – Isothermal EoS



Indirect Drive Tabletop Compression
***U**ltrafast **T**ime **D**omain **I**nterferometry*

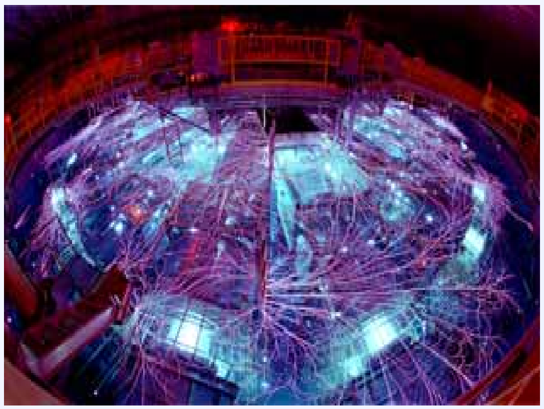
Ultrafast Compression – Dynamic EoS



Utilization of High-Throughput (Laboratory-Scale) Diagnostics for Faster & Cheaper Optimization & Validation of Detonation Performance, Safety, and Hydrodynamic Predictive Codes

Facility- and Large-Scale Experiments and Tests Validate or Invalidate Predictions of Extreme Condition Phenomena

Z-Pinch-SNL ~ 10 Mbar,
< 1000 K , < 3 us time scale



Castle Yanke test, 13.5 Mt, 1954

These *platforms* characterize the dynamic response of materials and systems

low-throughput impedes statistical analysis

NIF- LLNL, 1.8 MJ , 500 TW
< 100 Mbar, 100 M K,
10 ns time scale

Gun shots < 20 Mbar, < 25,000 K
100 ns – μs time scale

Small-scale platform R&D improves the confidence of S-E theoretical predictions

Colleague Acknowledgements

Experimental Team:

Mike Armstrong – Ultrafast Compression Experiments

Jonathan Crowhurst – High Pressure Experiments

Chris Grant – Isotropic HE Preparation and Experiments

Harry Radousky – Shock Wave Physics

Elissaios Stavrou – EOS of α – NTO, HP XRD Experiments

Theoretical Team:

Ryan Austin

Sorin Bastea

Larry Fried

Anisotropic HE Sample Assembly and Characterization Team:

Troy Barbee – Coating consultant

Lou Ferranti – Crystal orientation; Initial polish; < 10 mg xtals

Rick Gross – Final Polishing

Yong Han – HE nucleation/growth consultant

Ray Swan – Sample blocking; Intermediate polish

Nick Teslich Jr. – FIB; Ablator stoichiometry

Mark Wall – Al thickness measurement (Zygo) consultant

HE Materials:

Alan DeHope – Synthetic Chemist

Phil Pagoria – Synthetic Chemist

Mao Xi Zhang – Synthetic Chemist

TID Animations:

Ryan Chen

Kwei-Yu Chu

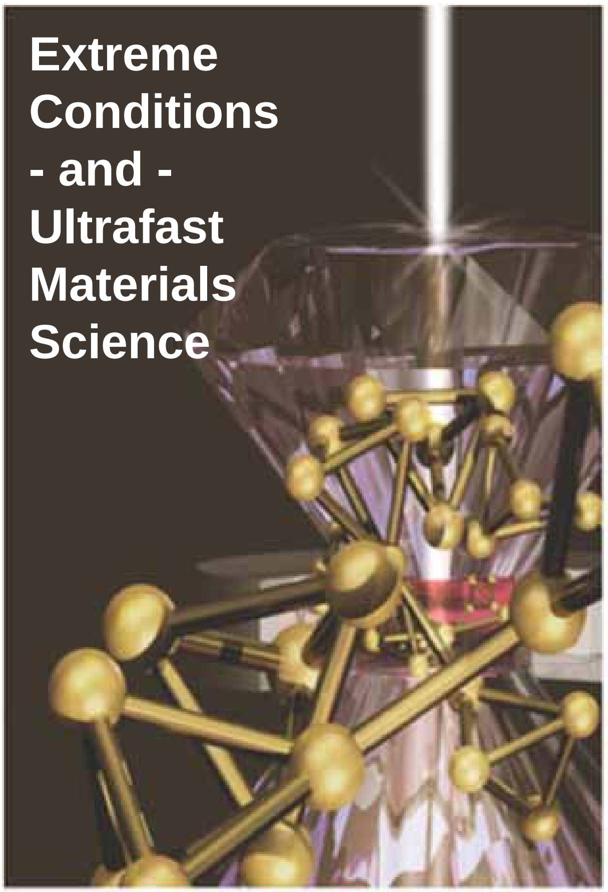
Sandia National Laboratory – ABQ
(JMP Sponsored Collaborations)

Alex Tappan – Isotropic PETN

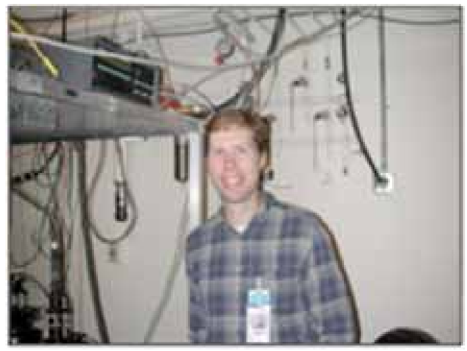
Rob Knepper – Thin Film XRD Characterization

The Experimental Reaction Dynamics Group at LLNL

October, 2015



Mike Armstrong ‘07



Jonathan Crowhurst ‘02

Elissaios Stavrou ‘14

Harry Radousky ‘12

One Can Apply Ultrafast Compression - Time Domain Interferometry (TDI) to Address Important Materials Questions

What is the unreacted EOS of shocked HE (P > Gas Gun Shots) ?

<u>Example</u>: polycrystalline PETN

How anisotropic is the dynamic response of shocked single crystals ?

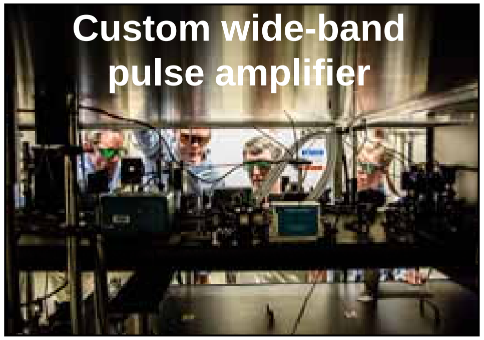
<u>Example</u>: β - HMX single crystals

What is the unreacted EOS of shocked polymer ?

<u>Example</u>: PDMS

Can we characterize the ignition dynamics of energetic materials ?

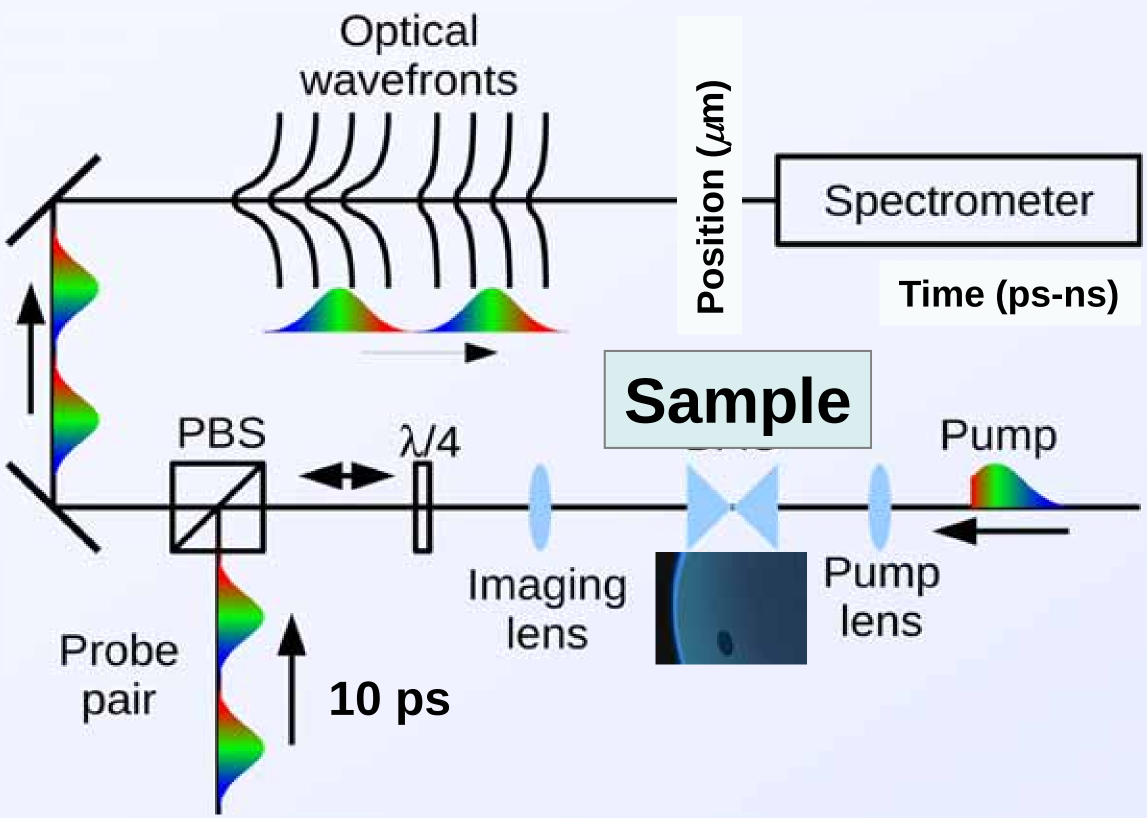
<u>Example</u>: H_2O_2



Ultrafast Tabletop Compression Experiments Address Important Knowledge Gaps

zaug1@llnl.gov 925-423-4428

Ultrafast Time Domain Interferometry (TDI) is a Method to Characterize Dynamic Response Phenomena



Ultrafast-TDI

Version 2.0

Footprint 60' square

Compression wave e.g., ramp, shock, generation and probe technique

1.5 Mbar shock stress in free-standing Al (< 3 μm thick, 400 ps); 18 mJ pulse

Time res. > 1 ps

Spatial res. > 2 μm

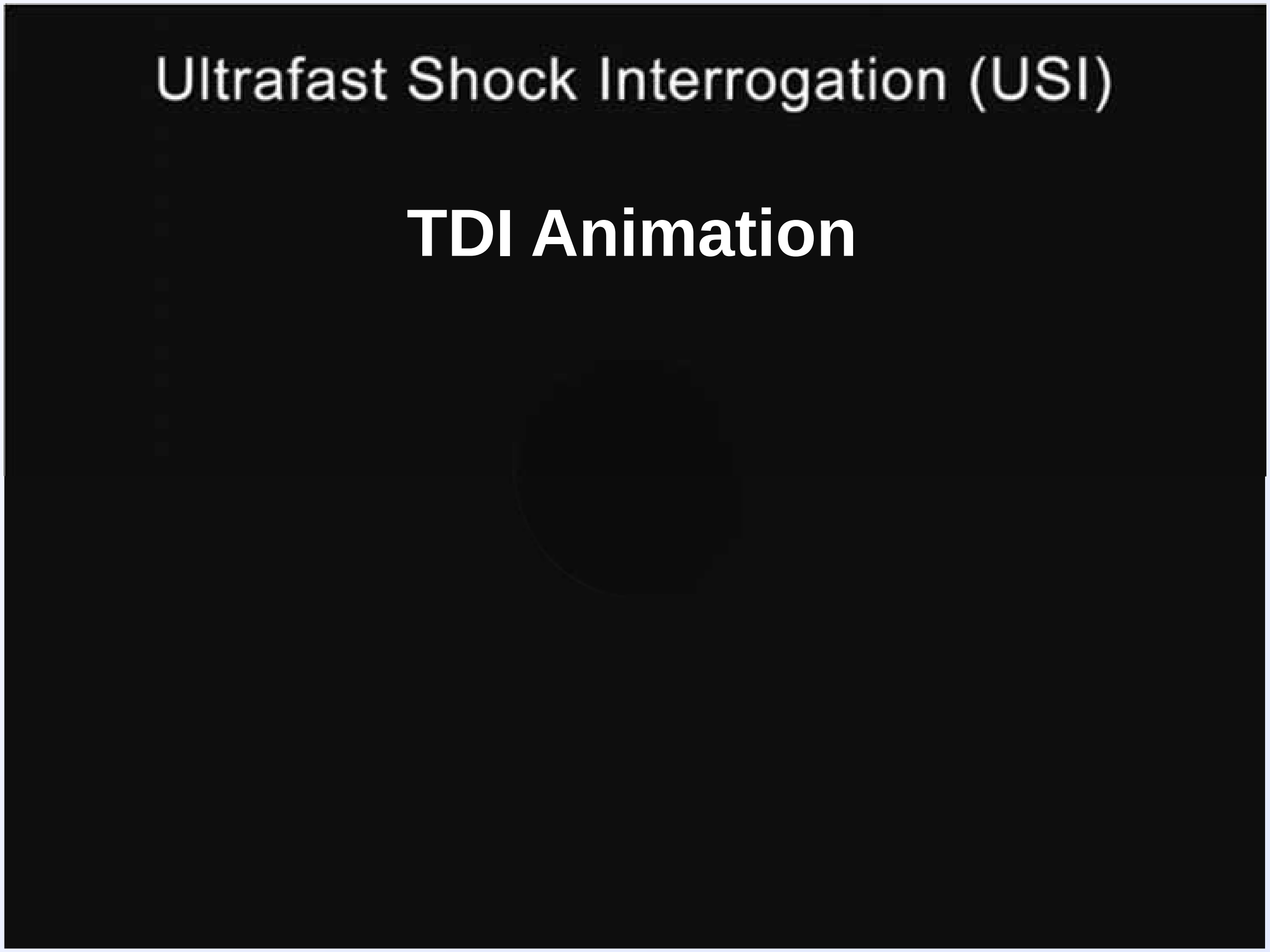
Duration < 1-2 ns

U-TDI + Analysis Examples: M.R. Armstrong et al. *JAP* 108, 023511, (2010)

M.R. Armstrong et al. *APL* 92, 101930, (2008)

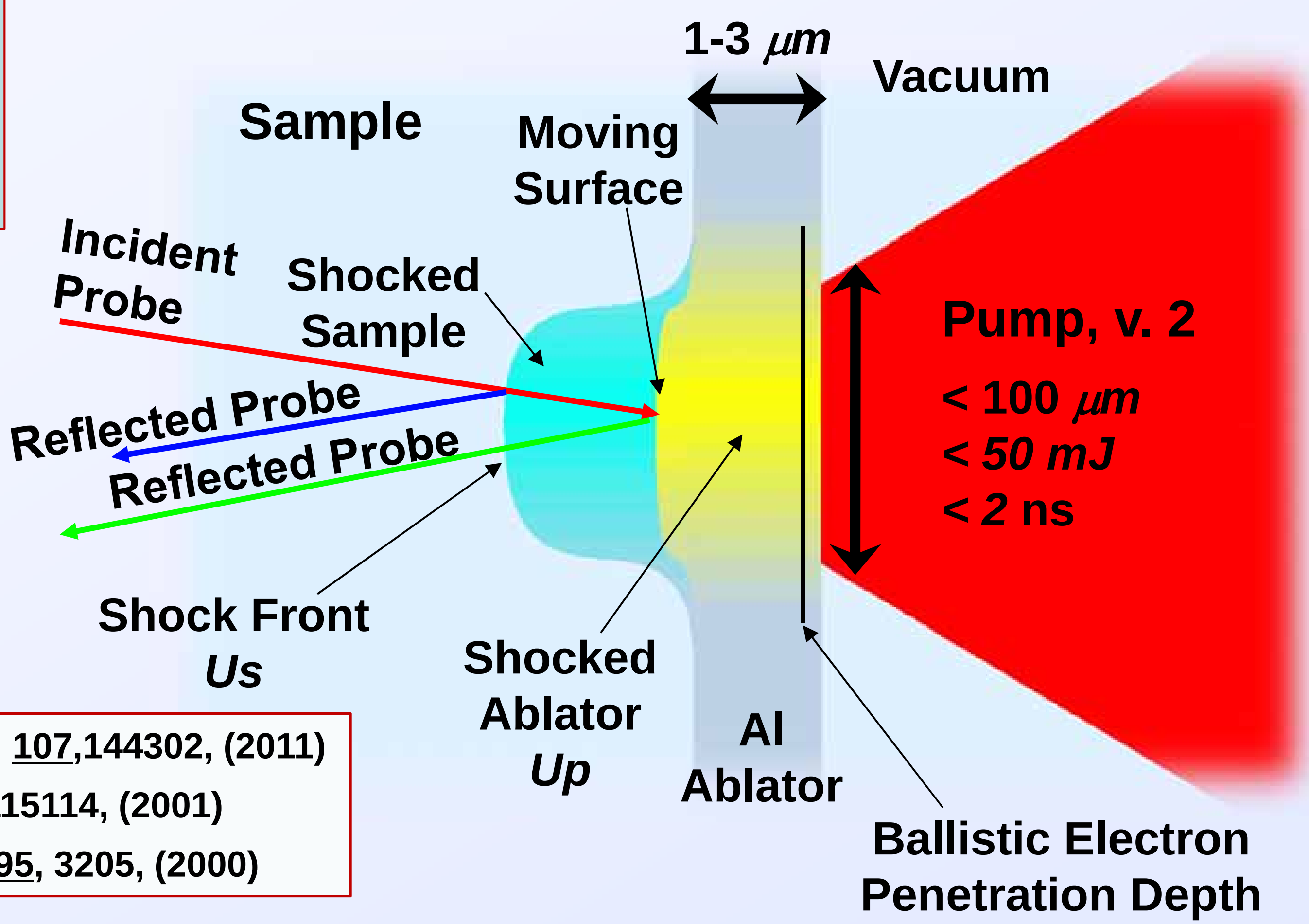
U-TDI is based on many other works

Ultrafast Time Domain Interferometry (TDI) is a Method to Characterize Dynamic Response Phenomena



Tabletop Shock Generation: A Small-Scale Experiment to Characterize Dynamic Properties

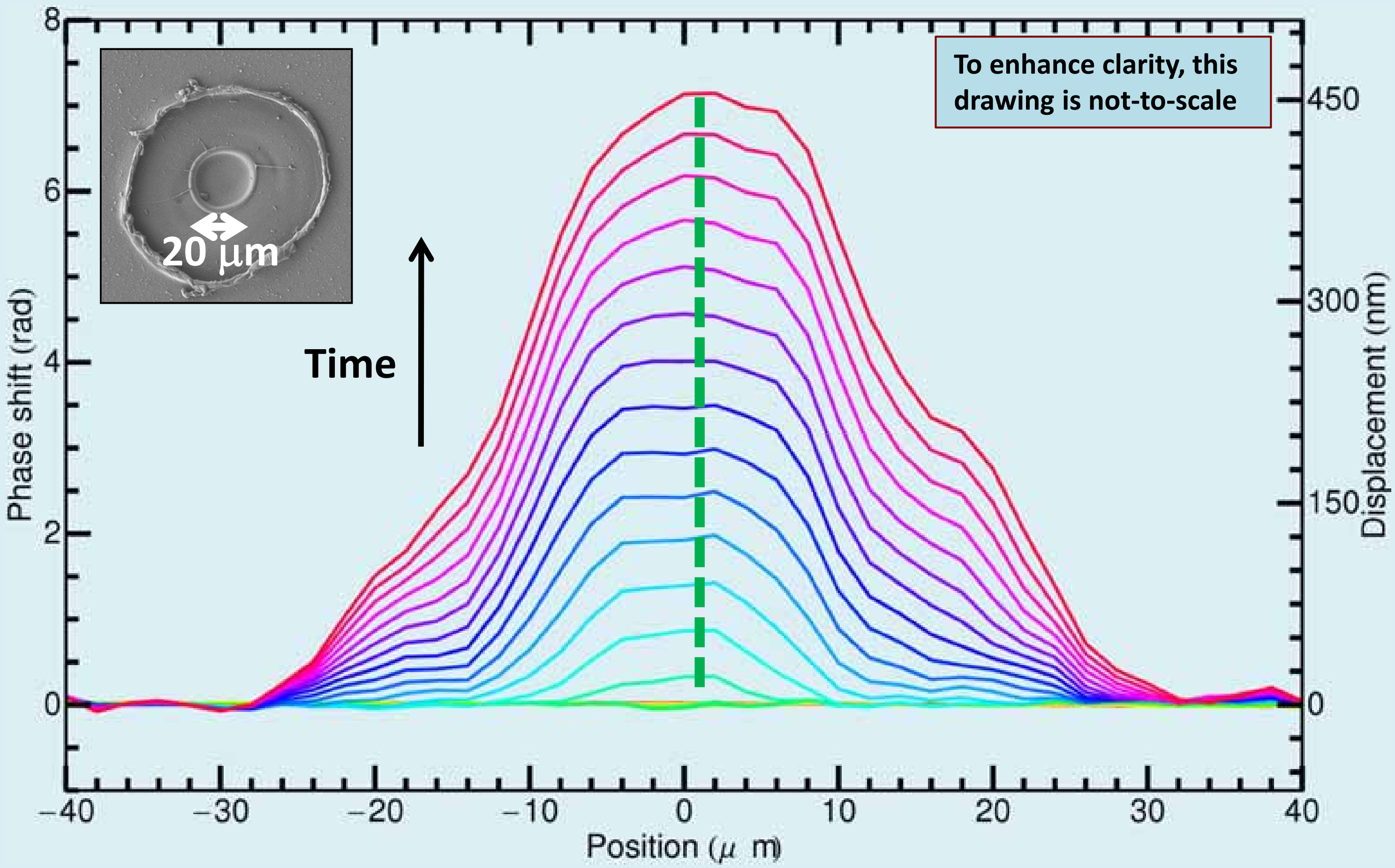
Aluminum is well-characterized on the ultrafast timescale*



* J.C. Crowhurst et al., *PRL* 107,144302, (2011)
* D.J. Funk et al., *PRB* 64, 115114, (2001)
* K.T. Gahagan et. al., *PRL* 95, 3205, (2000)

To enhance clarity, this drawing is not-to-scale

We Effectively Maintain 1-D Hydrodynamic Flow

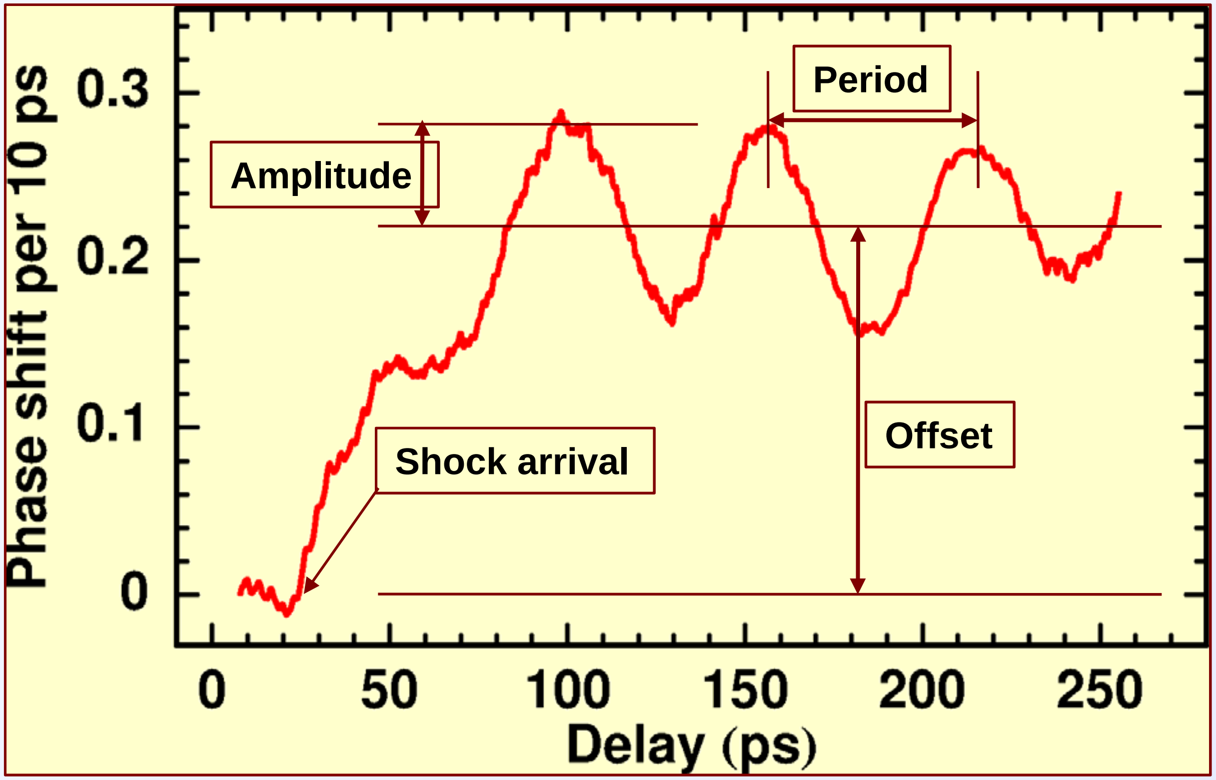


Free Surface Displacement from a 1 µm thick Al Ablator

The propagation distance over the duration of an ultrafast compression TDI experiment is approximately 0.1% of the radius of curvature of the shock front

zaug1@llnl.gov 925-423-4428

Three Interferometric Parameters Determined at the Shock Front Yield U_p, U_s, and n_s, the Index Behind the Wave



Assuming a steady shock wave, geometric relations connect interferometric parameters to shock state parameters, U_p, U_s, n_s

$$|r_s| = \frac{period}{2\pi\Delta t}\frac{|r_m|}{1+|r_m|^2}\, oscillation\ amplitude$$

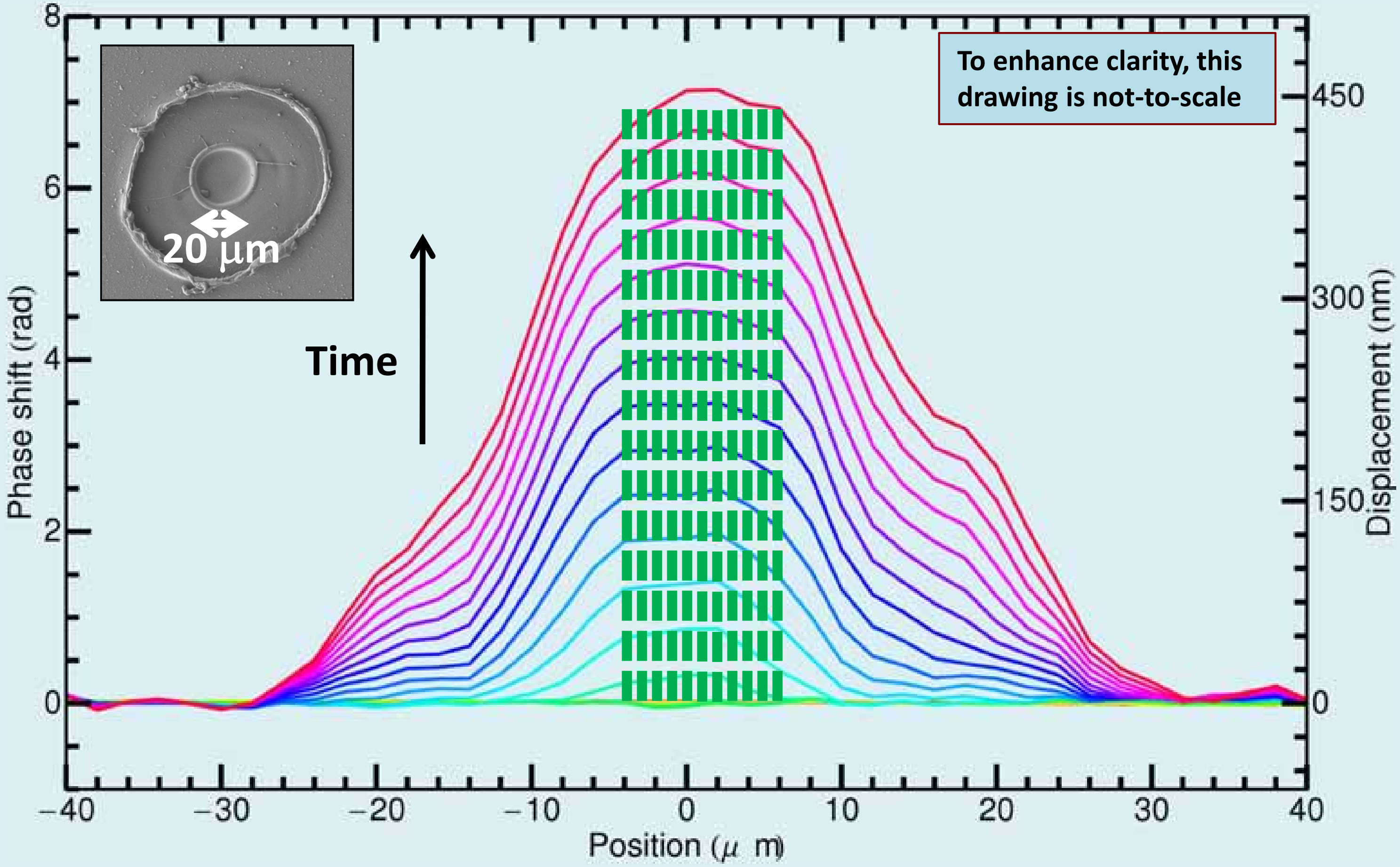
$$n_s = n_0\left(1 + \frac{2|r_s|}{1-|r_s|}\right)$$

$$v_s = \frac{\lambda}{2n_0}\left[\frac{offset}{2\pi\Delta t} + \frac{1}{period}\right]$$

$$v_p = \frac{\lambda}{2n_0}\left[\frac{offset}{2\pi\Delta t} + \frac{1}{period}\left(1 - \frac{n_0}{n_s}\right)\right]$$

- **Offset is analogous to phase-shift in VISAR, giving the velocity of a free surface**
- **Period is from interference between light reflected from the metal ablator and the shock front – rate at which the shock front propagates away from the *piston***

For Sufficiently Planar Shocks, One Can Determine Multiple Shock Hugoniot States From One Measurement



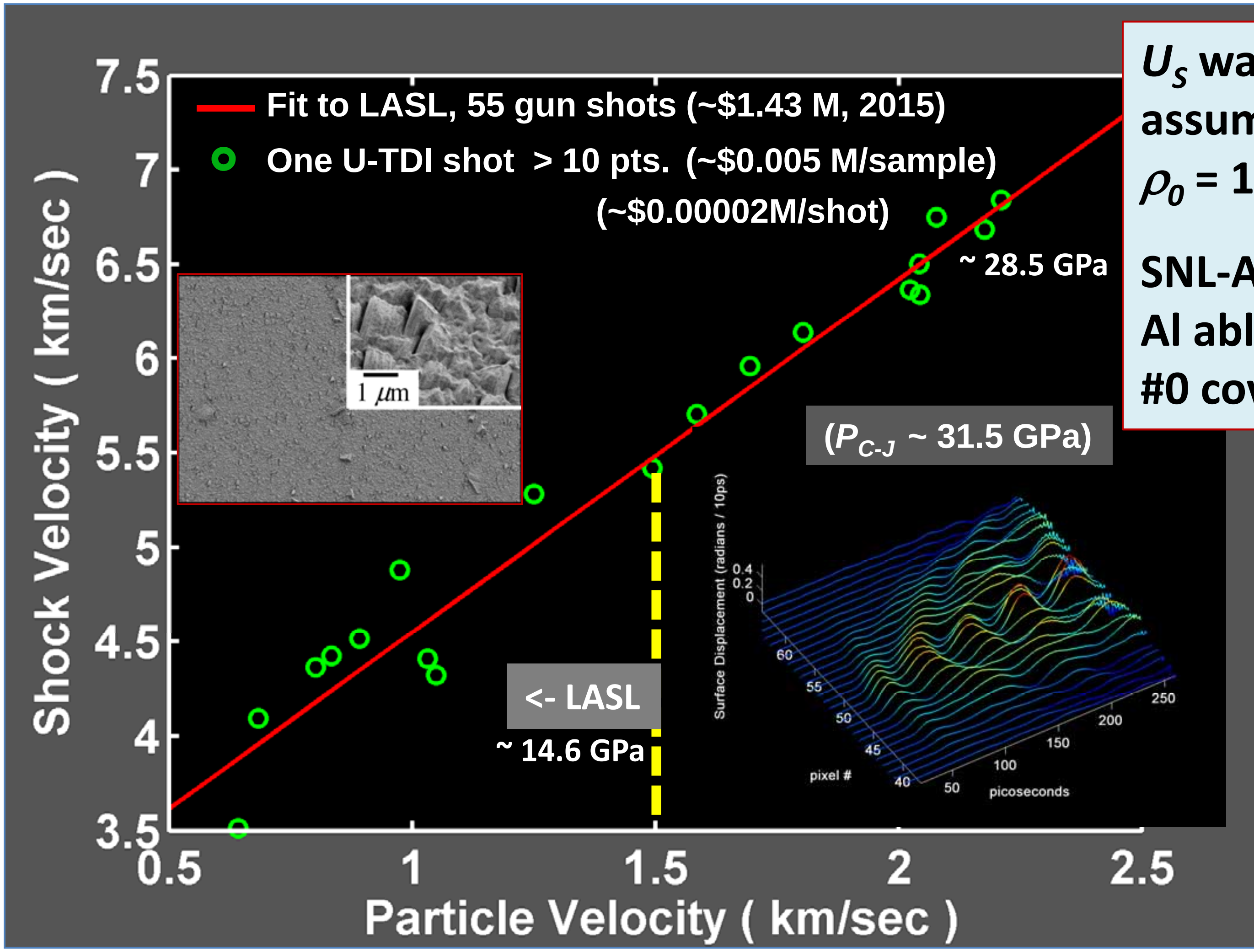
Free Surface Displacement from a 1 μm thick Al Ablator

The propagation distance over the duration of an ultrafast compression TDI experiment is approximately 0.1% of the radius of curvature of the shock front

zaug1@llnl.gov 925-423-4428

U-TDI is Complementary to Conventional Gun Platforms; it Enables Rapid Hugoniot Characterizations Closer to or Above C-J Pressures

PETN Principal Hugoniot Data from One U-TDI Measurement



U_S was determined assuming 100% TMD,
ρ_0 = 1.77 g/cm³, n_o = 1.552

SNL-ABQ PETN, 3.5 μm
Al ablator, 0.5 μm
#0 coverslip substrate

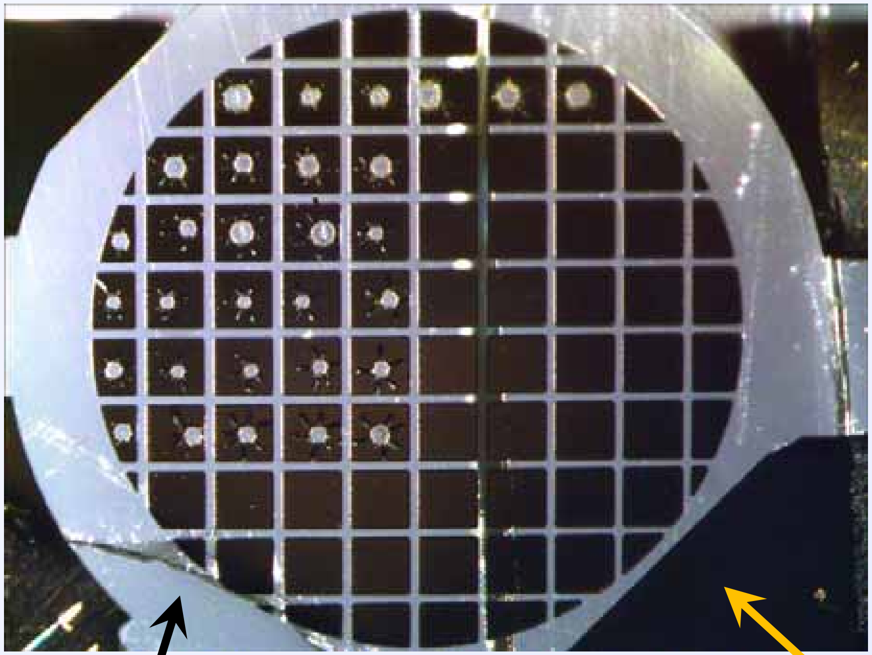
100's of shots/day

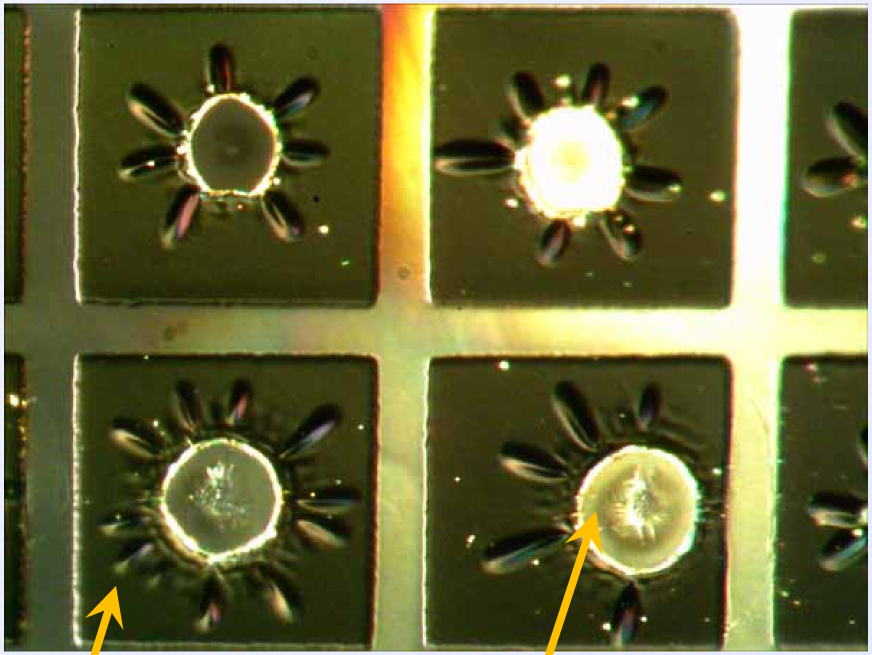
zaug1@llnl.gov 925-423-4428

1 μm of Al is Deposited onto a Single Crystal HE – a TEM Grid Serves as a Mask (125 μm x 125 μm)

β - HMX after 30 TDI shots (< 300 μJ, 350 ps)

There is visual evidence of shock initiated decomposition





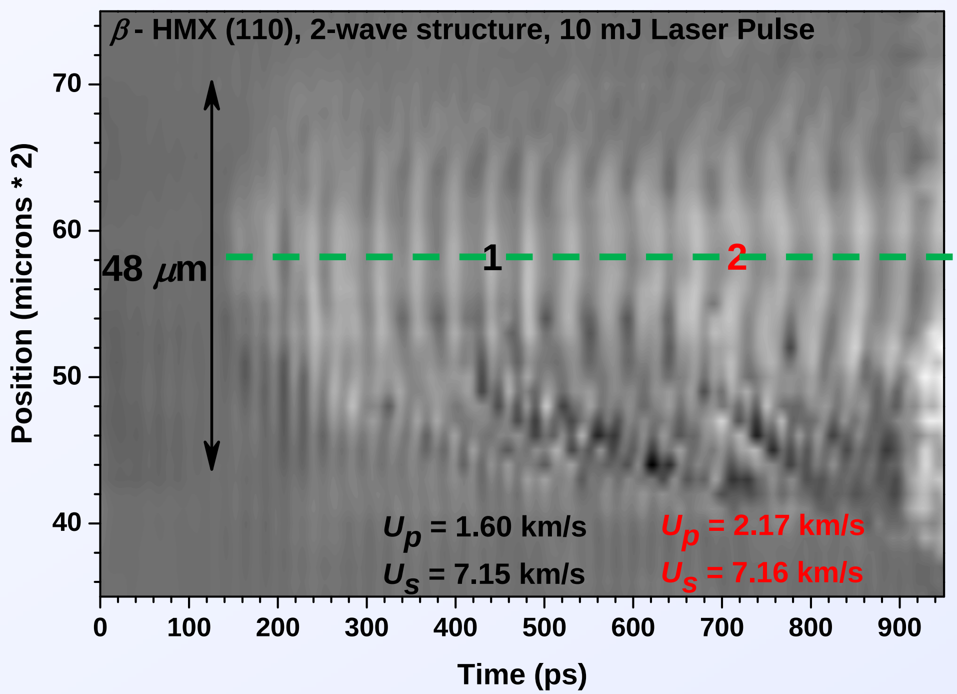
β - HMX

Al

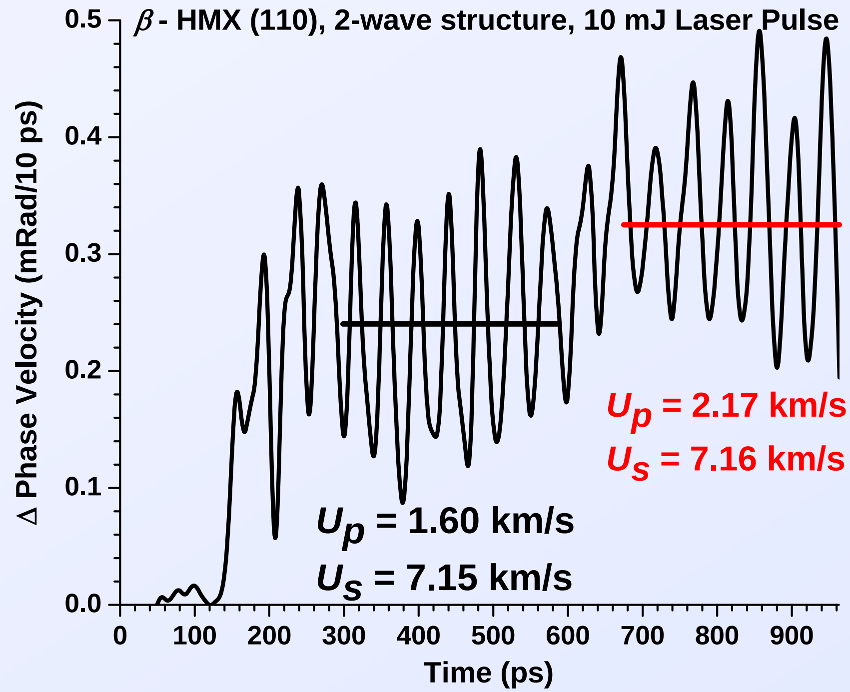
Ablated Al

2-Wave Phenomena Observed in (110) β - HMX, t > 350 ps

2D Contour Plot

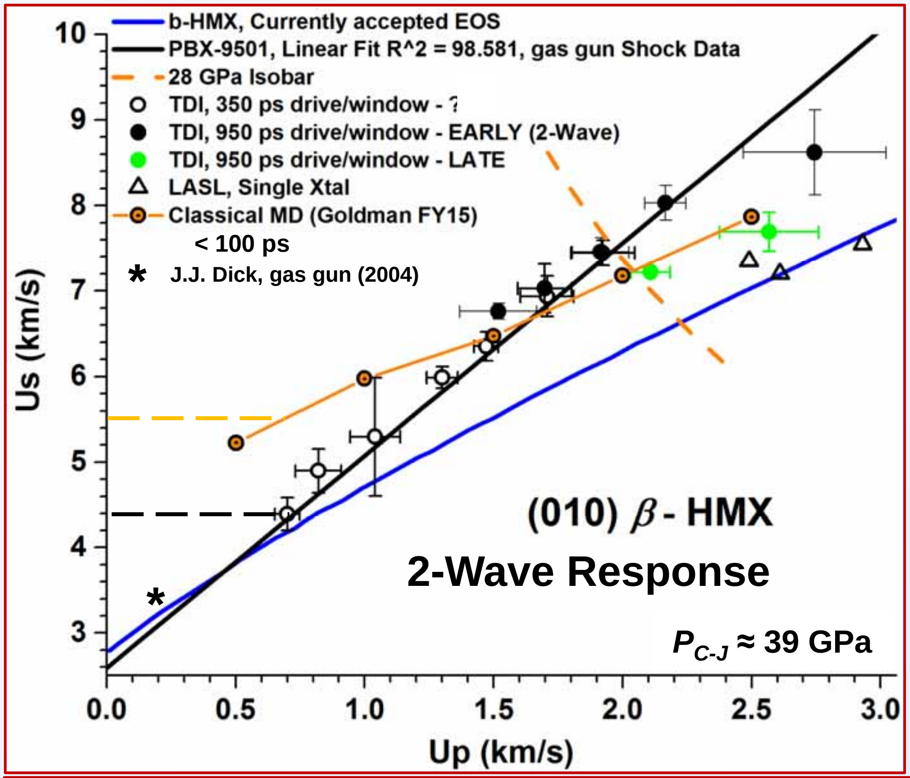


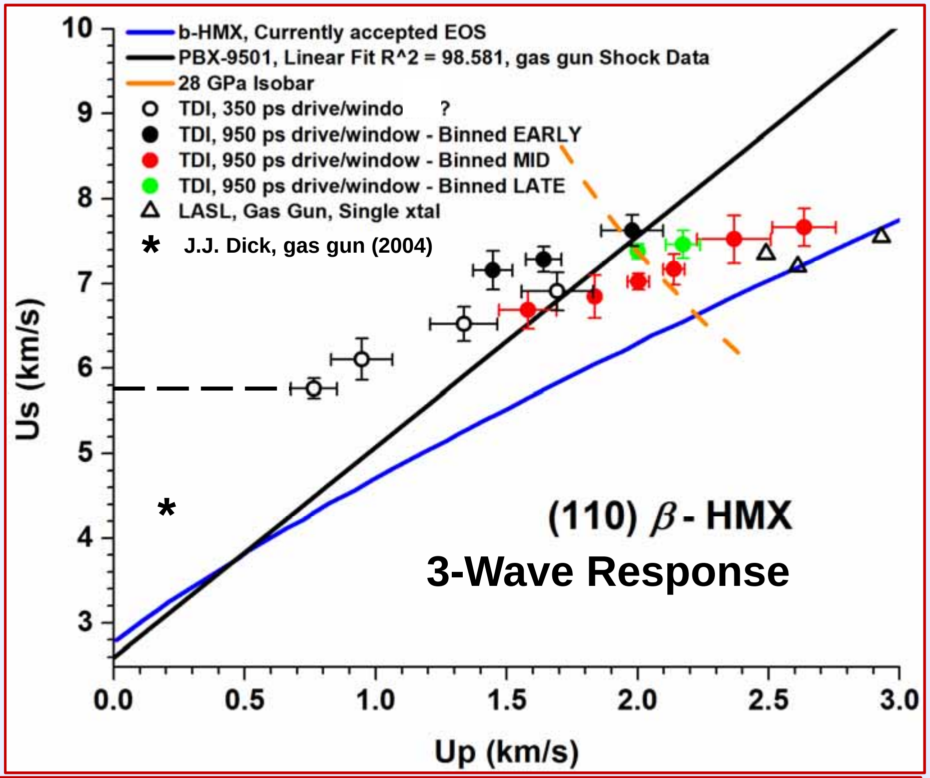
Integrated Signal Along 1 CCD Row



A ~ 36% increase in *Up* may be attributed to a decrease in volume, which may be attributed to a solid-solid phase transition or an endothermic Rxn

β-HMX Exhibits 2- and 3-Wave Responses to High Strain-Rate Shock Impacts on the 1 ns timescale





For the (010) impact ([101] direction), only brittle failure occurs – no twinning or slip stress relaxation occurs Palmer & Field, *Proc. Roy. Soc. Lond. A.* 383, 254 (1993)

For the (110) impact ([001] direction), twinning & cleaving occur before brittle failure – higher shock stress states will occur Rae, Hooks, & Liu, *IDS*-13, 293 (2006)

TDI Data from (010) and (110) β- HMX Single Crystals Reveal an Anisotropic Response to High Strain-Rate Compression

β- HMX (110) and (010) shock loaded crystals respond differently

J.J. Dick et al., JAP 96, 374 (2004) – From Lit. data:

Plastic relaxation mechanisms (twinning and cleaving) are operative for impacts driven into the (110) plane; hence, higher stress achieved prior to fracturing

For the (010) plane, brittle failure is the only failure mechanism; hence, higher stress elastic wave precursors occur prior to plastic wave propagation

We appear to observe a distinct transition from primarily elastic to primarily plastic wave propagation – given previous gas gun results

Are 3rd wave (110) data representative of reacted β– HMX ?

Previous DAC studies* report a phase transition, burn-rate discontinuities, and Raman spectral changes at ~ 26-28 GPa

*Yoo et al., *JCP* 111, 10229 (1999) – Cold Compression EOS Data, 4% volume reduction at ~ 28 GPa

Zaug et al., *APS Proc. SCCM* 1195, 420 (2009) – Discontinuities in HP Deflagration Rates and P-Dependent Raman Spectra

How do Ultrafast TDI Results Help Advance Modeling Efforts?

- **Wave speeds are controlled by the full stiffness tensor ***
  - TDI measurements may allow for refinements of the crystal stiffness tensor, which encompass bulk and non-bulk responses
- **Insight to plastic deformation (or lack thereof) on this time scale (100s ps)**
  - How do the measurements compare to plasticity kinetics determined from longer time scale experiments?
  - When do the shocked states reach equilibrium?
- ***Planned* simulation work**
  - Test crystal mechanics models against TDI data
  - Compute wave speeds
  - Investigate elastoplastic deformations

*** Most HE mechanics models may overlook these issues…**

Mechanical responses of low-symmetry crystals tend to be highly anisotropic relative to the loading axes

Many solids exhibit a sharp increase in flow stress when subjected to rapid change in strain rate

If plastic relaxation occurs on a similar time scale to pore collapse, high shear stresses will persist until they are relieved by dissipative inelastic deformation

U-TDI Experiments Conducted on PDMS Serve to Test DFT predictions: Results are Similar to Sylgard-184 Shock Data

Sylgard-184 consists of knotted strands of PDMS

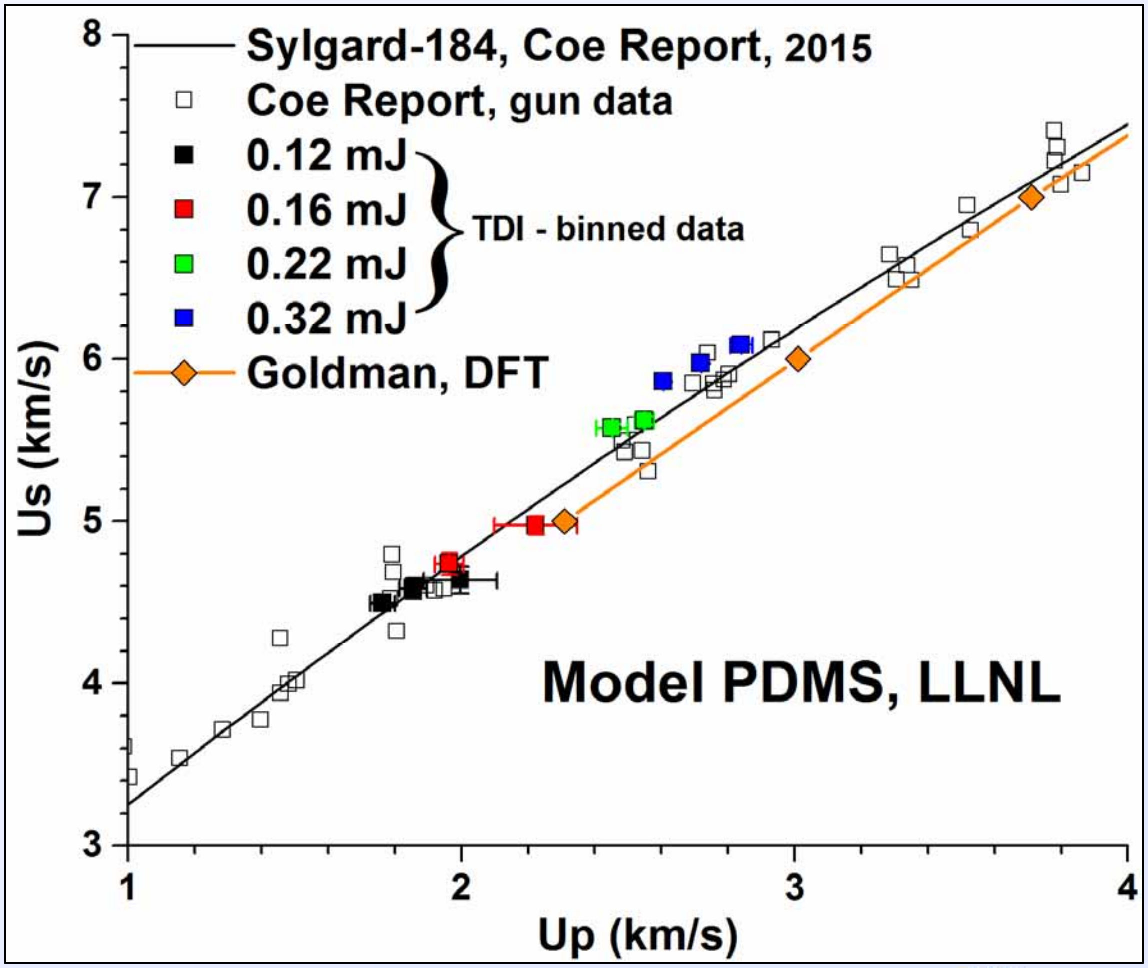
$\rho_{Sylgard} > \rho_{PDMS}$ (Δ=10%)

TDI data extend to 950 ps

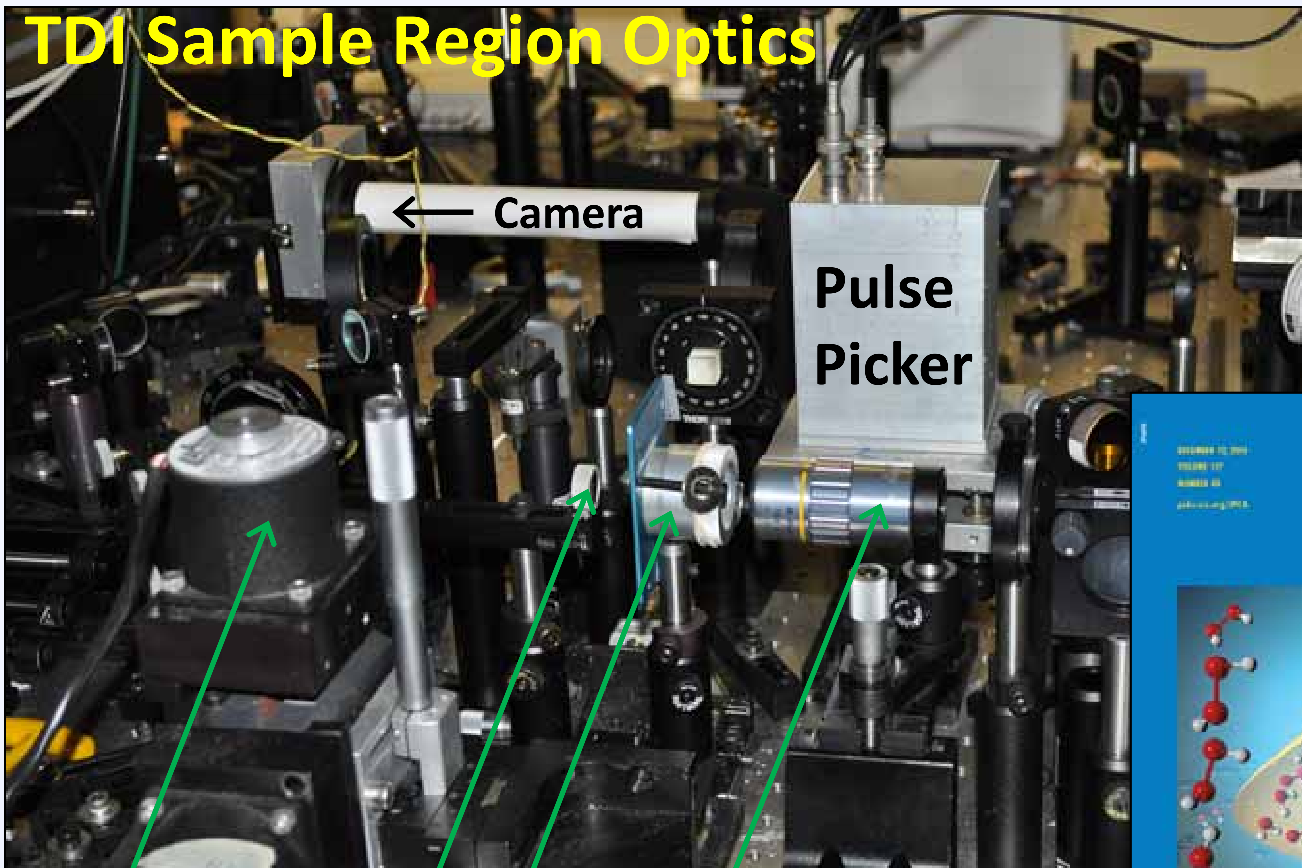
DFT results extend to 80 ps

- ➢ Freestanding samples nxt
- ➢ AM polymers to be investigated using TDI and DCS – 7 orders of magnitude in strain rate

Poly Dimethyl Siloxane (PDMS) Unreacted Shock Hugoniot



We Shock Energetic Fluids to Provide EOSs for Thermochemical Codes and to Validate Molecular Dynamics Simulations?



Sample Stage

Pump Focus

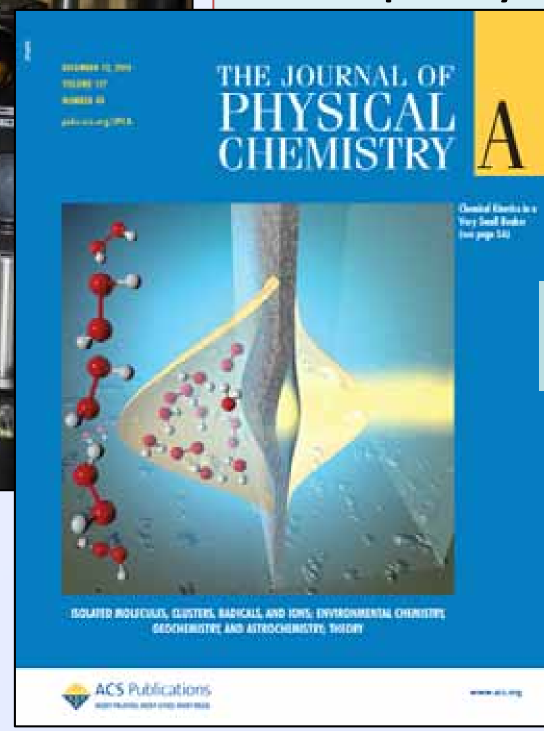
Sample

Probe Objective

Fluid is 100-750 um thick, 20 mm in diameter

AR coated probe window; ~33 mm objective stand-off

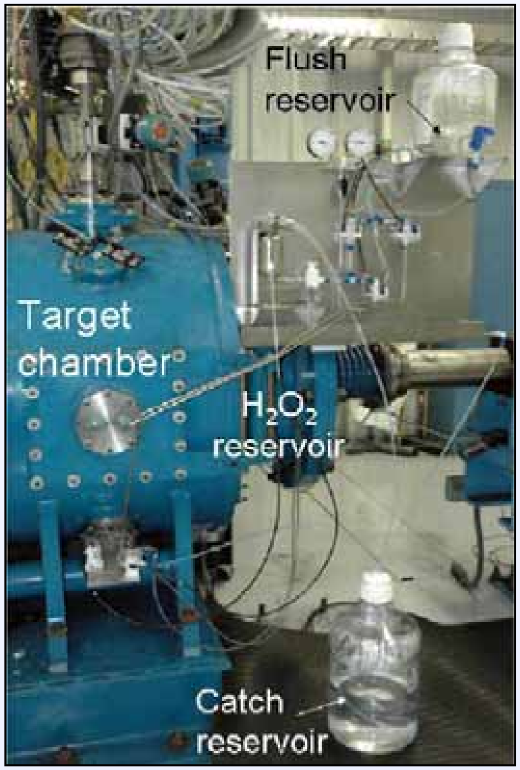
Motorized stage; hundreds of shots per day

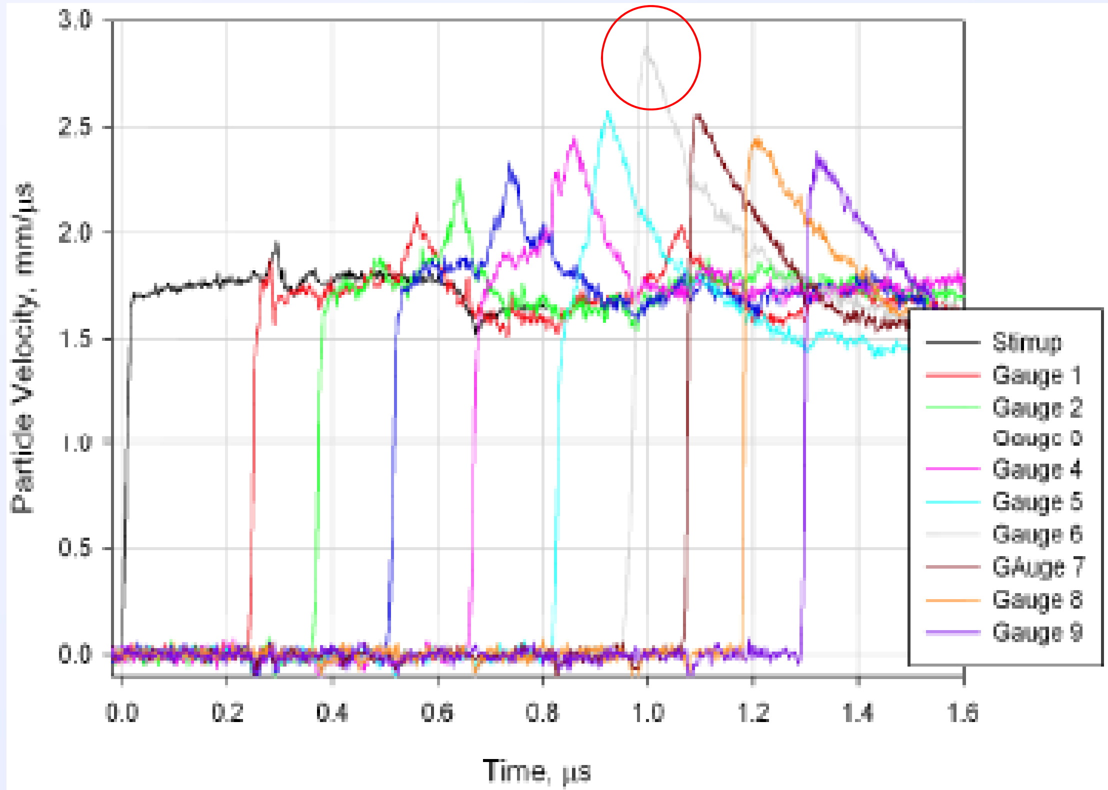


H_2O_2

Gas Gun Data from 98 wt% H_2O_2 Signals Decomposition on the μs Time Scale – DFTB Results Indicate Ignition in 75 ps

Time to ignition from DFTB (~ 75 ps) does not reconcile with two-stage gas gun shock results (~100,000 ps)

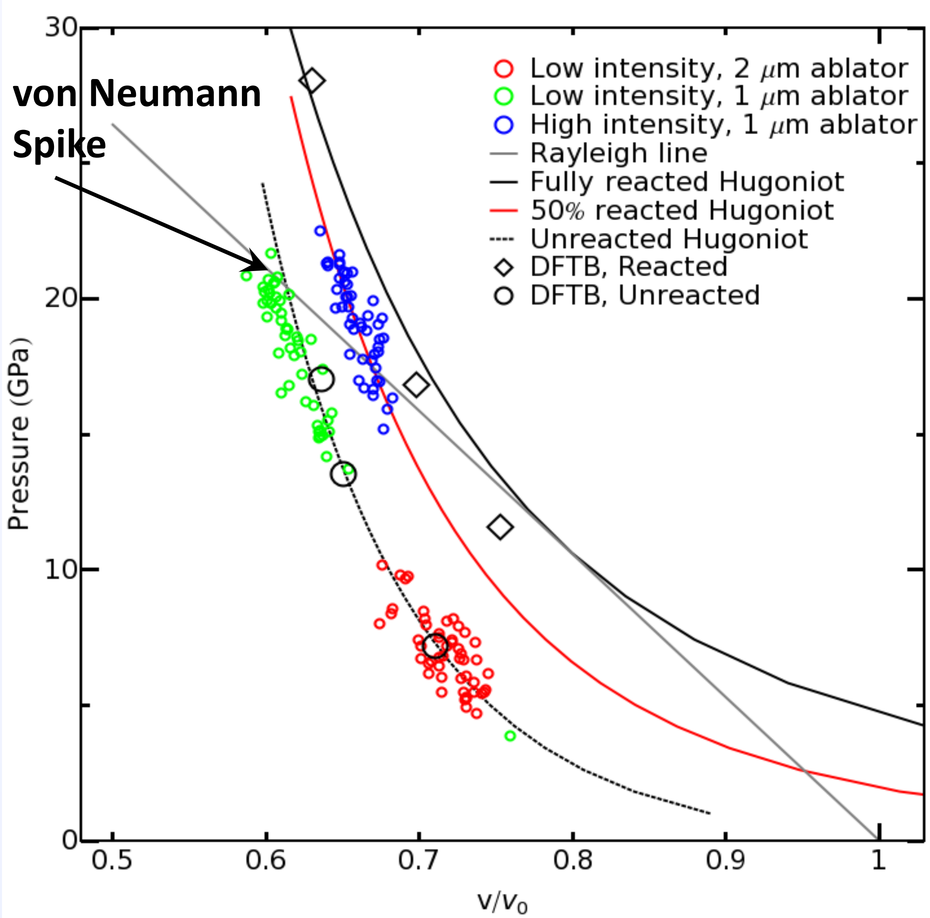




L.L. Gibson, B. Bartram, D.M. Dattelbaum, S.A. Sheffield, and D.B. Stahl, **A Remote Liquid Target Loading System For a Two-Stage Gas Gun,** *SCCM* pg. 135, (2009)

How do Ultrafast TDI Results Advance Modeling Efforts?

TDI | DFTB | Cheetah EoS results for H_2O_2



The Cheetah-code thermodynamic model (constrained by ISLS data) predicts Hugoniot states of shocked peroxide

1 atm. liq. *ρ*, *Pc*, and *Tc* values are reproduced well by the current Cheetah model

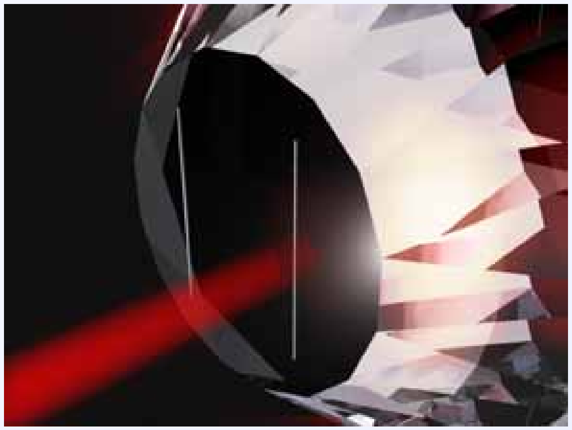
Identical pressures for some unreacted and reacted states?

M.R. Armstrong*, J.M. Zaug*, N. Goldman, I.-F.W. Kuo, J.C. Crowhurst, W.M. Howard, J.A. Carter, M. Kashgarian, J. Chesser, T.W. Barbee, and S. Bastea, **Ultrafast Shock Initiation of Exothermic Chemistry in Hydrogen Peroxide.** *J. Phys. Chem. A* 117, 13051 (2013). Cover Article

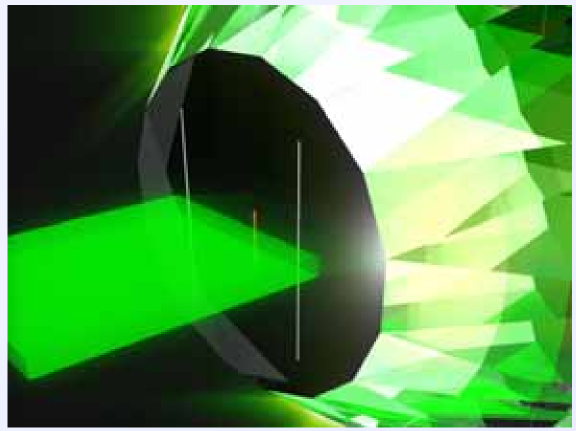
1st published experimental verification of MD shock results on the 10 ps time-scale

Ultrasonic Measurements Yield Adiabatic Sound-Speeds and for Anisotropic Materials Elastic Tensor Data

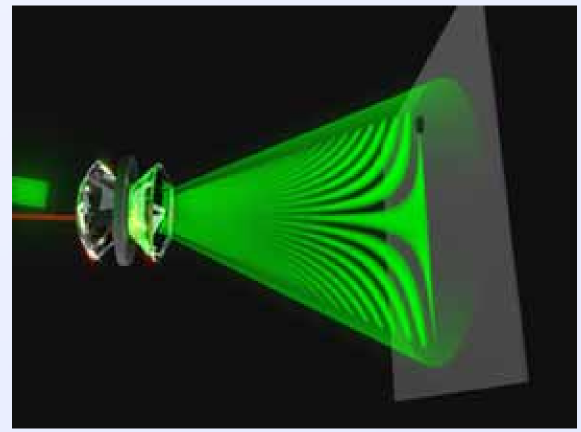
1. Launch acoustic wave

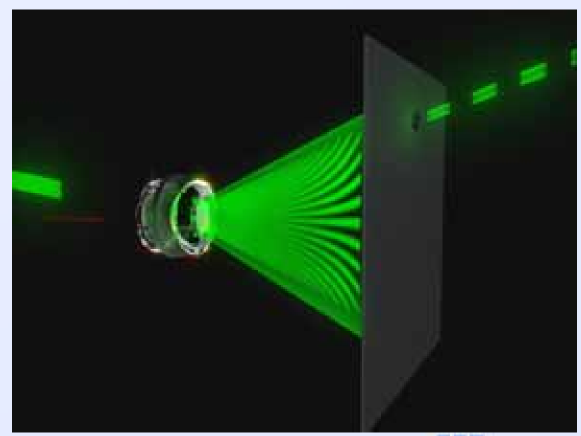


2. Probe acoustic wave



3. Time-Domain sig.





Laser Induced Ultrasonics Provide Data Required to Understand the Nature of Extreme Condition Chemical Reactions

Scientific Challenges and Opportunities

Elucidation of detonation and planetary evolutional processes

Custom Tools Enable us to Extend our Reach

ISLS laser-induced ultrasonics and vibrational spectroscopy

Speeds of Sound sparse P-T fluid data sets, less data on mixtures, describes the thermodynamic state of a fluid

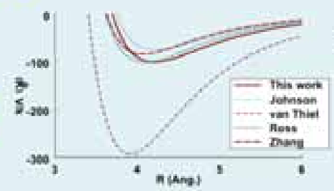
Accurately Parameterized Interatomic Potentials

The exp-6 model has been commonly used for high pressure equation of state modeling

$$V(r) = \frac{\varepsilon}{\alpha - 6}\left[6e^{\alpha(1-r/r_m)} - \alpha\left(\frac{r_m}{r}\right)^6\right]$$

ε = well depth
α = steepness of repulsion
r_m = size

Improved Thermochemical Models



Exp-6 models are not unique, despite the simplicity of the potential.

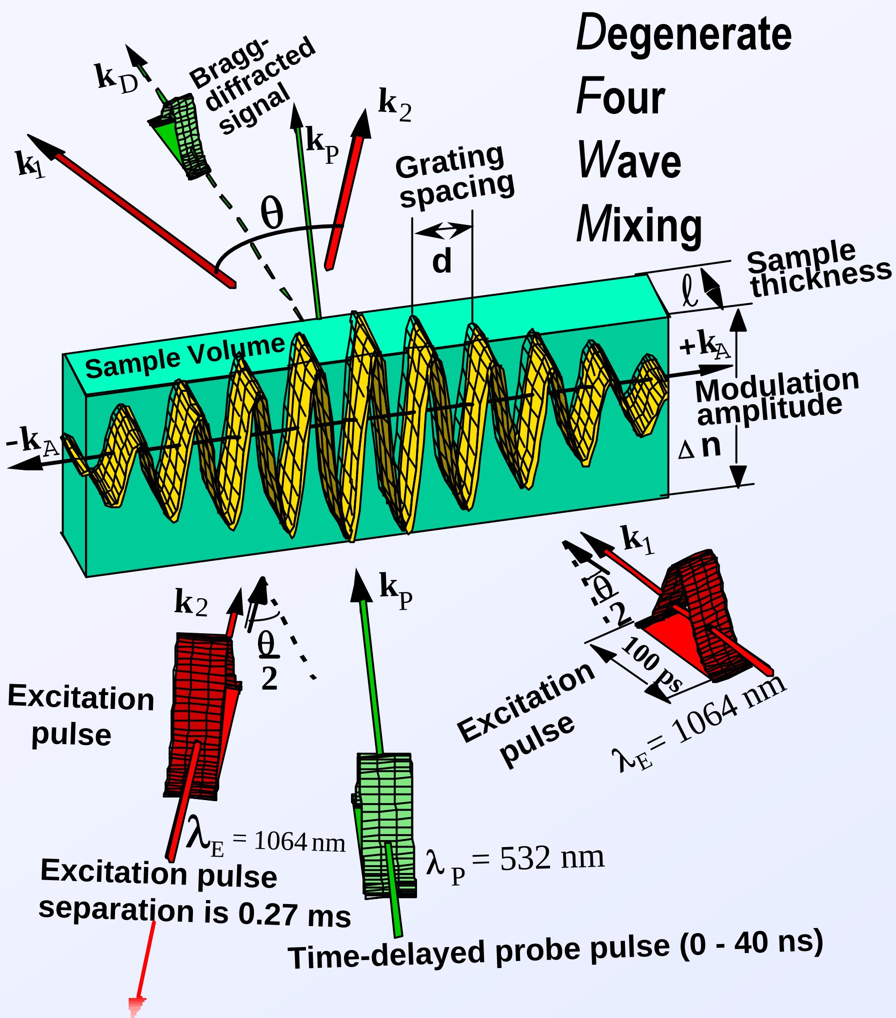
Proposed models for N_2 are shown to the left.

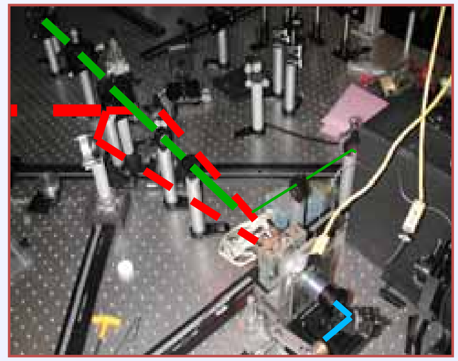
Improved Hydrodynamic Simulations

More confident predictions of extreme condition dynamic chemistry and physics processes

Impulsive Stimulated Light Scattering, (ISLS) is Used to Directly Measure Adiabatic Sound-Speeds – Samples at GPa ‘88*

Laser Induced Time Domain Ultrasonics

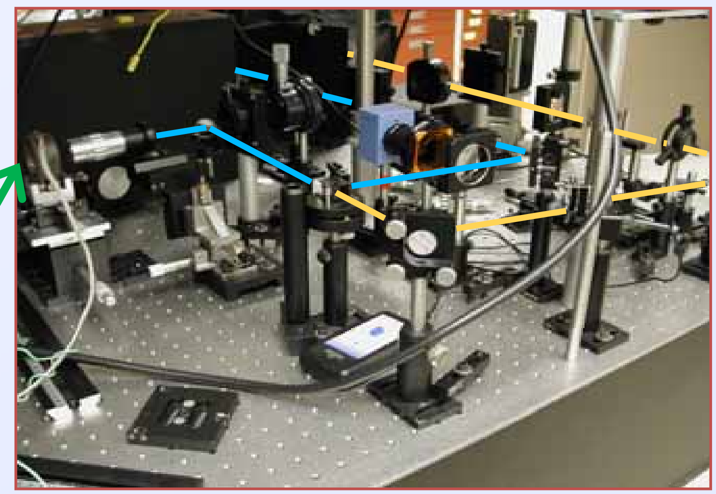




Minerals
Fluids – Mixtures
Metals – Alloys
Polymers
Energetic Materials
Seeded Plasmas

Confocal μ-Raman System to Characterize High P-T Chemical and Phase Stability

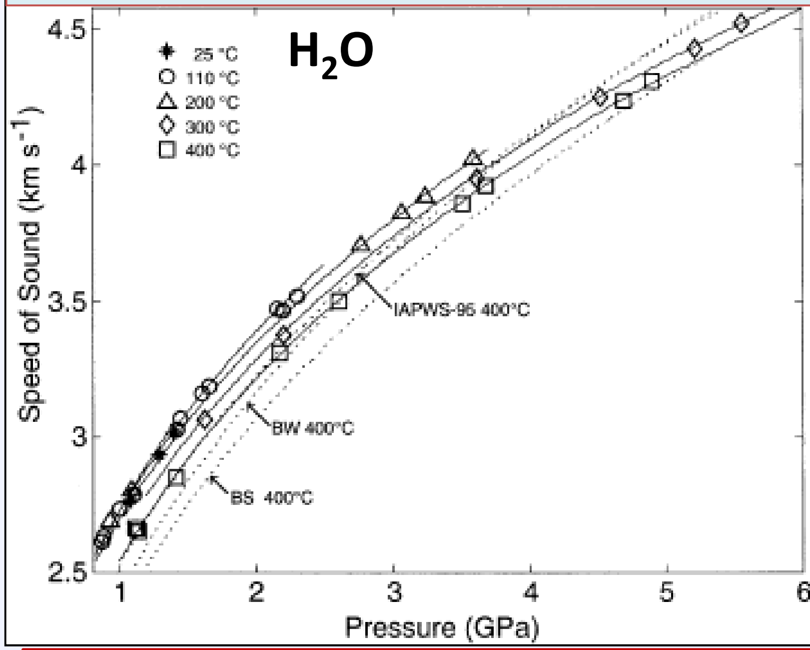
DAC

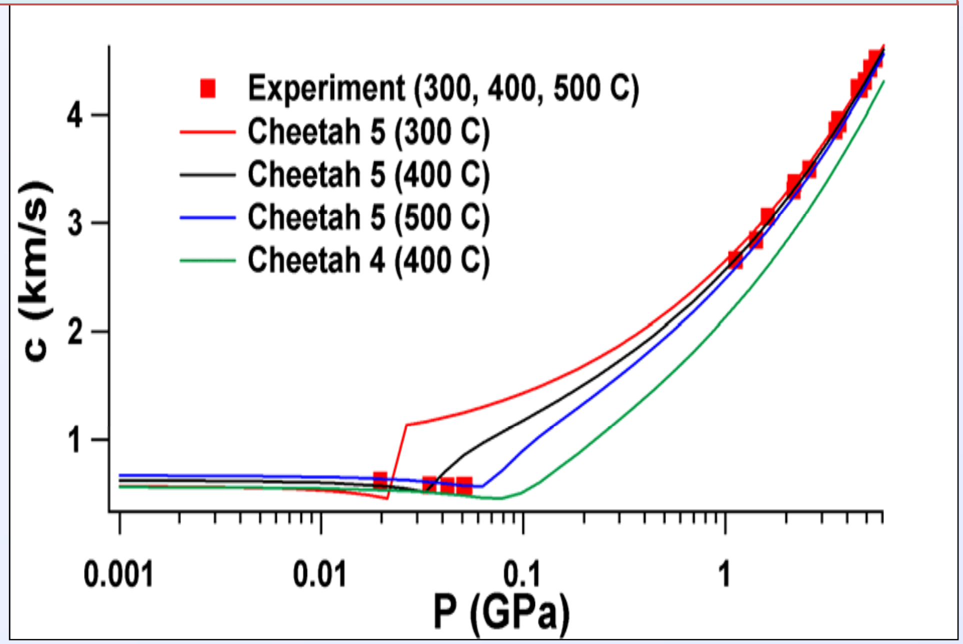


J.*M. Brown, L.J. Slutsky, K.A. Nelson, and L.T. Cheng , **Velocity of Sound and Equations of State for Methanol and Ethanol in a Diamond-Anvil Cell**, *Science* 241, 65, (1988)

Impulsive Stimulated Light Scattering, (ISLS) is Used to Directly Measure Adiabatic Sound-Speeds – Samples at GPa '88*

A significant reduction in exponential-6 misfits to water speeds of sound was achieved using ISLS data





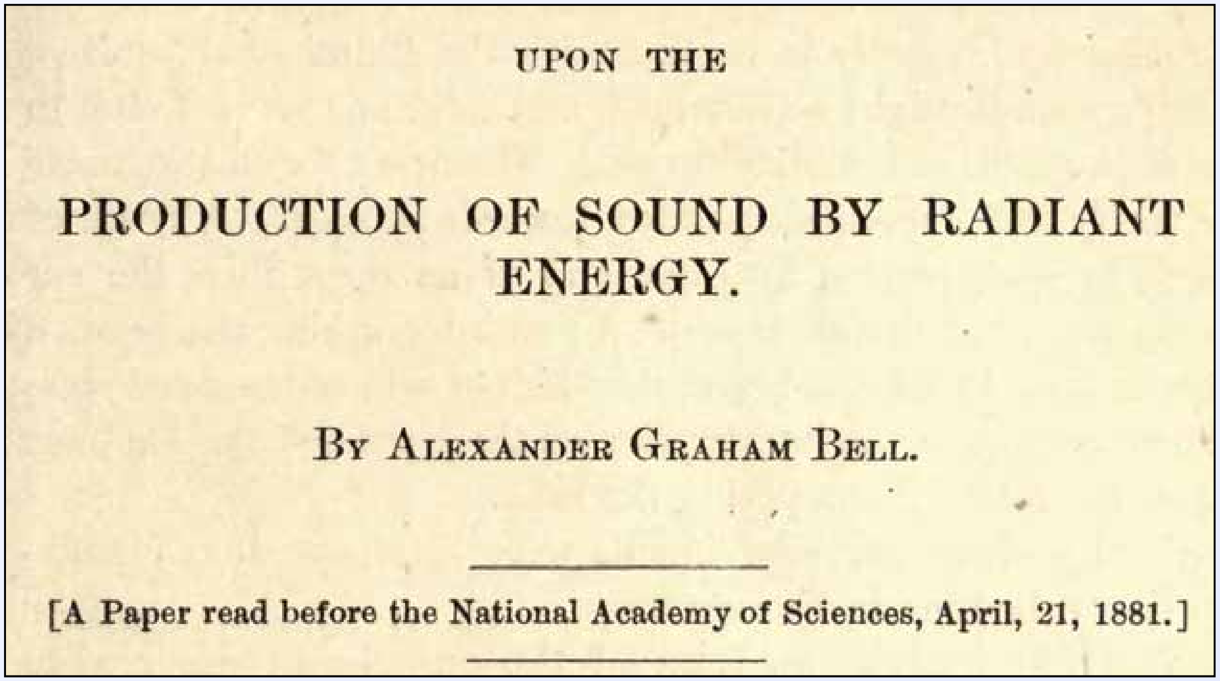
Predicted detonation velocities for HCNO explosives are improved:
Average error, D(km/s) = 1.38% (new H_2O model)
Average error, D(km/s) = 1.41% (old H_2O model)

Effort is then directed to improve or generate new intermolecular potentials

The Photoacoustic Effect was Adopted to Characterize any Detonation Product/Mixture at GPa Pressures

Photoacoustic Light Scattering (PALS) *e.g.* $\underline{HBO_2}$, CO, I_2

Sound Speeds -> Interatomic Pots. -> Det. Chem. Predictions


UPON THE

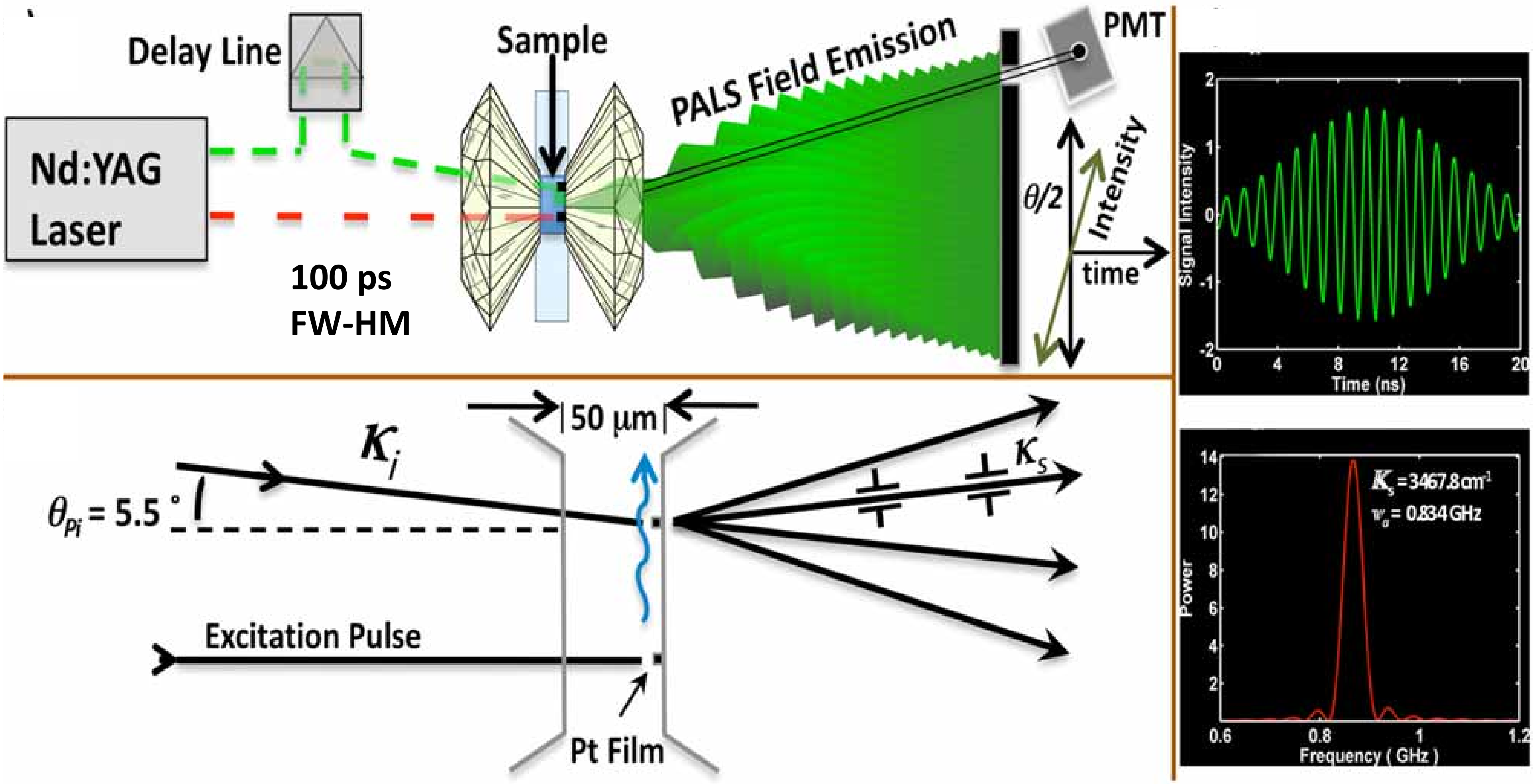
PRODUCTION OF SOUND BY RADIANT ENERGY.

BY ALEXANDER GRAHAM BELL.

[A Paper read before the National Academy of Sciences, April, 21, 1881.]

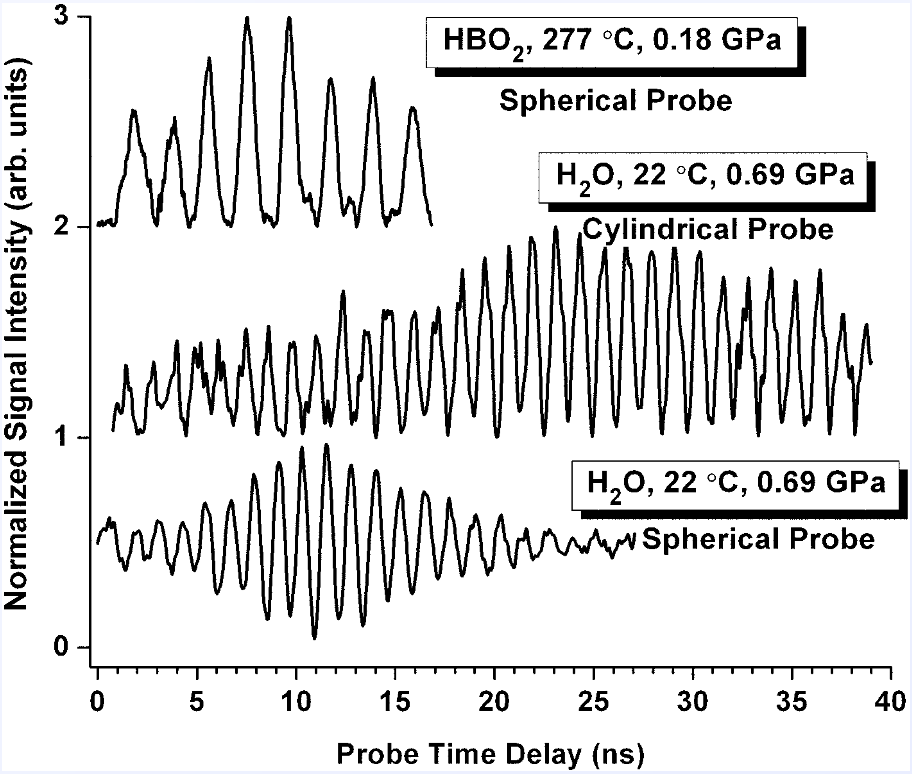
Merging old-school methods with ps pulsed laser technology and advanced signal collection/amplification techniques

PALS Signal is Heterodyne Amplified – Fixed Phase (Carrier) and Modulated Phase Signals Mix at a PMT Detector

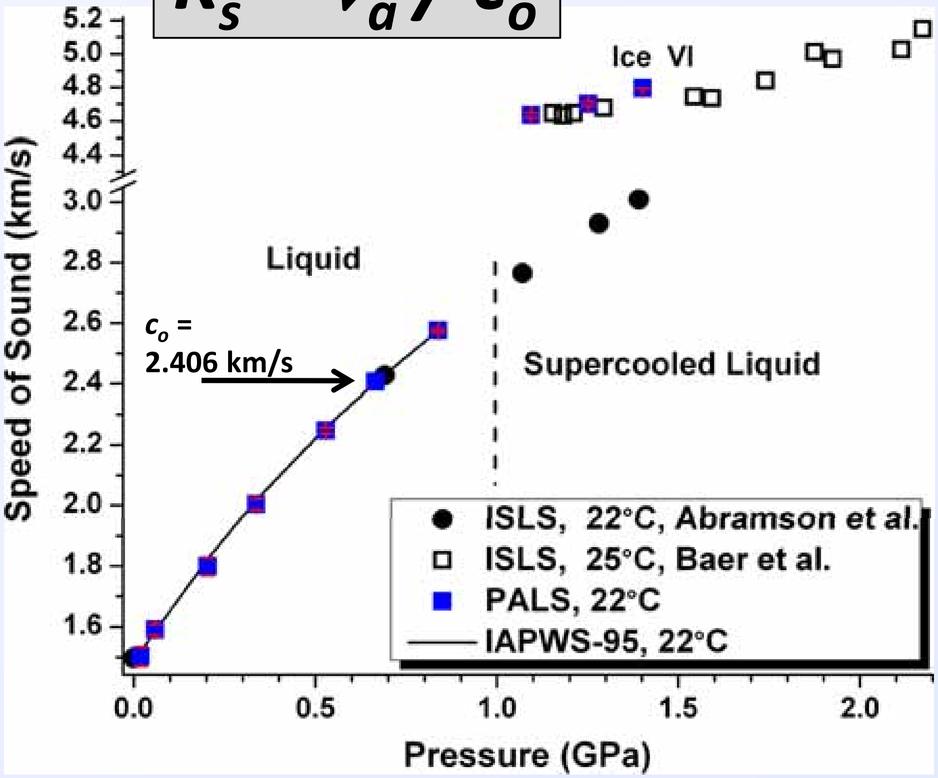


The Photoacoustic Effect was Adopted to Characterize any Detonation Product/Mixture at GPa Pressures

PALS Time Domain Traces



$$K_s = v_a / c_o$$



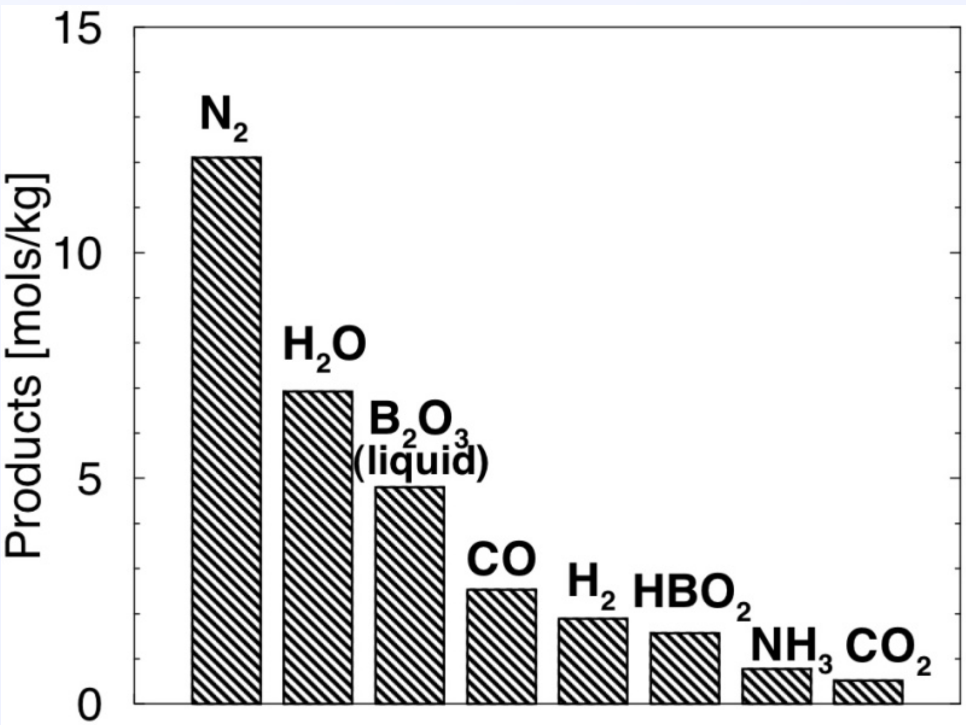
E.H. Abramson et al., **Equation of State of Water based on Speeds of Sound Measured in the Diamond-Anvil Cell**, *Geochimica Et Cosmochimica Acta* 68, 1827, (2004)

B.J. Baer et al., **Impulsive Stimulated Scattering in Ice VI and Ice VII**, *J. Chem. Phys*. 108, 4540, (1997)

“**Release of the IAPWS Formulations 1995 for the Thermodynamic Properties of Ordinary Water Substances for General and Scientific Use**” International Association for the Properties of Water and Steam, (1996)

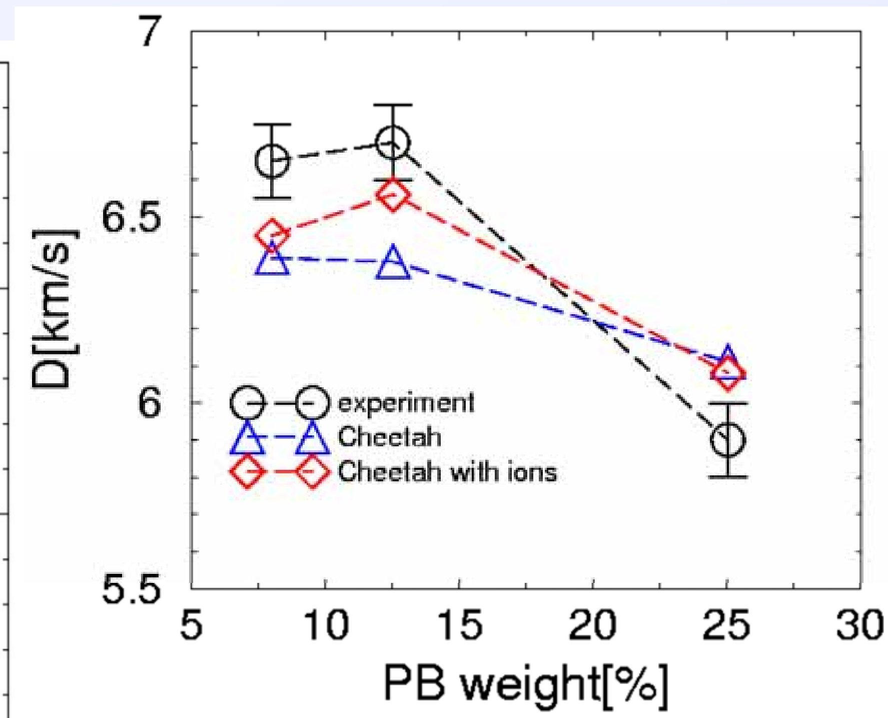
A Stringent Test of our Thermochemical Predictions is Made by Comparison to Experimental Detonation Velocities (tetranitromethane + pentaborane)

Cheetah predictions of Akimov et al.*, boron containing HEs: the applicability of our high P-T EOS model is confirmed



Chapman-Jouguet temperature ~ 6,000 K

$CN_4O_8 + B_5H_9$ Det. Vel.



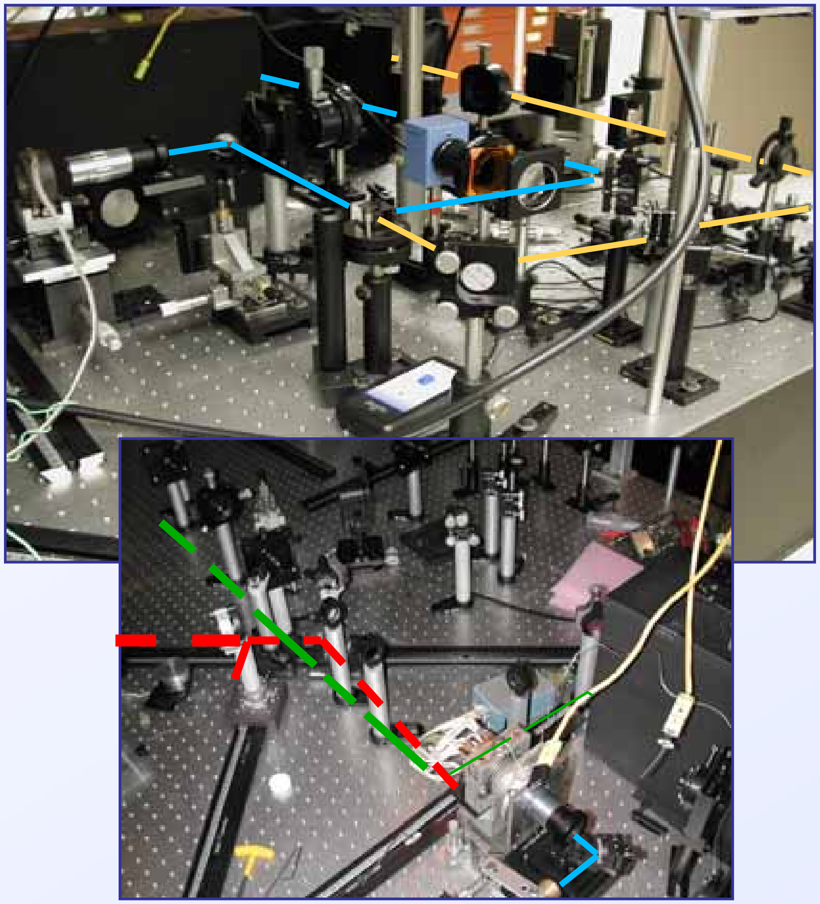
In addition we tested boron loaded PETN: results indicate boron oxidation kinetics play a key role in performance

***Akimov, L. N., et al., "Detonation of explosives containing boron and its organic derivatives" *Combustion Explosion and Shock Waves*, 8, 387 (1972)**

J.M. Zaug, S. Bastea, J.C. Crowhurst, M.R. Armstrong, and N.E. Teslich Jr., *J. Phys. Chem. Lett.* 1, 298, (2010).

PALS/BRPALS is a Versatile Tool used to Characterize any Dense Molecular Fluid including Mixtures

We embedded a confocal μ-Raman System



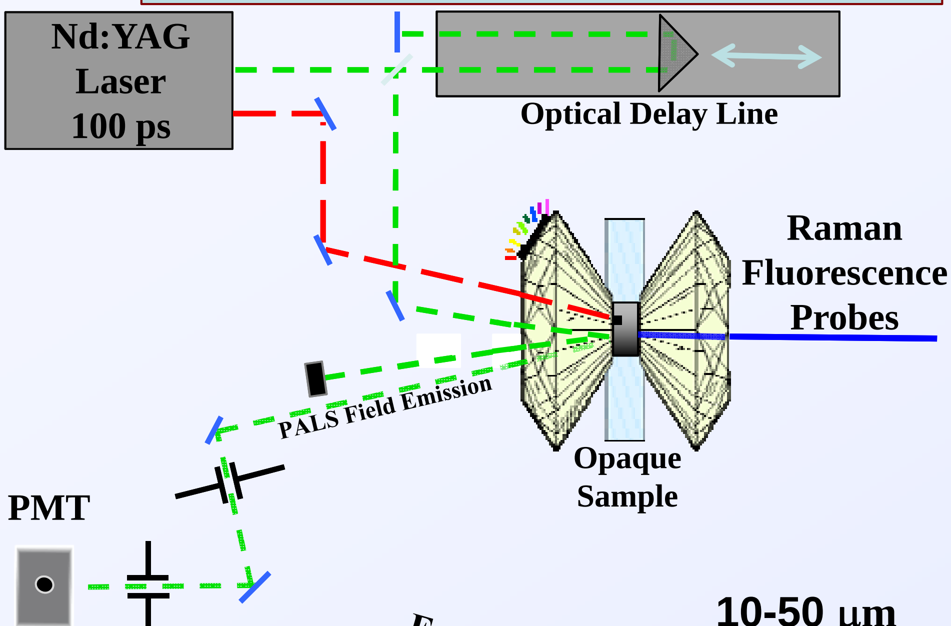
The serendipitous observation of an ISLS acoustic signal -with only one pump beam- lead to the development of DAC-PALS

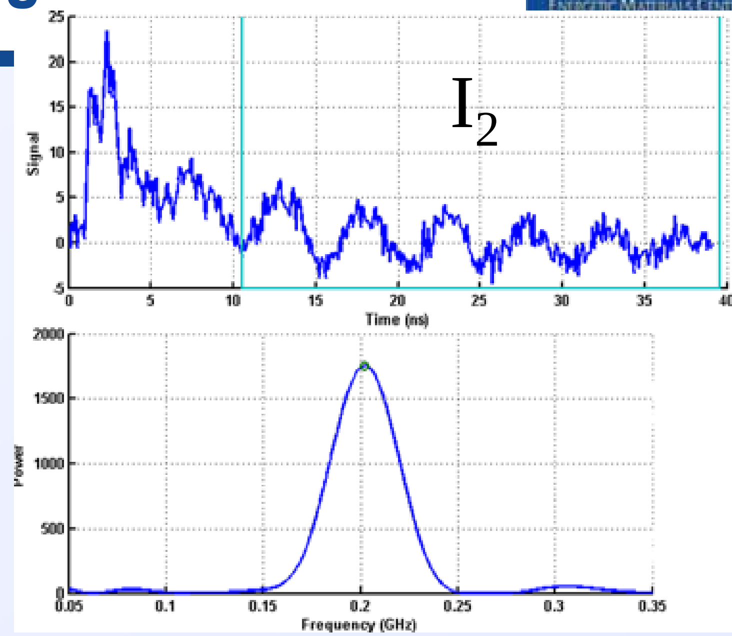
13 fluids have been characterized:

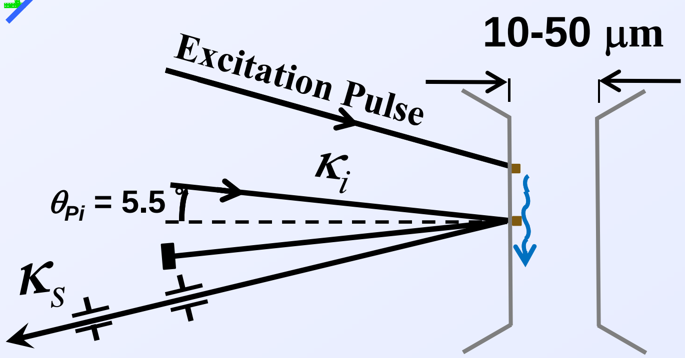
	P_{max} T_{max}	
Ar	5.0 GPa, 673K	
N_2	5.0 GPa, 473K	
C_2H_6	1.5 GPa, 295K	
C_3H_8	3.3 GPa, 473K	
C_7H1_6	4.8 GPa, 573K	
C_8H_{18}	3.4 GPa, 573K	
HCl	1.8 GPa, 473K	
CF_4	1.1 GPa, 598K	
SO_2	7.3 GPa, 873K	
HBO_2	0.5 GPa, 550K	
I_2	1.6 GPa, 700K	BRPALS
CO	3.0 GPa, 600K	
BiI_3	0.5 GPa, 790K	BRPALS

Speeds of Sound from Backreflected Photoacoustic Light Scattering - BRPALS

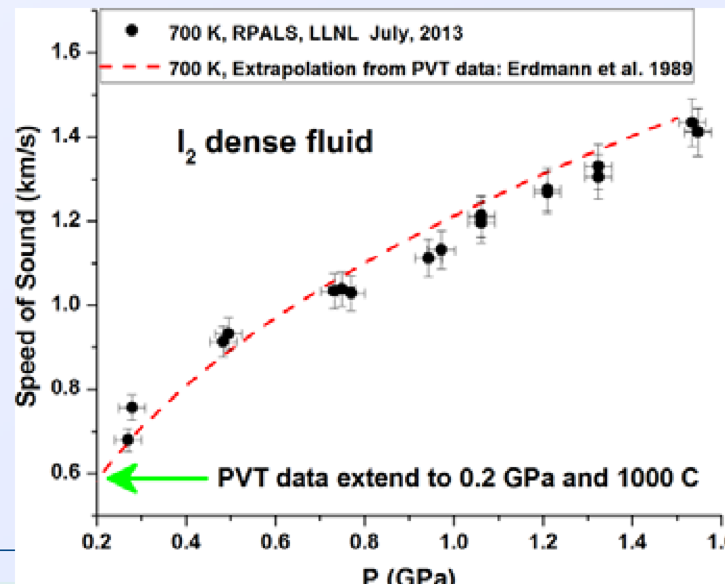
BRPALS was successfully applied to characterize optically opaque and photosensitive dense fluids







Au transducer and reflector, 4 μm wide, 86 nm thick



EOS determination through microscopy-interferometry measurements: A low symmetry energetic materials case study

Shock Compression of Condensed Matter meeting – Tampa FL

June 18, 2015

E. Stavrou*, J. M. Zaug*, S. Bastea, J. C. Crowhurst, M. R. Armstrong, and H. B. Radousky

Lawrence Livermore National Laboratory

LLNL-IM#-**789390**

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Lawrence Livermore National Security, LLC

And Now, it is Time for Something Completely Different

Quasi-static PVT EOS data provide ~ 90% of the information semi-empirical thermochemical codes need to accurately parameterize interatomic interaction potentials > detonation performance

What can be done if budgets and/or availability to specialized diagnostic platforms impede access to relevant knowledge?

…or existing diagnostics simply can't access the data?

Time to go old-school – again ?

We Need to Determine the EOS of Structurally Complex HEs or Polymers up to C-J Pressures, where Gas Gun and X-Ray Diffraction Experiments are Not Viable Options

EMC
ENERGETIC MATERIALS CENTER

XRD Issues

- **Low Z elements (C, H, N, O)**
  - **Low XRD intensity; the number of Bragg peaks diminish with pressure**
- **Low-symmetry and large molecules**
  - **Monoclinic or Triclinic Space Groups – challenging to refine at Room P-T**
  - **Crystal twining and/or cleaving *blows-up* Bragg peak refinements**
- **For most low-symmetry powdered materials, EOSs are accurately reported up to a max. pressure of ~ ½ $P_{C\text{-}J}$**
  - **TATB: L. L. Stevens *et al.* (2008) XRD V(P) EOS up to 13 GPa**
  - **PETN: B. Olinger *et al.* (1975) & J. A. Ciezak (2006) *et al.* V(P) up to 10 GPa**
  - **LLM-105: J. C. Gump *et al.* (2011) V(P) up to 6 GPa**

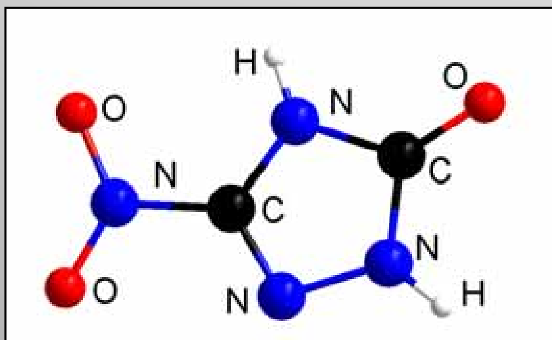
    *** E. Stavrou *et al.* V(P) up to 20 GPa – Oct. 2015 ***
  - **α - NTO: No high pressure data – impossible task using XRD; gg expensive**

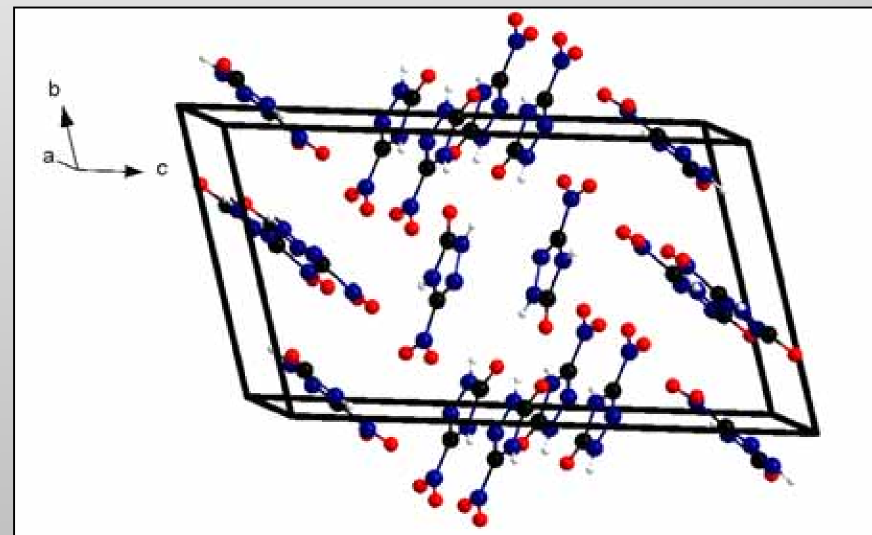
    5-nitro-2,4-dihydro-1,2,4,-triazol-3-one

One Important Case Example: α - NTO

- **Four-component twins with a triclinic symmetry (space group P-1)**
- **NTO - known energetic material in 1905; and 100 years later, the crystal structure was solved, (N. Bolotina *et al., Act. Cryst.* B61, 577, (2005)**
- **The most difficult type of molecular system (yet to be reported)**

5-nitro-2,4-dihydro-1,2,4,-triazol-3-one

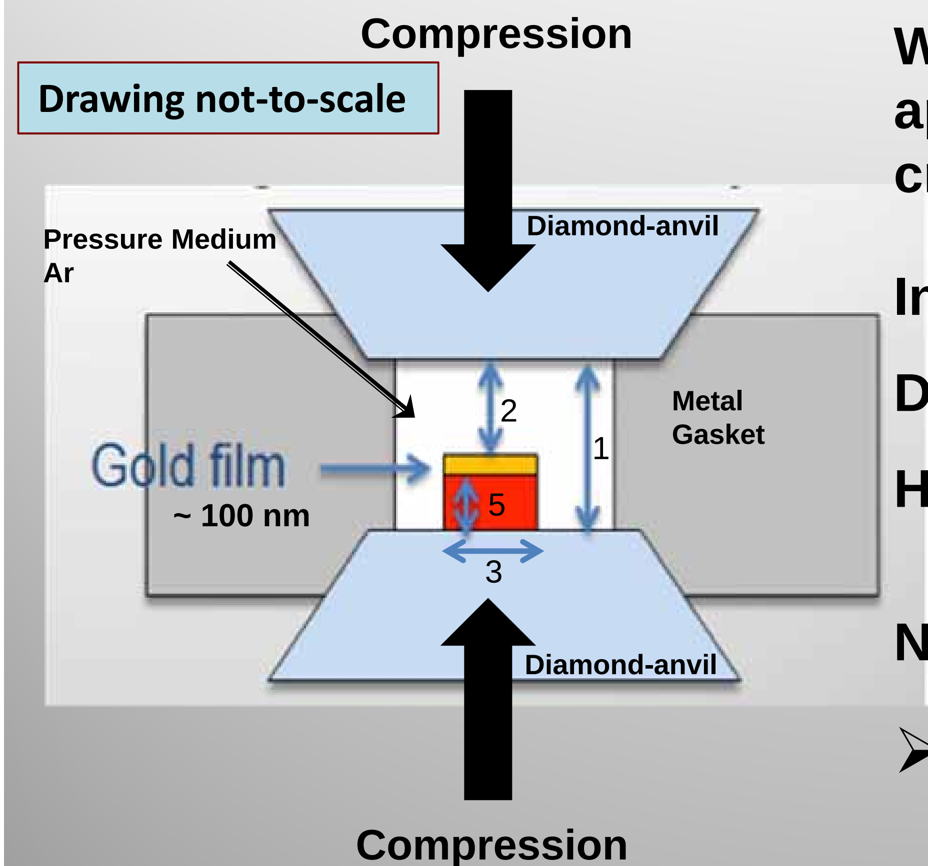




If gas-gun platforms are not available, indirect tabletop shock compression is not possible (due to sample prep.), and XRD is a non-starter – Then what?

A Versatile Technical Approach was Adopted/Developed to Measure *V*(*P*,*T*) Isothermal EOSs

Conventional Diamond-anvil Cells, (DACs) and white-light sources have been utilized to conduct *V*(*P*,*T*) measurements on isotropic materials *e.g.,* polymers



We further developed this method by applying a thin-film of Au to anisotropic crystals (HEs)

Initial DAC Chamber Dimensions:

Diameter ~ 300 μm

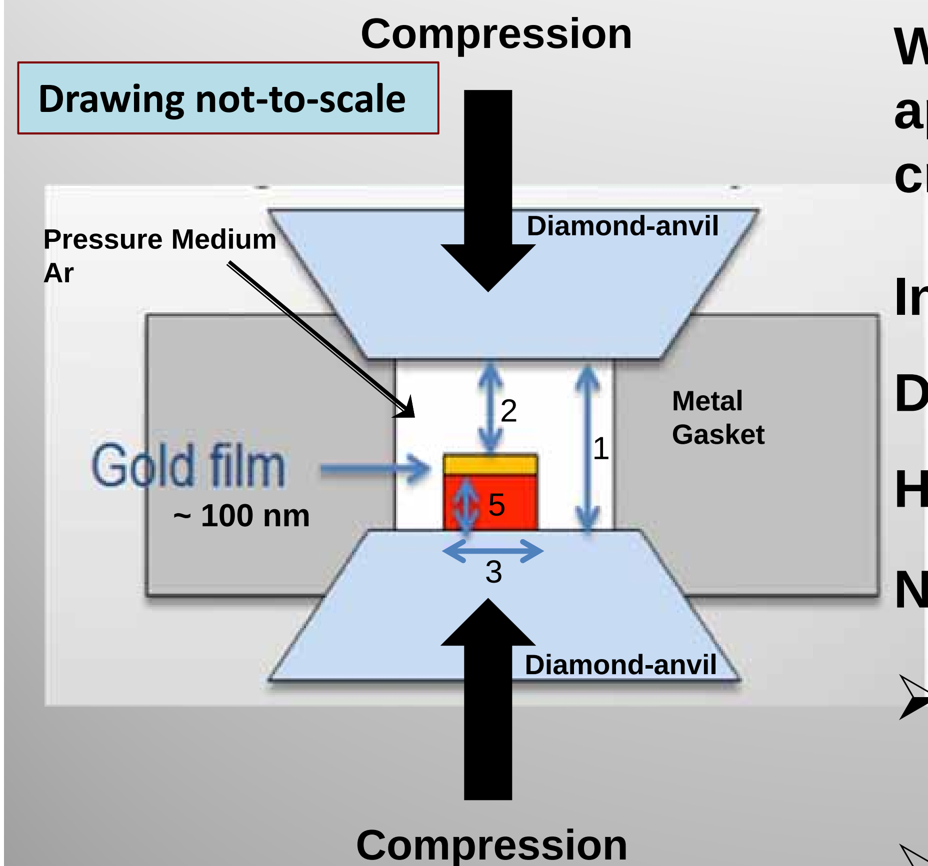
Height ~ 50 μm

Nominal specs. to achieve 40 GPa

- **Four required measurements to report *V*(*P*)**

A Versatile Technical Approach was Adopted/Developed to Measure *V*(*P*,*T*) Isothermal EOSs

Conventional Diamond-anvil Cells, (DACs) and white-light sources have been utilized to conduct *V*(*P*,*T*) measurements on isotropic materials *e.g.,* polymers



We further developed this method by applying a thin-film of Au to anisotropic crystals (HEs)

Initial DAC Chamber Dimensions:

Diameter ~ 300 μm

Height ~ 50 μm

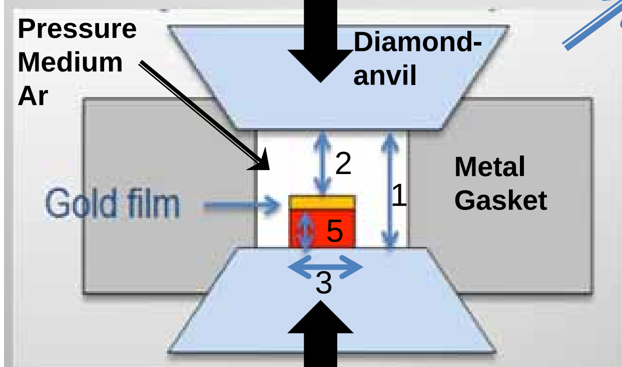
Nominal specs. to achieve 40 GPa

- **Four required measurements to report *V*(*P*); h_1, h_2, *A*, *P***
- **$n_{PTM}(P)$ is also required !**

A Versatile Technical Approach was Adopted/Developed to Measure *V*(*P*,*T*) Isothermal EOSs

Conventional Diamond-anvil Cells and white-light sources utilized to conduct *V*(*P*,*T*) measurements

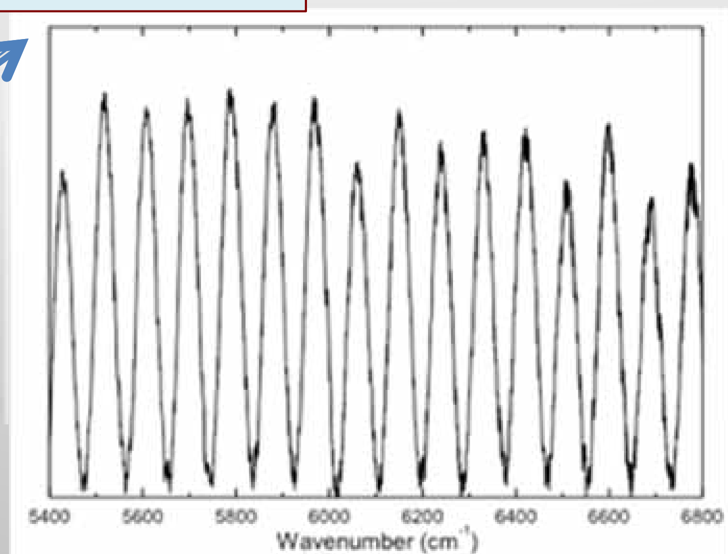
Compression



Drawing not-to-scale

Compression

***H*eight by Interferometry**



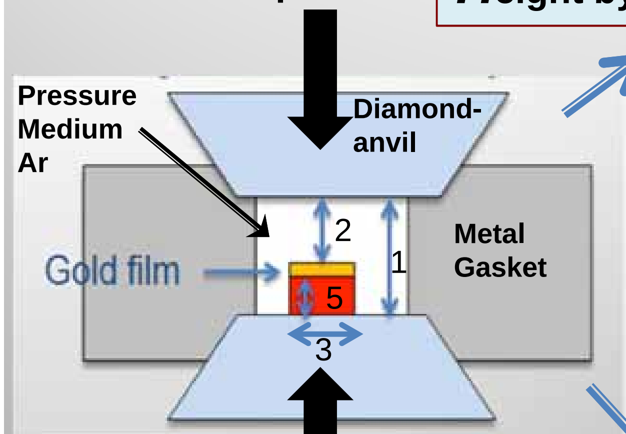
$$H(P,T) = [\ (h_1 - h_2\ )\ /\ n_{Ar}(P,T)\ ] - \text{Au height}\ (P,T)$$

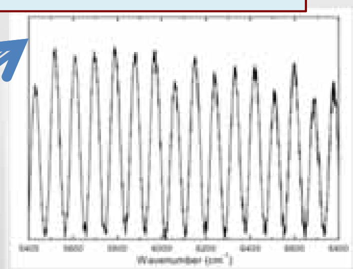
A Versatile Technical Approach was Adopted/Developed to Measure *V*(*P*,*T*) Isothermal EOSs

Conventional Diamond-anvil Cells and white-light sources utilized to conduct *V*(*P*,*T*) measurements

Compression

Height by Interferometry

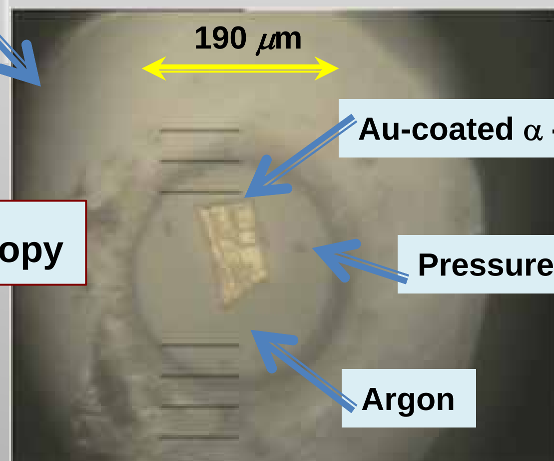




Drawing not-to-scale

Compression

Area by Microscopy



$$V = A*H, \quad \mu m^3$$

Q: How Many Ph.D.s does it take to snap a micrograph?

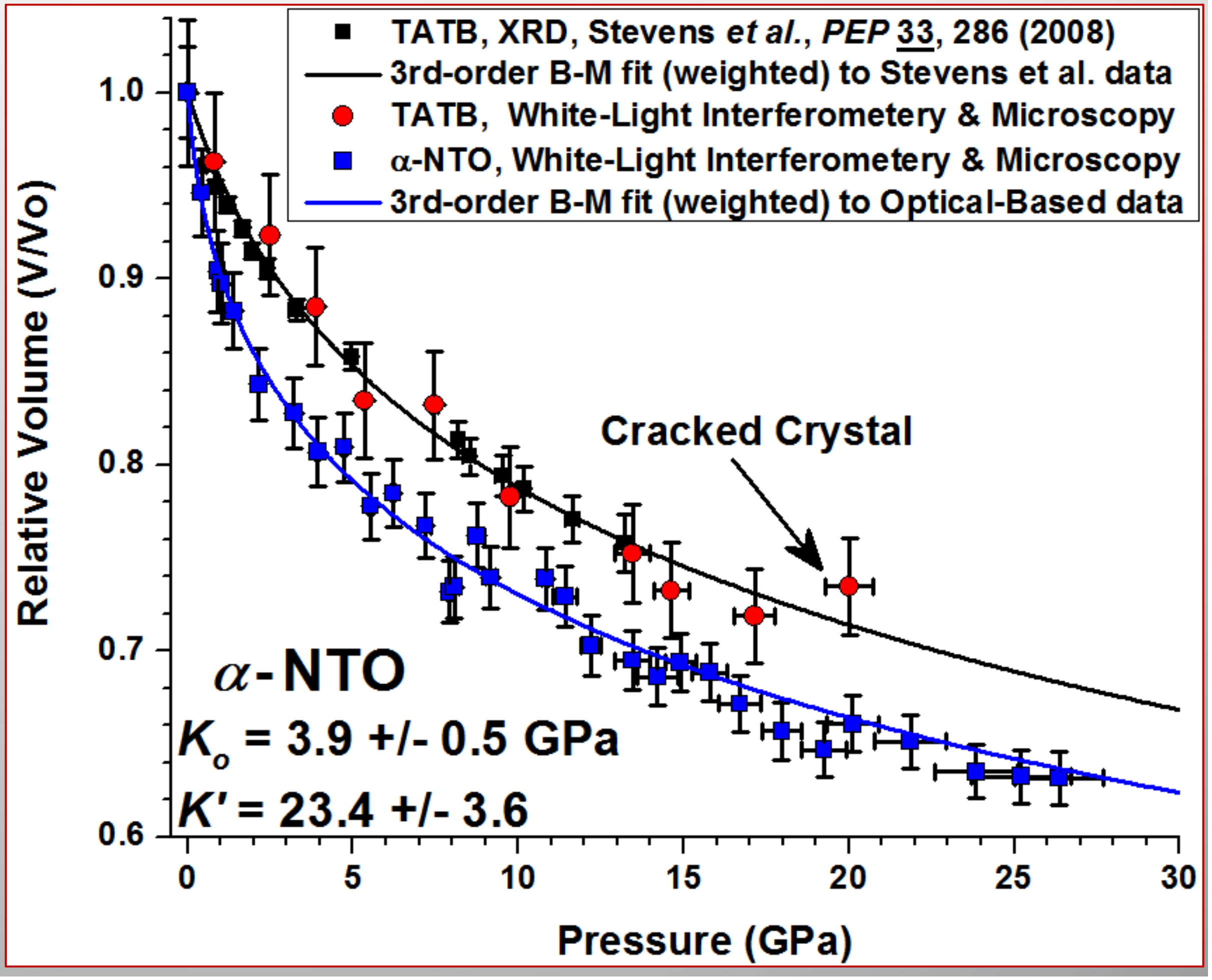
A: 3 – if it's a PR photo-opp.

A Versatile Technical Approach was Adopted/Developed to Measure *V*(*P*,*T*) Isothermal EOSs

These optical-based experiments permit high-pressure EOS measurements of low-symmetry HEs, polymers, and PBX* materials where existing methods do not work – <u>A new route to validate or improve simulations and S-E theory calculations</u>

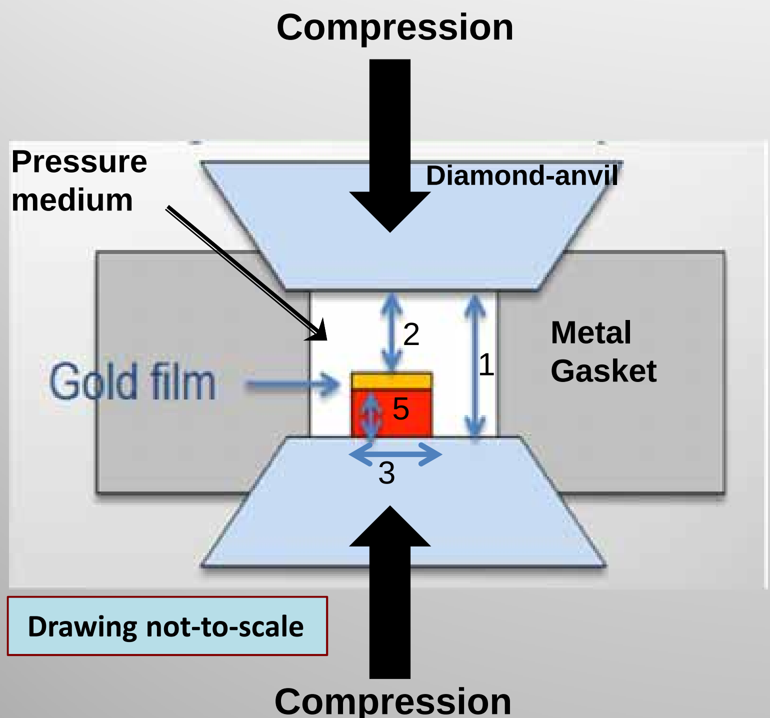
* Larger volume chamber required

Validation by a TATB Benchmark Study; α - NTO EOS to 27 GPa

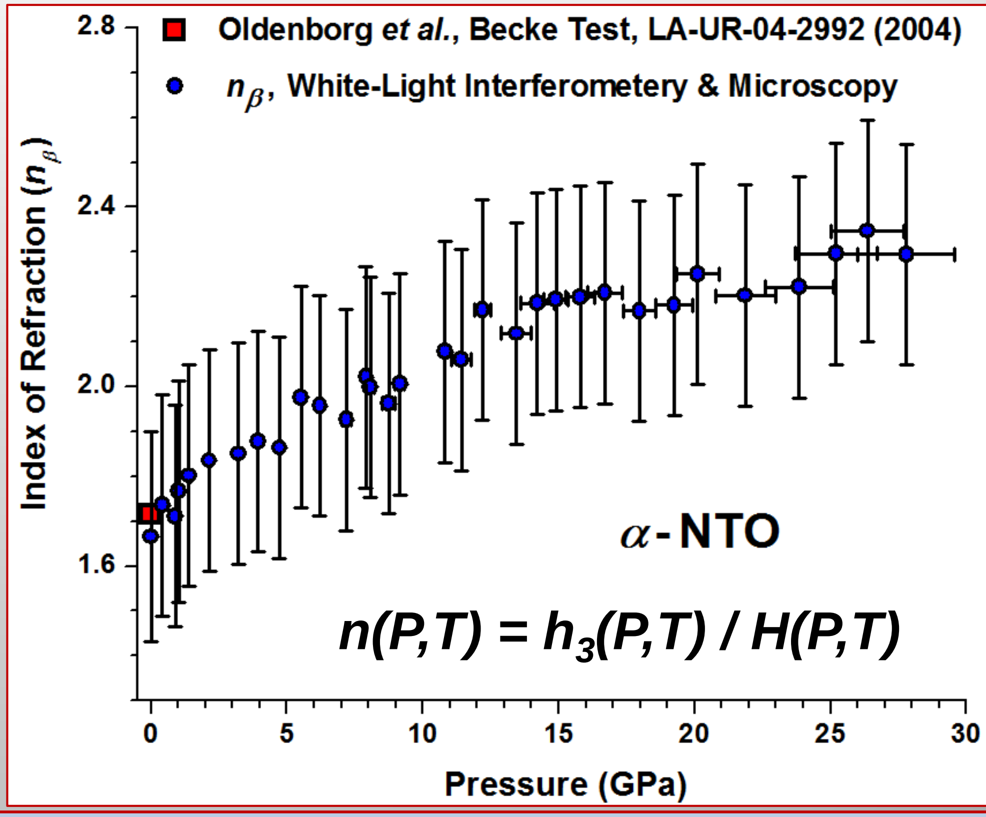


A Versatile Technical Approach was Adopted/Developed to Measure (an)isotropic *n*(*P*,*T*) – the 5th measurement

Conventional Diamond-anvil Cells and white-light sources utilized to conduct *V*(*P*,*T*) measurements



Pressure dependent indices of refraction are also determined



High-Pressure (GPa) molecular crystal and polymeric optical properties are virtually nonexistent in the Lit.

We Propose an Alternate Approach to Quasi-Static EOS Measurements

White-light interferometry plus microscopy to determine pressure dependent volumes of anisotropic crystals

***H**eight by Interferometry*

↓

V(P,T) = H*A ← ***A**rea by Microscopy*

Disadvantages:

- The approach becomes virtually unusable if a material undergoes a significant structural phase transition – may be able to reset V_o
- Each volume measurement requires hours of time (XRD requires mins.)
- Culets will cup (deform) at some threshold pressure – depending on diam.
- Require pressure transmitting pressure dependent refractive index data

Advantages:

- No pressure-induced texturing – intrinsic volume error remains constant
- Measurements conducted using inexpensive in-house diagnostics
- Enables the determination of anisotropic optical properties
- Low-power white-light will not induce decomposition; <u>a concern with XRD</u>

One Must Take Care to Minimize XRD Exposure (x,t) When Conducting EOS Studies of HE Materials

3rd Generation Synchrotron Radiation can (often) Decompose HE Materials e.g., see work by M. Pravica *et al.* UNLV and others

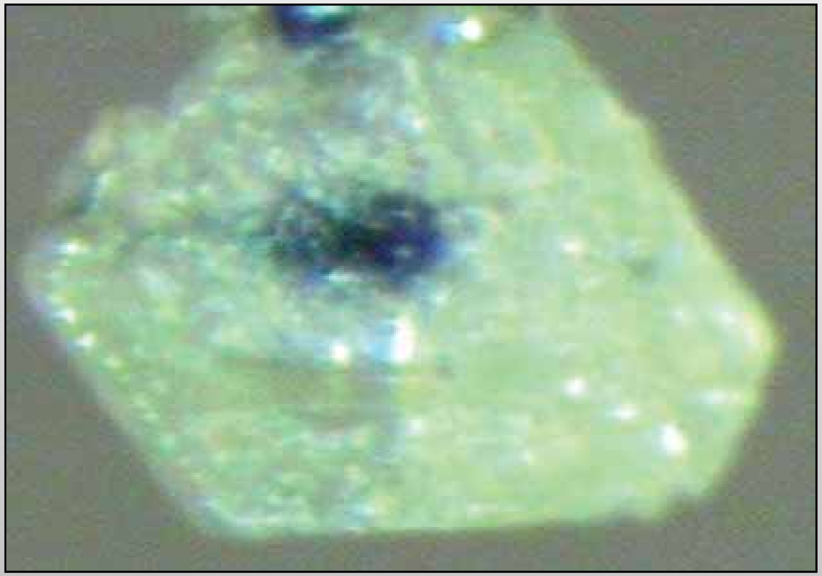


Figure 9. Typical decomposition observed in single-crystal TATB samples when exposed to ~10 keV X-ray radiation for a few minutes.

L.L. Stevens *et al. PEP* 33, 286, (2008)

Summary

- **We are enhancing our understanding of basic HE detonation physics and chemistry including ultrahigh strain rate mechanics**

- **HE detonation models based on Cheetah are improving in accuracy**

- **Focus will move from steady detonation to initiation, failure, safety, and characterization of AM materials and damaged materials**

The Future

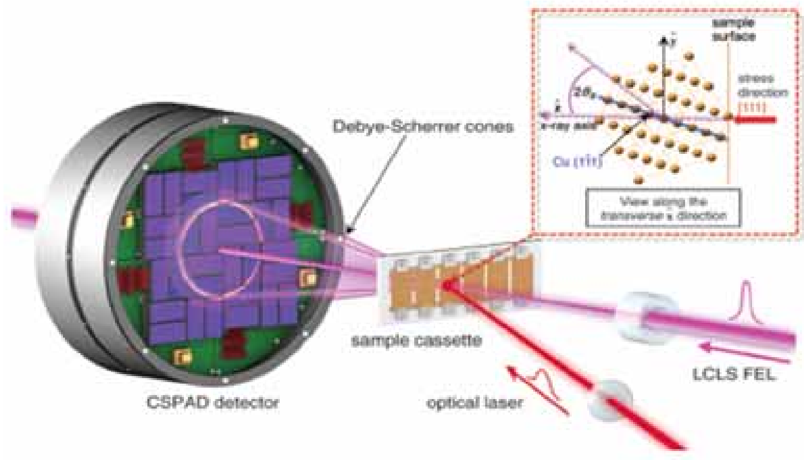
- **Development of relevant high-throughput diagnostics is a priority not just for basic R&D but also to nurture a more nimble National Defense Program**

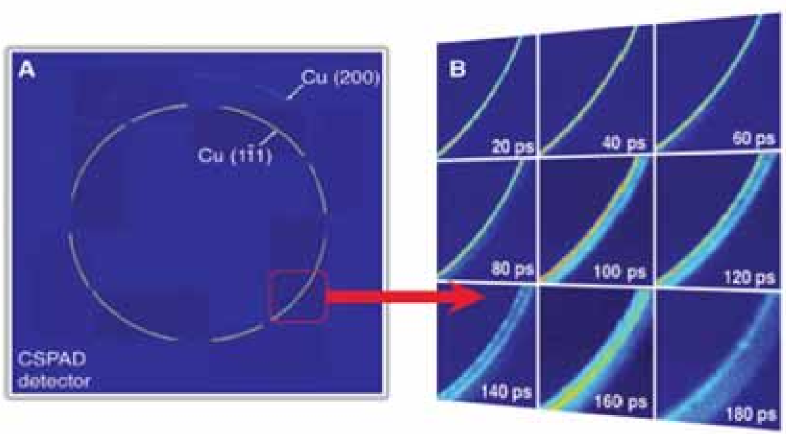
Ultrafast Tabletop X-Ray Imaging and Diffraction will Greatly Compliment TDI Hydrodynamic Measurements

X-ray Imaging

The Future

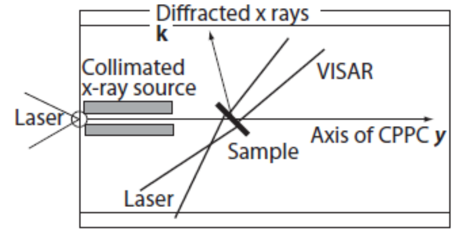
The LCLS enables direct visualization of lattice dynamics with 10 ps time resolution and 1 μm spatial resolution

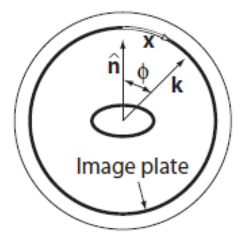




Milathianaki *et al. Science* 342, 220, (2013)

We aim to develop a tabletop laser x-ray imaging system synchronous with TDI measurements

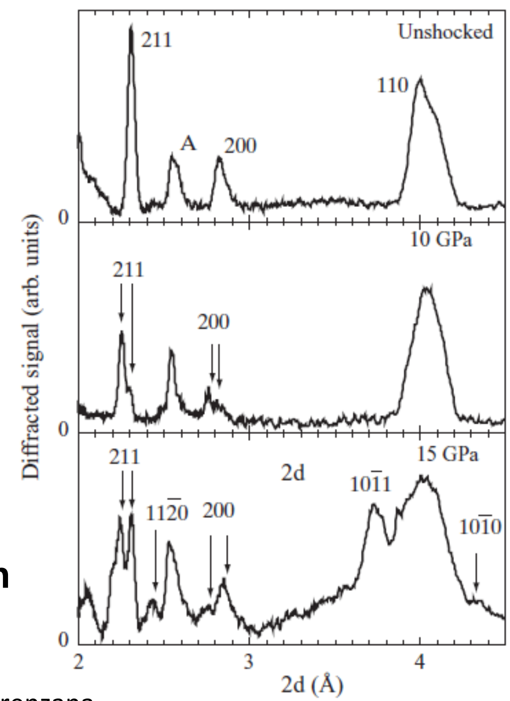




To begin, we would scale-down the approach by Hawreliak et al.*

- **2 J laser pump (1Hz)**
- **1 J X-ray backlighter**
- **~200 ps time resolution**
- **~70 shots (Z=Metals)**

1 ns X-ray patterns from shocked Fe*



* J.A. Hawlreliak, B. El-Dasher, and H. Lorenzana, *PRB* 83, 144114, (2011)

We Will Submit a Concept Paper to *APL*

X-ray Imaging

The Future

Cap. Inv. ~$2.5 M (Laser + Detector)

Small-Scale Consortium User Facility (concept)

Very High-Throughput X-Ray Data (1 Hz PRF)

Test-Bed for APS-DCS and/or LCLS Platforms

Thank You for Your Time and Interest

*** NOTE: We have a job opening - postdoctoral position**

Ultrafast Laser Systems including Spectroscopy e.g., TRIR

#100303 – go to the LLNL Careers Website

Our group collaborates with academic groups in a wide array of disciplines *e.g.* materials science, geophysics, geochemistry, synthesis of materials for industrial applications etc.

Lawrence Livermore National Laboratory

Lawrence Livermore National Laboratory