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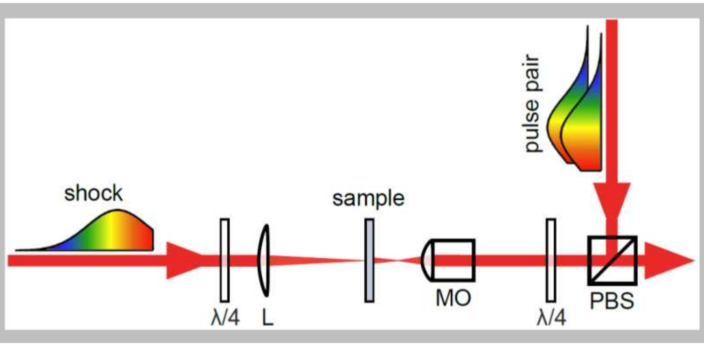

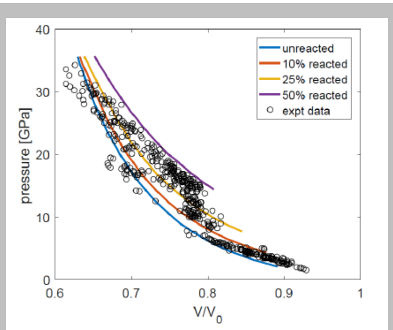

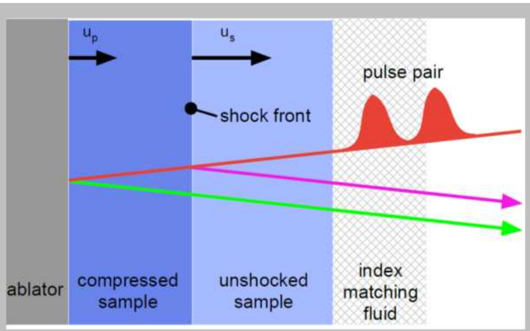

Ultrafast Shock Interrogation and Ultrafast Spectroscopy of Shocked Thin-Film Explosives

Samuel D. Park*, Michael R. Armstrong‡, Ian Kohl*, Joseph M. Zaug‡, Robert Knepper*, Alexander S. Tappan*, Sorin Bastea‡, and Jeffrey J. Kay*

*Sandia National Laboratories, ‡Lawrence Livermore National Laboratory

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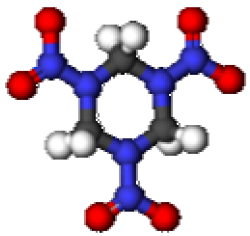
Shock Initiation of Energetic Materials

Project Purpose: Understanding Mechanisms of Shock-Induced Reactions

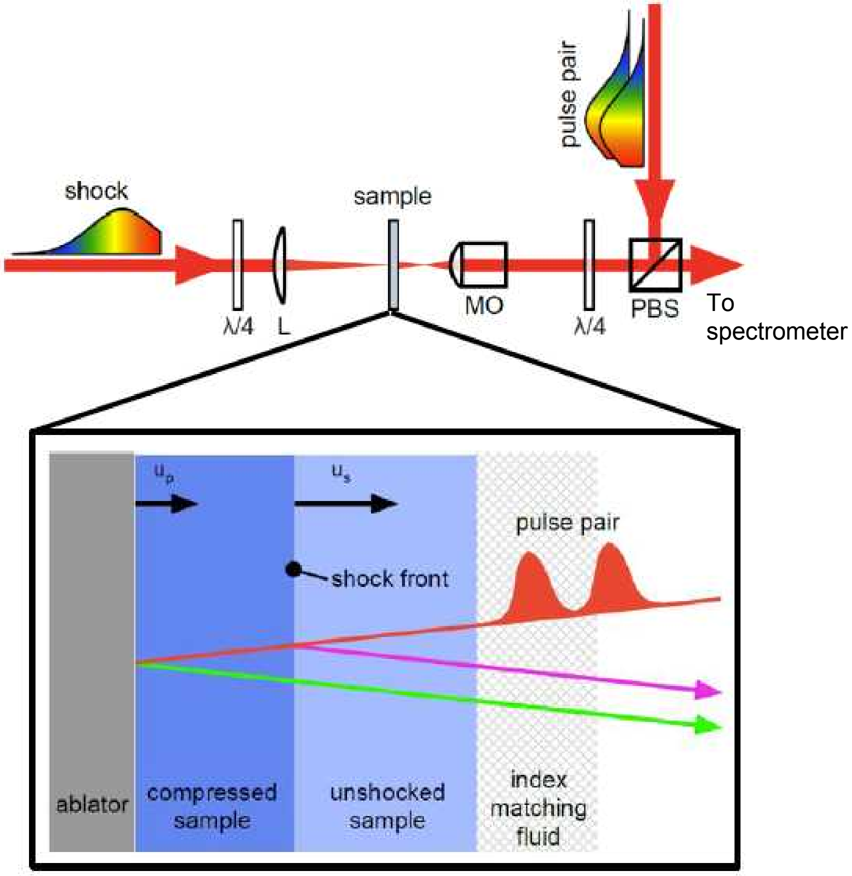


RDX

→ $H_2O + N_2 + CO + CO_2 + \ldots$

- We know "before" and "after", but not what happens in between
- The reaction mechanism is important, as it will help answer:
  - Why certain materials are more shock sensitive/less sensitive
  - How to predict margins/uncertainties for initiation
  - Dependence on material properties (particle size, microstructure, morphology)
  - Aging characteristics
  - Response in adverse environments
- We examined these mechanisms using ultrafast laser interferometry (ultrafast shock interrogation, USI) and ultrafast absorption spectroscopy.

Ultrafast Shock Interrogation



Ultrafast Shock Interrogation (USI):

Interferometric technique developed at LLNL[1]

Allows measurement of shock Hugoniot on ultrafast (ps) timescale.

Chirped pulses overlap in shocked sample; differences in index of refraction cause interference that is measured in frequency spectrum.

Features:

Ultrafast time resolution (ps)

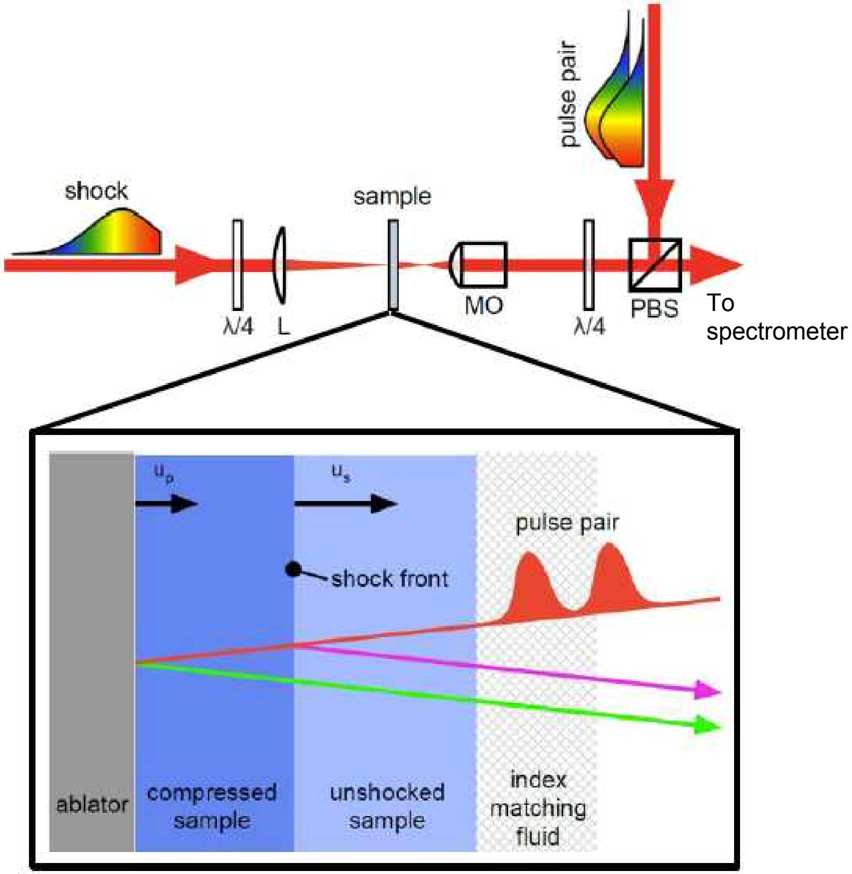
Rapid EOS acquisition (>20 shots/day)

Tabletop experiment

Works on any material transparent to probe

[1] M. R. Armstrong, J. C. Crowhurst, S. Bastea, and J. M. Zaug, *J. Appl. Phys.* **108**, 032511 (2010)

Ultrafast Shock Interrogation



Experimental Parameters

- Shock drive pulse characteristics: ~350ps, 800 nm center, 25 µJ – 1 mJ, ~10 ps risetime, ~35 µm spot size on target
- Interrogation pulse pair characteristics: ~350ps, 800 nm center, 1 µJ
- Sample characteristics: ~3-7 µm explosive on 1.5 µm aluminum on ~200 µm glass. Explosive density ~98-99% TMD

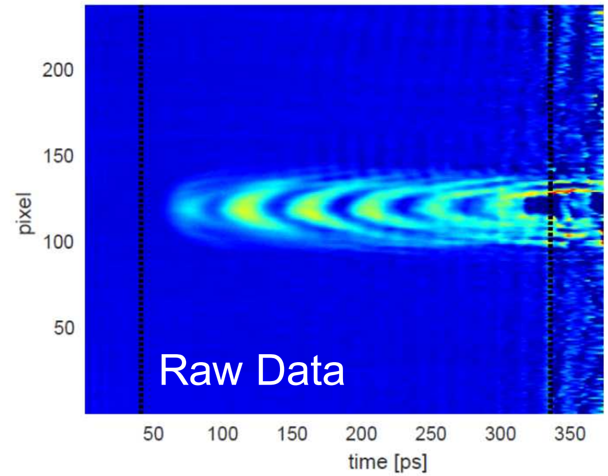
Ultrafast Shock Interrogation

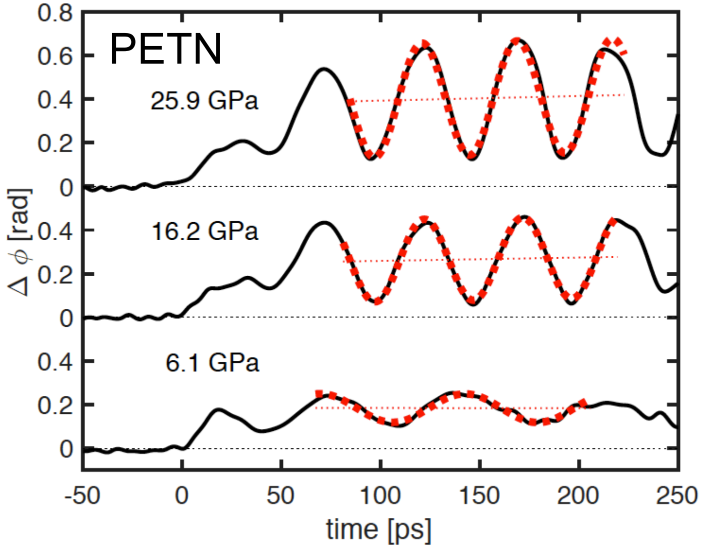
Interpreting USI Data

USI produces an interferometric signal that encodes the temporal evolution of the shocked sample

USI determines <u>shock velocity</u> and <u>particle velocity</u> from amplitude, period and offset[1] of the interferometric signal.

<u>Shock stress</u> and <u>density</u> are calculated from the shock velocity, particle velocity, and initial density[1].





[1] M. R. Armstrong, J. C. Crowhurst, S. Bastea, and J. M. Zaug, *J. Appl. Phys.* **108**, 032511 (2010)

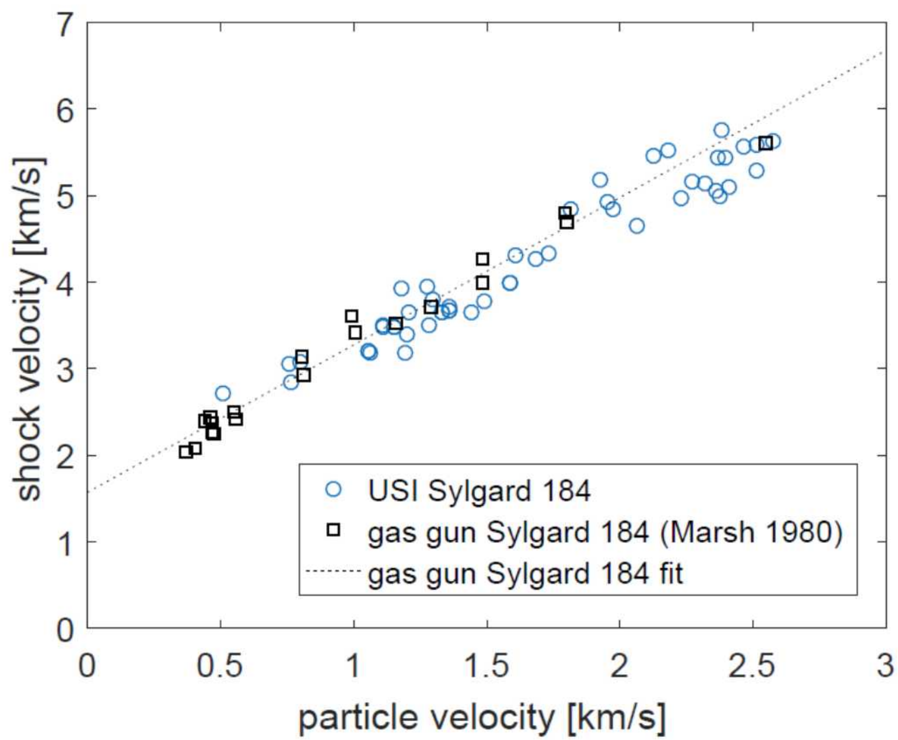
Ultrafast Shock Interrogation: Polymer

Validation Using Inert Materials

Apparatus tested using inert polymer (Sylgard 184)

Good agreement with previous gas gun data[1]

USI results are expected to coincide with gas gun data – no chemical or physical changes in material under shock compression



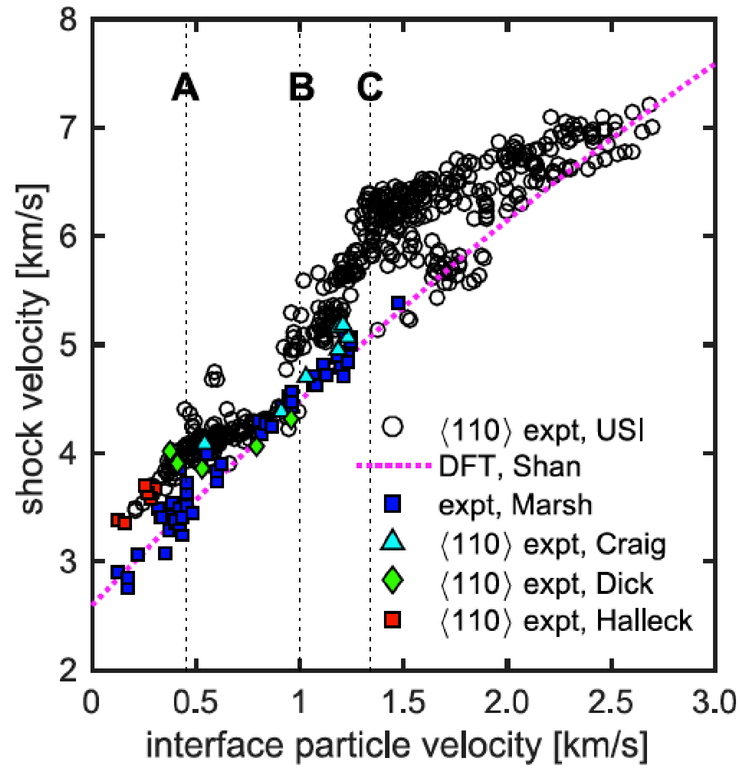
[1] S. P. Marsh, LASL SHOCK HUGONIOT DATA, University of California Press (1980)

Ultrafast Shock Interrogation: PETN

PETN

Vapor-deposited PETN samples have strongly preferred (110) orientation

USI results *should* match unreacted equation of state[1] if sample is not reacting



[1] T.-R. Shan, R. R. Wixom, A. E. Mattson, and A. P. Thompson, *J. Phys. Chem. B* **117**, 928 (2013)
[2] S. P. Marsh, *LASL Shock Hugoniot Data*, University of California Press (1980)
[3] T. R. Gibbs and A. Popolato, *LASL Explosive Property Data*, University of California Press (1980)
[4] J. J. Dick, *J. Appl. Phys.* **81**, 601 (1997)
[5] P. M. Halleck and J. Wackerle, *J. Appl. Phys.* **70**, 3572 (1991)

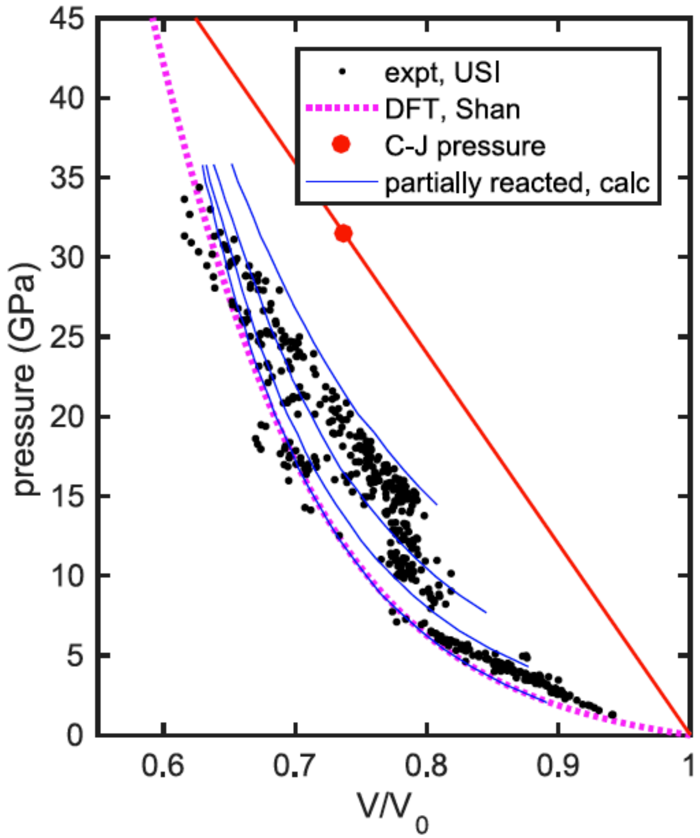
Ultrafast Shock Interrogation: PETN

PETN

Plotting P vs. V/V_0, we observe a steep rise between ~7-12 GPa

PETN <110> is sensitive and reacts from 8.5-12.5 GPa[1]

Appears to indicate exothermic chemistry



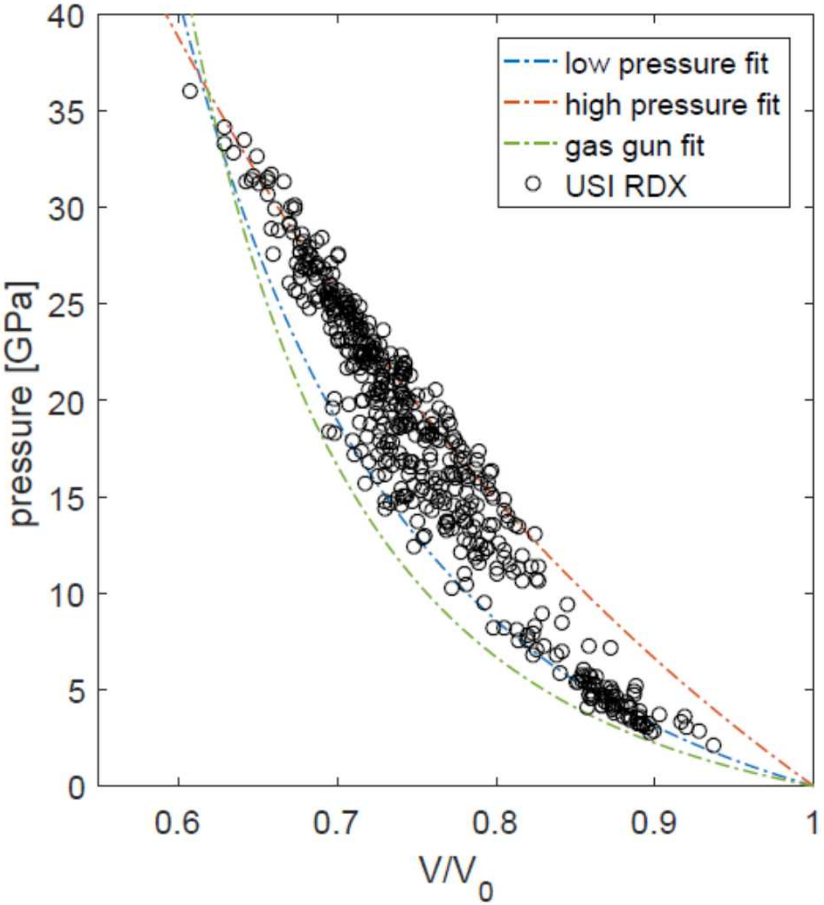
[1] J. J. Dick, R. N. Mulford, W. J. Spencer, D. R. Pettit, E. Garcia, and D. C. S. Shaw, *J. Appl. Phys.* **70**, 3572 (1991)

Ultrafast Shock Interrogation: RDX

RDX

Vapor-deposited RDX displays no strongly preferred crystallographic orientation

RDX displays similar behavior, not as pronounced (possibly due to orientation effects)



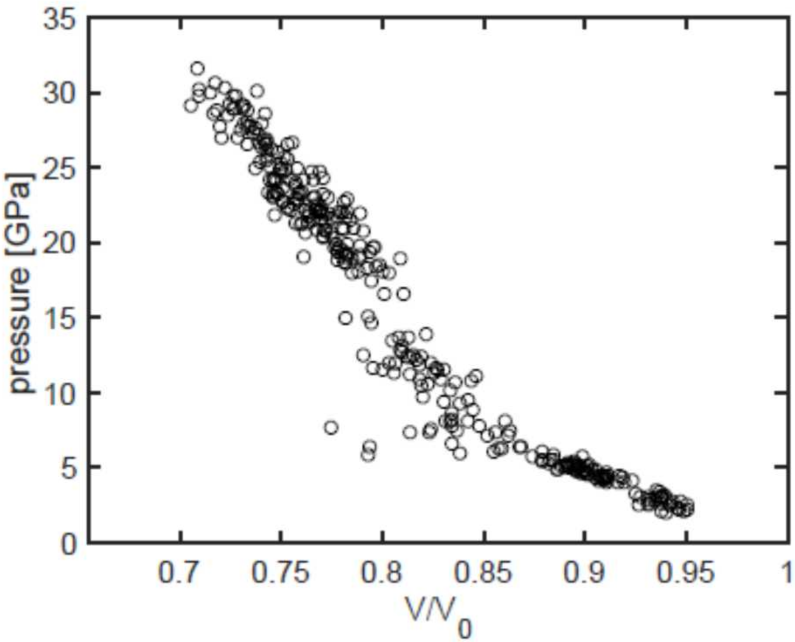
[1] J. J. Dick, R. N. Mulford, W. J. Spencer, D. R. Pettit, E. Garcia, and D. C. S. Shaw, *J. Appl. Phys.* **70**, 3572 (1991)

Ultrafast Shock Interrogation: CL-20

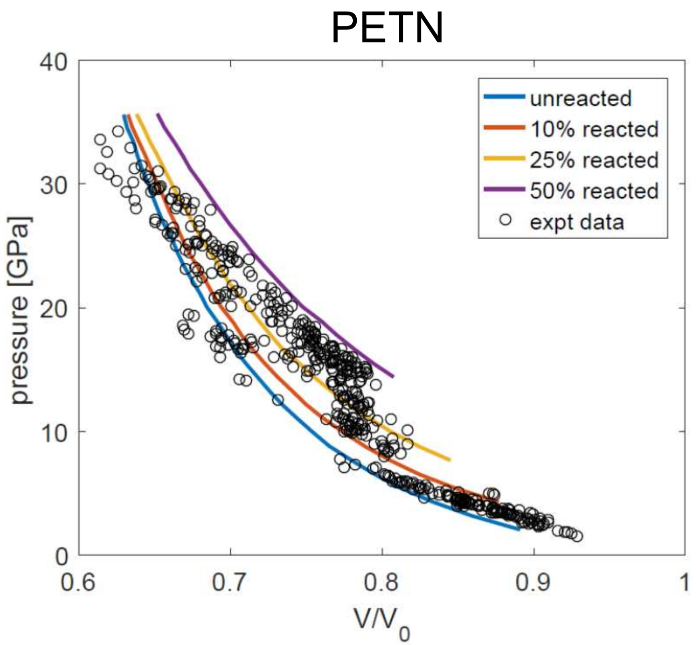
CL-20

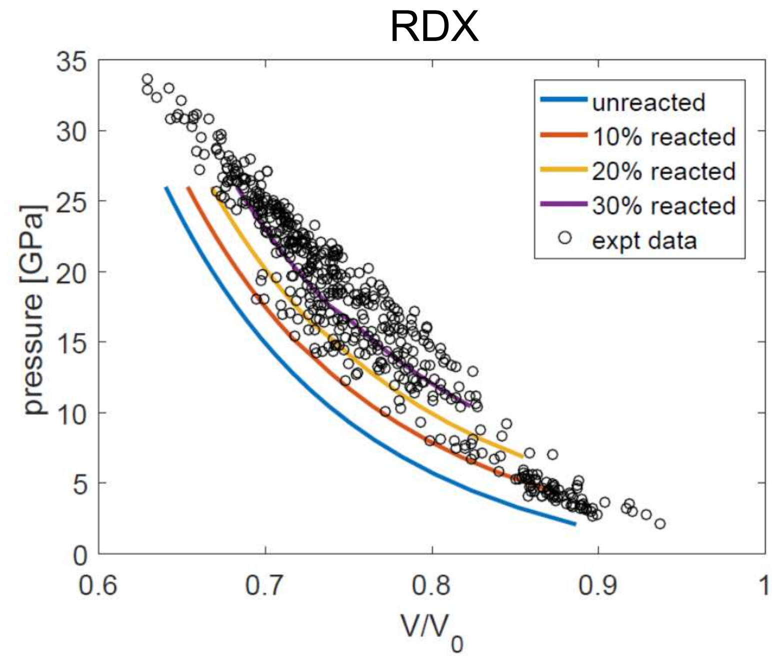
Vapor-deposited is highly oriented (111)

Visible discontinuity at ~12-17 GPa; qualitatively similar to RDX and PETN.



Ultrafast Shock Interrogation: Support for Exothermic Chemistry



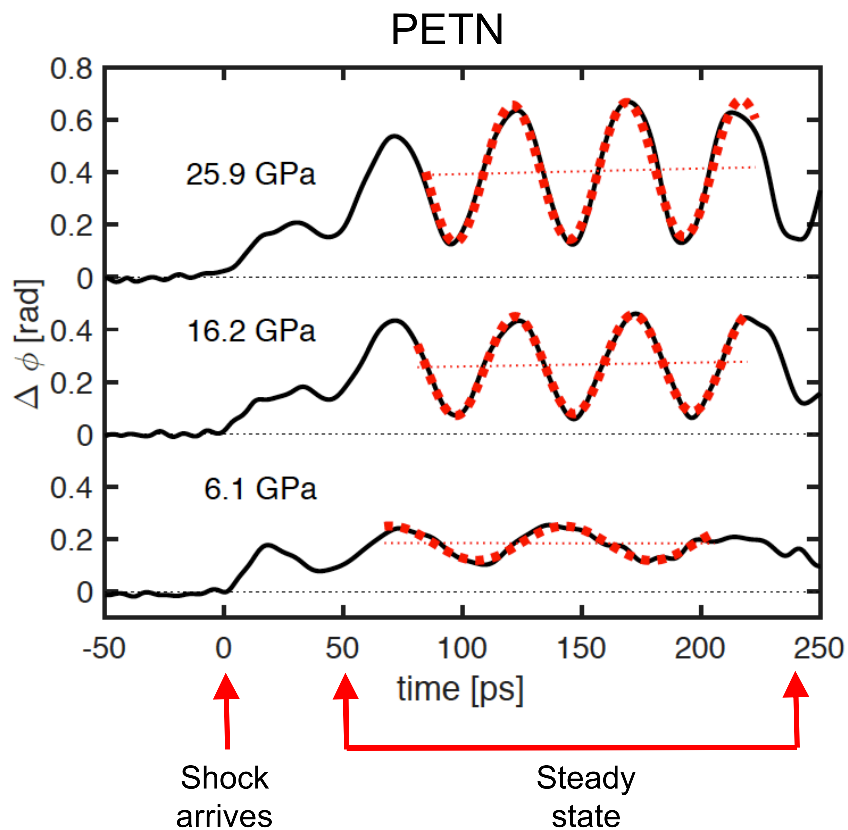


Calculated partially-reacted Hugoniots appear to support exothermic chemistry; calculated using CHEETAH and assume complete reaction of 10-50% of material

Recent MD simulations have also predicted reaction at a threshold shock velocity of 5 km/s for <110> PETN[1]

[1] T.-R. Shan, R. R. Wixom, A. E. Mattson, and A. P. Thompson, *J. Phys. Chem. B* **117**, 928 (2012)

Ultrafast Shock Interrogation: Timescales

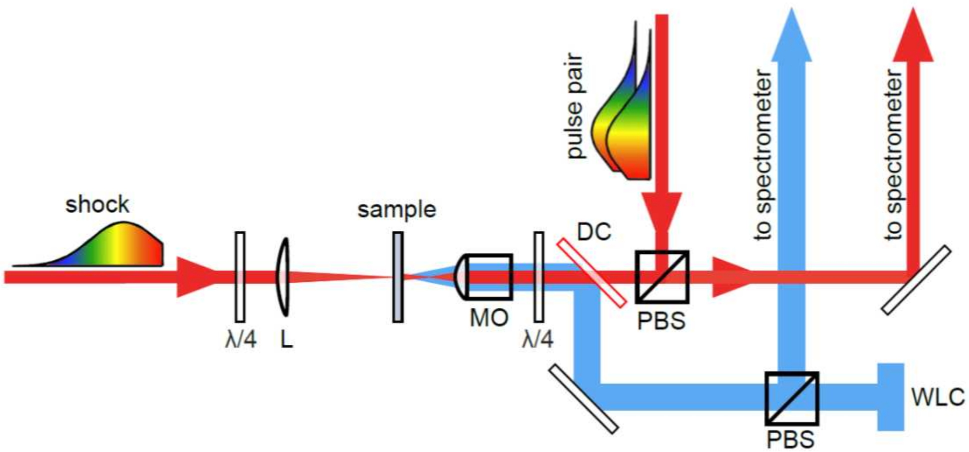


Measurement sets timescale for partial reaction – shocked material reaches steady state at ~50 ps, persists until end of measurement at ~300 ps

<u>This sets a window of ~50 ps during which reaction appears to occur.</u>

This is congruent with measurements of PETN reaction zones by R. Knepper: <50 ps.

Ultrafast Absorption Spectroscopy



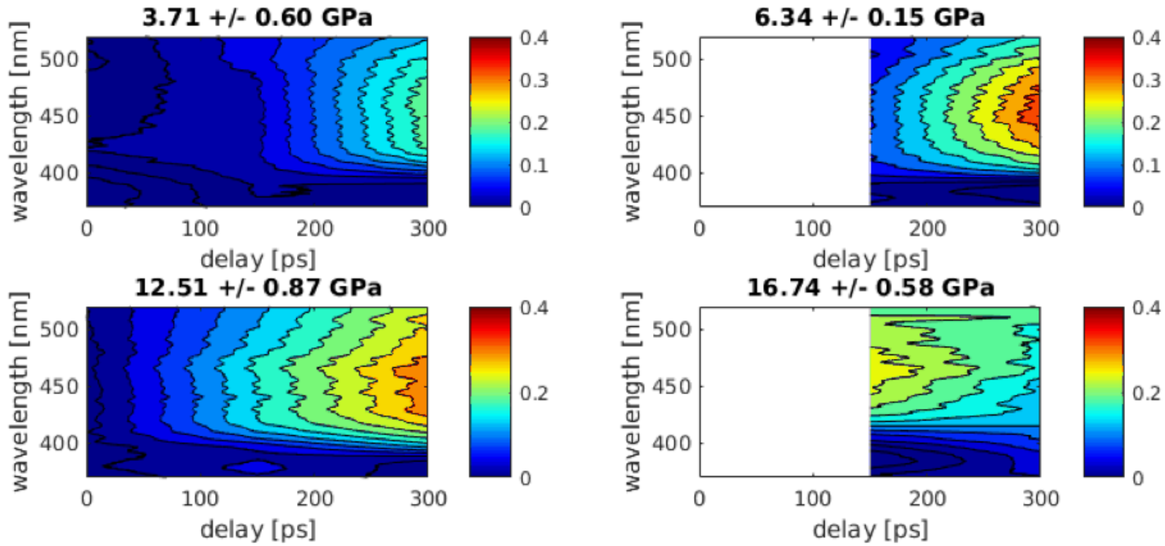
Ultrafast Absorption Spectroscopy

Uses broad-band supercontinuum (“white light”) to probe visible spectrum of material for evidence of reaction

White light generated by focusing 800 nm pulse into CaF_2

WL characteristics: 400-550 nm bandwidth, ~50 nJ, <1 ps duration

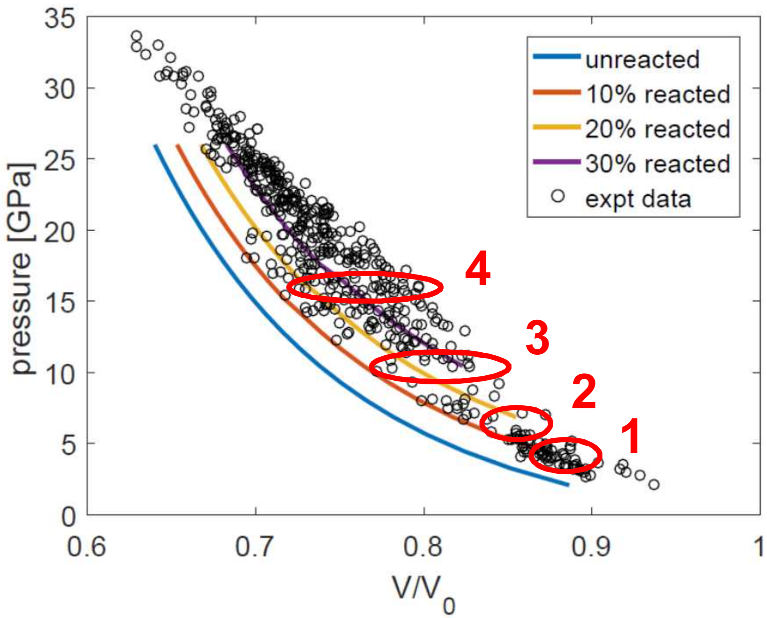
Ultrafast Absorption Spectroscopy: Shocked RDX

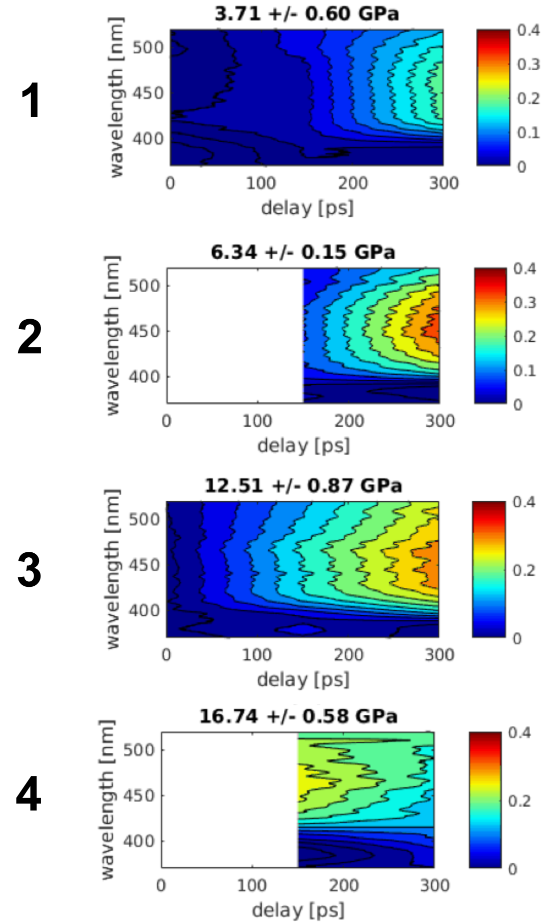


We observe broadband absorption that increases with time and shifts to earlier time at higher shock stress

Uncompressed RDX is transparent in this wavelength range; minimal absorption under severe compression

Ultrafast Absorption Spectroscopy: Shocked RDX





Absorption appears to be due to reaction products; free radicals (such as NO_2) will absorb visible light.

Conclusions

We have used USI to characterize shocked PETN, RDX, and CL-20 on ultrafast timescale

USI experiments indicate sharp pressure rises off of unreacted Hugoniot for all three materials

Observed pressure jumps are consistent with exothermic chemistry on <50 ps timescale

Ultrafast absorption experiments on RDX show broadband absorption in visible region (400-550 nm), consistent with reaction products

Acknowledgments

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Sandia National Laboratories

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Lawrence Livermore National Laboratory

LDRD
Laboratory Directed Research and Development

Sandia Laboratory-Directed Research and Development Program