SAND2018-6964C

Ultrafast Shock-induced Reactions in Pentaerythritol Tetranitrate Thin Films

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

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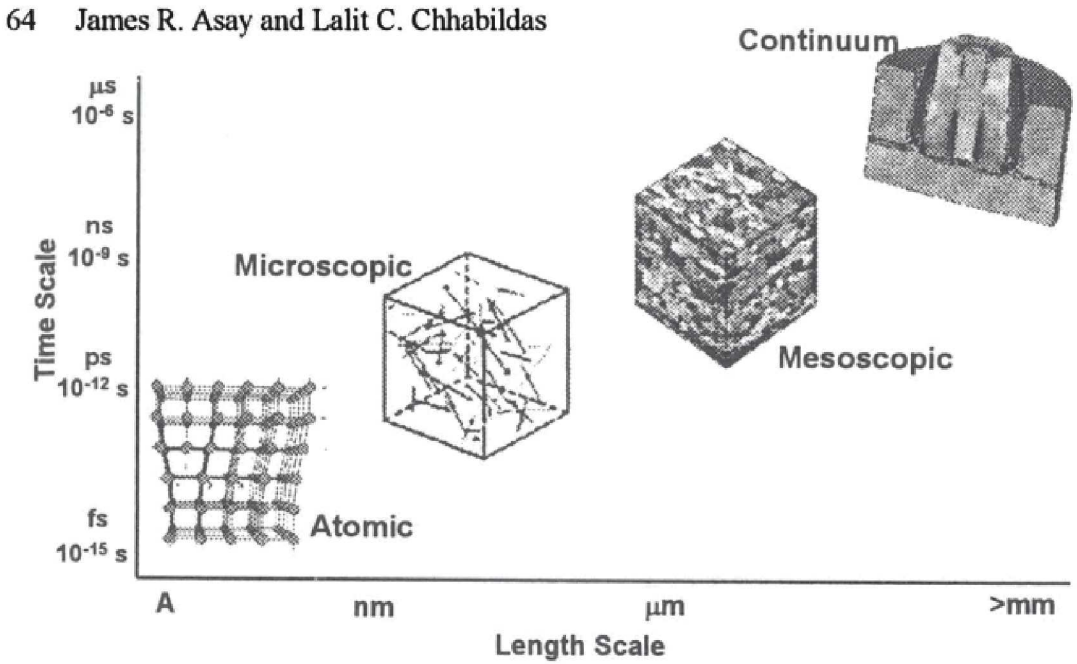
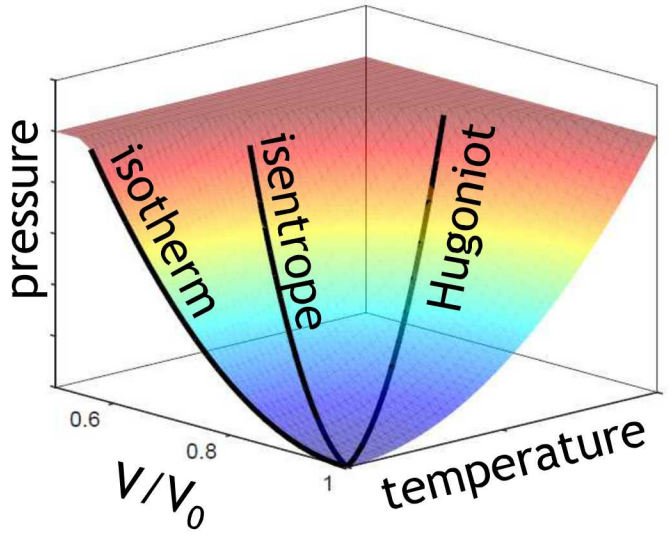
Shock Physics: A Multi-scale Problem





- Shock waves can **uniquely** generate conditions of extreme pressure-temperature in materials
- Deformation in a shock wave needs to be understood at all levels for a fundamental understanding

Microsopic Length Scales

Shock-induced shear bands in an energetic molecular crystal: Application of shock-front absorbing boundary conditions to molecular dynamics simulations

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Physicochemical Parameters of Nitramines Influencing Shock Sensitivity

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TNO Prins Maurits Laboratory, Research Group Pyrotechnics and Energetic Materials, P.O. Box 45, 2280 AA Rijswijk (The Netherlands)

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TNO Prins Maurits Laboratory, Division Munition Technology and Explosion Safety, P.O. Box 45, 2280 AA Rijswijk (The Netherlands)

- Microscopic events occurring behind a shock is nontrivial especially for solids
- Transition from mechanical to chemical energy remains an active area of study
- Understanding the timescale can elucidate possible initiation mechanisms in energetic solids

Ultrafast Shock Compression Study

Shock Induced Reaction Observed via Ultrafast Infrared Absorption in Poly(vinyl nitrate) Films

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Dynamic Experimentation Division, DX-2, Los Alamos National Laboratory, MS P952, Los Alamos, New Mexico 87545

Ultrafast Shock Initiation of Exothermic Chemistry in Hydrogen Peroxide

Michael R. Armstrong,* Joseph M. Zaug,† Nir Goldman, I-Feng W. Kuo, Jonathan C. Crowhurst, W. Michael Howard, Jeffrey A. Carter, Michaele Kashgarian, John M. Chesser, Troy W. Barbee, and Sorin Bastea

Ultrafast Chemical Reactions in Shocked Nitromethane Probed with Dynamic Ellipsometry and Transient Absorption Spectroscopy

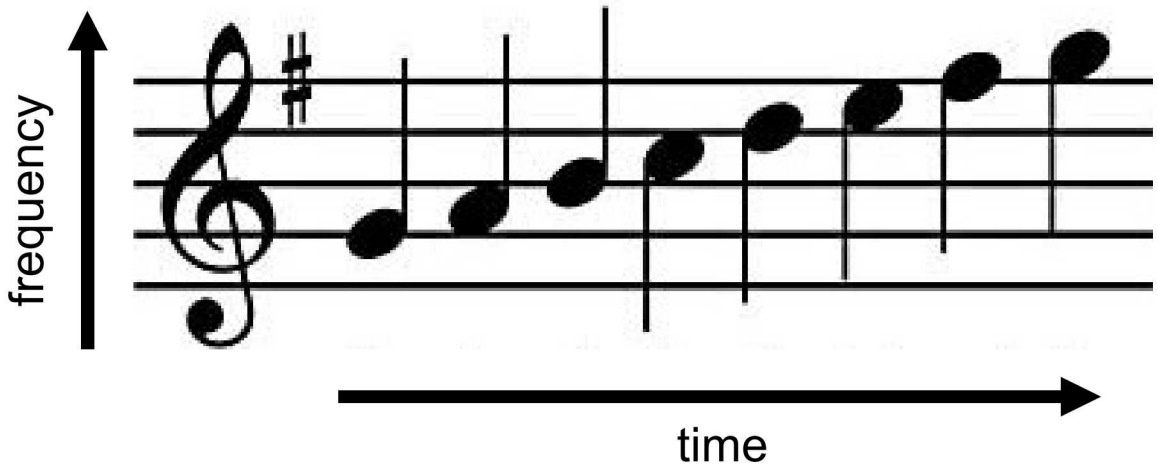
Kathryn E. Brown,* Shawn D. McGrane, Cynthia A. Bolme, and David S. Moore

The $\alpha \rightarrow \epsilon$ phase transition in iron at strain rates up to $\sim 10^9\ s^{-1}$

Jonathan C. Crowhurst,[a)] Bryan W. Reed,[b)] Michael R. Armstrong,[c)] Harry B. Radousky, Jeffrey A. Carter, Damian C. Swift, Joseph M. Zaug, Roger W. Minich, Nick E. Teslich, and Mukul Kumar

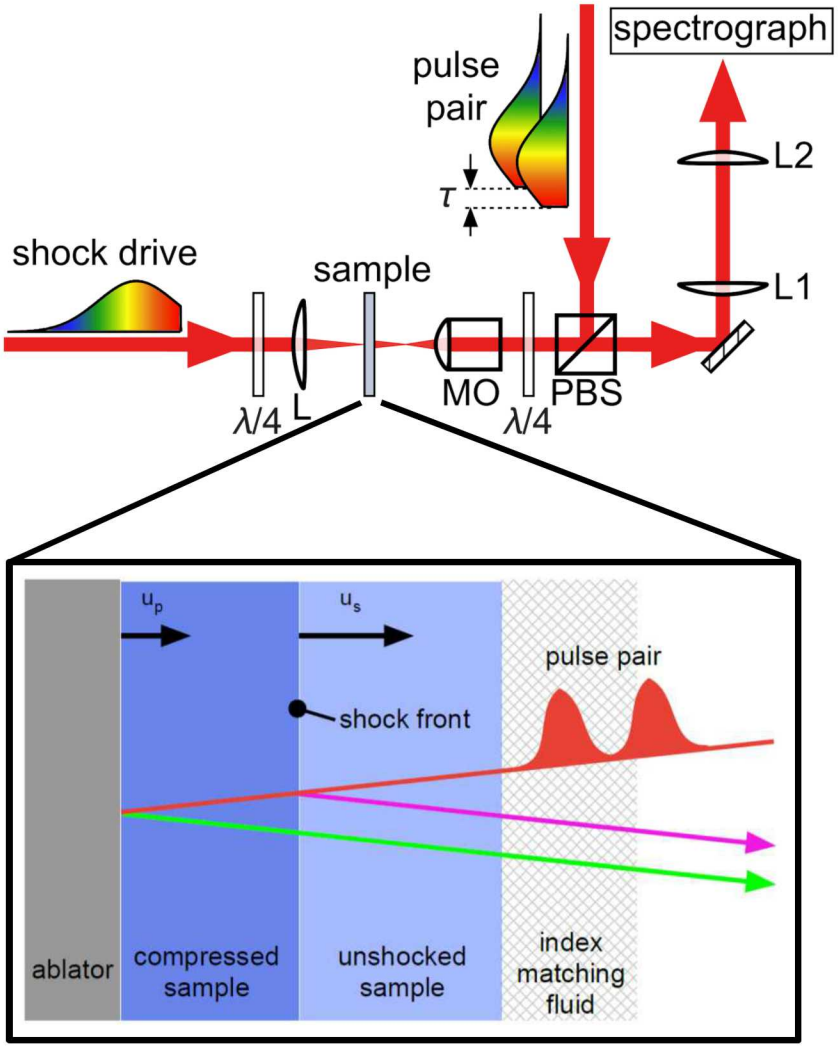
- Ultrafast experiments can provide insight towards phenomena such as chemical reactions and phase changes
- The capability of resolving the shock-to-detonation transition would be valuable for understanding the complex shock initiation mechanisms in energetic solids

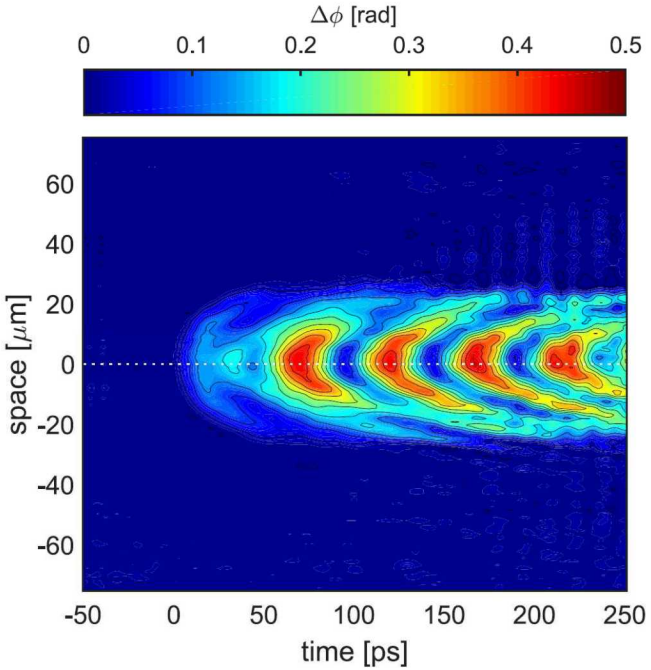
Achieving Picosecond Time Resolution

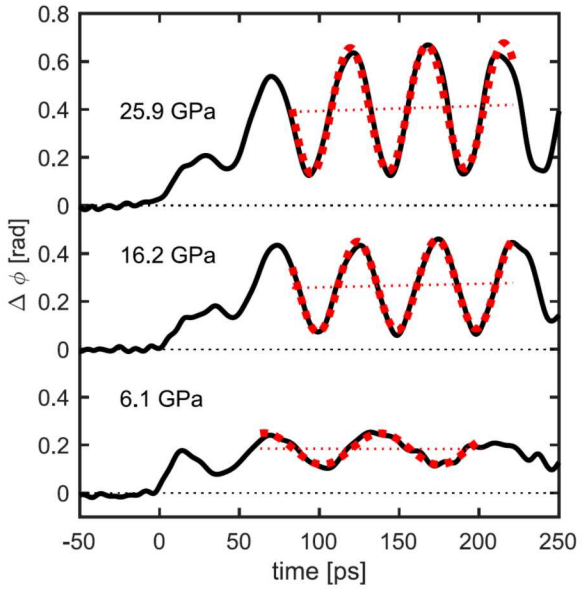


- Measure in time-frequency domain to probe the material during the initial stages of shock compression with fast time resolution
- Ultrafast pulses are linearly chirped to ~350 ps
- The linear chirp on the pulses maps the frequencies to a time axis, allowing the time-dependent phase difference to be measured with a spectrometer in a single shot

Ultrafast Shock Interrogation (USI)



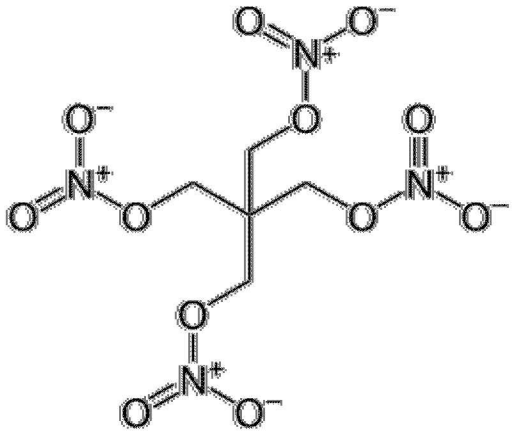




Armstrong *et al.*, *J. Appl. Phys.* (2010)
Bolme *et al.*, *J. Appl. Phys.* (2007)

Pentaerythritol Tetranitrate (PETN)

- Relatively high oxygen balance
- Thin reaction zone
- Many reports of the dynamics of initiation in PETN
  - Evidence of decomposition behind plastic wave
  - Reported intermediate velocity transitions – partial decomposition
  - Anisotropic shock sensitivity – steric-hindrance model

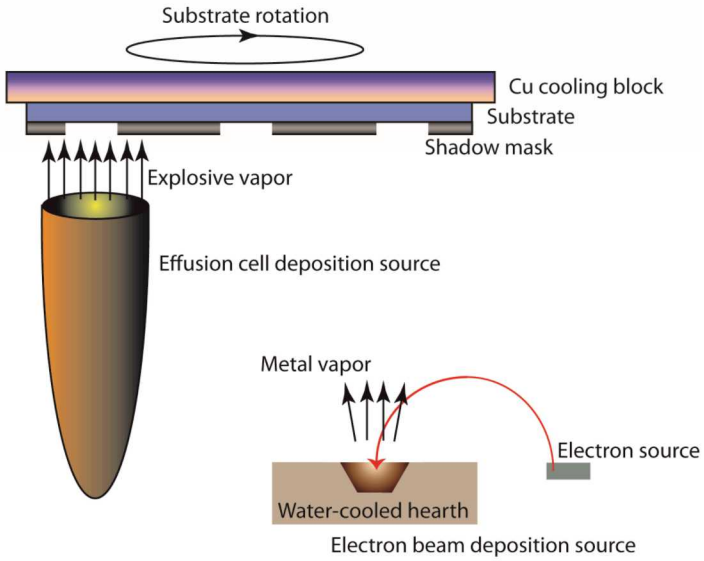


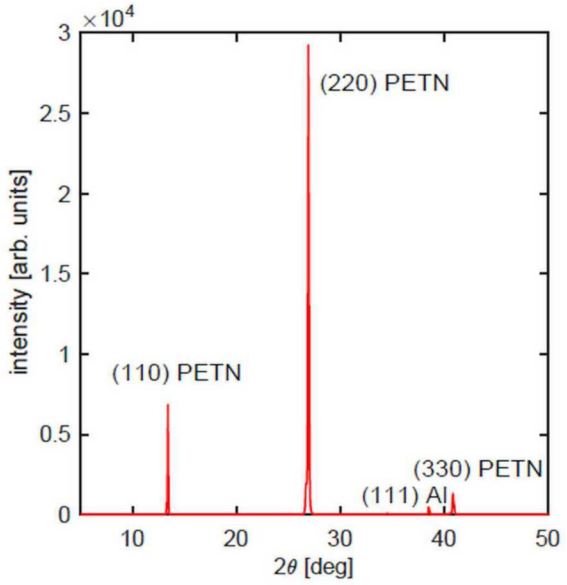
between the reaction zone and the shock front. Further improvement in the understanding of the PETN detonation wave front requires subnanosecond time resolution data.

Tarver *et al.*, *J. Appl. Phys.* (1997)
Halleck *et al.*, *J. Appl. Phys.* (1976)
Dick *et al.*, *J. Appl. Phys.* (1991)
Dick *et al.*, *Appl. Phys. Lett.* (1984)

PETN thin film samples

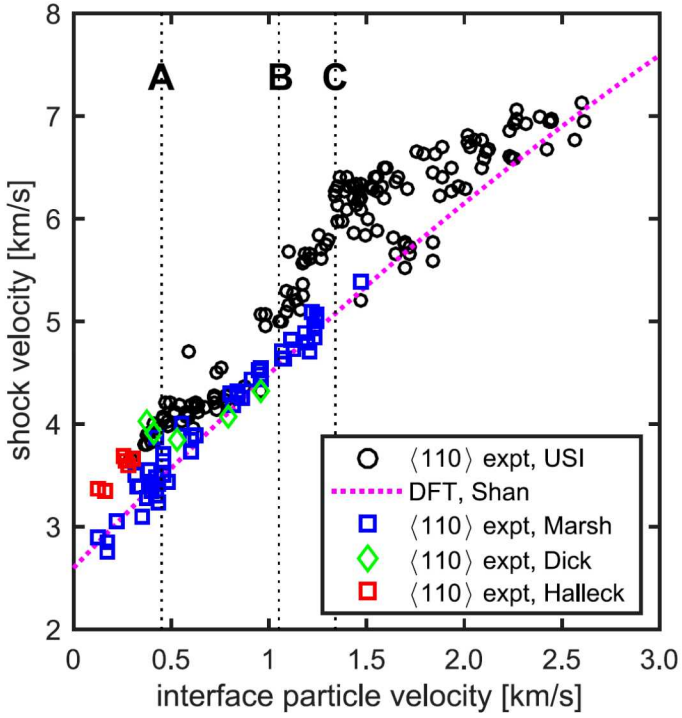
- Vapor-deposited PETN samples
- High density (≥ 97% unstrained crystal density – estimated from SEM images of fracture cross-sections)
- Very strongly textured with (110) planes parallel to the surface (measured using x-ray diffraction)

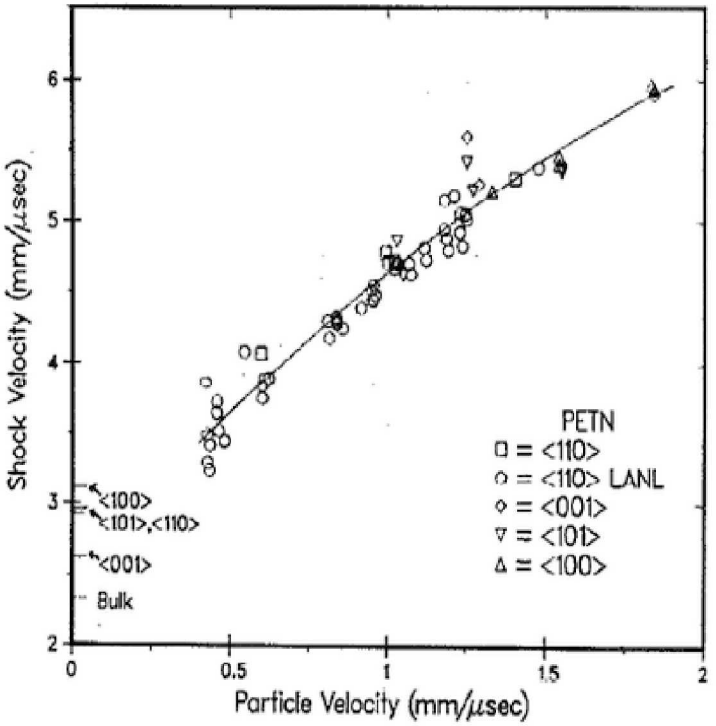




Knepper *et al.*, *J. Mater. Res.* (2011)

Ultrafast Shock-induced Reactions

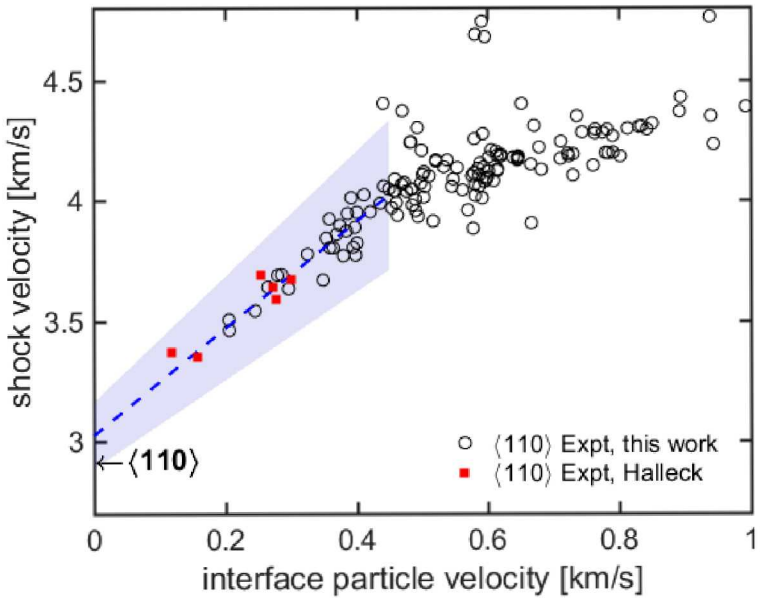


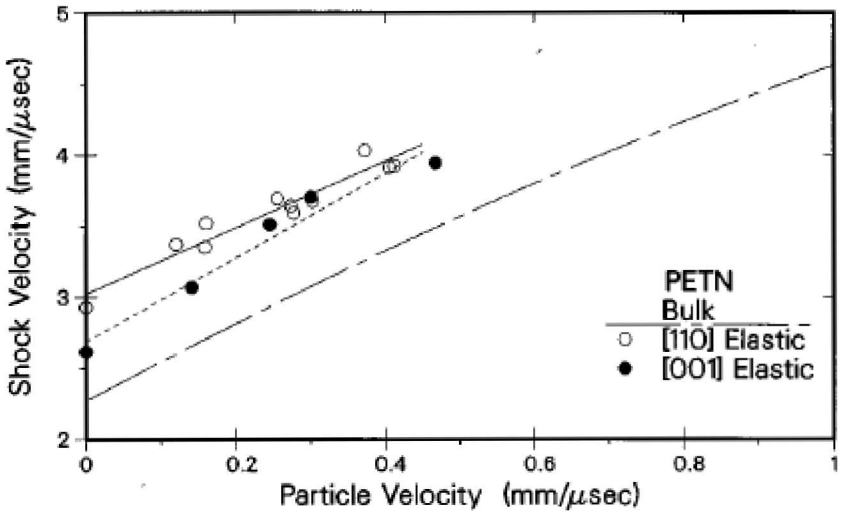


- Above u_p ~ 1.05 km/s, USI data lie well above the unreacted Hugoniot (greater than 4 standard deviations of the experimental uncertainty)
- Indication of **exothermic reaction** in which expanding gaseous products reduce the interface particle speed and increase the speed of the lead shock

Dick *et al.*, *J. Appl. Phys.* (1991)

<110> PETN Elastic Hugoniot

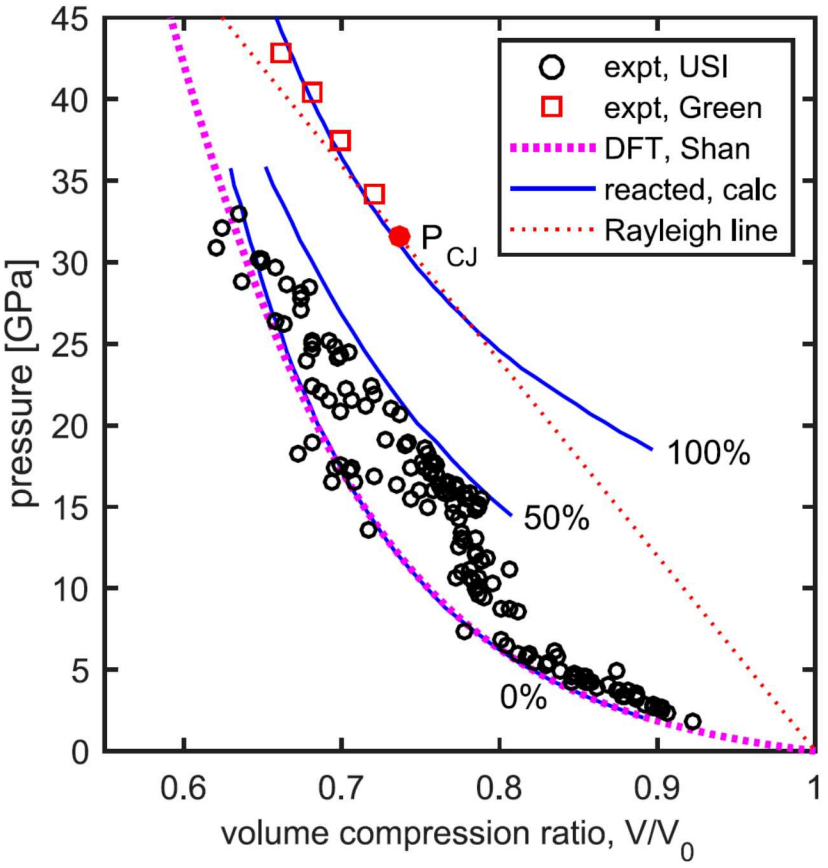




- Good agreement with literature results with the <110> elastic Hugoniot
- <110> longitudinal elastic sound speed of 2.93 km/s within the 95% confidence interval.

Halleck *et al.*, *J. Appl. Phys.* (1976)
Dick *et al.*, *J. Appl. Phys.* (1997)

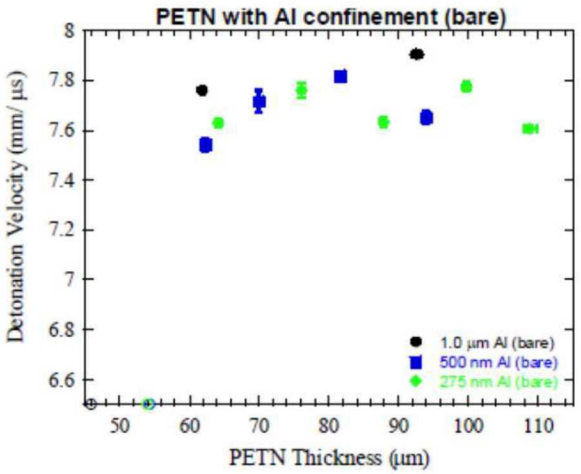
Ultrafast Shock-induced Reactions

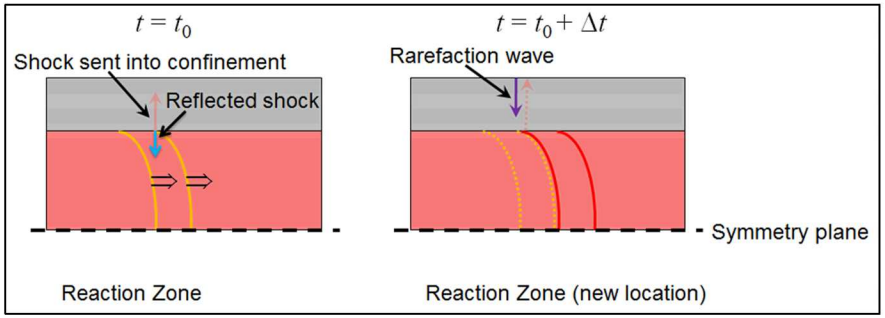


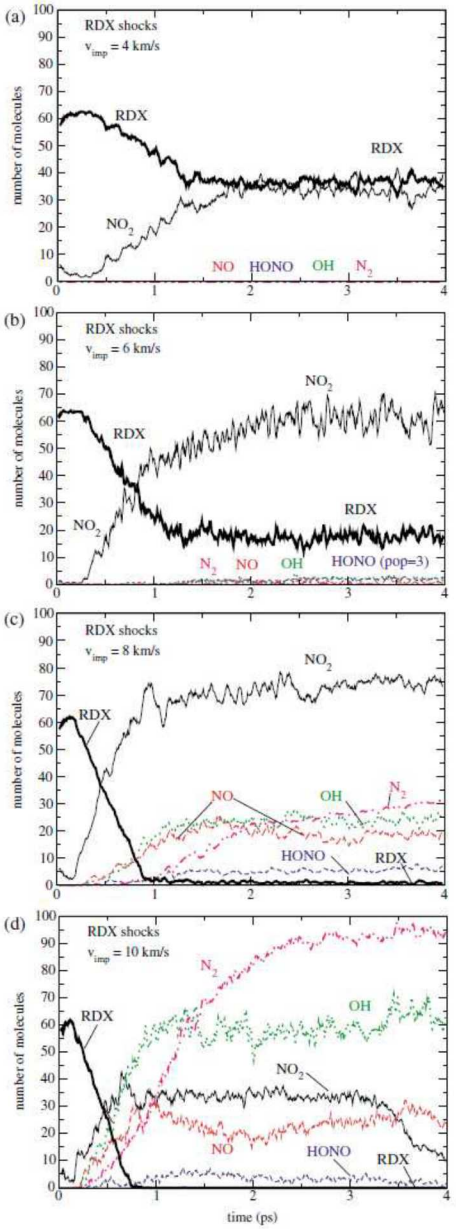
- Reported <110> sensitive from 8.5 – 12.5 Gpa
- Thermochemical calculations suggest that even on the timescale of the experiment, the higher pressure shots have induced considerable exothermic reaction
- Exothermic chemical reactions begin less than 50 ps after shock arrival

Dick *et al.*, *J. Appl. Phys.* (1991)
Green *et al.*, *Int. Symp. Pyro. Expl.* (1987)

Ultrafast Shock-induced Reactions





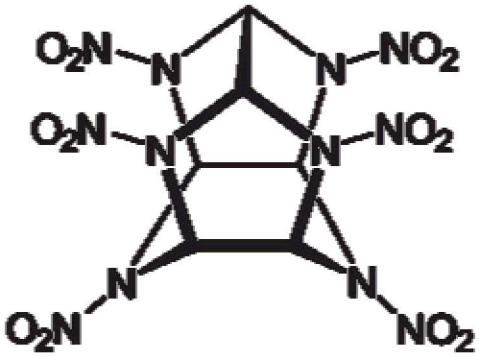


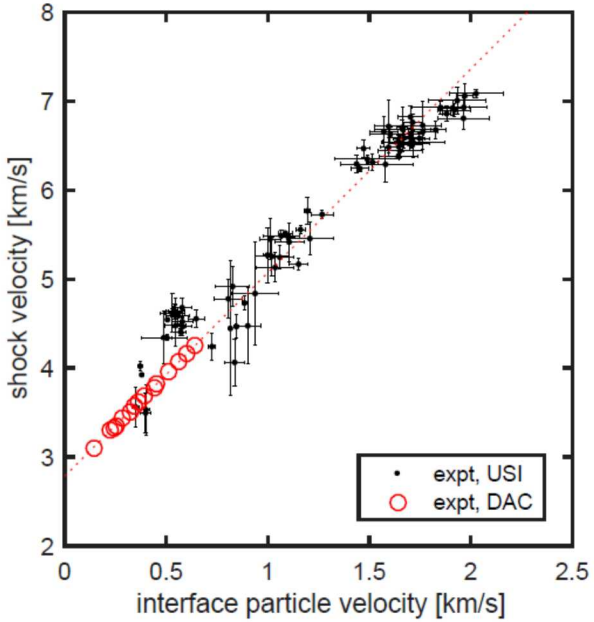
- Recently reported results from confinement thickness experiments with similar thin-film PETN samples suggest reactions occur <90 ps
- MD simulations may provide information about the kinetic mechanisms

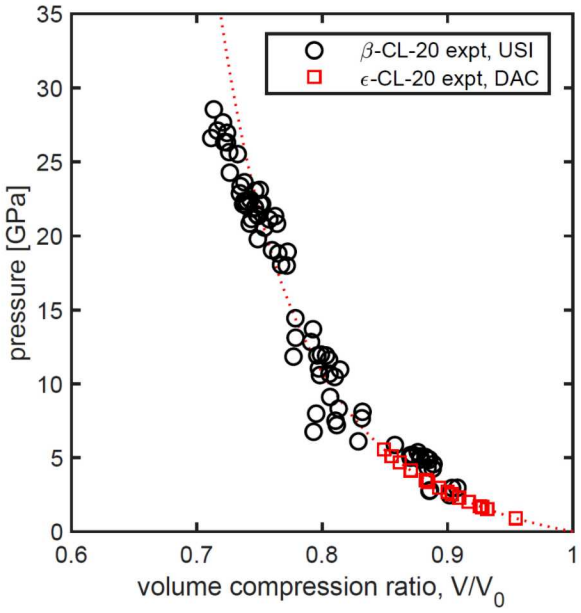
Strachan *et al.*, *Phys. Rev. Lett.* (2003)
Knepper *et al.*, *SCCM Proc.* (in press)

Preliminary Results on β-CL-20

- beta polymorph of CL-20
- preferred (111) orientation
- Compare to ε-CL-20 Hugoniot calculated from isothermal compression measurements (LLNL)
- Do not observe any indication of reaction outside of noise
- Possible indication of structural sensitivity?







Summary & Conclusions

- Direct measurement of the transient of the initial chemical reactions induced by shock in PETN thin films.
- Observe a large volume a large volume expansion in PETN that places state of the material well above the unreacted Hugoniot (~8 GPa and $V/V_0 \sim 0.78$)
- **Suggest that exothermic chemical reactions begin less than 50 ps after shock arrival**
- Possible indication of structural sensitivity from CL-20 experiments

Acknowledgments

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- Ian Kohl
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- Craig Tarver
- Brian Barnes
- Shawn Mcgrane
- Leanna Minier
- Ryan Wixom
- Michael Marquez
- Ronald Goeke

Joint Department of Defense/Department of Energy Munitions Technology Development Program

Sandia's Laboratory Directed Research and Development program

U.S. DEPARTMENT OF ENERGY

National Nuclear Security Administration

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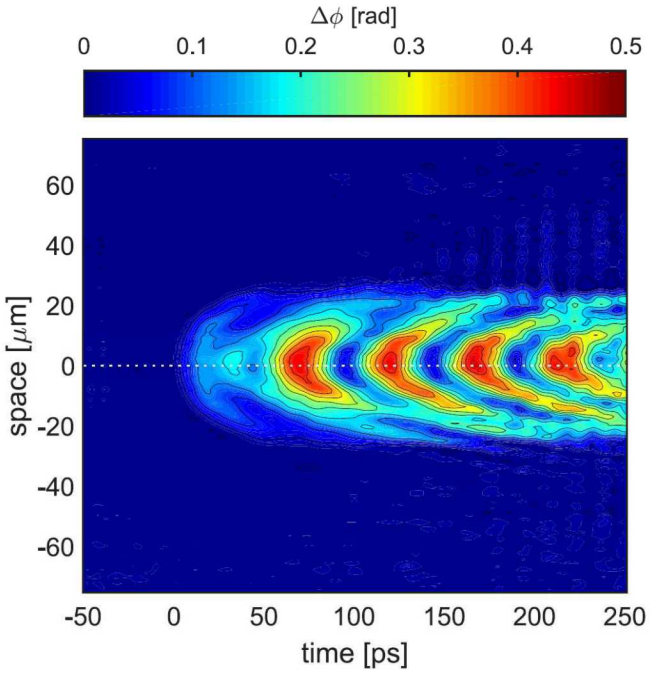
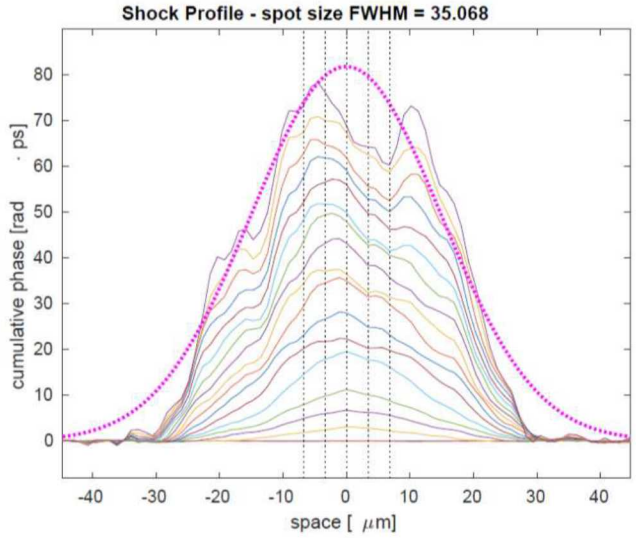
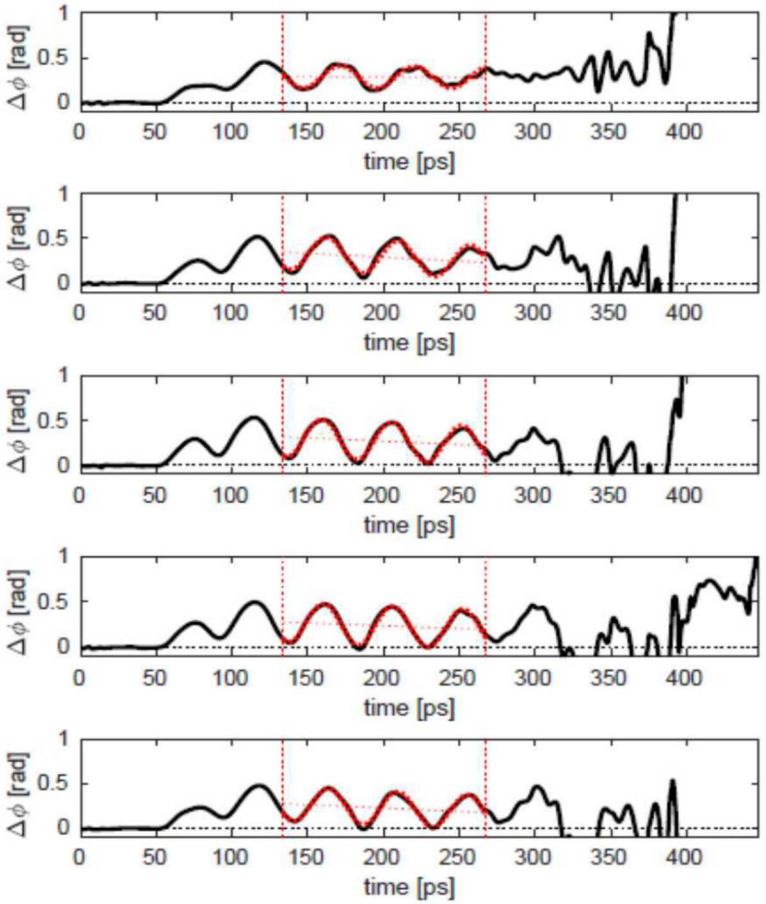
Begin extra slides

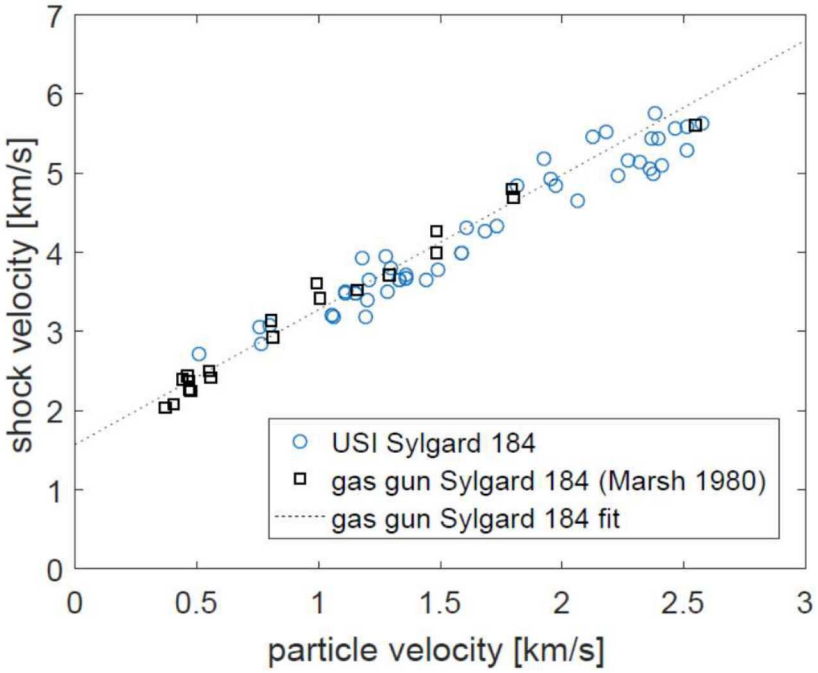

Δϕ [rad]
0
0.1
0.2
0.3
0.4
0.5
space [μm]
60
40
20
0
-20
-40
-60
-50
0
50
100
150
200
250
time [ps]


Shock Profile - spot size FWHM = 35.068
cumulative phase [rad · ps]
80
70
60
50
40
30
20
10
0
-40
-30
-20
-10
0
10
20
30
40
space [μm]

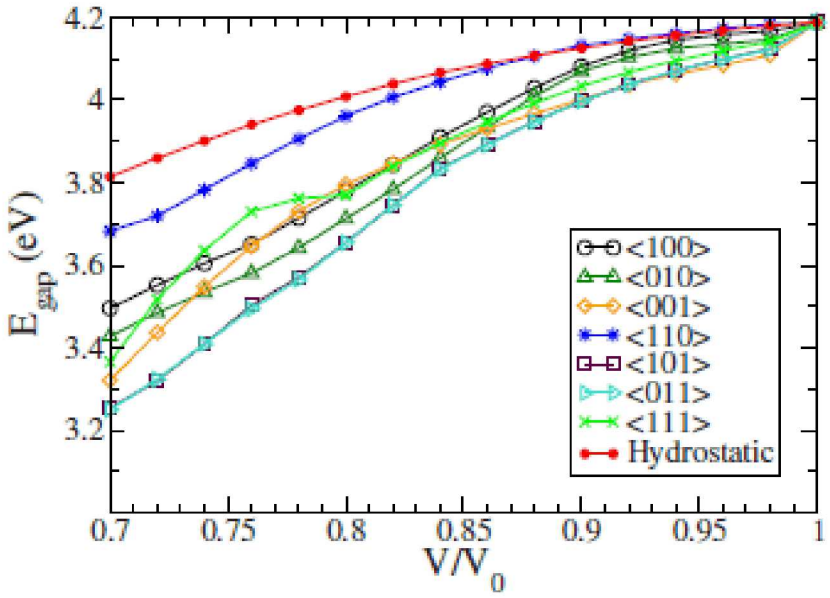

Δϕ [rad]
1
0.5
0
0
50
100
150
200
250
300
350
400
time [ps]

Sylgard 184

- Good agreement with previously reported gas gun results on polymer, Sylgard 184
- USI results are expected to lie on gas gun results because there are no expected chemical or physical changes in the material under shock compression



Marsh *et al.*, *LASL SHOCK HUGONIOT DATA* (1980)



- Red-shift of electronic bandgap during compression leads to increase in refractive index (Kramers-Kronig relation)
- Bandgap far away from pulse spectrum used in experiment
- Do not observe any absorptive losses in experiment

Conroy *et al.*, *Phys. Rev. B* (2008)

Assume highest absorbance – OD = 1.5

$\kappa = 0.000478$

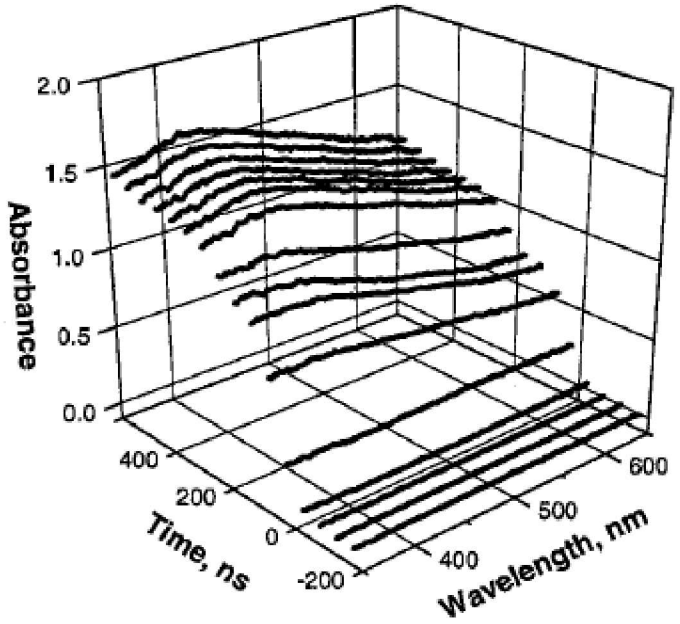
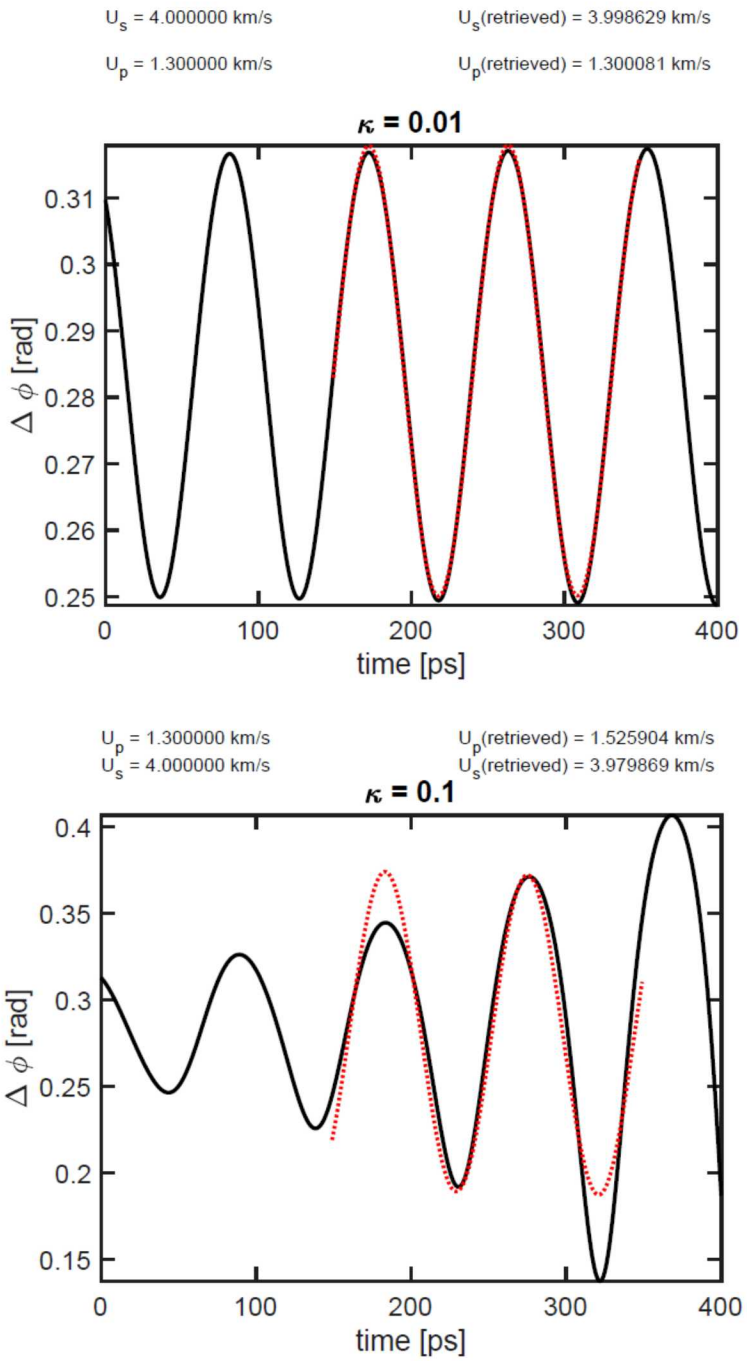
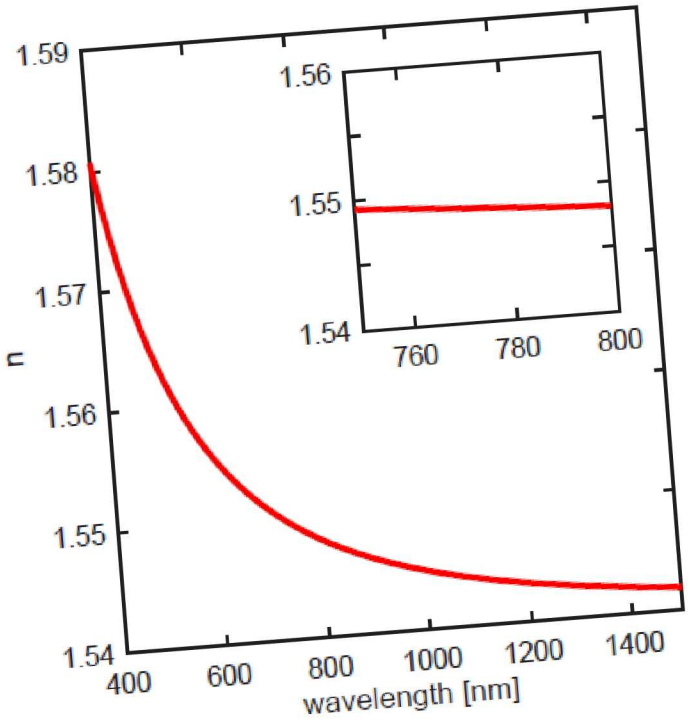


Figure 3. Time-resolved absorption spectra of PETN shocked to 5.1 GPa along the [100] direction (experiment A3). At 0 ns shock enters the sample.

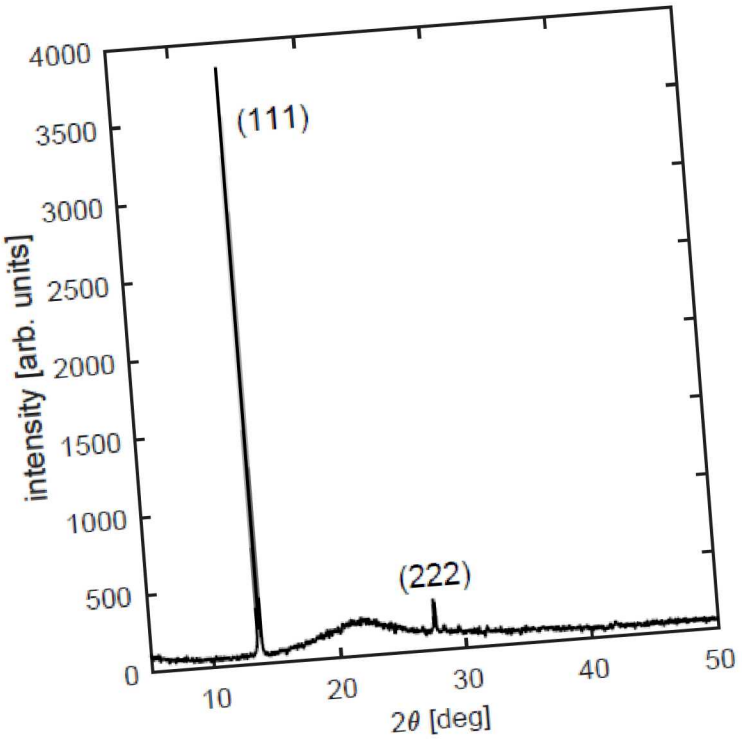


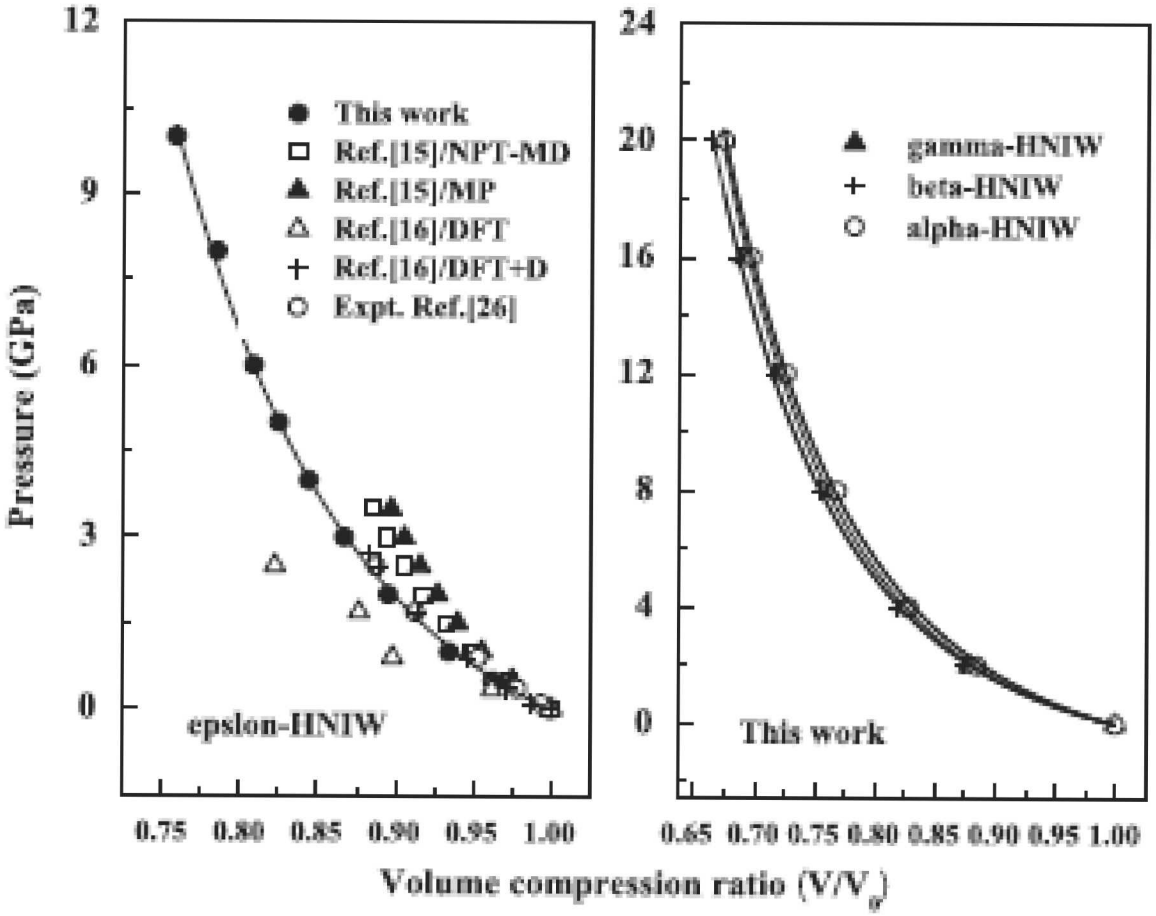
Dreger *et al.*, *J. Phys. Chem. B* (2002)

PETN


1.59
1.58
1.57
1.56
1.55
1.54
n
400
600
800
1000
1200
1400
wavelength [nm]
1.56
1.55
1.54
760
780
800

CL20


4000
3500
3000
2500
2000
1500
1000
500
0
intensity [arb. units]
(111)
(222)
10
20
30
40
50
2θ [deg]



Tan *et al.*, *Phys. B* (2011)