SAND2013-8993C

To be submitted for *Presentation at the 15th Detonation Symposium, San Francisco, CA, July 13-18, 2014*

Unreacted Hugoniot of hexanitroazobenzene (HNAB) determined by Ultrafast Shock Interrogation (USI)

Brook A. Jilek[1], Ian T. Kohl[1], Darcie A. Farrow[1], Junji Urayama[1], Robert Knepper[1], Sean P. Kearney[1],

Michael R. Armstrong[2], Jonathan C. Crowhurst[2], Joseph M. Zaug[2]

1. Sandia National Laboratories[1], Albuquerque, NM 87185, USA
2. Lawrence Livermore National Laboratory[2], Livermore, CA 94550, USA

Submitted to subject area: Advanced and Novel Experimental Techniques (primary); Detonation and Sub-Detonative Phenomena (secondary)

Abstract

Equation-of-state (EOS) models are based on measurement of the Hugoniot of a material. Ideal explosives are characterized by rapid reaction kinetics, which make Hugoniot measurements of the unreacted material difficult because particle velocities in the shocked material must be observed before reactions take place. The need for extreme time resolution limits the measurement of shock Hugoniots in ideal explosives to input pressures well below detonation pressures. For example, the standard reference, Los Alamos Scientific Laboratory (LASL)

---

[1] Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

[2] This research was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. DE-AC52-07NA27344.

Shock Hugoniot Data, reports Hugoniots for single-crystal PETN up to 14 GPa [1], far less than the 31.4 GPa C-J pressure of PETN of [2]. Recent efforts by Wixom to extend Hugoniot data to higher pressures via first-principles Density Functional Theory (DFT) modeling have shown good agreement with reported Hugoniots for PETN, but direct experimental measurement of higher-pressure Hugoniot states is needed for further validation [3].

Hexanitroazobenzene (HNAB) is an energetic material with properties that make it a model system to study the effects of microstructure on initiation. HNAB can be vapor-deposited as a fully dense amorphous film. If kept at room temperature, it crystallizes into a dense (99.4% dense) film with nanometer-scale pores [4]. In the amorphous state, it is homogeneous like liquid explosives (e.g. nitromethane) and lacks any pores to serve as nucleation sites for localized initiation of chemical reactions. Unlike liquid explosives, it is a solid, so there are much stronger intermolecular bonds. In the crystallized state, detonation studies can be done to compare its properties to those of the amorphous state to infer effects of pores and lattice structure on initiation.

We have implemented a diagnostic, Ultrafast Shock Interrogation (USI), originally developed by Benuzzi-Mounaix [5], recently used by Armstrong, *et al.* at Lawrence Livermore National Laboratory [6,7], and closely related to work by Bolme, *et al.* at Los Alamos National Laboratory [8], which can rapidly measure the unreacted Hugoniot of thin films of energetic materials. USI overcomes the time-resolution limitations of other diagnostics such as PDV and VISAR by encoding time information onto the spectrum of a probe pulse. A diagram of the experimental setup is seen below in Figure 1. A broadband (25 nm FWHM) femtosecond pulse is chirped so that the various wavelengths travel different path lengths and become separated in time, resulting in a probe pulse that is 300 picoseconds (FWHM) in duration. The blue edge is clipped in the beam stretcher to produce a pulse with a sharp rise-time. This chirped pulse is split into two portions: a higher energy pulse drives a shock into a target while a low energy pulse travels through a Michelson interferometer to create a pair of probe pulses with a relative time delay of ~10 picoseconds between them. The drive pulse is focused to a spot approximately 30 μm in diameter on the substrate side of a target, rapidly ionizing an aluminum layer on the other side

of the substrate and causing a shock wave to be driven into the sample material. The probe pulses are slightly focused by an objective lens to a spot approximately 100 μm in diameter so they cover not only the entire area shocked by the drive pulse, but unshocked reference regions as well. The objective lens images the face of the sample onto the slit of an imaging spectrometer. The CCD attached to the spectrometer records an interference pattern created by the path-length differences between corresponding wavelengths in the probe pulse pair.

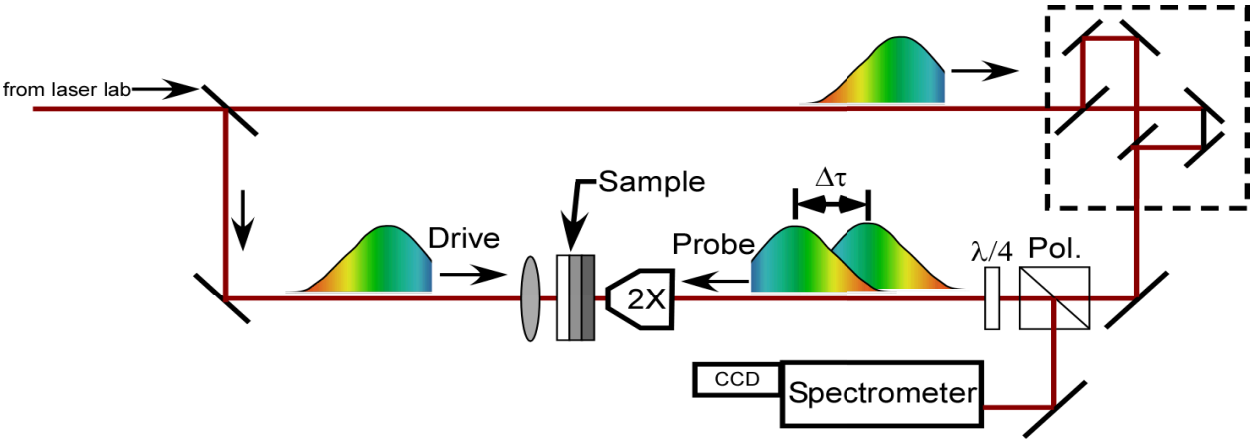

Figure 1. USI setup detailing the splitting of chirped pulses into drive and probe pulses and their subsequent measurement with spectral interferometry.

As time progresses, the phase difference accumulated between the two probe pulses changes as the second pulse is reflected from the moving ablator and the shock wave in the material a few ps later than the first pulse, curving the fringes seen on the spectrometer, as below in Figure 2. Thus a record of the ablator velocity (U_p) and the shock velocity in the material (U_s) can be made. This is done for a range of shock drive pressures to develop the unreacted U_s-U_p Hugoniot. We present Hugoniot measurements on the polymer Sylgard® and compare to LASL Shock Hugoniot Data. We also present, for the first time, measurements of the unreacted Hugoniot of hexanitroazobenzene (HNAB). Measurements made independently at SNL and LLNL on the same samples are compared to each other.

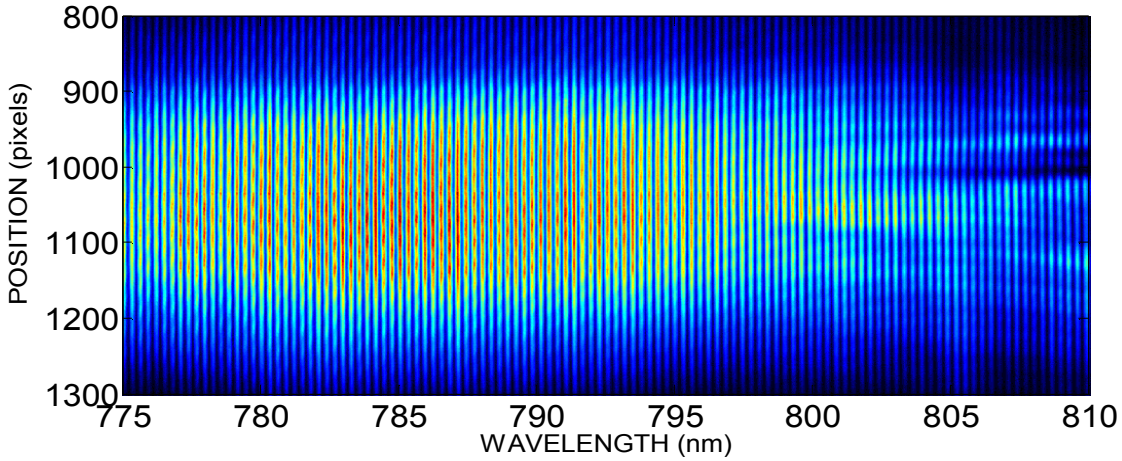

Figure 2. Example of fringes measured by spectrometer. Note the slight curvature of the fringes near row 1000 in the wavelength range of 800-810 nm due to movement of the metal layer as time progresses.

References:

[1] Marsh, S. P. LASL Shock Hugoniot Data. (University of California Press, Berkeley, 1980).

[2] Green, L., & Lee, E. Detonation Pressure Measurements on PETN. (13th International Detonation Symposium. Norfolk, VA. 2006).

[3] Wixom, R. R., & Mattsson, A. E. Sandia National Laboratories. SAND_2013-8812. Development of Ab Initio Techniques Critical for Future Science-Based Explosives R&D. (2013).

[4] Knepper, R., Browning, K., Wixom, R. R., Tappan, A. S., Rodriguez, M. A., & Alam, M. K. Microstructure Evolution during Crystallization of Vapor-Deposited Hexanitroazobenzene Films. Propellants, Explosives, Pyrotechnics. **37,** 459-467 (2012)

[5] Benuzzi-Mounaix, A.; Koenig, M.; Boudenne, J. M.; Hall, T. A.; Batani, D.; Scianitti, F.; Masini, A.; Di Santo, D. Chirped Pulse Reflectivity and Frequency Domain Interferometry in Laser Driven Shock Experiments. Phys. Rev. E, **60**, R2488–R2491 (1999).

[6] Armstrong, M. R., Crowhurst, J. C., Bastea, S., & Zaug, J. M. Observation of off-Hugoniot shocked states with ultrafast time resolution. (14th International Detonation Symposium. Coeur d'Alene, ID. 2010)

[7] Armstrong, M. R., Crowhurst, J. C., Bastea, S., & Zaug, J. M. Ultrafast observation of shocked states in a precompressed material. J. Appl. Phys. **108**, 023511. (2010).

[8] Bolme, C.A., McGrane, S. D., Moore, D. S., Whitley, V.H., & Funk, D.J. Single shot Hugoniot of cyclohexane using a spatially resolved laser driver shock wave. Applied Physics Letters. **93**, 191903 (2008)