

Development of spectral interferometry for shock characterization in energetic materials

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Abstract. Ultrafast laser diagnostics have opened new pathways for investigation of shock physics and initiation of energetic materials. Short laser pulses on the time scale of 100s of picoseconds can be utilized for direct laser drive and coupled with imaging, spectroscopic, and interferometric tools at femtosecond scales for studies of dynamic loading during shock transit. At Sandia National Laboratories, we are implementing diagnostic platforms which extend the recent development of Ultrafast Shock Interrogation (USI) by Armstrong et al. [1] for table top measurement of Hugoniot/Equation-of-state data and characterization of shock structure with micron spatial resolution and picosecond time resolution. We present bare aluminium ablator data and initial data for pentaerythritol tetranitrate (PETN) as well as describe our experimental setup.

1. Introduction

Mechanical and thermochemical mechanisms at play during shock passage are tightly coupled at extreme spatial and temporal scales on the order of microns and picoseconds; not experimentally accessible until the development of femtosecond lasers. With thin film samples deposited on metal ablators, carefully shaped broadband laser pulses can be utilized for direct laser drive with fully supported shocks for 100's of picoseconds [1-13] to even nanosecond time scales, enabling ultrafast observations of mechanical and thermochemical behavior during shock passage.

Direct laser drive experiments have been coupled with spectral interferometry [14] to characterize the response of dynamically loaded materials on the timescales of the shock passage itself. Ultrafast Dynamic Ellipsometry (UDE) has been developed at Los Alamos National Laboratory to combine two heterodyne interferometers at high and low angles of incidence to examine the polarization-dependent optical response of shocked samples [8, 10]. This approach provides spatially resolved histories of shock and particle velocities along with dynamic pressure. A simplified spectral interferometer using a single homodyne pulse pair at normal incidence has been demonstrated at Lawrence Livermore National Laboratory (Ultrafast Shock Interrogation-USI) [1]. Both the UDE and USI approaches offer the attractive possibility of Hugoniot equation-of-state measurements using economical tabletop laser setups and small amounts of explosive, which dramatically reduce safety considerations. Recent measurements have reproduced portions of the cyclohexane [10] and H_2O_2 [17] Hugoniot [18].

At Sandia, we are implementing the Livermore-developed USI approach for the characterization of laser-driven shock waves in energetic materials and for shock-Hugoniot measurements to complement gas-gun data. Here, we present the details of the Sandia USI instrument and briefly summarize the



canonical USI working principles outlined by Armstrong et al. [1]. We present preliminary results on shocked aluminum ablators and shocked PETN.

2. Spectral Interferometry/Ultrafast Shock Interrogation

The USI approach was implemented as described by Armstrong et al. [1] with the following exceptions: A commercial Ti: Sapphire amplifier (KM Labs Wyvern) provides 4 mJ pulses of 42 fs duration at 1 kHz repetition rate. The amplifier output spectrum is centered near 790 nm, with a bandwidth of 25 nm (FWHM). A user-selectable portion of the ~ 170 ps regenerative amplifier output energy is extracted before compression and further stretched in an external grating compressor, so that the blue edge of the spectrum leads the red. A fast time-domain rise is added to the pulse by clipping the dispersed spectrum within the compressor. A cross-correlation frequency resolved optical gating (XFROG) trace of the pulse shaper output using a 1.5 nm bandwidth-limited gate pulse is shown in figure 1. We measure a rise time of 25 ps, based on the 10%-90% intensity points of the spectrally integrated XFROG trace. Previous work [5] using an external stretcher, produced 10 to 20 ps rise shock waves in aluminum films. In the “overcompressed” pulse-shaper design used here, lack of a true Fourier plane in our external compressor may result in a more gradual rise to the clipped spectrum and time-domain pulse.

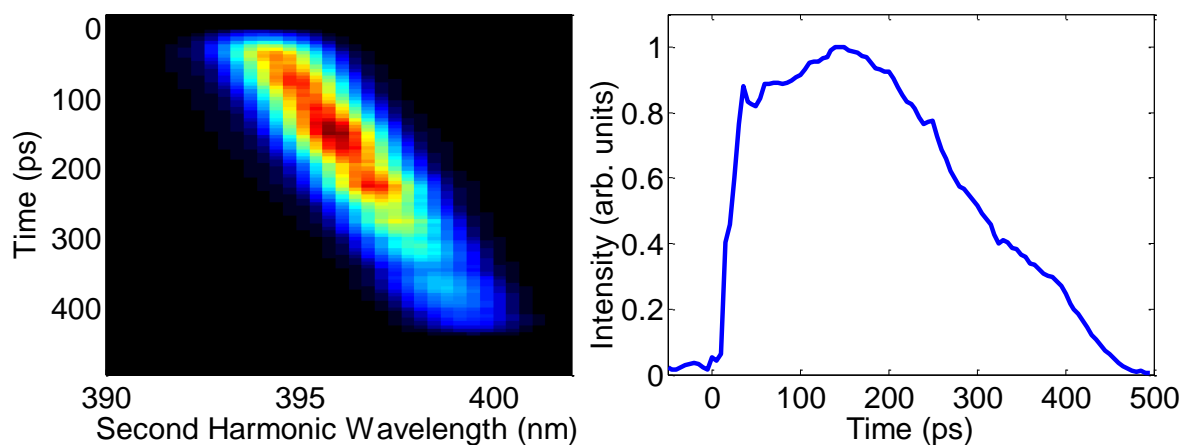


Figure 1. X-FROG trace (left) of the shock-drive pulse, (right) summed over all wavelengths.

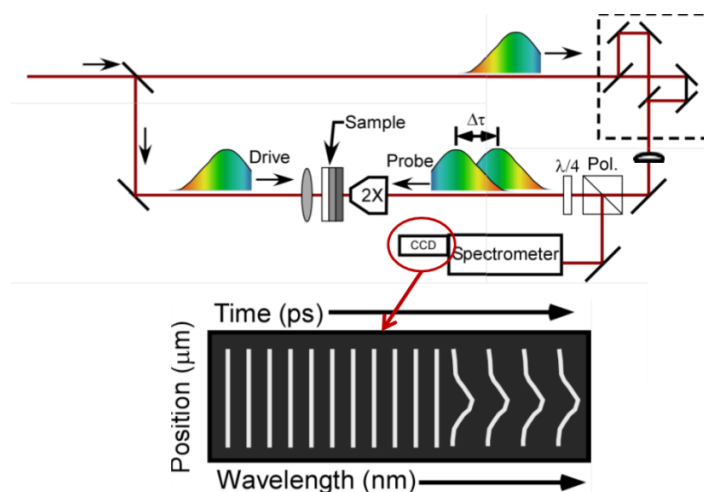


Figure 2. Schematic of USI experiment.

The bandwidth-clipped and stretched pulse is relayed to the sample in the arrangement shown in figure 2. The majority of the pulse energy is delivered to a pump beam directed onto the back side of the samples; 2 μm thick aluminum films deposited on a glass substrate bare or a film stack of 2 μm aluminum and a 5 μm layer of PETN on a glass substrate. The pump beam is focused at a numerical aperture (NA) ~ 0.03 using a $f = 150$ mm convex lens. The pump then focuses through the glass substrate to a ~ 30 μm spot at the back surface of the aluminum ablator, where the momentum exchange from the formation of laser-induced plasma drives a shock wave into the film. A small fraction of the pulse energy is split via a Michelson-type interferometer, where a delay is introduced to form a collinear probe-pulse pair that is directed to the sample through a polarizing cube with circular polarization imparted by a quarter-wave plate. We used delays of 11 ps for bare aluminum film measurements and 8 ps for PETN films. The probe-pulse pair is weakly focused at normal incidence onto the sample through the combined effect of a $2\times$ microscope objective of 0.07 NA and a $f = 500$ mm lens placed in the probe beam path. Introduction of the 500 mm lens permits proper imaging of the sample plane with a probe that is only weakly focused to ~ 100 μm , $3\times$ larger than the pump-beam spot that is centered within the probe. Back-reflected probe-beam energy is collected through the microscope objective and rejected by the polarizer-waveplate pair onto the entrance slit of a 0.3 m grating spectrograph, where the pulses are spectrally dispersed and combined onto a CCD camera placed at the spectrometer focal plane.

3. Results and Discussion

3.1 Bare Aluminum Films

A series of shots was recorded to characterize the performance of the USI setup for direct laser drive into bare aluminum films. Single-laser-shot spectral interferograms were recorded with drive-pulse energies of 400 and 800 μJ . By considering the path difference due to free-surface motion experienced by a near-monochromatic spectral slice, $\lambda(t)$, of the probe pulse, Armstrong *et al.* [1] showed that velocity of the reflector free surface, u_a , can be related to the measured phase shift, $\Delta\phi$, and the user-prescribed delay between the probe pulses, $\Delta\tau$, by

$$\Delta\phi = \frac{4\pi}{\lambda} u_a \Delta\tau \quad (1)$$

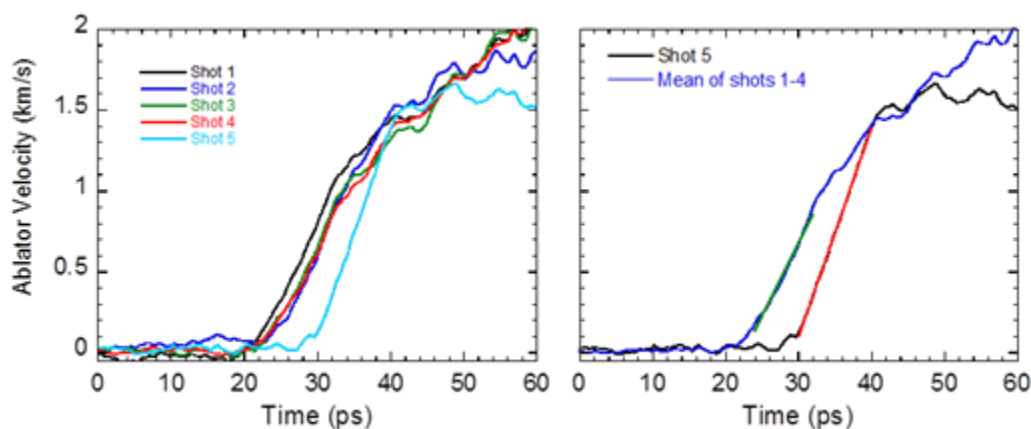


Figure 3. Bare aluminum ablator response at 400 μJ pump energy.

The resulting time histories of the shock-induced phase-shift profiles were converted to ablator velocity using equation (1). Free-surface velocity histories for pump energies of 400 and 800 μJ are shown in figures 3 and 4, respectively. Velocity-vs.-time histories for five consecutive single-laser-shot experiments are displayed at the left-hand side of the figures. These velocity data were taken from a single pixel row at spatial location near the center of the pump laser spot, where the velocities are maximized. Raw velocity data were smoothed with a 6 ps running average. For a 400 μJ pump, shots 1-4 exhibit highly repeatable behavior, with a rise from 0 to ~ 2 km/s in a 40 ps time window, however, the velocity appears to still be increasing at the end of the experiment, so that steady shock conditions are not yet reached. Data for shot 5 at 400 μJ pump exhibit a delayed shock-arrival time, with a steady-state velocity near 1.6 km/s achieved. The right-hand side of figure 3 highlights the difference in behavior at 400 μJ pump, with the average of shots 1-4 plotted alongside shot 5. The red and green lines on the right-hand plot represent linear fits to the data during the initial rapid rise, with acceleration of 0.09 km/s/ps observed for shots 1-4, where steady conditions were not realized, and 0.13 km/s/ps, a 45% increase in acceleration, for the fifth laser shot. At 800 μJ pump energy, two types of behavior are observed. The data for shots 1, 3, and 5, shown in figure 4, exhibit a rise to a near-plateau level of $u_a = 1.5$ -2 km/s before a second rise to 3 km/s by the end of the experimental window. Shots 2 and 4 display a more monotonic rise to a higher velocity of 4 km/s. The behavior of shots 1, 3, and 5 at 800 μJ pump is similar to the observation of very strong elastic precursors followed by a subsequent rise to a plastically deformed shocked state by Crowhurst *et al.* [11] and by Whitley *et al.* [12]. However, a steady state condition is not achieved at the end of the 60 ps experimental window, and more data would be needed to support this claim. Finally, it is noteworthy that the pump-pulse energy required to achieve significant acceleration of the Al films in these preliminary experiments was significantly higher than used by Armstrong *et al.* [1, 9, 13, 17], who observed effective laser drive at pump energies of 100 μJ and below. The numerical aperture of the pump-beam focusing in our experiments was discovered to be significantly smaller, which results in a longer beam waist and potentially induces parasitic losses from nonlinear processes within the glass substrate [20]. Recent experiments in our lab with more aggressive pump focusing have resulted in much more efficient laser shock drive.

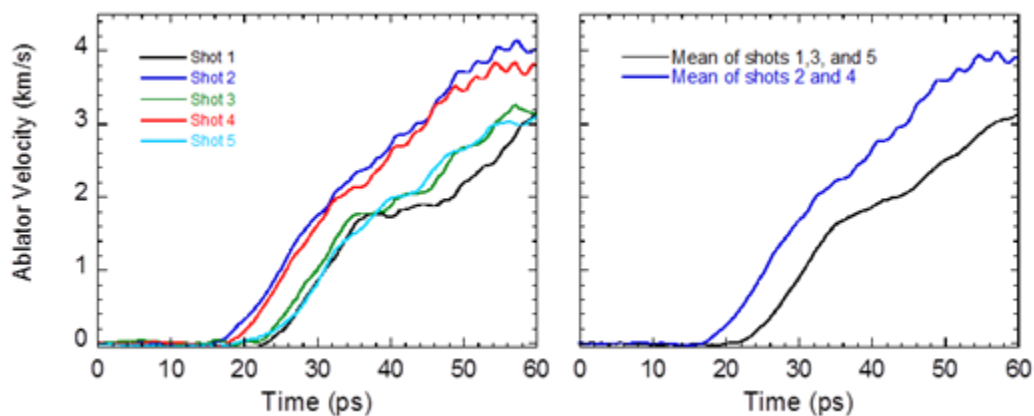


Figure 4. Bare aluminum ablator response at 800 μJ pump energy.

3.2 PETN Film

We have additionally performed preliminary USI experiments with a 5 μm PETN film deposited on the same type of aluminum ablators used for the results in section 3.1. The PETN films are vapor-deposited from an effusion cell source in the same deposition system immediately following the aluminum deposition without breaking vacuum. The PETN films are very dense and have a strong

(110) out of-of-plane crystal orientation. [21] In a shocked transparent material deposited on a reflective drive layer, Armstrong *et al.* [1] describe a canonical USI response, where an initial rise in the first ~ 20 -30 ps associated with the motion of the aluminum ablator is then followed by an oscillatory phase response. The preliminary result in figure 5 is similar to the canonical USI response. Armstrong *et al.*, considered the first-order reflections from an explosive-on-ablator sample with a shock front passing through the explosive film. They developed a model based on a rapid initial rise associated with the ablator motion behind the shock front and the dynamic optical etalon that is formed by the combined reflectivity of the shock-front and the ablator. The thickness of this etalon is proportional to $(u_s - u_p)$, which are the shock and particle velocities. The varying thickness of the etalon results in an oscillatory response of the sample's total reflectivity, with a period, amplitude, and offset that can be related to u_s and u_p , using equations provided in [1]. These estimates yield values of $u_s = 6.1$ km/s and $u_p = 2.2$ km/s. This value of u_s is in reasonable agreement (10%) of the result computed from PETN shock Hugoniot of $u_s = 2.81 + 1.75u_p$ [22]. We are currently widening the time window for USI on explosive films to capture a larger portion of the oscillatory portion of the trace and perform reliable shock-Hugoniot measurements.

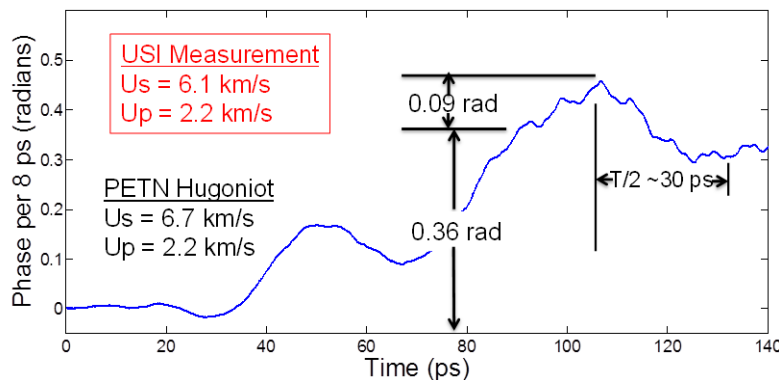


Figure 5. Preliminary USI phase-derivative history from a PETN-on-aluminum sample. The phase offset of 0.36 rad, amplitude of 0.09 rad and half period of ~ 30 ps are shown on the trace.

4. Summary and Conclusion

We are adopting the USI diagnostic developed by Armstrong *et al.* [1] for measurements of shock transit and Hugoniot equation-of-state data in explosive materials. Our preliminary measurements on shock breakout in bare aluminium films with direct laser drive exhibit well-resolved single-shot measurements at the lowest drive energies used, although steady breakout velocities were not generally achieved within the 60 ps experimental window. At higher drive energies, two types of shock-breakout behaviour were observed, with a monotonic 40 ps duration rise to a high velocity and behavior suggested from recent measurements [11, 12] which reveal strong elastic precursors in ultrafast laser-drive measurements on aluminium layers. More data are needed to confirm this observation. Preliminary data on a PETN film are also presented, which reveal the canonical USI response outlined in [1], albeit with a short experimental time window. We are currently implementing wide-format detectors to increase the time-duration of our experiments.

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