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Single shot ultrafast dynamic ellipsometry (UDE) of laser-driven shocks in single crystal explosives

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Abstract. We report on the first experiments to measure states in shocked energetic single crystals with dynamic ellipsometry. We demonstrate that these ellipsometric techniques can produce reasonable Hugoniot values using small amounts of crystalline RDX and PETN. Pressures, particle velocities and shock velocities obtained using shocked ellipsometry are comparable to those found using gas-gun flyer plates and molecular dynamics calculations. The adaptation of the technique from uniform thin films of polymers to thick non-perfect crystalline materials was a significant achievement. Correct sample preparation proved to be a crucial component. Through trial and error, we were able to resolve polishing issues, sample quality problems, birefringence effects and mounting difficulties that were not encountered using thin polymer films.
Keywords: RDX, PETN, Ultrafast dynamic ellipsometry.
PACS: 47.40.-x, 47.40.Nm

INTRO

Laser-generated shocks and diagnostics are extremely useful for studying small amounts of difficult-to-obtain material. Sub-millimeter regions can easily be probed, allowing for multiple measurements to be performed on a single sample with precise temporal and spatial synchronization between shock and diagnostics. We have developed and implemented a technique of generating shocks using a common chirped pulse amplified laser. Spectrally modifying a chirped laser pulse results in a drive shock with a material limited rise time and a sustained constant pressure for hundreds of picoseconds. This shock can be used to generate conditions similar to those found in gas-gun flyer plate experiments and potentially measure material Hugioniots with proper diagnostics.

With pressure loading from the shocks occurring in picoseconds, we need diagnostic techniques that have sub-picosecond to picoseconds time resolution and sub-nanometer sensitivity to surface motion. Dynamic ellipsometry(DE) has sufficient temporal and spatial resolution to probe the shock generated by chirped laser pulses. DE measures the time and space-resolved phase shifts of light incident on a shocked surface. Analysis of the phase shifts allows one to monitor the evolution of shock velocity, the particle velocity, the shocked index of refraction, and the pressure of a material during the hundreds of picoseconds that the shock traverses the sample.

This technique was originally developed to measure shocks in thin spin-coated polymer films, such as PMMA. Making minor changes in the system, we have adapted the technique to measure shocks in single crystal explosives of RDX, HMX and PETN. We report the unreacted Hugoniots determined using DE for single crystals of RDX and PETN.

EXPERIMENTAL PROCEDURE

A modified flashlamp-pumped Ti:Sapphire femtosecond laser was used to both generate the shock drive and to produce the probe pulses used to measure the shocked states of the crystals. Due to space constraints, we refer the reader to prior publications by McGrane et al for the shock drive generation and Bolme et al for descriptions of the dynamic ellipsometry used to map the family of shock states. Shock drive energies ranges from 0.5 to 8 mJ/pulse, corresponding to pressures ranging from 1 to 20 GPa.

A considerable amount of effort was dedicated to producing the highest quality surface finish with minimal sample damage. Thin slabs of material were sliced from larger crystals using a diamond impregnated wire saw. The cross-sectional area of the slabs are ~7mm x 7mm for these experiments. The slabs are cut to ~1mm thick with a 2° wedge between faces. The wedge diverts the reflection off the front surface of the crystal out of the collection pathway. This eliminates most problems associated with interference effects between the front and rear reflections off the crystal.

Molecular crystals like RDX and PETN are brittle and have a propensity to crack under the strain of polishing. The edges of the crystals also chip during the polishing and will create large scratches across the polished surface. Thus, there is a tradeoff between increased surface quality and increased likelihood of significant surface damage the longer each sample is polished. The technique we found to produce a workable surface quality with a minimal chance of damaging the sample was: (i) cut the sample slowly with a diamond saw to produce a surface smooth enough that we could skip the course polish, (ii) polish by hand with 0.05 um polishing compound mixed with water until few surface scratches remain when viewed with a microscope using a 50x objective. That correlates with approximately 30 minutes of polishing per side.

The polished sample was glued to a solid brass holder centered over a through hole in the block slightly smaller than the size of the crystal. 1-2 um of aluminum was deposited on the shock drive side of the sample through the hole in sample holder. A thin film of PMMA was evaporated out of toluene onto the shock drive surface in order focus the laser energy into the sample more efficiently[1]. The crystal and holder were mounted onto a kinematic fixture on a X-Y stage that allows us to move the sample from shot to shot in order to do multiple experiments on a single sample. Before data is taken, the overlap of the shock drive and two probe beams was checked to ensure the same is probed by all beams. In addition, RDX is birefringent[2] creating an additional interference pattern as the two birefringent images interfere with one another. Two of the crystal direction have almost identical indices of refraction for the β and g directions at ~1.62, but the α direction is 1.60. We rotate the crystal until the beam is only passing through the β-γ plane, eliminating the birefringent interference.

RESULTS AND DISCUSSION

A typical result of the processed phase and reflectivity is shown in

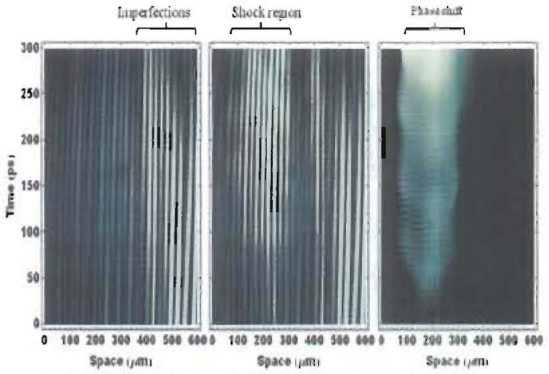

Figure 1. Here, the ordinate axis is the 250 μm spatial image across the sample surface and the abscissa is time. The p-polarized phase and reflectivity show prototypical features found in these experiments. The change in phase is Gaussian-shaped, due to the shape of the drive pulse, and increases in time up to 300 ps. The reflectivity shows little change, mainly because the aluminum layer determines the bulk of the reflectivity and does not change significantly at these pressures.[3]

The s-polarized images should be nearly identical to p-polarized images at 28°. The s-

phase, however, shows a significant phase shift from 60-90 ps, a 'phase flash', then returns to the Gaussian rise. The s-reflectivity shows a significant increase from 25-90 ps, drops to almost zero, and then recovers after 150 ps. These are features that have not been seen before and currently do not fit within the context of our model. The features only appear in s-polarization and generally occur earlier in time with higher laser energy used for shock generation. We are currently working to determine the causes of the features and if they are real or are an experimental artifact.

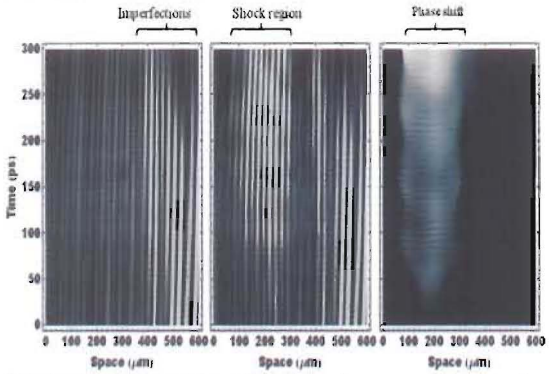

Figure 1. Comparison of the interferogram taken before and during a shock event. The figure on the right shows the phase shift generated by the shock.

With data taken at 0.5-1mJ drive pulse, the 'phase flash' occurs after several hundred picoseconds. During the first few hundred picoseconds, the S- and P-polarized images show the expected thin film behavior. For fitting purposes, a 1mJ data set was chosen that had phase flash issues after 200ps. The first 200 ps of data were used to fit for the shock parameters, as shown in Figure 2.

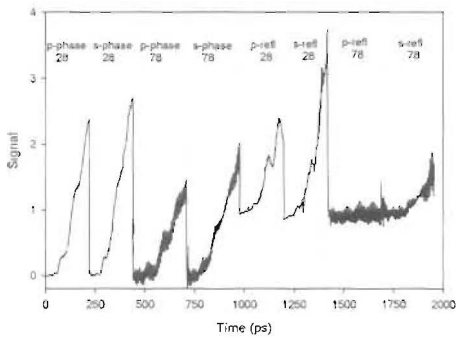

Figure 2. Set of ellipsometric data from 1.0 mJ shot showing the eight data sets that are simultaneously fit.

Fits to all eight data sets were poor. There are several likely reasons for this. As long as the high angle and low angle cameras are viewing identical regions, the fits should match all eight data sets. If there is a misalignment in the viewed regions, then each camera will see a different pressure from the Gaussian-shaped shock. In these cases, the high angle and the low angle cannot be fit simultaneously and instead must be fit separately. A second problems lies in the low angle reflectivity. The low angle reflectivity in both polarizations increased by a factor of three, as shown in Figure 2. The initial reflectivity of the idealized system is 90%, so the thin film equations can, at best, increase the reflectivity by only 10%. The experimental reflectivity was not fit because the thin film equations cannot fit the low angle reflectivity increase. Thus, the data was fit using low-angle phase or high-angle phase. The fits will be less constrained when all eight data sets are not used in a simultaneous fit, but will still produce good results.

To get multiple pressure points, data was chosen at 100%, 90%, 80%, 70%, and 60% of center of Gaussian. This data is shown in Figure 3. The exact value chosen is not important because the fits will still produce values for Us and up that should lie on the Hugoniot. So, the inconsistent increase in phase change between the various curves shown in Figure 3 is not concerning.

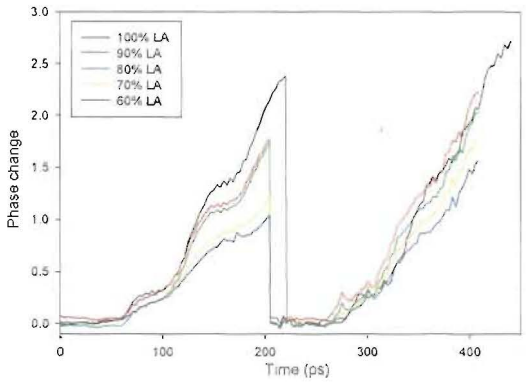

Figure 3. Low angle phase change at different pressures

A typical fit of the low angle phase data to the thin film ellipsometric equations is shown in Figure 4. There is good overall agreement between the shape of the fit and the data, but the phase change is overpredicted for p and underpredicted for s. We believe that the s-phase data may be affected by the phase flash and is abnormally high. When the data is fit, the abnormally high s-phase will cause the p-phase fit to predict higher than expected values.

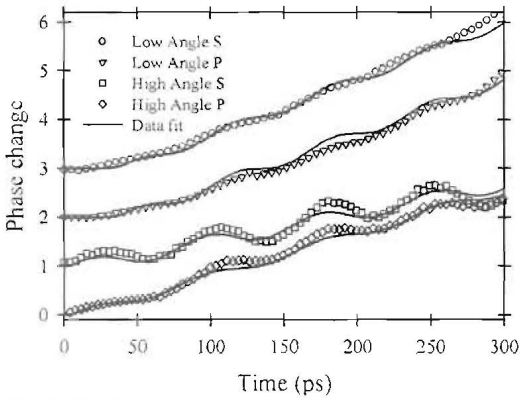

Figure 4: Comparison of the phase and fit of the low angle phase change.

Fits of the phase for the five different data sets were produced and used to calculate the pressure. Figure 5 shows the P-up fit in comparison with results from Ramos obtained using gas gun flyers and from a molecular dynamics calculation by Cawkwell. It is surprising that MD calculations using a few hundred nanometers of material on a picoseconds timescale compares with ellipsometry on 0.0625 mm^3 of material on hundreds of picoseconds time scales and flyer plate experiments on 500 mm^3 of material on hundreds of nanoseconds . Obtaining and comparing values across *four* decades in time and *six* decades in volume is a significant achievement in itself.

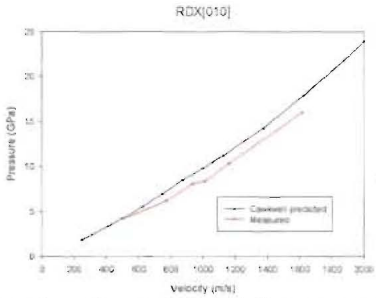

Figure 5: Comparison of Hugoniot of RDX[010] predicted by MD calculations with Hugoniot measured using dynamic ellipsometry.

The values obtained by spectroscopic ellipsometry are consistently lower than values found in flyer plate experiments and values predicted by Cawkwell. One reason for the difference is related to the volume of the probed material: the ellipsometry probes smaller, more pristine areas of the sample than gas gun experiments. The ellipsometry probes optically homogenous material with flaws smaller than ~ 5um in the probed region. Flyer plate experiments use the entire sample and therefore encounter numerous intrinsic flaws in the sample over the duration of the experiment. So, the material being probed between the two experiments is not identical. Additionally, the ellipsometry probes time frames long before any chemical reactions take place. The gas-gun experiments probe much later in time after reactions have potentially begun. So, the state of the material may be significantly different between the two experimental techniques. However, the MD predictions, which use perfect non-reacting material, compare more favorably with the gas-gun values than the ellipsometry values. The potential fitting issues in ellipsometry could be a factor. More work needs to be done to understand what causes the difference in values produced by ellipsometry from those produced by gas-gun flyer plates and MD calculations.

CONCLUSIONS

We report on the first experiments to measure states in shocked energetic single crystals with dynamic ellipsometry. We have successfully

demonstrated that these ellipsometric techniques can produce a reasonable Hugoniot using small amounts of material. The adaptation of the technique from uniform thin films of polymers to thick non-perfect crystalline materials was a significant achievement. Correct sample preparation proved to be a crucial component. Through trial and error, we were able to resolve the polishing issues, sample quality problems, birefringence effects and mounting difficulties that were not encountered using thin polymer films.

Comparisons with pressures produced using these ellipsometric techniques with gas-gun flyer plate experiments and MD calculations show a good correlation between the three techniques.

Several issues still need to be resolved. The ellipsometric values are consistently lower than values produced by other methods. The fitting of the phase data needs to be improved. Lastly, many figures show anomalous phase and reflectivity changes. A 'flash' is seen in the phase for a short period of time before it recovers to original phase. The reflectivity increases several times before dropping to 0 at roughly the same time as the phase recovers. These effects are only seen in the s-polarized data. The cause is unknown and more data will be taken to determine if it is real or an experimental artifact.

Future work will involve measuring RDX samples cut along the [210] and [001] orientations and PETN.

ACKNOWLEDGEMENTS

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