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Title: Ultrafast Dynamic Ellipsometry and Spectroscopies of Laser Shocked Materials

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Ultrafast Dynamic Ellipsometry and Spectroscopies of Laser Shocked Materials

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Abstract: Ultrafast ellipsometry and transient absorption spectroscopies are used to measure material dynamics under extreme conditions of temperature, pressure, and volumetric compression induced by shock wave loading with a chirped, spectrally clipped shock drive pulse.

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1. Introduction and Motivation

A shock wave can uniaxially compress a material by tens of percent, raise the temperature by several hundred to a few thousand Kelvin, and create molecular level distortions that may alter electronic structure. The drastic changes occur in the time it takes the shock front to traverse the molecule at velocities approaching 10 nm/ps. The broad and diverse applications of shock physics have led to extensive continuum level understanding, however, most of the molecular level physics and chemistry detail remains invisible. How does a shock affect a molecule in a solid? How is the energy of the shock coupled into the molecule?[1] What chemical reactions occur? How fast do they occur? What is the molecular temperature? Are the molecules in their ground electronic state? Are the electronic states seriously distorted, excited, or ionized at the shock front?[2,3,4,5,6] What is the effect of shock orientation relative to the molecular structure?

These types of molecular level phenomena are typical of observations accessible to ultrafast laser spectroscopies. We discuss our method of generating appropriate shock states[7] synchronized to single shot spectroscopies[8] capable of dealing with the destructive nature of the shock process and demonstrate typical experimental results on a few material systems.

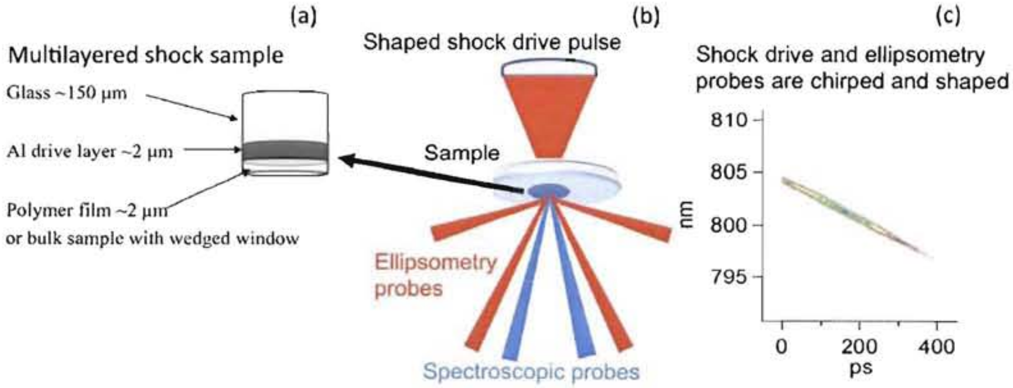

FIGURE 1. Sample geometry (a) shown in reference to the shock drive and reflected probe lasers (b). The shock diameter is ~100 μm, allowing thousands of shots on a 25 mm diameter sample. The shock drive and ellipsometry probe pulses are chirped and spectrally clipped. The time dependent spectrum and intensity is measured by X-FROG (c).

2. Experiment

A 110 fs, 25 mJ, 10 Hz chirped pulse amplified Ti:sapphire laser is modified by spectral clipping the red edge and separating the subsequently amplified chirped and compressed pulses with a beamsplitter.[7] The chirped pulse is used to drive a 300 ps sustained pressure shock, and to probe the shock dynamics with single shot ultrafast dynamic ellipsometry.[9,10] The compressed pulse is used to generate a supercontinuum for transient absorption spectroscopy. A schematic of our shock experiment, typical sample configuration, and cross correlation frequency resolved optical gating (X-FROG) measurement of the chirped pulse is shown in Figure 1. Ultrafast dynamic ellipsometry measures the shock and particle velocities, as well as refractive index changes that occur due to shock loading. Shock compression of transparent materials leads to partial reflection off the shock front that influences the signatures seen in both the ellipsometry and the spectroscopy. We analyze these signatures with time dependent thin film equations[9,10,11] to obtain quantitative information about the shocked material dynamics on time scales of 0-300 ps.

3. Results

Transient absorption spectra are recorded in single shot mode, as shown in Fig. 2 for a sample of polymethylmethacrylate (PMMA). The results shown are typical of shock-compressed samples in that large oscillations in reflectivity versus wavelength occur at any given time, due to the partial reflection from the shock front. Fitting these data together with the ultrafast dynamic ellipsometry data allows a complete picture of the motion of the shock, the compression of the material, and increases in absorption due to changes in electronic properties. Similar results on shocked single crystals, polymers, and liquids will be discussed along with the accompanying ultrafast dynamic ellipsometry results.

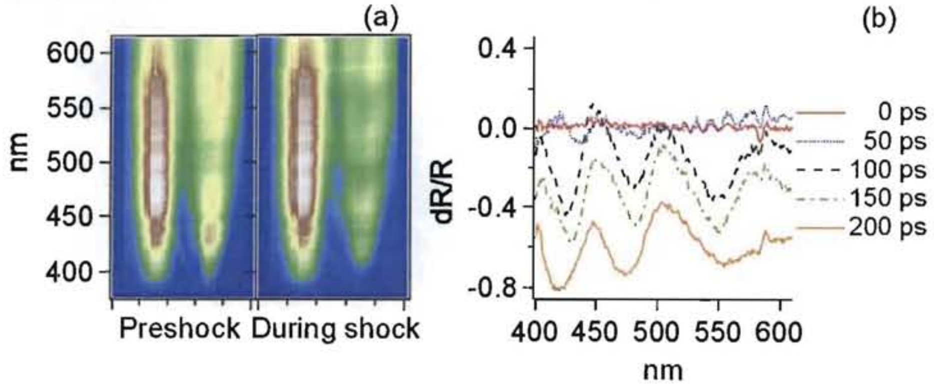

FIGURE 2. Two dimensional raw data of reference spectra (left) and sample spectra (right) are shown (a) before and 100 ps after shock loading of a sample of 1380 nm PMMA on 2000 nm of Al. A subset of the date recorded as a function of time after shock is shown in (b). The oscillations are primarily due to thin film interference from partial reflection off the shock front. An broadband decrease in reflectivity is also apparent as a function of time.

4. References

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