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FY07 LDRD Final Report Optical properties as a real-time in situ materials diagnostic at extreme conditions

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FY07 LDRD Final Report

Optical properties as a real-time *in situ* materials diagnostic at extreme conditions

LDRD Tracking Code: 05-ER-030

Jeffrey H. Nguyen, Principal Investigator

ABSTRACT

The purpose of this three-year two-directorate LDRD-ER project is to develop the use of optical properties as a diagnostic tool to obtain materials information, including inferred crystal structures, in real time from state-of-the-art experiments at the LLNL light-gas gun and other facilities that probe materials at extreme pressures and temperatures. Characterizing the state of materials at 10- to 100-picosecond time resolution is critical for dynamic experiments studying phenomena such as the time evolution of phase transitions. Not only do real-time *in situ* measurements of optical properties have the intriguing potential to address this important issue, but there is also a general paucity of realistic alternatives. During the time this LDRD was funded, we measured accurate optical quantities [e.g. complex dielectric constants $\epsilon_{1,2}(\lambda, T, P)$ and reflectivity $R(\lambda, T, P)$] in conjunction with calculations of these same quantities. This direct comparison between theory and experiment enables the association of the observed optical properties with other materials properties of interest (crystal structure, liquid-versus-solid, etc.) by establishing spectroscopic fingerprints for each crystalline phase as well as the liquid.

BACKGROUND

The development of functionally graded density impactors (02-ERD-033) opened up new capabilities for tailoring the thermodynamic path (i.e. controlling T and P

FIG. 1: Incident plane in an ellipsometric experiment. Linearly polarized incoming light is elliptically polarized upon reflection off the sample.

FIG. 2: Calculated optical spectra for aluminum at T = 0 K, 100 K, 200 K, 300 K, 400 K, 550 K, 750 K, 900 K, 950 K, 1400 K and 1800 K [6]. Above the melting temperature of aluminum (938 K), the peak near 1 eV goes away.

in real time) of materials in a gas gun experiment [1, 2]. This new capability allows us to study the dynamics of previously inaccessible materials properties such as the time-scale and hysteresis of phase transitions. Additionally, these new experiments emphasize our need to develop a capability to characterize material properties in real time. A velocimeter (such as VISAR) typically fulfills that role but it has its limits: VISAR essentially measures the change in density, but yields no identifying structural information about the material. Even though many difficult and time-consuming challenges lie ahead, the development of X-ray diffraction techniques under dynamic compression is promising [3, 4], and it should be noted that X-ray diffraction data will be *complementary* to that proposed here. Optical measurements will yield *in situ* inferred crystal structure and electronic band structure data that will enhance our understanding of materials under extreme conditions. In addition, an optical diagnostic capability will not be limited to any one experiment or experimental geometry—it will be supremely portable, and thus can be equally well utilized in high-power laser experiments at NIF, the Z-machine, and other facilities, and in dynamic compression experiments at gas guns including JASPER.

FIG. 3: Calculated broadband optical properties, $\epsilon_2(\lambda)$, for the α and ϵ phases of Fe.

RESEARCH ACTIVITIES

There are two essential components to this effort: experimental and theoretical. The experimental set-up mainly consists of a sub-nanosecond ellipsometer, capable of working with both laser as well as white-light sources [5]. The input light source is linearly-polarized, but the output light is elliptically polarized due to the optical properties of the materials under study (see Fig. 1). Spectrally-resolved data will help characterize materials properties; time-resolved data will track changes in these properties in real time. Both of these capabilities are essential in tailored dynamic compression experiments. In particular, the time-resolved capability is necessary to characterize multiple phase transitions or to measure the time scale of phase transitions themselves.

On the theoretical side, we developed the capability to calculate the optical absorption and reflectivity spectra of liquid metals and hot (partially disordered) solids. This effort involves coupling electronic structure methods with molecular dynamics simulations of high-temperature atomic structure. Prominent features in the absorption spectra of solids often involve transitions between delocalized electronic band states. Such states involve many atoms and therefore the accurate computation of these spectral features is only possible by including large numbers of atoms in the computation. The loss of spatial periodicity in a liquid or hot solid makes these calculations particularly difficult. However, it is precisely this loss of periodicity that causes otherwise sharp spectral features to broaden and change shape, giving the optical spectra the structure- and temperature-dependence we seek. (i.e. spectroscopic fingerprints, see figs. 2 & 3).

FIG. 4: Preliminary results for dielectric constants of α -Sn as it is shocked into a liquid state. Window effects are being investigated and will be used to correct the dielectric constants after shock-loading.

RESULTS AND TECHNICAL OUTCOME

We have employed this ellipsometer to measure nanosecond-resolution optical properties of Fe, Al, Bi and Sn at extreme conditions at $\lambda = 532 \text{ nm}$ (Fig. 4), and nanosecond-resolution broadband optical properties of Au and Al at ambient conditions (Fig. 6). The Fe samples were shocked to pressures exceeding 20 GPa. At these pressures, the Fe samples underwent the $\alpha \rightarrow \epsilon$ phase transition. The observed changes in the optical properties of Fe at $\lambda = 532 \text{ nm}$ are in agreement with theoretical calculations (Fig. 3). We also shock-melt Bi and Sn samples from their ambient solid states to observe the change in their optical properties upon phase transitions. The resulting changes in the dielectric constants for Bi and β -Sn (fig. 4) are consistent with calculations. Similar shock experiments on Al exhibit no change in optical properties at $\lambda = 532 \text{ nm}$. These results are consistent with calculations at that wave length (fig. 2) [6]. The aluminum experiments also provided us a *null* result that we can use to contrast with results from the Fe, Bi and Sn experiments.

In the experiments described above, we demonstrated that our miniaturized ellipsometer [5] can be used to measure the optical constants of Fe, Bi, and Sn (at $\lambda = 532 \text{ nm}$) as they transform under shock loading ($\alpha \rightarrow \epsilon$ in Fe, solid $\rightarrow$ liquid in Bi and Sn) with sub-nanosecond temporal resolution. However, the ultimate goals are to characterize the phases these materials transformed into. This requires measurement of optical properties at multiple wavelengths. To this end, we have measured the **broadband** optical properties of Au and Al with nanosecond resolution at ambient conditions (Fig. 6). In

FIG. 5: Intensity of transmitted polarized light. Shock front arrives at near $t=0$. Intensity decreases due to light-scattering off newly formed ice particles [7]. Sinusoidal variations can be attributed to pressure-induced birefringence of ice [5].

addition, we employed the single-wavelength technique on a much more complicated tailored dynamic compression experiment [1]. We have for the first time ever observed pressure-induced birefringence as water is being converted to ice VII under quasi-isentropic compression (Fig. 5). On the theoretical side, we completed calculations of optical absorption across the solid-liquid transition in Al [6], as well as the various phases of Bi and Fe (fig. 3). The deliverables that we have accomplished can be summarized as follows:

1. Real-time observation of phase transitions in metals (Fe, Bi, Sn) under shock-loading (Fig. 4).
2. Calculations of band structures of various phases of Fe (Fig. 3) and Bi.
3. Real-time observation of water solidification under quasi-isentropic compression (Fig. 5).
4. Measured broadband optical properties of Au and Al with nanosecond resolution at ambient conditions (Fig. 6).
5. Publication of calculated optical absorption across the solid-liquid transition in Al [6]. Publications of other results are being prepared.

EXIT STRATEGY

The primary deliverable associated with this LDRD is a diagnostic capability for analyzing and measuring *in situ* and real time structures of materials as they are subjected to extreme temperatures and pressures in experiments such as those at JASPER. The real-time nature

FIG. 6: (Top) Raw streak camera data for Al optical properties. Wavelength (nanosecond resolution time) in the vertical (horizontal) direction. (Bottom) Broadband optical property of Al taken from a slice of the above data. It shows a similar trend to that exhibited by calculations (Fig. 2).

of this diagnostic enables us to infer structural quantities **as they are changing**. This information is not only essential for the understanding of experiments done on very disparate time scales (from sub-ns laser shocks to minutes or hours in a diamond-anvil cell), but is crucial to developing a theory of dynamic phase transitions for incorporation into existing hydrodynamic simulation codes. We are planning to improve and port this technology to various experimental platforms including JASPER and NIF. We hope to employ the diagnostics to probe the temporal aspects of phase transitions at extreme conditions.

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