Femtosecond dynamics of the laser-induced solid-to-liquid phase transition in aluminum

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Abstract: We present femtosecond time-resolved broadband measurements of the reflectivity of aluminum during the laser-induced solid-to-liquid phase transition. Our experiments show that this transition is a thermal process, settling an existing controversy.

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The dynamical properties of the electrons and the lattice in solids define many basic properties of these materials. Excitation with intense ultrashort laser pulses is one way to induce non-equilibrium processes and observe the resulting dynamics. Laser-induced melting that results from a rise of the lattice temperature above the material's melting point, called thermal melting, can readily be observed with laser pulses of duration longer than 10 ps. After the development of femtosecond lasers, a different, non-thermal melting mechanism was reported in semiconductors [1-4]. This mechanism takes place when the ions rapidly gain kinetic energy after the laser-induced electronic excitation and rearrange themselves in a liquid-like, disordered configuration before the lattice has enough time to reach the melting temperature. Non-thermal melting is usually completed within hundreds of femtoseconds after photoexcitation.

In this paper we present an optical pump-probe study of the laser-induced solid-to-liquid phase transition in aluminum. We employ a broadband time-resolved reflectometry technique that enables the measurement of the optical properties of aluminum during the solid-to-liquid phase transition over a wide frequency range, from 1.7 to 3.5 eV, with a time resolution of 40 fs. Our findings show that the laser-induced solid-to-liquid phase transition in aluminum is a thermal process that takes place on the picosecond timescale. The observed optical properties over this broad frequency range do not exhibit a subpicosecond timescale as previously reported [5]. Our results agree with those in Ref. [6], in that the phase transition is a thermal process, mediated by heat transferred from the excited electronic population to the lattice through electron-phonon coupling.

Pump–probe experiments were performed on a 1-μm thick polycrystalline aluminum sample using 800-nm pulses from a multipass amplified Ti:sapphire laser. A white-light probe pulse (1.7 – 3.5 eV) and a CCD spectrometer were used to measure the transient reflectivity. The sample was translated between successive pump laser pulses so that each pump pulse excited an undamaged area. Two sets of reflectivity measurements were taken with the probe beam at angles of incidence of 68.4° and 58.7°. The detection system was calibrated to obtain absolute reflectivity values.

Figure 1 shows the response of the reflectivity of aluminum across a spectral range of 1.8–3.5 eV following single pulse excitation with a fluence of $3.6F_{th}$, where F_{th} = 2.1 KJ/m^2 is the lowest fluence for which we observe sample damage. The angle of incidence of the probe is 68.4°. At negative time delays (when the probe arrives before the pump pulse), the reflectivity data agree with ellipsometric measurements of the aluminum sample. At positive time delays (after the arrival of the pump pulse), we observe that the reflectivity starts to drop across all frequencies within our spectral range as the solid-to-liquid phase transition is initiated. The transition is complete after 1.9 ps, as can be seen by the darkened flat region in Fig. 1.

Figure 2 shows the average of the solid-to-liquid transition time over the frequency range of the white-light probe for all eight fluences and two angles of incidence for which we took measurements. None of the data sets exhibits a femtosecond timescale. Our results show that over the entire fluence range studied the laser-induced solid-to-liquid phase transition in aluminum takes 1.5–2 ps, confirming that the transition is thermal.

Detailed results about the transition timescale and the optical properties of liquid aluminum will be presented . We will discuss the differences between our findings and the results in the previous optical [5], and electron diffraction studies [6].

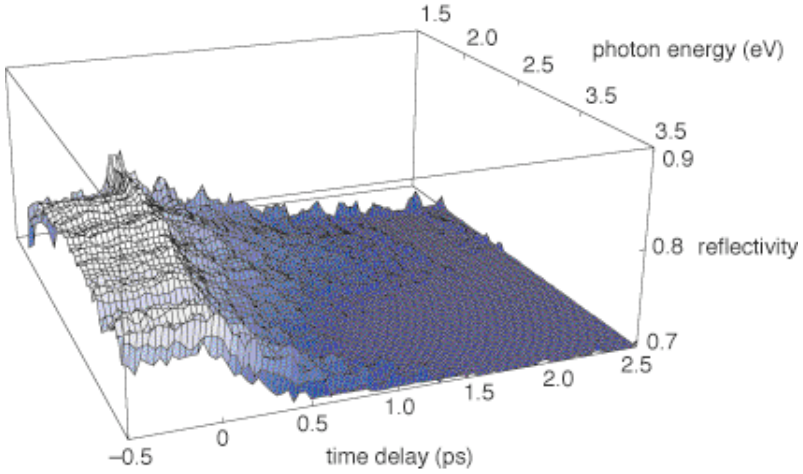

Figure 1: Time-resolved reflectivity dynamics of aluminum following single-pulse photoexcitation across the frequency range 1.8–3.7 eV. The peak absorbed fluence of the incident laser pulse is $3.6F_{th}$.

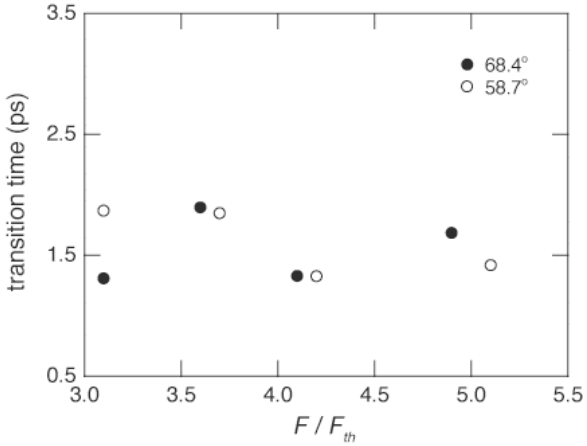

Figure 2: Average transition times extracted from reflectivity measurements similar to the ones shown in Fig. 1 for different peak absorbed excitation fluences and incidence angles of 68.4° (filled circles) and 58.7° (open circles). Transition times of reflectivity curves in the range 2–3.3 eV were averaged for each fluence.

References

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