

Turning Aluminum Liquid in Picoseconds

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The interactions between the electrons and the lattice in solids define many basic properties of these materials. Exciting the electrons in a solid with intense ultrashort laser pulses is one way to induce non-equilibrium processes, observe the resulting dynamics and obtain direct information on electron-lattice interactions. An ultrashort laser pulse can rapidly heat electrons to temperatures on the order of 10^3 K, while leaving the lattice near room temperature.

This transient two-temperature system tends to reach quasi-equilibrium within a few picoseconds via electron-phonon interactions, as well as electron diffusion out of the excited region. For sufficiently intense laser pulses, structural changes such as melting and ablation take place. Thermal melting, which results from a rise of the lattice temperature above the material's melting point, can be readily observed with picosecond laser pulses.

After the development of femtosecond lasers, a non-thermal melting mechanism was reported in semiconductors. This mechanism occurs when the laser-induced electronic excitation rearranges the positions of the ions in a liquid-like disordered configuration before the lattice reaches the melting temperature. Non-thermal melting usually occurs within hundreds of femtoseconds after photoexcitation, in contrast to the picosecond time scale of thermal melting.

The first metal for which non-thermal melting was observed was aluminum. It was reported that the material's dielectric constant at 800 nm reaches the value for liquid aluminum 500 fs after excitation of solid aluminum with a femtosecond laser pulse, a time scale that is significantly shorter than the picosecond time scale for lattice thermalization.¹

This conclusion was later called into dispute, after researchers conducted electron-diffraction experiments of optically excited, thin aluminum films and

found a 3.5-ps time scale for the solid-to-liquid phase transition, indicating that it is a thermal process.²

We recently measured the reflectivity of aluminum during the laser-induced solid-to-liquid phase transition over a broad wavelength range, from 350 to 730 nm, with a time resolution of 65 fs.³ The figure shows the response of the reflectivity at 590 nm for four laser intensities above the melting intensity I_m .

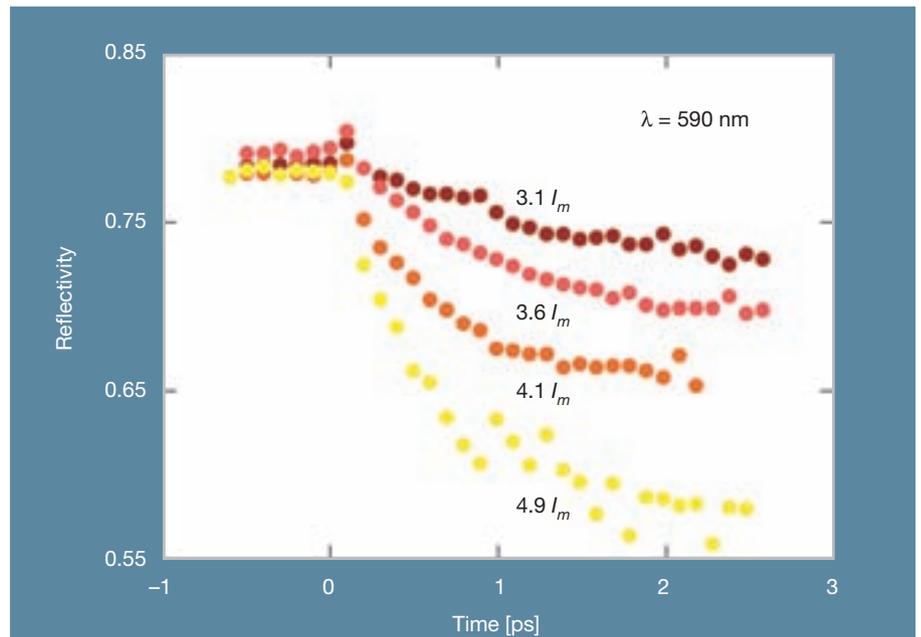
At negative times—that is, before the arrival of the excitation pulse—the reflectivity remained constant at its equilibrium value. At positive times, after the arrival of the excitation pulse, the reflectivity started to drop as the solid-to-liquid phase transition began. At all excitation intensities, it took 1.5–2 ps for the transition to be complete. We observed the duration for this transition over the entire range of wavelengths over which reflectivity was measured.

Our results unambiguously settle the argument: The phase transition in optically excited aluminum is thermal, and is mediated by heat transferred from the excited electronic population to the lattice through electron-phonon interactions. Only in semiconductors, where covalent bonding causes strong electron-lattice correlations, is it possible to observe non-thermal melting. ▲

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References

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Time-resolved reflectivity dynamics of aluminum following single-pulse photoexcitation at 590 nm for different absorbed intensities of the excitation pulse.