

Laser-induced melting of silicon observed by picosecond time-resolved x-ray absorption spectroscopy

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INTRODUCTION

Time-resolved x-ray spectroscopy of the Si L-edges is used to probe the electronic and atomic structure of silicon heated by an ultrafast laser pulse. The high temporal resolution of these measurements allows observation of the liquid phase at a stable density and temperature. The cross section at the Si L_{2,3} edge becomes dramatically reduced during a time scale of 5ps or less. Furthermore, there is a ~1.6 eV shift in the L₁ edge.

RESULTS

The melting transition from solid covalently bonded semiconductors to metallic liquids has attracted considerable attention due to the peculiar nature of the liquid phase, which has coordination numbers of 6-7 in contrast to most liquid metals that have coordination numbers in the range of 10-12. Several optical techniques have been used to investigate the laser-induced transition from solid to liquid Si due to femtosecond pulses. Although these experiments strongly suggest a fast semiconductor-to-metal transition, the short-range electronic and atomic structure of this liquid phase remains largely unknown, even for equilibrium liquid Si.

Time-resolved x-ray absorption spectroscopy is one way to obtain information about the transient liquid phase induced by a laser pulse. We performed picosecond time-resolved x-ray absorption measurements in a transmission mode on laser-irradiated amorphous Si foils. The experiments were performed at beamline 5.3.1 of the Advanced Light Source (ALS). A broad spectrum of soft x-ray light from a synchrotron bending magnet first reflects from a grazing incidence focusing mirror set to focus the beam to a 300 um diameter spot on the foil. Before reaching the foil, the beam reflects from a pair of parallel carbon-coated mirrors that serve as a low-pass photon energy filter, absorbing photons with critical angles smaller than the incidence angle. The filtered x-rays then strike the foil in the center of a 400 um diameter laser-excited region. After passing through the foil, the x-rays travel through a dispersive grating spectrometer that spatially separates the transmitted light into a spectrum ranging from 70eV to 300eV.

The resulting soft x-ray spectrum is detected in one of two ways. For temporal resolution of about 70~ps, we use a pump-probe technique with a gated set of microchannel plates (MCP's) coupled to a phosphor screen and CCD camera. The gate width of the plates is 20ns, which is sufficient to isolate a single 70ps x-ray pulse from the ALS filling pattern. By adjusting the relative timing of the laser and x-ray pulses, we obtain the spectrum as a function of time after excitation. Better temporal resolution is achieved with an ultrafast x-ray streak camera. In principle, this detector can resolve times of less than 1~ps for the entire spectrum in a single x-ray pulse.

The absorption spectrum before laser excitation agrees with previous measurements on solid Si up to a scaling factor. A large edge at 100~eV is a superposition of the L₂ and L₃ edges, spin-orbit split edges which are not separately distinguishable due to the 1~eV resolution of our spectrometer. At ~150eV the smaller L₁ edge appears, and at larger energies small EXAFS oscillations are present. Energies beyond 285eV were inaccessible due to absorption from the carbon K-edge.

The absorption spectrum taken ~100 ps after laser excitation (taken using the pump-probe techniques) shows that melting induced by the laser causes a 50% reduction in the magnitude of the L_{2,3} edge, and the edge is broadened to ~2eV. In addition the near-edge features of the L_{2,3} are modified, the L₁ is shifted to lower energies by 1.6 eV, and the EXAFS oscillations past the L1 edge seem to disappear. These features do not change significantly for longer pump-probe delays until approximately 10ns after excitation. At this time, the overall transmission of the foil begins to increase, an effect and time scale consistent with the Si liquid breaking up into droplets and leaving the probed region. The absorption changes are being compared to expectations based on model calculations.

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