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Ultradast Absorption Spectroscopy of Aluminum Plasmas Created by LCLS using Betatron X-Ray Radiation

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Ultrafast absorption spectroscopy of aluminum plasmas created by LCLS using betatron x-ray radiation

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Abstract

This document summarizes the goals and accomplishments of a six month-long LDRD project, awarded through the LLNL director Early and Mid Career Recognition (EMCR) program. This project allowed us to support beamtime awarded at the Matter under Extreme Conditions (MEC) end station of the Linac Coherent Light Source (LCLS). The goal of the experiment was to heat metallic samples with the bright x-rays from the LCLS free electron laser. Then, we studied how they relaxed back to equilibrium by probing them with ultrafast x-ray absorption spectroscopy using laser-based betatron radiation.

Our work enabled large collaborations between LLNL, SLAC, LBNL, and institutions in France and in the UK, while providing training to undergraduate and graduate students during the experiment. Following this LDRD project, the PI was awarded a 5-year DOE early career research grant to further develop applications of laser-driven x-ray sources for high energy density science experiments and warm dense matter states.

Background and Research Objectives

The nascent field of High Energy Density (HED) science aims to resolve grand challenges to understand how the Sun works, how the planets were formed, how to harness fusion energy, or how to explain the mechanisms of stellar explosions. It is now possible to recreate astrophysical conditions of extreme temperatures and pressures at large scale laser and x-ray free electron laser (XFEL) facilities such as the Linac Coherent Light Source (LCLS). However, such conditions are transient in nature and can be extremely difficult to probe. Phenomena such as shock physics, opacity, ultra-relativistic laser-matter interactions and x-ray-matter interactions are central to this field of physics, but their investigation often requires diagnostics that are massive, costly and limited in temporal or spatial resolution. Laser-wakefield accelerators (LWFAs), relying on intense laser fields to drive plasma waves and subsequently accelerate particles, can produce bright x-ray sources at a fraction of the cost but with a 1000-fold increase in time resolution, and they have not yet been fully exploited for applications in HED science, especially at large-scale facilities. Betatron x-ray radiation, a broadband, femtosecond, collimated x-ray source produced when relativistic electrons oscillate in a LWFA, has been successfully developed at LLNL with the support of a previous LDRD project (13-LW-076) [1].

The goal of the present LDRD project (a few months in FY16) was to support beam time at the LCLS Matter under Extreme Conditions (MEC) end station. The goal of the experiment was to use betatron x-ray radiation to probe an aluminum plasma heated by the x-ray free electron laser, and to study the ultrafast electronic transitions of a solid density aluminum plasma by means of x-ray absorption spectroscopy. The plasma can be produced with the LCLS x-ray beam at the MEC end station via K-shell photo-absorption and probed with laser-based betatron x-ray radiation. Because of its unique colocation with a multi-TW, femtosecond laser system, the MEC end station at LCLS is currently the only facility capable of hosting this experiment.

Our work demonstrates a new diagnostic capability to measure the transient charge state of materials with a high temporal accuracy. We obtained femtosecond time-resolved x-ray absorption spectra of heated matter at well-defined density and temperature conditions, giving access to its electron temperature, ion temperature, and charge state as the sample relaxes back to equilibrium. The plan was to use ultrafast single shot x-ray-absorption near-edge spectroscopy (XANES) around the Al K-edge (1.56 keV) to probe the electronic structure modifications of aluminum at conditions of density (~1X solid density) and temperature (1-5 eV) uniquely accessible at the LCLS.

Due to lower than expected performance of the short pulse optical laser at the LCLS-MEC end station, the betatron x-ray source did not produce sufficient x-ray photons near the Al K-edge (1.56 keV) for us to perform the desired experiment. Instead, we performed our measurements using the same technique, at the iron L-edge (0.707 eV), on iron heated by LCLS to a few eV temperatures. With proper data accumulation and acquisition, we were able to demonstrate the possibility of using betatron x-ray radiation for ultrafast absorption spectroscopy of plasmas created by LCLS.

Scientific Approach and Accomplishments

The experiment we conducted at LCLS MEC is presented in Figure 1. The LCLS x-rays (7.5 keV, 3 mJ, 70 fs) are focused onto a solid iron foil (100-300 nm) to bring it to temperatures of a few eV. Simultaneously, the MEC short laser pulse (1 J, 40 fs) is focused onto a gas cell to accelerate electrons and produce betatron x-rays. The radiation is collected by a 3.4 degrees grazing incidence ellipsoidal mirror (multilayer coating) and focused onto the iron samples. The spectrum of the betatron x-ray beam transmitted through the iron foils is analyzed with an imaging grating spectrometer, and compared with a reference spectrum.

The technique used to analyze evolution of the iron sample temperature is X-ray Absorption Near Edge Structure (XANES). At room temperature, a metal, such as iron, exhibits sharp absorption edges. For example, the iron K-edge (7.1 keV), corresponds to a 1s-4p transition, while the iron L-edge corresponds to a 2p-3d transition. Iron is a transition metal, with a partially filled 3d band. When it is heated to warm dense matter states (a few eV), electrons in the 3d band are excited above the Fermi level, which changes the occupation states, and therefore the slope of the L-edge absorption spectrum. By measuring the slope of the iron L-edge with XANES, it is possible to access the sample temperature.

Simulations, performed by T. Ogitsu at LLNL (Figure 2), show the sensitivity of the XANES spectrum to electron temperature T_e at the iron L-edge. These calculations use a theoretical model derived from first- principles density function theory and local density approximation (DFT-LDA). This code can calculate the fine structure near the edge with high accuracy in the XANES region. It first calculates the absorption spectrum at room temperature: the Fermi distribution for 300 K is applied to the solid x-ray absorption cross section. For the initial state (before LCLS heating), a face-centered cubic solid at 300 K is assumed. Following the laser pulse, liquid atomic configurations are used, with electron temperatures up to a few eV and solid densities. Data that have been obtained at the ALS synchrotron at the L-edge of warm dense copper [2] and warm dense iron [3] (temperature ~ 1 eV, optical laser heating, 2 ps temporal resolution) are showing good agreement with these calculations.

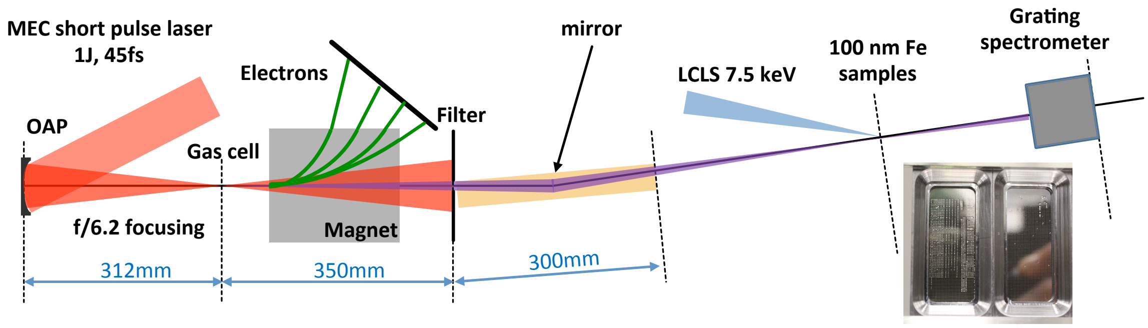

Figure 1. LCLS experimental setup, showing the betatron x-ray source, produced by a short (1J, 40 fs) laser pulse focused onto a gas cell and refocused on the iron samples with an ellipsoidal mirror. Transmitted x-rays through the heated sample are analyzed with a grating spectrometer.

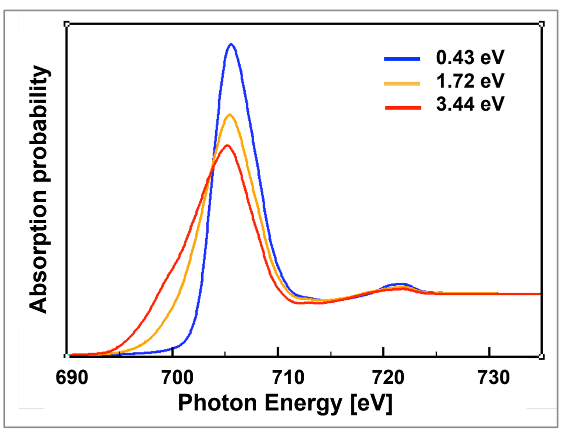

Figure 2. DFT-MD simulations of the Fe-Ledge absorption spectrum as a function of the sample temperature (simulations courtesy of T. Ogitsu, LLNL).

Impact on Mission

Our projects and its results are well aligned with LLNL's missions. This novel combination of an x-ray-matter interaction experiment with a femtosecond x-ray probe from laser-produced plasmas will elucidate the mechanisms of ion-electron equilibration and energy transport in HED plasmas, which is relevant to the LLNL Inertial Fusion Science and Technology mission focus area. The physics of developing laser-driven light sources and using them for applications is also directly tied to two LLNL core

competencies: HED science and Lasers and Optical Science and Technology.
The high visibility and success of this work resulted in a prestigious DOE early career award ($ 2.5 million over 5 years) being awarded to the PI. This project, supported by the DOE office of fusion energy sciences, will seek to develop more applications of laser-driven betatron x-rays for high energy density science experiments and warm dense matter states. Graduate students and postdocs participated to the experiment, and several of them are also going to work for the LLNL HED programs and missions.

Conclusion

In conclusion, we demonstrated a new diagnostic capability of warm dense matter states at LCLS. The warm dense iron was produced with the LCLS x-ray beam at the MEC end station via K-shell photo-absorption and probed with laser-based betatron x-ray radiation. We used ultrafast single shot x-ray-absorption near-edge structure (XANES) around the iron L-edge (0.707 keV) to probe the electronic structure modifications of iron at conditions of density (~1X solid density) and temperature (1-5 eV) uniquely accessible at the LCLS. This novel combination of an x-ray-matter interaction experiment with a femtosecond x-ray probe from laser-produced plasmas will open up new possibilities to inform and design future high x-ray intensity experiments and to improve current opacity and radiation transport models that are essential for high energy density (HED) science.

Our work enabled large collaborations between LLNL, SLAC, LBNL and institutions in France (CELIA, LULI, LOA) and in the UK (Imperial College London), while providing training to undergraduate and graduate students during the experiment. Following this LDRD project, the PI was awarded a 5-year DOE early career research grant ($ 2.5 Million over 5 years) to further develop applications of laser-driven x-ray sources for high energy density science experiments and warm dense matter states [4,5]. As a result, we expect to continue similar efforts at LCLS by making the betatron x-ray source more robust and by increasing its yield and photon energy to access other materials and conditions.

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