Laser and Timing Electronics for Time-resolved XAFS Measurements of Laser-melted Germanium

D. L. Brewe,[1] E. A. Stern,[1] K. M. Beck,[2] S. M. Heald,[2] Y. Feng[2]
[1] University of Washington, Seattle, WA, U.S.A.
[2] Pacific Northwest National Laboratory, Richland, WA, U.S.A

Introduction

Understanding the kinetics of melting and phase transformations is fundamental to progress in the design and manufacture of solid-state devices. The melting of Ge is particularly interesting because of its complex behavior. For example, the density of the liquid is greater (by 4.7%) and has a larger number of nearest neighbors (six vs. four). Previous investigations [1-6] have probed the melting kinetics with Bragg diffraction, which is insensitive to changes after long-range order disappears. X-ray absorption fine structure (XAFS) spectroscopy, however, does not require long-range order and can therefore monitor local structural changes in the disordered (i.e., liquid) phase directly. With this in mind, we've developed time-resolved XAFS capability on the picosecond timescale for investigating the melting kinetics of Ge by using a femtosecond laser to melt the Ge. The apparatus is also well-suited for other types of pump-probe experiments, and it utilizes a laser firing at the APS P0 clock rate (~ 272 kHz), which is about two orders of magnitude higher than that of typical laser systems used in time-resolved XAFS experiments on this timescale. This allows us to collect data at a rate similar to that of typical non-time-resolved experiments. This report describes the laser and timing system and the initial XAFS results from laser-heated Ge.

Methods and Materials

The Ti:sapphire laser produces ~ 250-fs pulses with ~ 4 uJ/pulse at 800-nm wavelength. It is capable of a repetition rate equal to the 272-kHz P0 clock rate at the APS, allowing us to collect data for a given bunch each time around the ring. Synchronization of the laser to the x-rays from the ring is accomplished by phase-locking the mode-locked seed laser (Mira 900F) to an 88-Hz signal derived from the ring RF signal and triggering the main laser amplifier (RegA 9000) with the P0 signal. X-ray pulses are detected by an APD (I0) and a plastic detector (sample fluorescence), and pulses from the bunch with the desired timing are selected by passing the detector outputs through analog gates, also triggered by P0. Relative timing is changed by adjusting the phase of the 88-Hz reference signal and/or advancing the RegA trigger to select different seed pulses from the Mira for amplification.

The results shown below were obtained in pulse-counting mode by using fast discriminators for the pulses passing through the gates. We are currently implementing a system that uses lock-in detection of the gated detector pulses, which will allow us to increase our signal significantly, because we won't be limited by the saturation effects produced by pulse-pileup in pulse-counting mode. The timing resolution is limited by the x-ray bunch width and the accumulated jitter of the electronics components. The effective timing resolution is estimated to be on the order of 100-200 ps. A block diagram of the electronics and laser system is shown in Fig. 1.

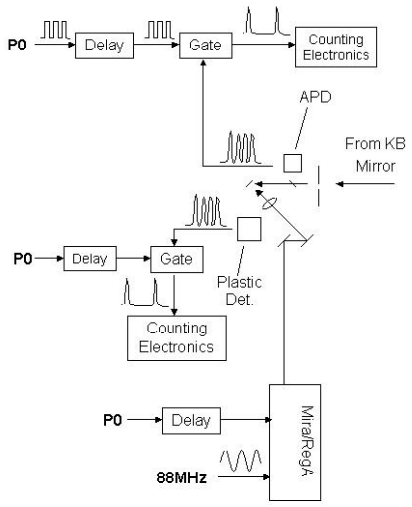

FIG. 1. Schematic of the timing electronics and laser system.

The Ge sample consisted of a self-supported 200-nm film, produced by depositing the film on a silicon substrate and etching the substrate from the back of the film. Initially the film was amorphous, but it crystallized during initial heating with the laser. The incident x-ray beam was focused with KB mirrors to a spot on the sample approximately 10 μm in diameter. The laser was initially focused to a spot on the sample approximately 50 μm in diameter, but because of alignment problems due to imprecise actuators on the mirror that steers the laser beam onto the sample, the laser was defocused to a size several times larger to ensure that the x-ray beam was

probing a portion of the sample that was heated by the laser.

Results

We were able to obtain good-quality time-resolved XAFS data from the Ge sample heated by the laser to approximately 700K. The k^2 fourier transform of the EXAFS from the sample at 700K is shown in Fig. 2. The temperature was estimated by analyzing the data to obtain the mean-square relative displacement and applying the Einstein model, which has been shown to represent the temperature dependence of Ge EXAFS well [7]. The temperature estimate is also in agreement with a recent experimental study of Ge at high temperatures [8].

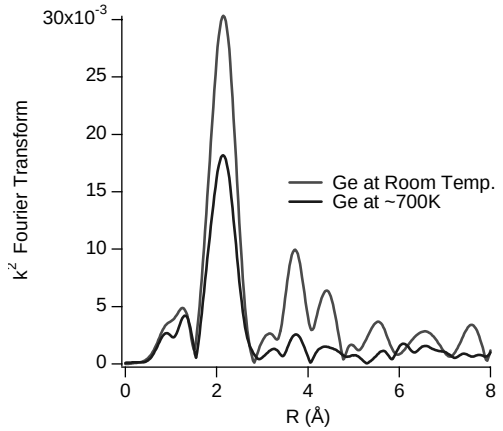

FIG. 2. Fourier transform of Ge at 700K and room temperature.

The sample was damaged when attempts were made to heat it much above 700K, presumably because of thermal stresses. This effect was most likely exacerbated by the necessity of increasing the total laser power incident on the sample in the defocused beam that was used to make up for the lack of precision in aligning the laser beam. We are in the process of upgrading the mirror actuators, and this should improve the alignment precision and lower the total laser power necessary to heat the sample. However, it's also possible that the self-supported film will not withstand the stresses at higher temperatures, and we are fabricating samples with substrates that don't alloy with Ge, as Si does.

The sample was also somewhat oxidized at higher temperatures, which is indicated by Fig. 3, which shows the k^2 EXAFS before and after heating the sample. A change in the EXAFS at lower k values indicates the presence of lower-Z neighbors, most likely oxygen. A change in the XANES (not shown) was also consistent

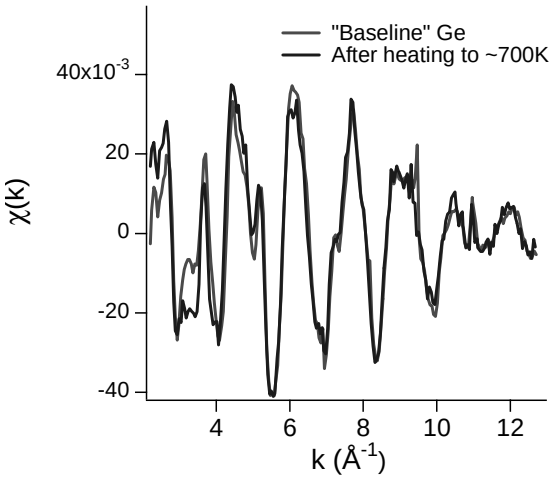

FIG. 3. The k^2 EXAFS of Ge before and after heating to 700K.

with the presence of a small amount of GeO_2. A sample chamber to surround the sample with a reducing atmosphere is under construction.

Figure 3 also serves to illustrate the performance of the overall system. The data in the figure were collected in approximately 1-1/2 h. It should be pointed out that even though the sample was pure Ge, it was quite thin, with an absorption step of ~ 0.02. The total signal was also reduced by filters to remain within the photon-counting regime. The use of lock-in detection in the next iteration of the apparatus will remove this restriction and allow further improvement in data collection efficiency.

Discussion

We've developed an apparatus for obtaining time-resolved XAFS with high efficiency for use in laser pump-probe experiments on a picosecond time scale by using a high-repetition-rate laser to increase the effective flux by more than two orders of magnitude over typical pulsed laser systems used for time-resolved x-ray experiments. We've used the apparatus to obtain time-resolved EXAFS of laser-heated Ge. Data quality is quite good for data collection times similar to non-time-resolved experiments.

Acknowledgments

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