

# Ultrafast Soft X-ray Absorption Spectroscopy

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The development of reliable femtosecond solid-state lasers brought new possibilities into time-resolved spectroscopy to monitor the nuclear motion of molecules, crystal lattices and other out-of-equilibrium structures. However, usually it is very difficult to map the experimental observations to the structural dynamics. Therefore, experimental approaches are needed that can overcome the limitation of optical studies for structural determination, while the high temporal resolution of femtosecond lasers is maintained. Structural techniques such as X-ray diffraction (XRD) or X-ray absorption spectroscopy (XAS) deliver much more direct information about the structure<sup>1</sup>. In this contribution we report on the generation of soft x-ray pulses via high harmonic generation (HHG) and their first use for time resolved XAS to investigate the structural dynamic of amorphous silicon with a temporal resolution of about 20fs. To our knowledge this is highest temporal resolution ever demonstrated in XAS.

HHG sources have been limited so far to the 100eV range, but recently we were able to demonstrate an extension of the cut-off to nearly 1keV<sup>2</sup>. Using very short driving laser pulses, the spectrum becomes continuous near the cut off and will be ideally suited for absorption spectroscopy. Due to the coherent generation process, the XUV pulses are always shorter than the driving laser pulses. The output of our CPA Ti:sapphire laser (1mJ, 20fs, rep rate 1 kHz) has been focused into a Ne gas jet. The generated XUV radiation hits a 100nm thick silicon film, consisting of randomly oriented micro-crystallites (amorphous silicon a-Si). The XUV signal has been safely above the noise level up to energies of about 500eV opening the way to EXAFS (extended x-ray absorption fine structure). A small fraction of the laser beam is split off and pumps the sample at fluences which are nearly two orders of magnitude below the damage threshold.

The IR pump pulse modifies the conduction and valence band state density in the a-Si, via single and two-photon absorption. The dynamic evolution of the density of states can be monitored by recording changes in the fine-structure of the soft-x-ray absorption spectrum around the L-edge. We varied the delay in a range between -300fs and 100ps and identified a fast and a slow time constant. To follow structural changes it is much more convenient to measure the absorption far above the edge, which is known in the literature as EXAFS. In a second set of measurements we recorded spectra in a range of 150 to 300eV for delays of -300 to 800 fs in 20fs steps. From these data we estimate the atomic distance and found a variation with delay. At a fixed energy a Fourier-transform of the time series can be interpreted as phonon spectrum. The obtained spectrum is in reasonable agreement with measured phonon spectra of a-Si reported in the literature.

We demonstrated time resolved x-ray absorption spectroscopy with a resolution in the sub-20 fs range around the L- edge (100 eV) of amorphous silicon (XANES) and gathered information beyond the L-edge about the atomic structure with EXAFS. Our setup can be easily adapted for other materials such as Carbon, and open the way to gain insight into fast dynamical processes of large organic molecules.

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<sup>1</sup> C. Bressler, M. Chergui, Chem. Rev. **104** 1781-1812 (2004)

<sup>2</sup> E. Seres, J. Seres, K. Krausz, Ch. Spielmann Phys. Rev. Lett. **92** 163002, 2004