Time-resolved x-ray absorption spectroscopy using high harmonic radiation

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The development of reliable femtosecond solid-state laser brought new possibilities into time-resolved spectroscopy. For the first time it became possible, in principle 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, X-ray absorption spectroscopy (XAS), or X-ray photoelectron spectroscopy deliver much more direct information about the structure. The key to the successful realization was the development of laser driven x-ray sources. In this contribution we report on the optimized generation of soft x-ray pulses via high harmonic generation. We will show that we are able to generate broad continuous spectra ideally suited for XAS. High-order harmonic (HH) generation is a suitable way to generate coherent, well collimated electromagnetic radiation in the extreme ultraviolet and soft-x-ray region [1]. However, the observed low conversion efficiency, especially at higher photon energies is a serious limitation for several applications. Applying non-adiabatic self-phase-matching, quasi-phase-matching [2], and x-ray paramteric amplifcation[3] overcomes these limitations. They allowed the generation of coherent radiation up to few keV's [2] with suitable photon flux for the spectroscopic applications. The XUV signal has been intense enough in an energy range up to 3500 eV opening the way to EXAFS (extended x-ray absorption fine structure). In a first proof-of principle experiment we followed structural changes in Silicon after excitation with an intense laser pulses probed above the L and K absorption edge. The modulation in the EXAFS spectrum is in reasonable agreement with measured phonon spectra of amorphous-Si reported in the literature[4].

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