High harmonic generation for time-resolved photoelectron spectroscopy

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In this contribution we will present our approach to elucidate the evolution of the electronic structure of matter during ultrafast processes such as phase transitions and chemical reactions. We aim at mapping the valence electron rearrangements in real time as atoms are moving. Valence band photoelectron spectroscopy is probably the most direct way to map the valence electronic structure. In order to follow the electronic structure evolution on the femtosecond time scale of atomic motion ultrashort vacuum ultra violet (VUV) pulses are needed for time-resolved valence band photoelectron spectroscopy. We use high-order harmonics of a femtosecond laser as generated in a gas cell [1] or capillary [2] to produce up to 10^{10} photons/s at a photon energy of 23 eV.

With a stable and reliable source for femtosecond time-resolved spectroscopy in mind, different ways of generation, monochromatization, focussing and flux determination of femtosecond VUV pulses will be discussed. We use diffraction gratings with grazing incidence reflection mirrors [1], transmission zone plates [2] or reflection zone plates to monochromatize and focus the radiation. The flux is determined on an absolute level with calibrated detectors. Assets and drawbacks of the different approaches will be discussed in the light of their application for ultrafast surface dynamics.

As an example for the application of this approach we present the ultrafast dissociation of Br_2 molecules in the gas phase [3]. A laser pump pulse initiates dissociation and, for the first time, the entire occupied valence electronic structure is followed from the excited molecule to the free atoms.

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