Thesis SPAM

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L'Orme des Merisiers - Amphithéâtre Claude Bloch (bât. 774)

«Generation and Application of Attosecond Pulses»

To capture electronic rearrangements inside a molecule or during chemical reactions, attosecond (as, 1 as =10–18 s) time resolution is needed. To create a light pulse with this duration, the central frequency has to be in the XUV range and cover several tens of eVs. Moreover, the frequency components have to be synchronized. The so called High Harmonic Generation (HHG) in gases fulfills these requirements. During this process, a high intensity laser pulse is focused in a gas jet, where its electric field bends the potential barrier of an atom/molecule allowing an electron wave packet (EWP) to tunnel ionize. Following the electric field of the laser, the EWP gets accelerated, gaining a large kinetic energy that may be released as an attosecond burst of XUV photons in the event of a re-collision with the ionic core. This recolliding EWP probes the structure and dynamics of the core and encodes the information in the attosecond emission. More precisely, this "self-probing" scheme gives access to the complex valued recombination dipole moment (RDM) of the molecule, that encodes the electronic structure and dynamics, and to the nuclear autocorrelation function, that encodes the nuclear dynamics. The advanced characterization of the spectral amplitude, phase and polarization of the harmonic radiation thus provides a big amount of information. In principle, it is even possible through a tomographic procedure to reconstruct the radiating molecular orbital.

The objective of my thesis was two-fold. First, by implementing advanced characterization techniques of the harmonic amplitude, phase and polarization, we studied the electronic structure of N2 and the laser-induced multi-channel tunnel ionization. We performed the tomographic reconstruction of molecular orbitals and revealed the ionization channel-dependent ultrafast nuclear dynamics. Second, we studied the reflectivity and dispersion of recently designed chirped XUV mirrors that can shape the temporal profile of attosecond pulses. With these mirrors, we could control the spectral phase over 20 eV and compensate the GDD of the harmonic emission or introduce a TOD. We also proposed a novel attosecond pulse shaper.

Everyone is welcomed on the buffet after the defense