Dr Wolfi Helml

Physics Dept, Technical University of Munich, DE

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Towards attosecond X-ray experiments at free-electron lasers

With the advent of ultrashort laser pulses in the femto- and recently also in the attosecond range via high harmonic generation (HHG), lab-based XUV and soft X-ray laser sources promise a novel way for the investigation of dynamics on the natural time-scale of the motion of electrons. On the other hand, free-electron lasers (FELs) have heralded the regime of high-intensity coherent X-ray radiation for single-shot studies on non-reproducible systems, time-resolved measurements on interactions involving inner-shell electronic states and X-ray diffraction and imaging experiments. Bringing together ideas from these two exciting new scientific fields allows to combine the attosecond temporal resolution and pulse control of optical lasers with the high photon flux and versatility of FELs.

In this talk, I will give a quick overview of our attempts to increase the photon energy [1] and intensity [2] of table-top HHG sources at the Max-Planck-Institute of Quantum Optics (MPQ) and Technische Universität München (TUM) in Garching, Germany. Then, I will present photoelectron streaking spectroscopy [3,4] at an FEL as a promising route to further bridge the gap between both worlds of X-ray science. In 2014, we demonstrated a non-invasive, single-shot measurement of the X-ray pulse duration at the Linac Coherent Light Source (LCLS) [5] at Stanford in California, USA. This scheme is independent of photon energy, decoupled from machine parameters and provides an upper bound on the shortest FEL X-ray pulses available today, determining their duration to be on average no longer than 4.4 fs. Analysing the pulse sub-structure indicates a small percentage of the FEL pulses consisting of individual high-intensity spikes to be on the order of hundreds of attoseconds [6]. Recently, we developed a completely novel and extremely powerful approach for the characterization of the temporal and spectral FEL pulse structure—that is the application of the attoclock technique [7,8] to the field of SASE free-electron lasers. The method of angular-resolved streaking [3,7] relies on exciting photoelectrons in a gaseous target with the FEL pulse in the presence of a near-circularly polarized infrared (IR) field and angle-resolved detection of the dressed photoelectrons witha multi-time-of-flight (TOF) spectrometer. This instrument consists of a ring-like array of 16 TOF detectors arranged in a circle perpendicular to the propagation direction of the FEL and the co-linear streaking field [9].
For the first time, we were able to measure 16 simultaneous streaking spectra at the LCLS with various settings of the FEL. The experimental results provide direct information about the full time–energy distribution of the X-ray pulses with attosecond resolution on a single-shot basis, including X-ray pulse duration, intensity substructure and chirp. At the same time, the relative strength of the streaking effect gives information about the arrival time shift of the pulses with respect to the laboratory frame from shot to shot. As a side effect, we are also monitoring the elliptically polarized IR field (phase and amplitude) for randomly changing phase values in single snap-shots, from which the full 3D distribution of the streaking laser field can in principle be reconstructed.
These findings can assist the development of envisioned attosecond FEL pulse shaping [e.g. 8] and will pave the way to ultrashort-pulse X-ray laser systems and accompanying pump/probe experiments.
References

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