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Towards attosecond streaking at X-ray free-electron lasers

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Since the generation of the first sub-femtosecond extreme-ultraviolet (XUV) pulses in 2001 [1], the frontiers of ultrafast science have been rapidly expanded by attosecond time-resolved techniques. Notably, attosecond streaking spectroscopy has been used to reconstruct the electric field of light pulses, clock core-hole decay lifetimes, and even measure photoemission delays as short as tens of attoseconds [2-5]. Whilst the majority of these measurements have been made using table-top extreme-ultraviolet (XUV) sources based on laser-driven high-harmonic generation (HHG), there have been contemporaneous advances in large-scale sources of X-ray radiation. Across the natural sciences, X-ray free-electron lasers (XFELs) have become established tools for pioneering experiments that exploit their intense, ultrafast X-ray pulses. In addition to the extreme brilliance and high photon energies accessible at these large-scale facilities, XFELs are now able to generate X-ray pulses with attosecond duration [6]. This could herald a new era in ultrafast science, with experimentalists able to simultaneously capitalise upon the extreme time resolution offered by attosecond pulses without the restrictions on photon energy tunability suffered by HHG sources.

However, applying established attosecond streaking techniques at XFELs is challenging. Unlike XUV sources, XFELs cannot inherently synchronise their pulses with those of an external probe laser, leading to timing jitter on the scale of tens or hundreds of femtoseconds. Furthermore, most XFEL facilities do not currently have access to carrier-envelope-phase-stable laser pulses, which are typically a prerequisite for attosecond streaking spectroscopy. Despite the increasing availability of attosecond XFEL pulses, these two challenges have generally kept attosecond-resolution experiments out of reach for XFELs.

Here, we present a novel technique to overcome these limitations and unlock the full potential of attosecond XFEL pulses for ultrafast science. Dubbed 'self-referenced attosecond streaking', the method takes advantage of inherent fixed properties of the experimental sample or geometry, using these to reconstruct timing and phase information. In this way, the technique sidesteps the usual strict timing and phase requirements for attosecond streaking spectroscopy. It is anticipated that this will render a hitherto challenging class of experiments –those that demand extreme time resolution and short-wavelength X-rays –to be realised at XFELs.

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