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Phase cycling and coherent spectroscopy in the XUV domain

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In the visible spectral range, coherent nonlinear spectroscopy is an important concept for the real-time study of ultrafast dynamics in complex quantum systems [1]. Likewise, theorists have suggested numerous applications in the XUV and X-ray domain [2]. However, corresponding experiments have been hindered by severe technical challenges involved with XUV/X-ray optics. These include the demand for sub-cycle phase stability and background-free detection of weak nonlinear signals based on phase matching/phase cycling schemes.
We present a new concept facilitating XUV/soft X-Ray interferometry combined with phase cycling. The method relies on precise manipulation of the timing and phase properties of the fundamental laser field driving harmonic generation. This enables interferometric XUV-pump, XUV-probe experiments with high sensitivity while omitting any modifications of the XUV beamline. The approach is demonstrated for high harmonic generation (HHG) in a gas cell [3] and at a seeded free-electron laser (FEL) [4]. In the tabletop HHG experiment, narrow-bandwidth harmonics around 14 eV in argon and krypton are characterized with high resolution through fringe-resolved linear XUV autocorrelation measurements [3]. At the seeded FEL, XUV wave packet interferometry is performed to probe the coherence decay of an atomic inner-valence excitation (28eV) in real-time [4]. Furthermore, Interatomic Coulombic Decay (ICD) dynamics in the HeNe dimer are studied. The combination of wave packet interferometry and phase cycling opens up a palette of methodologies in a single experiment, ranging from high-resolution absorption spectroscopy to time-resolved photoelectron spectroscopy and multidimensional coherent methods.

References:
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