HU1100141

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Direct interferometric measurement of the atomic dipole phase in high-order harmonic generation

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For low gas densities and negligible ionization, the so-called atomic dipole phase, connected with the electronic dynamics involved in the generation process, is the main source of phase modulation and incoherence of high-order harmonics. To accurately determine these laser-intensity-induced phase shifts is therefore of great importance, both for the possible spectroscopic applications of harmonics and for the controlled generation of attosecond pulses.

In a semiclassical description, only two electronic trajectories contribute to generate plateau harmonics during each pump optical half-cycle. Electrons appearing in the continuum by tunnel ionization may follow two different quantum paths, namely a *long* (l) and a *short* (s) trajectory before recombination. According to the SFA approximation, the harmonic of q^{th} order acquires a phase proportional to the electronic classical action, and simply given by:

$$\varphi^j_q(r,t) = -\alpha^j_q I(r,t) \qquad \text{with} \qquad j = l, s$$

where α^j_q are non-linear phase coefficients, roughly proportional to the time that the originating electron spends in the continuum before recombination. The space and time variation of the laser intensity $I(r,t)$, causes just a little phase modulation for the s-trajectory harmonic component, while the l-trajectory component becomes strongly chirped and spatially defocused; this gives rise to two spatially-separated regions having different temporal coherence.

Here we report the first direct measurement of such atomic dipole phase in the process of high-order harmonic generation. Differently from previous measurements based on the analysis of the frequency chirp of harmonic pulses, here phase shifts are measured in the most natural way, i.e., by interferometry. Two phase-locked pump pulses generate two phase-locked harmonic pulses in two nearby positions in a gas jet; one of them is used as a fixed phase reference while the generating intensity of the other is varied. The shift of the XUV interference fringes observed in the far field then gives a direct estimate of the intensity-dependent dipole phase. Besides being a conceptually much simpler kind of measurement, our approach has the important advantage of being able to clearly discriminate between the contributions of the two different quantum paths leading to harmonic emission.

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