Probing electronic wavefunctions by all-optical attosecond interferometry

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Attosecond spectroscopy enables real-time observations of electron dynamics on their natural time scale (1 as = 10^{-18} s) [1]. It is based on steering electron motion by the electric field waveform of strong laser pulses. High-harmonic generation (HHG), the mechanism underlying the production of attosecond pulses in the extreme ultraviolet (XUV) provides an built-in spectroscopic pump-probe measurement with extremely high spatial and temporal accuracy. Here, under the influence of a strong laser field an electron is liberated from an atom by tunneling ionization, propagates in the laser field and is driven back to the parent ion. Recollision and recombination of the electron with the ion leads to the emission of photons in the extreme ultraviolet (XUV) regime. The amplitude and phase of the emitted XUV radiation encodes all parts of the light-matter interaction in the recollision process, enabling attosecond self-probing spectroscopy.

Here we present two applications of attosecond self-probing spectroscopy. In the first study [2], we initiate HHG with an XUV pulse instead of tunneling ionization, enabling us to measure and control the XUV photo-ionization dynamics in the presence of a strong infrared (IR) field in amplitude and phase. The existence of multiple quantum paths leading to the same final state within XUV-initated HHG naturally leads to quantum interference; this *in-situ* interferometer is controlled by the temporal delay between the IR field and the XUV pulse, enabling us to fully reconstruct the ionization process in the dressed atomic system. Our scheme opens the door to measurements of inner-shell multielectron dynamics by the self-probing scheme.

In the our study [3], we implement an *ex-situ* XUV interferometer in order to establish a powerful alternative to photoelectron spectroscopy. In the latter, the ionized electron wavefunction carries information on the structure of the bound orbital, the ionic potential as well as the photo-ionization dynamics itself. While photoelectron spectroscopy resolves the absolute amplitude of the wavefunction, retrieving the spectral phase information has been a long-standing challenge. Here, we transfer the electron phase retrieval problem into an optical one by measuring the time-reversed process of photoionization – photorecombination – in HHG (Fig. 1ab).

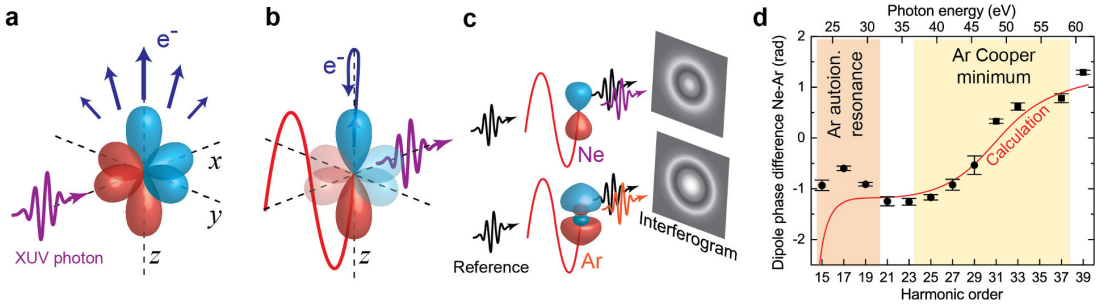

Fig. 1: a, Photoelectron spectroscopy. The emitted electron wavefunction carries the full spectroscopic information, but integrates over all angles and initial orbitals. **b**, Self-probing spectroscopy. IR-driven tunneling ionization selects one initial orbital; the returning electron is recombining through a single scattering angle and transfers its phase to an XUV photon. **c**, *Ex-situ* XUV interferometer. Interfering a reference XUV field (black) with XUV fields from either Ar or Ne leads to an interferogram, revealing their relative phases. **d**, Recombination phase difference of Ne and Ar with phase signatures of an autoionizing resonance and the Cooper minimum.

Here, we demonstrate all-optical interferometry of two independent phase-locked attosecond light sources and measure the difference in the recombination phase of different atomic species (Fig. 1c). Our scheme enables us to directly determine the scattering phase shift in simple quantum systems such as helium and neon, over a large energy range (Fig. 1d). In addition, the strong-field nature of attosecond pulse generation resolves the dipole phase around the Cooper minimum in argon through a single scattering angle, along with phase signatures of multielectron effects. Our study bears the prospect of probing complex orbital phases in molecular systems as well as electron correlations through resonances subject to strong laser fields, and also enables studies of chiral phenomena in the XUV, such as circular dichroism [4].

References

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