Revealing attosecond photoionization dynamics using Rainbow RABBIT

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The generation of attosecond light pulses has opened the possibility to track the fastest dynamics in matter, in particular the electron dynamics that naturally occur on such a short timescale. Attosecond spectroscopy is thus able to shed new light on fundamental electronic processes involved in a variety of physical, chemical and biological reactions. A recent example is photo-electron emission for which attosecond delays between photon absorption and electron emission are becoming measurable in rare gas atoms, molecules or solids. When ionization occurs in the vicinity of a resonance, the dynamics is strongly perturbed and cannot be simply characterized by a group delay [1,2].

In a recent series of experiments, we were able to reconstruct the full ionization dynamics through Fano resonances [3] by measuring the scattering phase and amplitude of the released electron wavepacket, using a spectrally-resolved RABBIT, so-called Rainbow RABBIT [4]. This allowed reconstructing the complete autoionization dynamics of helium around sp2+, including the resonance buildup, and evidenced how photoelectron wavepackets are born and morph into asymmetric Fano profiles [4,5]. The Rainbow RABBIT was then used to study more complex cases, where multiple resonances are simultaneously excited in helium or neon resulting in the emission of very structured electron wavepackets, or when a resonance is coupled to two energy-shifted continua corresponding to different spin-orbit components like in argon. It also proved useful in disentangling the overlapping contributions of the 3s and 3p subshells of argon and accessing their relative ionization delays over a large energy range (34-70 eV) covering the Cooper minima in both subshells. All these measurements thus provide stringent tests for multielectron theories aiming at an accurate description of intrashell and intershell correlation. To provide reliable data for theory, the question of the coherence in the RABBIT spectrograms should be addressed: we have generalized the Mixed-FROG approach to cope with the complexity brought about by various sources of decoherence [6].

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