Time delay anisotropy in photoelectron emission from isotropic helium

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Time delay anisotropy in photoelectron emission from isotropic helium

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Synopsis In contrast to expectations, we observe that the photoionization time delay from the $1s^2$ spherically symmetric ground state of He depend on the electron emission direction with respect to an external reference. We attribute the observed anisotropy to the interplay between different final quantum states, which become accessible once two photons are involved in the photoionization process. This is a universal effect, which needs to be taken into account for any study dealing with photoionization dynamics.

Attosecond electron dynamics have been studied with different pump-probe techniques each employing an extreme ultraviolet (XUV) pump-pulse to ionize the target system and an infrared (IR) probe-pulse, which subsequently probes the dynamics of the liberated electron. Thereby, relative timing information between electrons originating from different states within the same atom or different atoms can be extracted.

Alternatively, relative emission timing of electrons from the same initial state within the same target system but at different emission angles θ can also be studied. We define θ as the electron emission direction relative to the XUV-pump polarization axis.

Since photons carry a spin angular momentum of 1, the absorption of a single photon can excite electrons from an initial state into two different final states: $n_i l_i \rightarrow E_f l \pm 1$, with n (l) representing the principle (orbital angular momentum) quantum number and E the photoelectron energy. As a consequence, if the ground state is spherically symmetric only one final state can be reached. However, as soon as two photons are involved in the ionization process, two different final states $1s \rightarrow E_i p \rightarrow E_f s/E_f d$ become accessible (Fig. 1a). Their interplay may give rise to anisotropic time delays.

Here, we present a rigorous experimental and theoretical investigation of the angle-dependent photoemission time delay of electrons removed from the spherically symmetric $^1S^e(1s^2)$ ground state of He.

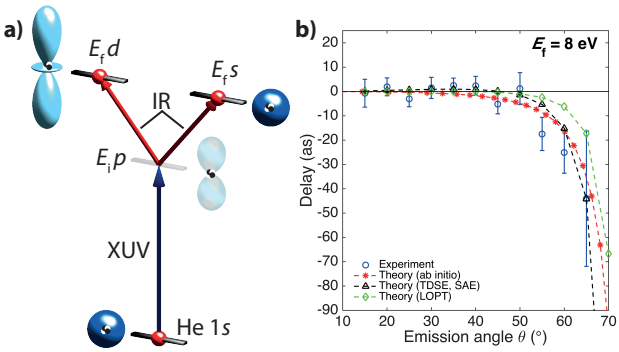

Figure 1. a) Accessible final states upon XUV-IR ionization: $E_f s$ and $E_f d$. b) Angular dependence of experimental (blue points) and theoretical (dashed lines) data for a photoelectron energy of 8 eV.

We measure a significant angular variation of the photoionization time delay, which can be as large as 50 attoseconds (Fig. 1b). Our observation sheds light on a new general aspect for attosecond measurements resolving electron dynamics triggered by ultra-short pulses. Indeed, so far most measurement techniques that resolved attosecond electron dynamics have been based on a two-photon process: one photon to initiate and another one to probe the dynamics of the system. Therefore, the effect studied in this work is universal and has important implications on the electron dynamics in more complex systems such as e.g. molecules and condensed matter.

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