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Attosecond delays in photoionization of diatomic and polyatomic molecules

V Loriot[1], A Marciniak[1], S Nandi[1], M Hervé[1], G. Karras[1], E Constant[1] and F Lépine[1*]

[1]Institut Lumière Matière, UMR 5306 – UCBL – CNRS, Villeurbanne Cedex, 69622, France

Synopsis We have investigated the photoionization dynamics of several molecular systems using attosecond interferometric techniques based on ultrashort extreme ultraviolet (XUV) pulses. For diatomic molecules such as N_2, we can get a detailed picture of the photoionization process by measuring the attosecond delays. For polyatomic molecules such as naphthalene, such measurements can provide the average timescale over which photoionization processes at different electron energies can occur in complex quantum systems.

After photoionization of an atom, the resulting photoelectron evolves in the combined field of the ionic core and the remaining electrons. For molecular photoionization, in addition to these effects the interplay between the electronic and nuclear degrees of freedom can also influence the photoelectron. Attosecond interferometry techniques, e.g., RABBITT (Reconstruction of Attosecond Beating by Interference of Two-photon Transitions) [1] can provide snapshots of such electron dynamics in their natural timescale inside parent molecules [2].

In our experiment, one part of a near infrared (NIR) pulse (λ~800 nm) was used to generate high-order harmonics in the XUV regime. The remaining part of the NIR pulse was used as a dressing field either directly or after frequency-doubling (λ~400 nm). Following photoionization of the target molecule by a dressed XUV field, the photoelectrons were detected using a Velocity Map Imaging (VMI) spectrometer. The angle-resolved photoelectron spectra show redistributions of photoelectron signal which oscillate (Fig. 1) with the delay between the XUV and the dressing fields at twice the frequency of the fundamental. The phase of the oscillation contains the imprint of the dispersion dynamics of the photoelectron wavepacket scattered in the molecular potential.

For N_2, the 400 nm pulse was used as the dressing field (hence, Blue-RABBITT) to significantly reduce the spectral congestion in the photoelectron spectra [3], compared to standard RABBITT-measurements [2]. The corresponding phase-differences (Φ_A-Φ_X) between the *A*- and the *X*-states in the N_2^+ ions are shown in Fig. 1. The overall decreasing trend in (Φ_A-Φ_X) values along the measured photoelectron energies is qualitatively well reproduced by the theoretical calculations [4] for single photon ionization of N_2 molecules.

For polyatomic molecules such as naphthalene, many different ionization pathways lead to the same electron kinetic energy due to photoionization of several close-lying electronic states by the broadband XUV pulse. Using 800 nm pulse as the dressing field, the phase of the molecular photoionization can still be extracted. The phase provides a measure of the average timescale over which an electron with a given kinetic energy can escape the molecular potential. This shows the applicability of attosecond interferometry techniques for complex systems, and can serve as a starting point towards studying quantum transport phenomena in two-dimensional materials.

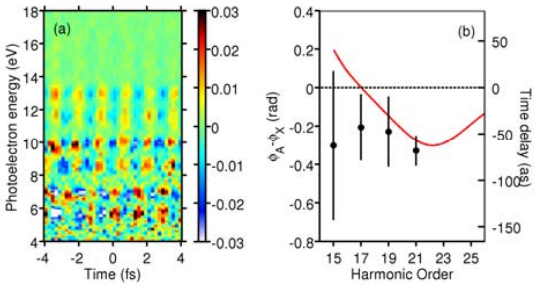


Figure 1. (a) Redistribution of photoelectron signal for photoionization of N_2. (b) Attosecond delay-differences between *A* and *X*-states of the N_2^+-ion, compared with that for single photon ionization of N_2, taken from [4].

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* E-mail: franck.lepine@univ-lyon1.fr

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