Photoemission delays in 2D model molecules

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Synopsis Photoelectron spectroscopy is one of the main techniques used to probe matter since the understanding of the photoelectric effect. Within the last decade, a new aspect has been given to spectroscopy, revisiting it in the time domain. It allows one to resolve the electron dynamics at the attosecond (10^{-18} s) time scale. Achieving such resolution experimentally brought one to establish innovative set up, and theoretical background is needed to validate, predict and interpret measurements. Much progress has been made in the attoscience field, this work is a step forward in understanding the electron dynamics for molecular continuum states at the attosecond timescale.

The recent progress in ultra-short laser pulses generation allows one to resolve temporally phenomena that was thought to occur instantly: photoionization processes. Photoemission delays can be observed through pump-probe experiments (RABBIT, streaking) [1–4]. In diluted systems, these delays are expressing the detailed influence of the parent ion on the electron. Therefore, they are highly depending on the species, but also on the channel the electron is ionized through for given species. They will also rely on the chirality for a molecule, the latter being involved in manifold natural phenomena [5].

In quantum physics, the dynamics of an electron is encoded in the phase of its wave function, and the dynamics are expressed in terms of group delay [6]. The ionization continuum being degenerated, there are multiple ways to describe it. The Selected Continuum Wave Function (SCWF) model [7] embeds all the information needed to describe the dynamics of photoemission, and provides an unambiguous and straightforward interpretation of the delays that have been recently discussed in the framework of attosecond science.

For a single-photon transition from a bound state $|\psi_{ini}\rangle$ with energy E_{ini}, the SCWF is defined in the framework of the first-order perturbation theory at a given energy E as $|\phi_{E,\mathrm{SCWF}}\rangle = \sum_\nu \langle\phi_{E,\nu}|\hat{\mathrm{d}}|\psi_{ini}\rangle|\phi_{E,\nu}\rangle$ where $\hat{\mathrm{d}}$ is the dipole operator coupling the initial and final states, and $\{|\phi_{E,\nu}\rangle\}$ an arbitrary orthonormal eigenbasis degenerated for the energy E. The SCWF model has already been verified with a time-dependent wave packet propagation in one-dimensional systems in [7]. Here, the two-dimensional approach allows to retrieve the angular dependent delays. Time-independent computations have been run to investigate ionization in an H_2^+ model molecule for a single-photon transition (Figure 1). The photoemission delays obtained from the time-independent approach are to be compared with a time-dependent propagation of a state at the same energy in order to validate the formalism of the SCWF. This work is the first step towards the extension to more realistic molecules.

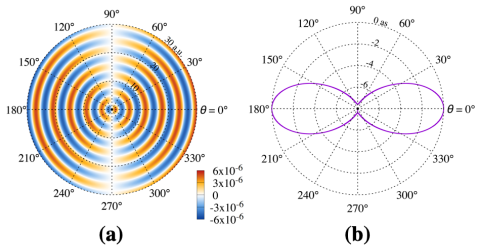

Figure 1. **(a)** The Selected Continuum Wave Function of a 2D model H_2^+ electron ionized in a single-photon transition at $\omega = 1$ a.u. and polarized linearly along the molecular axis. **(b)** Angular photoemission delays relative to the delay computed along the molecular axis ($\theta = 0°$) for the same transition.

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