Electron streaking in the autoionization region of H$_2$

**Alicia Palacios**[*][1]**, Alberto González-Castrillo**[*]**, Fernando Martín**[*†]

[*] Departamento de Química, Módulo 13,Universidad Autónoma de Madrid, Cantoblanco 28049, Madrid, Spain

[†] Instituto Madrileño de Estudios Avanzados (IMDEA) en Nanociencia, Cantoblanco, 28049 Madrid, Spain

Synopsis We use a UV-pump/IR-probe scheme, combining a single attosecond UV pulse and a 750 nm IR pulse, to explore laser-assisted photoionization of the hydrogen molecule in the autoionization region. The electron energy distributions exhibit unusual streaking patterns that are explored for different angles of the electron ejection with respect to the polarization vector and the molecular axis. Moreover, by controlling the time delay between the pulses, we observe that one can suppress the autoionization channel.

We present a theoretical study of H$_2$ ionization in the region of the doubly excited states by using a combination of a single attosecond UV pulse and a femtosecond IR pulse [1]. The IR field has a relatively low intensity and can not ionize the molecule by itself, simplifying the physics to a laser-assisted photoionization problem. Moreover, a well-known picture arises when a large-bandwidth UV pulse ionizes the target in a structureless region of the electronic continuum spectrum: the attosecond streaking phenomenom [2, 3]. If the UV pulse is significantly shorter than the IR pulse, the photoelectron is emitted at a well-defined time with respect to the IR field and the latter will then modify the electron motion in the continuum. We will explore how the presence of both autoionization and nuclear motion modifies this simple picture. We use an ab initio theoretical method to obtain total and energy differential probabilities for both dissociative (H+H$^+$+e$^-$) and non-dissociative (H$_2^+$+e$^-$) channels. We first investigate the behavior of the nuclear kinetic energy distributions and the electron-proton coincidence maps as a function of the time delay between the pulses. Most features can be explained in terms of perturbative transitions involving a UV photon and up to three IR photons. We found that autoionization is supressed when the IR pulse reaches the target after the UV absorption and with a delay shorter than 3-4 fs. This is due to the IR field depopulating the doubly excited states before they have time to decay into their background continua. At longer time delays, autoionization occurs before an the IR pulse arrives, and thus leaves its signature in the energy-differential distributions [4]. We later study the streaking patterns where the photoelectron is expected to be driven by the IR field, shifting its momentum in time following the vector potential

($A_{IR}(t)$). This simple picture, initially developed for atomic targets, is based on a single-active-electron approximation [2, 3]. We observe that when electrons ejected in both dissociative and non-dissociative ionization channels are not resolved (left panel in the figure), the distributions follow this simple pattern. However, this is no longer true for dissociative ionization alone (right panel in the figure), where the interferences with the autoionization process (H$_2^{**}$ → H + H$^+$+e$^-$) destroy this simple picture. This opens the question about how to extract ionization times from a streaking signal where non-direct processes such as autoionization are present.
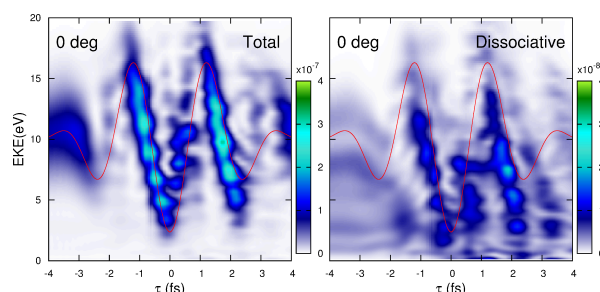
Figure 1. Electron energy distributions as a function of the time delay for ejection along the laser polarization axis. Left panel: total ionization. Right panel: dissociative ionization, which accounts for less than 10% of the total ionization (notice the different scales). Consequently, autoionization will strongly modify this channel and its streaking pattern. Red line: $-A_{IR}(t)$.

References

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[1]E-mail: alicia.palacios@uam.es