Attosecond chirp-encoded dynamics of light nuclei

S. Haessler*, [1], W. Boutu*, S. Weber*, J. Caillat†, R. Taïeb†, A. Maquet†, P. Breger*, B. Carré* and P. Salières*

*CEA Saclay, IRAMIS, Service des Photons, Atomes et Molécules, 91191 Gif-sur-Yvette, France
†UMPC Univ. Paris 06, CNRS, UMR 7614, LCPMR, 11 rue Pierre et Marie Curie, 75231 Paris, France

Synopsis We use the attosecond emission from light molecules to probe the ultrafast nuclear dynamics occurring between the ionization and recombination steps. By measuring the phase of the XUV emission, we have direct access to the mapping of harmonic frequency to the excursion time, which represents the delay after which the evolved molecular ion is probed. The small difference in the harmonic phase between H_2 and D_2 calculated theoretically is consistent with our experimental results.

An experimental method for tracking ultrafast nuclear re-arrangements termed PACER (probing attosecond dynamics by chirp encoded recollision) has been proposed [1] and demonstrated experimentally [2] a few years ago. The three steps, ionization, acceleration and recombination, commonly used to describe high harmonic generation (HHG), are considered as a pump, a delay-stage, and a probe process. In the case of H_2 and D_2, the molecular ion has a larger equilibrium internuclear separation than the neutral, so that during the excursion of the continuum electron, the molecular ion expands. The positive chirp of the re-colliding continuum electron wavepacket, assumed to be the same as that measured for rare-gas atoms [3], was exploited to map different pump-probe delays to the different harmonic frequencies.

Using the RABITT (Reconstruction of Attosecond Beating by Interference of Two-photon Transitions) [5] technique, we measure the group delay of the harmonic emission, which, for rare gas atoms, has been shown to be equal to the electron re-collision times, also called emission time [3]. As shown in figure 1, the emission times measured for argon, H_2 and D_2 under the same conditions are very similar. This experimentally confirms for H_2 and D_2 molecules the theoretical time-frequency mapping predicted by a strong field approximation (SFA) theory including nuclear dynamics [1] and verifies one of the basic assumptions made for PACER.

We then study the small difference in the measured group delay for H_2 and D_2. Within the SFA, the phase difference of the high-harmonic emissions from the two isotopes is given by the phases of the nuclear overlap integrals $C(\tau) = \int \chi_0(R)\chi(R,\tau)\mathrm{d}R$, where R is the internuclear distance, τ is the excursion time, mapped to harmonic order, and $\chi_0(R)$ and $\chi(R,\tau)$ are the nuclear wavefunctions of the neutral and the molecular ion, respectively [4]. A fully quantum mechanical calculation of $C(\tau)$ yields a small phase difference between H_2 and D_2, which encodes the nuclear dynamics just as the harmonic intensities considered in [1, 2]. Our experimental results are compatible with this phase difference.

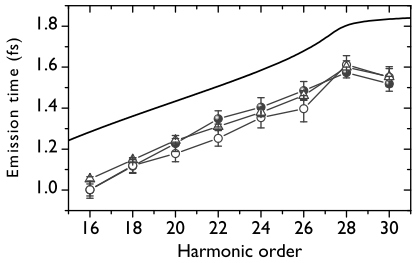

Fig. 1. Measured group delay or emission times for H_2 (◯), D_2 (△) and Ar (•). The line shows recollision times calculated with the SFA for Ar and an intensity of 1.2×10^{14} W cm^{-2}. The ≈ 200 as offset between theory and experiment is attributed to a macroscopic effect.

References

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[1] E-mail: stefan.haessler@cea.fr