When nuclei catch up with electrons

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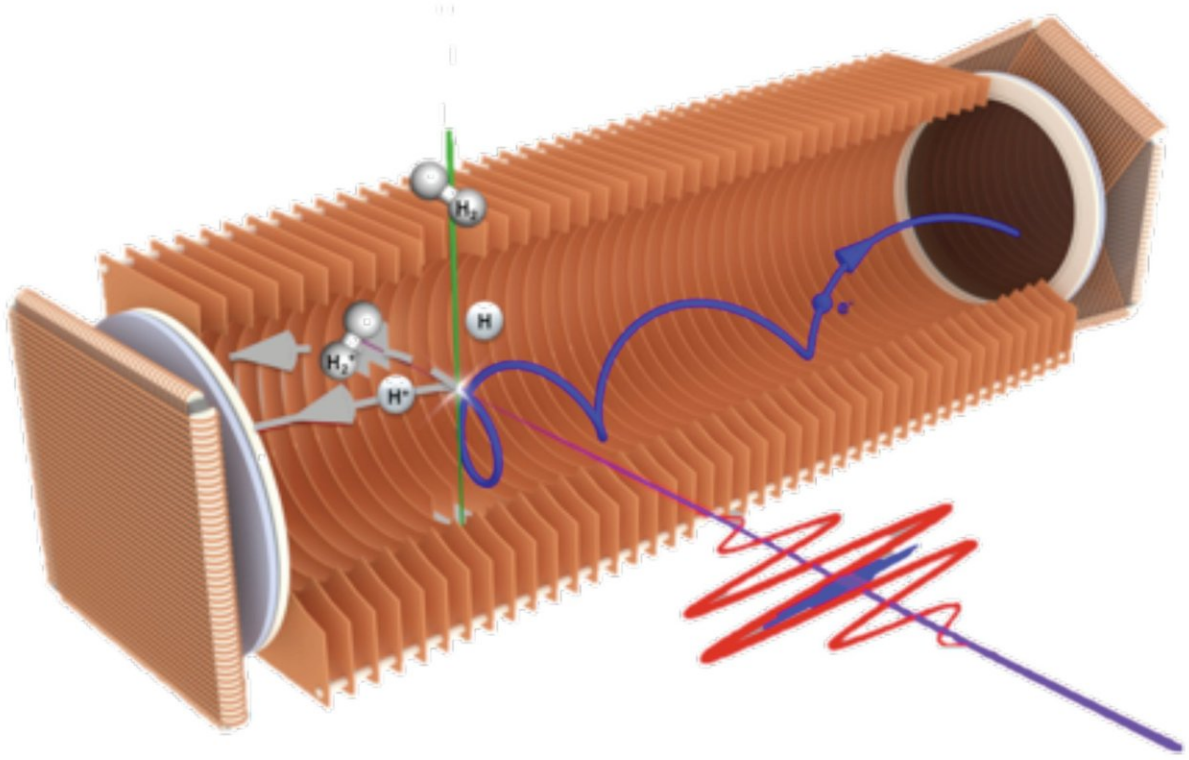

In the AttoCOLTRIMS apparatus, the three-dimensional motion of electrons (blue elliptical arrow) and ions (H2+ and H+, grey arrows) can be detected in coincidence. The combination of an extreme-ultraviolet (XUV, blue) and a long and rather weak infrared (IR, red) pulse in a pump-probe set-up provides the basis for studying the attosecond dynamics of the H2 molecule. Credit: Ultrafast Laser Physics group, ETH Zurich

In an attosecond study of the H2 molecule, physicists at ETH Zurich

found that for light atomic nuclei, as contained in most organic and biological molecules, the correlation between electronic and nuclear motions cannot be ignored.

The main goal in attosecond science is understanding the dynamics of quantum-mechanical systems on their natural timescale. Molecules, are among the most interesting systems to investigate, having a very high degree of complexity, particularly when compared to atomic systems. The few attosecond experiments performed on molecules to date have provided valuable insight into electron dynamics. In these studies, the dynamics of the nuclei around which the electrons evolve was assumed to be "frozen," given that nuclei are much heavier than electrons and therefore move more slowly. However, even in the attosecond time regime, the approximation that electronic and nuclear motion are decoupled from one another is often unjustified. In particular, in molecules composed of light atomic species, the nuclear motion can be as fast as electron dynamics, resulting in a strong coupling between the two.

A team led by Dr. Laura Cattaneo and Prof. Ursula Keller in the Department of Physics at ETH Zurich has now studied the lightest and smallest of all molecules, H2, and explored what happens when nuclear and electronic motion occur on a comparable time scale. As they report in an article published today in *Nature Physics*, they found that in molecules, ionization delays can significantly depend on the kinetic energy of both the photoelectron and the nuclei. (Ionization delays are the time between the absorption of a photon and the emission of an electron during photoionization.) This finding extends the concept of ionization delays introduced for atomic systems. Variations of ionization delays with nuclear kinetic energy can be as large as variations with electronic kinetic energy. This implies that whenever light atoms are involved in the molecular ionization process, the outgoing electron wave packet cannot be disentangled from the nuclear wave packet.

These measurements at the attosecond timescale are based on an experimental approach developed earlier in the Keller group. In the so-called AttoCOLTRIMS apparatus (see the figure), attosecond metrology is combined with the imaging technique COLTRIMS, in which the correlated properties of the fragments of a molecular reaction can be recorded. This experimental capability was combined with nearly exact ab initio theory, performed by collaborators at the Universidad Autonoma de Madrid (Spain), to describe both electronic and nuclear motions, as well the coupling between them.

The significance of this work goes well beyond the simple H2 molecule. Hydrogen atoms are present in most organic and biologically relevant molecules. Understanding the effects and contributions from coupled electron and nuclear dynamics present in such systems should therefore provide fundamental insight that will be important in various fields of research.

More information: L. Cattaneo et al, Attosecond coupled electron and nuclear dynamics in dissociative ionization of H2, *Nature Physics* (2018). DOI: 10.1038/s41567-018-0103-2

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