

# Imaging ultrafast molecular wave-packets using a single or a pair of UV pulses

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**Synopsis** In this talk, it is shown how to emulate a conventional pump-probe scheme using a single frequency-chirped ultrashort UV pulse to obtain a time-resolved image of molecular ultrafast dynamics. The chirp introduces a spectral phase in time that encodes the delay between the pump and the probe frequencies contained in the pulse. By comparing the results of full dimensional ab initio calculations for the  $\text{H}_2^+$  molecule with those of a simple sequential model, it is demonstrated that, by tuning the chirp parameter, two-photon energy-differential ionization probabilities directly map the wave packet dynamics generated in the molecule. As a result, one can also achieve a significant amount of control of the total ionization yields, with a possible enhancement by more than an order of magnitude.

Free-electron-laser facilities and high-harmonic generation techniques implemented in tabletop setups can nowadays provide intense ultrashort UV pulses with durations in the femtosecond and attosecond range [1, 2]. One of the more awaited capabilities offered by such pulses is to use them to monitor and control electronic and nuclear dynamics, for example within a UV-UV pump-probe scheme. While some progress in this direction has been made [3, 4, 5, 6], there are many technical challenges still to overcome, such as the limited intensity of the pulses and the fact that few optically active elements exist in the (extreme) ultraviolet. This precludes, for example, the use of pulse-shaping techniques and coherent control approaches that can be applied at optical and infrared frequencies to produce an “optimal” pulse for a desired photo-induced physical process or chemical reaction [7, 8]. Consequently, most experiments performed so far with attosecond UV pulses rely instead on an intense infrared pulse for either the pump or the probe step, which can significantly distort the system and alter the dynamics. The present work proposes an alternative approach: the use of a single chirped UV pulse, which can be realized in a lab, to emulate a conventional UV-UV pump-probe scheme.

In this talk, recent theoretical work demonstrates that by changing a single parameter, the spectral chirp of an ultrashort UV pulse, one can achieve a significant amount of control over molecular multiphoton ionization, changing the total ionization yield by more than a factor of ten. More importantly, it is shown how to emulate a conventional pump-probe setup to obtain direct time-resolved imaging of ultrafast molecular dynamics. The spectral chirp is experimentally tuneable both in high harmonic generation and with free electron lasers [9, 10, 2]. Although similar concepts were previously employed by Yudin et al. [11], it was only demonstrated for a simple super-

position of two bound states in the hydrogen atom. Here, the molecular (vibrational and electronic) wave packet in a small molecule is reconstructed, by being simultaneously pumped and probed by a single chirped UV pulse. A quadratic frequency chirp is chosen, which is experimentally achievable even in broadband pulses with Fourier-limited durations as short as a few hundreds of attoseconds [2, 9, 10], and ab initio simulations on the hydrogen molecular ion are performed. The full-dimensional time-dependent Schrödinger equation is solved numerically, including both electronic and nuclear degrees of freedom [12]. Finally, a sequential model is introduced to demonstrate the direct mapping of the pumped wave packet into the energy distribution of the charged fragments after the Coulomb explosion of the molecule. Although the  $\text{H}_2^+$  molecule is employed as benchmark target, the method should also be suitable to probe wave packet dynamics in more complex molecules.

## References

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