

# Sub-cycle Electron Dynamics Probed by Isolated Attosecond Pulses

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**Abstract:** Isolated attosecond pulses with low photon energies ( $\sim 12$ -40 eV) are used to probe sub-optical-cycle dynamics in laser-perturbed atomic and molecular targets using attosecond transient absorption spectroscopy, revealing dynamic wavepacket interferences and coupled electron-nuclear dynamics.

**OCIS codes:** (020.2649) Strong field laser physics; (300.6530) Spectroscopy, ultrafast

## 1. Introduction

Attosecond spectroscopy has so far focused on time-resolved measurement of the free electrons ionized by an isolated attosecond pulse in the presence of an external laser field. However, observation and control of attosecond dynamics in chemical processes requires access to the bound electron and nuclear motion which dictates the chemical reaction dynamics and outcomes. Here, we present attosecond transient absorption [1] measurements of bound state dynamics in helium atoms and hydrogen molecules, uncovering fast laser-induced dynamics in the electronic and vibrational wavepacket motion which evolve on the attosecond to few-femtosecond timescale.

## 2. Experiment and discussion

In the experiments, isolated attosecond pulses with spectra spanning from below 12 eV to more than 40 eV were generated from xenon gas by driving the generalized double optical gating (GDOG) [2] with  $\sim 5$  fs driving laser pulses. The attosecond pulse then passed through an aluminum ( $\omega_x > 15$  eV) or indium ( $11 \text{ eV} < \omega_x < 17$  eV) foil filter to block the residual NIR laser and to select the spectral components which overlap with the bound states of helium and hydrogen, respectively. The attosecond pulse was then recombined with a delayed portion of the few-cycle NIR laser, and both were focused together to a gas cell (1.5 mm inner diameter) filled with helium, neon, or hydrogen gas. In the absence of the NIR laser, absorption lines corresponding to the  $1snp$  excited states of helium and the  $D^1\Pi_u \leftarrow X^1\Sigma_g$  band of vibrational levels were observed with spectral resolution better than 50 meV.

Figure 1 shows the delay-dependent absorbance of helium (left) and hydrogen (right) in the vicinity of the states of interest. Negative delays indicate that the attosecond pulse arrives on the target before the NIR pulse. For both targets, the absorption changes dramatically near zero delay, and fast modulations with a periodicity approximately equal to half of the laser oscillation period are observed. Notably, different electronic states and different vibrational states within a given electronic configurations respond differently from one another.

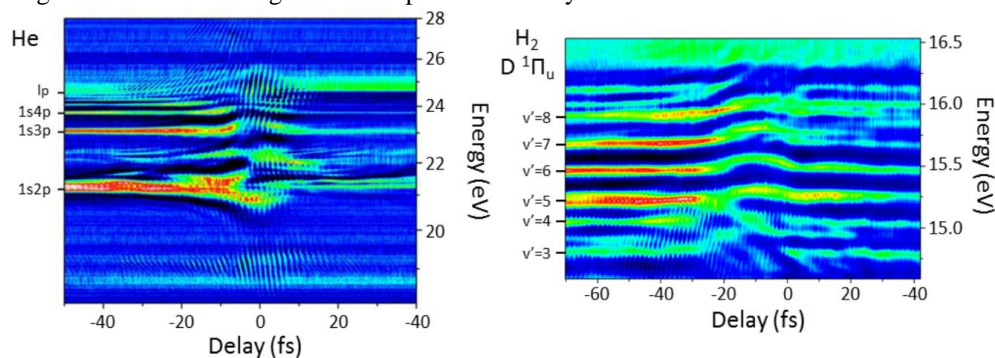


Fig. 1. Delay-dependent absorbance of helium (left) and hydrogen (right).

## 3. Conclusion

In attosecond transient absorption, dynamic changes in the absorption reflect the ultrafast electronic and nuclear motion. Such measurements, using isolated attosecond pulses with low photon energies, present a promising route towards time-resolving a chemical reaction. This material is based on work supported by the Army Research Office and the National Science Foundation under Grant Number 1068604.

## 4. References

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