

# Attosecond Transient Absorption in Molecular Hydrogen

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**Abstract:** Isolated attosecond pulses are used to probe laser-perturbed hydrogen molecules using attosecond absorption spectroscopy. We observe dynamic features in the delay-dependent absorption on both the electronic and nuclear timescales for the first time.

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## 1. Introduction

The rapid development of attosecond sources promises to enhance our ability to probe and control the fastest dynamics of electrons in matter. Attosecond transient absorption spectroscopy (ATAS) has recently been demonstrated to time-resolve electron dynamics on the few-femtosecond and even sub-optical-cycle timescales in atoms [1-3]. In such experiments, a broadband isolated attosecond pulse excites a coherent wave packet which is a superposition of many electronic states, allowing attosecond time-resolved measurements in bound states of atoms. On the other hand, attosecond spectroscopy in molecules has focused on time-resolved measurement of the free electrons/ions ionized by the combination of an attosecond pulse and the dressing laser field [4]. Such studies can provide only indirect evidence for the electronic and nuclear dynamics in the bound energy levels, which are often critical to ultrafast chemical processes. However, the application of the transient absorption spectroscopy technique to molecular targets has so far proved challenging due to the lack of attosecond sources in the vacuum ultraviolet (VUV, ~6-20 eV) spectral region. In this work, we present the first attosecond transient absorption measurements of bound state dynamics in hydrogen molecules, uncovering fast laser-induced dynamics in the electronic and vibrational wave packet motion which evolve on the attosecond to few-femtosecond timescale.

## 2. The experimental setup

The experiments were conducted by using the attosecond transient absorption spectroscopy technique. In the experiment, infrared pulses with 5-fs pulse duration and 730-nm central wavelength were generated from a 1 kHz repetition rate Ti:Sapphire laser and a hollow-core fiber compressor. The beam was split into two parts using a broadband beam splitter. In one arm, isolated attosecond pulses were generated by using the generalized double optical gating technique (GDOG) [5] in a 1-mm-thick cell filled with xenon gas. The spectral range of the attosecond pulses was selected using an indium foil filter with transmission that extends from below 11 eV to above 20 eV. In the other arm, the beam passed through an optical delay line and then recombined with the attosecond pulses collinearly. Both pulses were focused into a second gas cell which was filled with hydrogen gas. The transmitted VUV spectrum was recorded with a flat-field grating spectrometer as a function of the time delay between the attosecond pulse and few-cycle NIR pulse.

## 3. Results and discussion

Figure 1(a) shows the measured transmission spectrum of hydrogen molecules as a function of the time delay between the two pulses with a dressing laser intensity of  $\sim 10^{13}$  W/cm<sup>2</sup>. Negative delay indicates that the attosecond pulse arrives on the target before the NIR pulse. In the negative delay region, XUV pulses first excite electronic and nuclear wavepackets from the ground state via vibronic transitions. Absorption lines corresponding to the D←X, C←X bands are observed to change dramatically in the presence of the dressing NIR laser. These absorption lines shift as a function of the time delay, similarly to the sub-cycle AC Stark shifts observed in atoms. Furthermore, fast modulations with a periodicity approximately equal to half of the laser oscillation period are observed in the negative delay region in both the D←X and C←X bands.

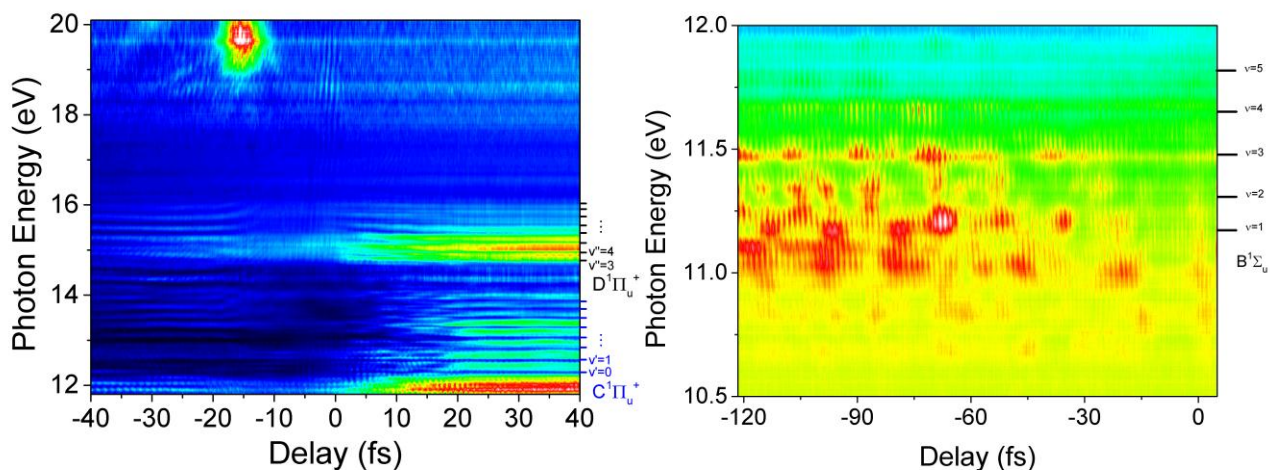


Figure 1. Measured delay-dependent transmission spectra of hydrogen molecules in the vicinity of the  $D \leftarrow X$  and  $C \leftarrow X$  bands (a), and  $B \leftarrow X$  bands (b).

At negative delays, the experiment results also reveal a relatively weak half-laser-cycle modulation of the absorption which appears near 17-20 eV. Such modulation was previously observed in helium atoms and was interpreted as quantum pathway interferences between electrons ionized directly by absorption of an XUV photon and indirectly by excitation of a bound excited state followed by absorption of one or more NIR photons [3]. More interestingly, emission structures (enhanced transmission) appear in the energy spectrum between 18-20 eV, repeating on a timescale of  $\sim 15$ -20 fs which is close to the vibrational period of the  $D^1\Pi_u$  state. We observe a half-cycle interference oscillation superimposed on these repeated structure, indicating the simultaneous measurement of the electronic and nuclear dynamics.

By carefully tuning the attosecond pulse spectrum to the lowest photon energies, we can further observe the absorption spectrum in the vicinity of the  $B \leftarrow X$  vibronic transitions, as shown in Figure 1(b). In this energy region, alternating absorption and emission structures appear in the vicinity of the  $B^1\Sigma_u$  states with a relatively slow periodicity of  $\sim 10$ -15 fs, along with a superimposed fast modulation of half the laser cycle period.

#### 4. Conclusion

In conclusion, we measured the transient absorption of an isolated attosecond pulse by laser-dressed bound states of hydrogen molecules. The absorption lines shift in the presence of the dressing laser as a function of the time delay. Absorption features corresponding to both electronic and nuclear dynamics are observed with sub-femtosecond precision. 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.

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#### 5. References

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