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ISBN

9781943580187

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Publication Date

2016-07-18

DOI

10.1364/UP.2016.UTh4A.39

Peer reviewed

Probing Ionization Dynamics with Attosecond Transient Absorption Spectroscopy

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Abstract: Attosecond transient absorption experiments are performed to study strong-field ionization dynamics in xenon and methyl bromide. Results in Xe depict half-cycle ionization steps that exhibit an apparent time delay between spin-orbit ionization channels.

OCIS codes: (320.7150) Ultrafast spectroscopy; (320.7120) Ultrafast phenomena

Attosecond transient absorption spectroscopy (ATAS) is a powerful technique for probing valence electron dynamics in a variety of systems [1], ranging from simple atoms [2] to condensed phase material [3]. In one variety of ATAS, a few-femtosecond near infrared (NIR) pump pulse is used to initiate a photo-chemical process and the dynamics are probed with a time-delayed attosecond extreme ultraviolet (XUV) pulse. The attosecond pulse excites core-to-valence transitions in the atom or molecule, leading to the absorption of the XUV photons. These so-called “absorption edges” exhibit element and charge specificity and can therefore disentangle complex, multi-electron dynamics.

In this presentation, strong-field ionization dynamics in Xenon and methyl bromide (CH_3Br) are investigated using ATAS. In the Xe ATAS experiment, a 5 fs NIR pulse ionizes either the $5p_{3/2}$ or $5p_{1/2}$ valence electron of the atom, yielding two distinct spin-orbit ionization pathways. Next, a time-delayed attosecond pulse centered at 60 eV [4] probes the creation of Xe^+ by promoting the $4d$ electron to fill the $5p$ vacancy. In the experiment, two prominent XUV absorption peaks are observed at 55.4 eV and 56.1 eV, corresponding to the excitation $4d_{5/2} - 5p_{3/2}$ and $4d_{3/2} - 5p_{1/2}$ respectively. The experimental set-up and pump-probe scheme are shown in Fig. 1.

Figure 1 The experimental set-up for ATAS in Xe. The inset depicts the pump-probe scheme for observing strong-field ionization dynamics in Xe.

The strong-field ionization dynamics of Xe are traced out by measuring the absorption profile of the two spin-orbit channels as a function of the NIR-pump and XUV-probe time delay. The corresponding ATAS spectrogram is shown in Fig. 2. The absorption profiles from both spin-orbit channels exhibit coherent build-up in amplitude with a half-cycle periodicity (1.3 fs). This is characteristic of strong-field ionization. To further demonstrate this behavior, the integrated absorption profile of each spin-orbit state is shown in the inset of Fig. 2. These plots clearly depict local maxima in the electron hole signal. This result stresses the importance of strong-field dynamics beyond the pure quasi-static picture, where the NIR-induced polarization of the neutral ground state plays an important role. Interestingly, the half-cycle oscillations also exhibit an apparent time delay of around 200 as between the two spin-orbit channels. While the origin of this experimental time delay between spin-orbit ionization steps is not exactly understood, ongoing work using time-dependent configuration interaction singles [5] is being performed to resolve the origin of the delay as well as to determine the limitations of ATAS on these extreme timescales.

The method of ATAS is extended to probe ionization dynamics in molecular systems. Preliminary work demonstrates that a spin-orbit wavepacket is observed when probing the bromine absorption edge in strong-field ionized methyl bromide. With this method, the nature of electronic coherence and its entanglement with nuclear degrees of freedom such as vibrations and rotations can be investigated.

Figure 2 The ATAS spectrogram for strong-field ionization dynamics in Xe. Integrating over the absorption lineshapes leads to the effective spin-orbit hole populations shown in the inset.

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