Dissociative ionization processes of D₂ molecule investigated with a-few-pulse attosecond pulse train

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Synopsis Dissociative ionization processes of D₂ molecule are investigated with the pump-probe measurement using a-few-pulse attosecond pulse train. Delay-dependent momentum images of D⁺ originated from the probe process in the singly-charged manifolds recorded with the velocity map imaging ion spectrometer exhibit clear modulation patterns reflecting the existence of autoionization processes excited by multiphoton absorption in the pump pulse in addition to the non-resonant direct ionization processes.

When the molecules are irradiated with the attosecond pulses, the vibrational and electron wavepackets are generated and their temporal evolutions can be monitored by recording the momentum images of fragment ions [1, 2]. In addition to the observation of quantum wavepackets, the attosecond pulses can induce the autoionizing processes. Since the excitation to the autoionization states accompanies the simultaneous excitation of more than one electrons, the autoionization process intrinsically accompanies the electron correlation. In the present study, we investigated the dissociative ionization processes of D₂ molecule with the pump-probe measurement using a-few-pulse attosecond pulse train to observe the autoionizing processes excited by the multiphoton absorption processes.

A-few-pulse attosecond pulse train (APT)(\(T_{\text{env}} = 4\) fs, \(T_{\text{pulse}} = 400\) as) is generated through the high harmonic generation process by focusing the output of sub-15 fs driving laser to a Xe gas cell. The generated APT is spatially-divided into two by the beam splitter composed of two Si plane mirrors. The pair of attosecond pulses is focused by a SiC concave mirror on the molecular beam of D₂ injected from a piezo valve integrated in the repeller electrode in the velocity map imaging ion spectrometer. The fragment ions D⁺ generated at the focal region are extracted by parallel electrodes toward the MCP/phosphor assembly. By moving one of Si mirrors, the delay between the pump and probe pulses is scanned every 36 as from \(-7\) fs to \(+7\) fs.

The delay-dependent kinetic energy (KE) distributions of D⁺ arranged by integrating over the ejection angle of the fragment ions satisfying the parallel and perpendicular to the laser polarization are shown in Fig. 1. The modulation patterns appearing in the KE distribution of the parallel component shown in Fig. 1(a) are ascribed to the probe processes of vibrational wavepacket formed in the 1σ\(_g\) state in D₂⁺ by the excitation to 2σ\(_g\) state [1].

On the other hands, the intensity modulations appearing in the KE distribution of the perpendicular component shown in Fig. 1(b) are assigned to the excitation to the autoionizing states \(Q_2 \Pi_u\) by three-photon absorptions leading to the dissociation into H(1s) and H(\(n = 2\)), respectively [3]. Since the intensity modulation in Fig. 1(b) covers much broader KE region than the spectral bandwidth of harmonics in the APT, the features are ascribed to be the excitation to the autoionizing processes in which the photoelectron can carry the certain energy leading to the broad kinetic energy distribution of fragment ions. The period of intensity modulations coincide with the optical period of fundamental and 3rd order harmonic contained in the APT.

Figure 1. Delay-dependent kinetic energy distribution of D⁺. (a) Parallel to the laser polarization, (b) Perpendicular to the laser polarization.

References
[1] Y. Nabekawa et al., 2016 Nat. Commun. 7 12835