High-order Harmonics Fourier Transform Spectroscopy of Two-Photon Dissociative Ionization of Hydrogen Molecules

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Abstract: We have investigated two-photon dissociative ionization processes induced by high-order harmonic generation pulses with Fourier transform spectroscopy. Delay-dependent kinetic energy distribution of fragment ions reveals dissociative ionization dynamics of hydrogen molecules.

OCIS codes: (300.6500) Spectroscopy, time-resolved; (320.7150) Ultrafast spectroscopy

1. Introduction

High-order harmonics (HH) pulse enable us to investigate multi-photon process in vacuum ultraviolet wavelength region. From measuring the interferometric autocorrelation traces of HH pulses with molecular responses, we identify not only optical field of attosecond pulse train, but also molecular dynamics, such as ionization and/or dissociation mechanism. In the case of nitrogen molecules and carbon dioxide molecules, attosecond pulse train's characteristic has appeared in the measured correlation traces [1, 2]. In the case of deuterium molecules (D_2), however, the autocorrelation traces of the single order were obtained through two-photon dissociative ionization [3]. The observed optical fields were the fundamental laser field, the 3rd-, and 5th-order harmonic field and there was no sign of the excitation process by the 7th- or higher order harmonic field.

In the present work, we investigated the two-photon dissociative ionization of D_2 using HH Fourier transform spectroscopy to detect the excitation process of D_2^+ by high-order harmonic field. The fundamental laser field, the 3rd-order, or 5th-order harmonic field tends to excite D_2^+ at the internuclear distance region close to outer turning points. On the other hand, higher excitation photon energy is expected to excite D_2^+ at the short internuclear distance region close to the inner turning point. In the measured kinetic energy distribution of D^+, we observed the fragment ions having the higher kinetic energy than that we reported previously [3, 4], and studied the excitation dynamics of fragment ions observed in this work.

2. Experimental

The HH pulse was generated in a xenon gas cell by focusing of the fundamental driving laser pulses with a pulse-duration of 13 fs and a center wavelength close to 800 nm [5]. The HH pulse was consisted from the multiple odd order harmonics of up to 21st-order. The pulse durations of HH are estimated to be shorter than 10 fs by measuring the interferometric autocorrelation. The HH pulse was spatially divided into two replicas by the beam splitter which was made with a pair of silicon mirrors. The resultant HH pulse pair was focused onto molecular beam of D_2 by a concave mirror, which was made of silicon carbide, with a focal length of 100 mm. Fragment deuterium ions D^+ produced through dissociative ionization process with HH pulses. To perform a delay-resolved measurement of dissociative ionization of D_2 induced by the HH pulse, the relative delay between the two replicas were scanned by moving the one silicon mirror of the beam separator with a piezo-actuated stage. Fragment momentum distributions of D^+ were analyzed using velocity map imaging (VMI) ion mass spectrometer. We executed signal counting analysis for image data in order to detect low probability events.

3. Results and Discussion

When the HH pulses was focused to D_2, the momentum distribution of D^+ is obtained as shown in Fig. 1(a). The three pairs of arcuate components appearing inner in the distribution have been reported. Owing to increase molecular density by improvement of molecular beam introduction, we succeeded to observe the three additional large arcuate components. The kinetic energy spectrum of D^+ is obtained by integrating the momentum distribution over the polar angle measured from the polarization direction of the HH pulses, as shown in Fig. 1(b). The kinetic energy spectrum of D^+ exhibits six components whose peaks located at 0.4 eV, 1.4 eV, 2.6 eV, 3.7 eV, 5.4 eV, and 7.1 eV. The three components, which are located at 0.4 eV, 1.4 eV, and 2.6 eV, are produced via sequential two-photon process that the dissociation occurs by another excitation pulse after ionization.

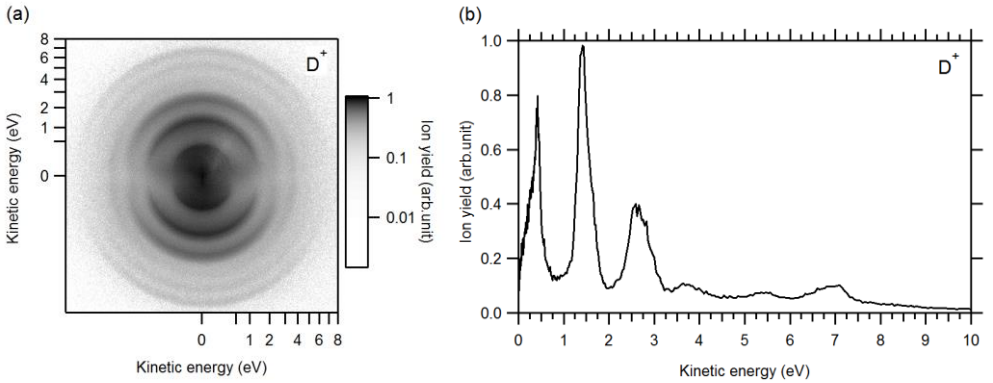

Fig.1 (a) Momentum distribution of D^+ obtained by velocity map imaging mass spectrometer. The polarization direction of the HH pulses is aligned to the vertical axis. (b) Kinetic energy spectrum obtained by integration of the distribution over polar angle measured from the polarization direction of the HH pulses.

To investigate how the harmonic fields interact with D_2, we performed HH Fourier transform spectroscopy on D_2 by measuring image data with scanning the delay between the HH pulse pair. By arranging the kinetic energy spectra obtained at each delay in order of increasing the temporal delay, we constructed the delay-dependent kinetic energy spectrogram shown in Fig. 2(a). The three components at 0.4 eV, 1.4 eV, and 2.6 eV clearly exhibit fringe patterns of the optical frequencies of fundamental, the 3rd-, and the 5th-order harmonics, respectively. On the other hand, the two components at 3.7 eV and 5.4 eV exhibit peaks at a half cycle intervals of the fundamental frequency similar as attosecond pulse train. The delay dependence of the highest kinetic energy component is not clear.

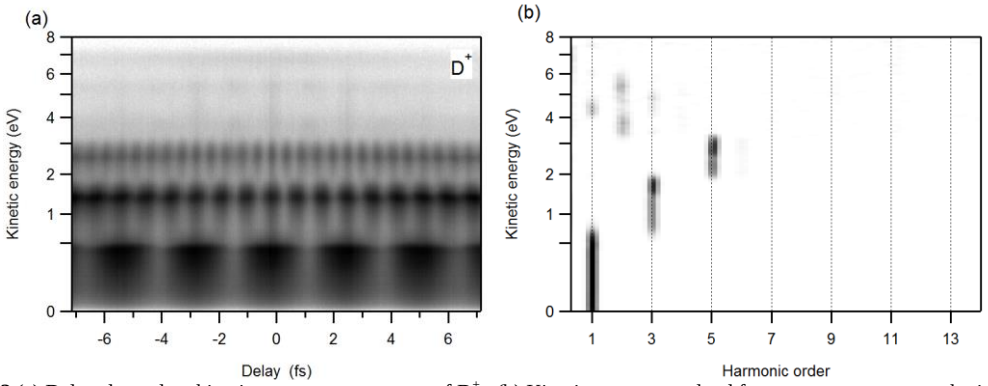

Fig.2 (a) Delay-dependent kinetic energy spectrogram of D^+. (b) Kinetic-energy-resolved frequency spectrogram obtained by carrying out Fourier transform of the spectrogram Fig. 2(a).

By carrying out Fourier transform of the kinetic energy spectrogram in Fig.2 (a), the kinetic-energy-resolved frequency spectrogram is obtained shown in Fig. 2(b). The second harmonic frequency peak appears in each kinetic energy region of 3.7 eV and 5.4 eV. In the case of N_2 in which high-order harmonic fields are involved in two-photon dissociative ionization, the second and fourth harmonic frequency peaks are observed. This similarity suggests that dissociation with the high kinetic energy is induced by high-order harmonic fields, such as the 7th-, 9th-, and 11th-order harmonics. By performing time-resolved measurement and Fourier analysis of the vibrational wavepacket evolution in the D_2^+ $1s\sigma_g$ potential energy curve after ionization, we will further understand the dissociation dynamics of high kinetic energy components reported in this work.

4. References

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