Attosecond Frequency Resolved Momentum Imaging of Two-photon Dissociative Ionization Dynamics of Nitrogen Molecule

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Abstract: Two-photon dissociative ionization processes of nitrogen molecule are investigated with attosecond nonlinear Fourier transformation spectroscopy. The frequency resolved momentum images extracted from delay dependent momentum images showed attosecond nonlinear response of nitrogen molecule.

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1. Introduction

When molecules are utilized as a nonlinear medium for autocorrelation measurement of ultrashort laser pulses, the nonlinear response of medium should be clarified for determining the temporal profile precisely if the spectral bandwidth of the laser pulse is broad to cover several electronic states of molecules. The frequency spectra extracted by Fourier transformation of the observed interferometric autocorrelation traces (IACs) of attosecond pulse train (APT) can be interpreted as a convolution of the electric waveform of APT and the nonlinear response of molecule to APT [1, 2]. Since the angular distribution of fragment ion is one of the sensitive probe to extract the symmetry of electronic states in the decomposition process [3], the angular distribution of fragment ions originated from two-photon absorption processes leading to the correlation signal in the IAC traces should reflect the information of electronic state.

In the present work, we observed IAC of a-few-pulse APT generated with a sub-15 fs, 100 Hz laser system using two-photon dissociative ionization processes of N_2 and extracted the molecular response by frequency resolved momentum imaging (FRMI) method.

2. Experimental setup

The intense a-few-pulse APT was generated by focusing (f = 5 m) the output of Ti:S femtosecond laser system (800 nm, 12 fs, 100 Hz) onto a Xe gas cell. The generated APT was spatially divided into two with two Si plates and the reflected harmonic beams are introduced into a single-shot velocity-map imaging (VMI) ion spectrometer. The replica of APT pulses was focused (f = 100 mm) onto a molecular beam of N_2 introduced into a vacuum chamber by a piezo valve integrated into the repeller electrode in the VMI apparatus. By moving one of the two Si plates on a single axis piezo stage every 20 nm, the temporal delay Δt between the two split APT was varied with a step of 35.6 as from -7 fs to $+7$ fs. In each delay, we recorded 2D momentum images of N^+ originated from two-photon dissociative ionization processes ($N_2^+ \rightarrow N^+ + N$) by temporally gating a MCP/Phosphor detector with 100 ns duration.

3. Results and discussion

The IAC traces using the ion signal (I-IAC) and the IAC traces using the fragment ion anisotropy (A-IAC) are obtained from the delay-dependent 2D momentum images. The A-IAC spectrogram was obtained by calculating $< \cos^2\theta >$ valve as a function of the delay Δt and the kinetic energy of fragment ion E. The resultant I-IAC and A-IAC spectrograms are shown in Fig. 1(a) and 1(b), respectively and there are fine characteristic modulation patterns. The recorded autocorrelation traces are converted into the frequency domain spectra by a Fourier transformation, and the results are shown in Fig. 1(c) and 1(d). The peaks in odd order frequencies of fundamental light field are originated from the interference fringes of the harmonic field. On the other hand, the peaks in even order frequencies are assigned to difference frequency between the adjacent and the next adjacent harmonic field modes to form the envelope of APT.

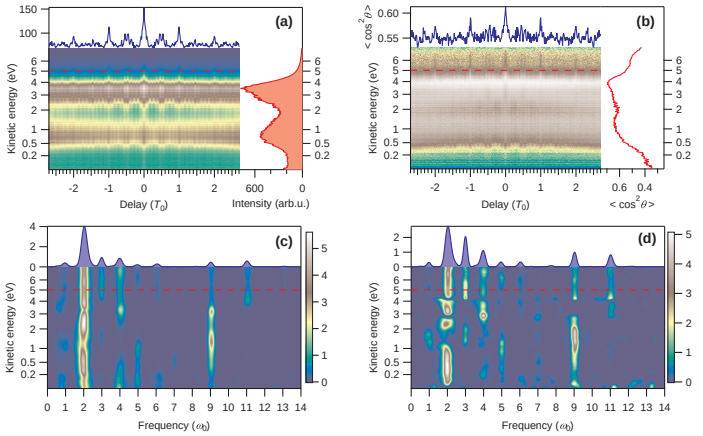

Fig. 1. (a) IAC trace obtained using the ion signal. (Right) kinetic energy distribution. (Top) I-IAC trace at E = 5 eV. (b) IAC trace obtained using the anisotropy of fragment ions. (Right) The anisotropy as a function of kinetic energy. (Top) A-IAC trace at E = 5 eV. (c) Frequency spectrogram of I-IAC with frequency spectrum at E = 5 eV. (d) Frequency spectrogram of A-IAC with frequency spectrum at E = 5 eV.

The observed frequency domain spectra has a clear dependency on the kinetic energy of the fragment ions exhibiting the nonlinear response of N_2 molecule in the short wavelength region. The advantage of nonlinear Fourier transformation spectroscopy is that only delay dependent signals originated from two-photon processes can be extracted in the frequency spectrogram. Therefore, the recorded momentum image of fragment ions on the detector can be decomposed into frequency resolved momentum images as shown in Fig. 2. These momentum images show clear correlations between two-color harmonics leading to the non-sequential two-photon processes.

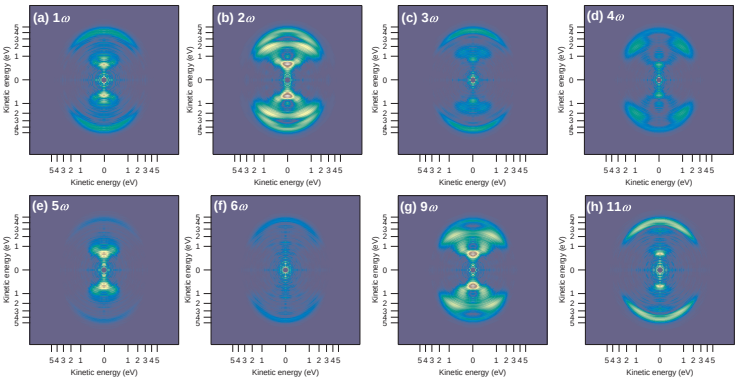

Fig. 2. Frequency resolved momentum images of fragment ion N^+. (a) 1ω, (b) 2ω, (c) 3ω, (d) 4ω, (e) 5ω, (f) 6ω, (g) 9ω, and (h) 11ω.

4. Conclusion

Two-photon dissociative ionization processes of nitrogen molecule are investigated with a-few-pulse attosecond pulse train. Two types of the interferometric autocorrelation traces (I-IAC, A-IAC) are obtained and the nonlinear responses of nitrogen molecule are extracted as frequency resolved momentum images.

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