Simultaneous Observation of Vibrational Wavepackets of Nitrogen Molecule in Neutral and Singly-charged Manifolds

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Abstract: Vibrational wavepackets are launched at neutral and single-charged electronic excited states of nitrogen molecule by sub-10 fs high harmonics. The evolution of vibrational wavepackets are tracked by sub-10 fs high harmonics.
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1. Introduction

When molecules are exposed to a short wavelength light with short pulse duration, vibrational wavepacket (VWP) in the electronic excited states of neutral molecules and/or the ionic electronic states of molecules can be initiated [1]. The manipulation of VWPs can be considered as one of the essential approaches for controlling chemical reactions in diatomic or polyatomic molecules. In most of cases, VWPs are prepared in the ionic electronic states of molecules by a pump pulse and a probe pulse is used for tracking the evolution of the VWPs by detecting the resultant fragment ions. In this experimental configuration, it is difficult to launch VWPs in the electronic excited states of neutral molecules because of shortage of DUV and/or VUV light sources with sub-10 fs pulse duration.

In the present work, we observed the VWPs of nitrogen molecule (N_2) launched by sub-10 fs XUV pulses generated with a sub-15 fs, 100 Hz laser system [2] and their evolution are monitored by sub-10 fs XUV pulses composing of attosecond pulse train (APT) by detecting momentum image of fragment ion N^+ originated from sequential two-photon ionization processes with velocity map imaging ion spectrometer.

2. Experimental setup

The APT was generated by focusing the output of Ti:S femtosecond laser system (800 nm, 12 fs, 100 Hz) loosely onto a Xe gas cell by a concave mirror (f = 5 m) in a vacuum pulse compressor. The generated APT was spatially divided into two with two Si plates and propagated into a single-shot detection capable velocity map imaging (VMI) ion spectrometer. The replica of APT pulses was focused by a SiC concave mirror (f = 100 mm) onto a molecular beam of N_2 injected by a piezo valve integrated into the repeller electrode in the VMI spectrometer. By moving one of the two Si plates on a single axis piezo stage every 1120 nm, the temporal delay Δt between the two split APT was varied with a step of 2 fs from -10 fs to $+400$ fs. In each delay, we recorded 2D momentum images of N^+ originated mainly from two-photon dissociative ionization processes ($N_2^+ \rightarrow N^+ + N$) by temporally gating a MCP/Phosphor detector with 100 ns duration. By tuning the central wavelength of fundamental laser pulse, the central wavelength of harmonics are blue-shifted about 1.4 %.

3. Results and discussion

The fragment ion signals and its anisotropy parameter $< \cos^2 \theta >$ are plotted as a function of the delay Δt and the kinetic energy of fragment ion E in Fig. 1(a) and Fig. 1(d), respectively. The modulation patterns shown in Figs. 1(a) and 1(d) correspond to the time evolution of vibrational wavepackets created either in the electronic excited states of N_2 or in the electronic excited states of N_2^+. After subtraction of DC components ascribed to the signals originated from one arm of APT, the spectrograms shown in Figs. 1(b) and 1(e) are obtained. Even though the modulation depth for the anisotropy parameter shown in Fig. 1(e) is small, the sign and tendency of modulation coincides with the modulation of fragment ion intensity shown in Fig. 1(b). The periodic modulation patterns shown in Fig. 1(b) and 1(e) are quantitatively analyzed by performing a Fourier transformation and the resultant frequency spectrograms are shown in Figs. 1(c) and 1(f). These modulation periods are ascribed to the periods of vibrational wavepackets.

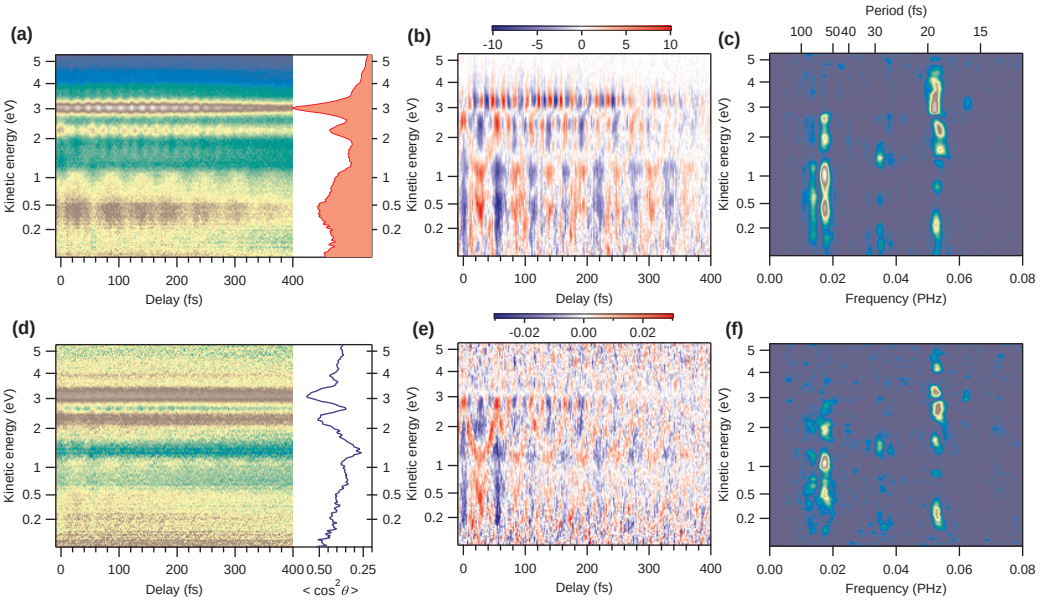

Fig. 1. Time evolution of vibrational wavepackets of N_2. [Intensity] (a) $I(E,\Delta t)$, (b) $\Delta I(E,\Delta t)$, (c) $I(E,\omega)$. The right panel in (a) shows the delay averaged kinetic energy distribution of N^+. [Anisotropy] (d) $A(E,\Delta t)$, (e) $\Delta A(E,\Delta t)$, (f) $A(E,\omega)$. The right panel in (d) shows the delay averaged $<\cos^2\theta>$ where the angle θ is measured from the laser polarization direction.

The modulation period $T_{vib} \sim 57$ fs at the kinetic energy $E < 1.5$ eV can be assigned to the time evolution of VWP launched at the electronic excited states ($a'^1\Sigma_u^-$, $b'^1\Sigma_u^+$) of neutral nitrogen molecule by 7th or 9th order harmonics. On the other hand, the modulation period $T_{vib} \sim 18$ fs at the kinetic energy $E > 1.5$ eV can be assigned to the time evolution of VWP launched at the electronic excited states ($A^2\Pi_u$, $B^2\Sigma_u^+$) of singly charged nitrogen molecule by 11th or 13th order harmonics. In both cases, 11th or 13th order harmonics would work as a probe pulse to generate the fragment ion N^+ via dissociative ionization ($N_2^+ \rightarrow N(^4S^o)+N^+(^3P)$).

As the kinetic energy E increases from 2.0 eV to 2.5 eV, the modulation period T_{vib} becomes larger. This would be explained by the fact that the population in high vibrationally excited levels leads to longer vibrational period.

The modulation period $T_{vib} \sim 30$ fs would be ascribed to the coexistence of two kinds of VWPs leading to the same kinetic energy E since the period coincides with the beat frequency of two VWPs.

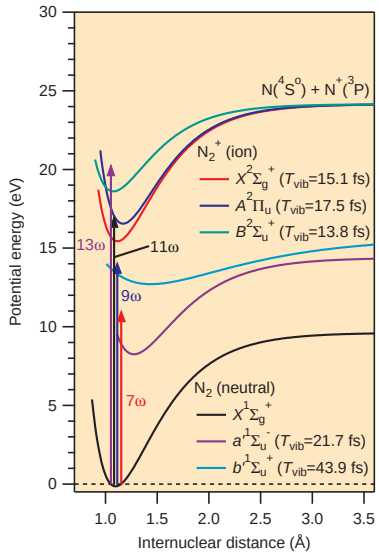

Fig. 2. Scheme for generating VWPs.

4. Conclusion

Sequential two-photon dissociative ionization processes of nitrogen molecule are investigated with a-few-pulse attosecond pulse train. The vibrational wavepackets in neutral/singly-charged electronic excited states are prepared by harmonics in VUV-XUV region and the evolution is monitored by inducing dissociative ionization by absorbing sub-10 fs high harmonics.

References

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