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Tracking Vibrational Wavepackets of Nitrogen Molecules by XUV-Pump XUV-Probe with Momentum Imaging

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1. Introduction

When molecules are excited by broadband ultrashort pulses, vibrational wavepacket (VWP) will be generated. Observation and manipulation of VWP is one of the key steps for understanding chemical reactions of molecules. So far, the VWPs are mainly prepared in singly charged manifolds because of the lack of short VUV-XUV pulse and their time evolutions are mainly tracked by a few-cycle intense infrared pulses [1, 2]. Even the pulse duration of infrared pulse can be much shorter than vibrational period of molecules, the interaction with intense infrared pulse might lead to the mixing of electronic states and the deformation of molecular geometry. Therefore, the time evolution of VWP should be monitored by one-photon absorption process of light whose pulse duration is much shorter than vibrational period.

In this purpose, high harmonic pulse generated from sub-15 fs laser system is considered to be one of the ideal light sources to prepare VWPs and tracking their time evolutions. In this work, we investigated VWPs of nitrogen molecule by XUV-pump XUV-probe method with recording momentum image of fragment ion N^+ originated from the dissociation process ($N_2^+ \rightarrow N^+ + N$) as a function of delay of two XUV pulses.

2. Experiments

Intense attosecond pulse train (APT) is generated via high harmonic generation process by focusing the output of sub-15 fs laser system to a xenon gas cell [3]. The resultant APT is propagated to a beam separator composed of two Si plates and is spatially divided into two. By moving one of the Si plates mounted on the high-precision piezo stage, the delay between two harmonic pulses is scanned every 2 fs from 0 to 400 fs.

The spatially divided harmonic pulses are introduced to velocity map imaging ion spectrometer and focused to a molecular beam of nitrogen injected from a piezo valve integrated into repeller electrode by a SiC concave mirror. The generated fragment ion N^+ is projected to microchannel plate/phosphor screen assembly and the fluorescence image on the phosphor screen is captured by a high-speed sCMOS camera.

3. Results and discussion

The fragment intensity is plotted as a function of the delay of two harmonics and the kinetic energy of fragment ions in Fig. 1(a). The intensity modulation as a function of delay is quantitatively analyzed by Fourier transformation of Fig. 1(a). The resultant spectrogram shown in Fig. 1(b) shows that there are at least two distinguished frequency peaks.

The slow modulation (T_{vib} ~ 55 fs) is ascribed to the VWP of the electronic excited state $b^1\Pi_u$ in neutral manifold formed by the absorption of harmonics whose order is greater than or equal to 9 since the XUV photon energy is not enough to populate to the electronic excited states in singly-charged manifold to explain the observed slow modulation. On the other hand, the fast modulation (T_{vib} ~ 18 fs) is ascribed to the VWP launched in $A^2\Pi_u$ state in singly-charged manifold excited by harmonics whose order is greater than or equal to 11. In both VWPs, the time evolution is monitored by exciting to the repulsive potential curves by another arm of XUV pulse.

In addition to the two main VWPs, VWPs are launched in $b'^1\Sigma_u^+$ (neutral manifold) and in $X^2\Sigma_g^+$ and $B^2\Sigma_u^+$ (singly-charged manifold). The final states for each VWPs are assigned from the kinetic energy of fragment ions and angular-dependent frequency spectrogram.

4. Conclusion

Vibrational wavepacket of nitrogen molecule is launched both in neutral manifold and in singly-charged manifold by XUV pump pulse, and their time evolutions are tracked by XUV probe pulse. The final states for each vibrational wavepackets are assigned from the angle-resolved frequency spectrogram. This work was a part of APSA project commissioned by MEXT.

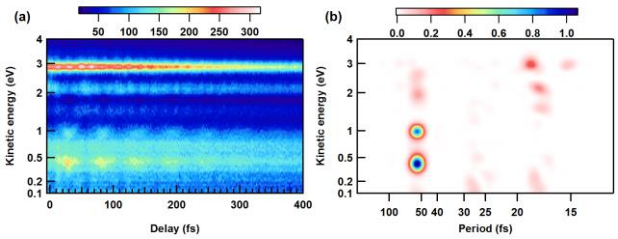

Fig. 1: (a) Delay dependent kinetic energy distribution of fragment ion N^+. (b) Frequency spectrogram obtained from (a).

References

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