Interferometric attosecond spectroscopy of molecules

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We have developed an attosecond nonlinear Fourier transform spectroscopy (NFTS) based on the interferometric autocorrelation (IAC) measurement of an attosecond pulse train (APT) using molecules as a nonlinear medium for investigating the ultrafast molecular dynamics [1]. In the attosecond NFTS, the nonlinear molecular responses are encoded in the IAC traces depending on the molecular species, the fragment ions and the kinetic energy distributions.

When the envelope of APT is shorter than the vibrational period of molecules, in other words, the number of attosecond pulses in the envelope is a-few, the vibrational wavepacket of molecules can be traced in the real-time manner. We have investigated with the quantum wavepackets of diatomic molecules with the combination of a-few-pulse APT and the velocity map imaging ion spectrometer (VMIS). It was identified that the settling time of $\sim$1 fs is required to compose the initial vibrational wavepacket in H_2^+ and D_2^+ [2]. In the VMIS, the information on the final electronic states in the probe process is encoded in the angular distribution of fragment ions. It was demonstrated that the branching of competing dissociation pathways between the $2p\sigma_u$ and $2p\pi_u$ states in H_2^+ was controlled within sub-10 fs by separating the H^+ signals based on the ejection direction of fragment ions [3].

In the case of multielectron diatomic molecules such as N_2, the APT can excite more than one bound electronic states simultaneously both in the neutral and singly-charged electronic states. By detecting the delay-dependent momentum images of N^+ generated by the probe APT pulse, the temporal evolution of multiple vibrational wavepackets were observed simultaneously, suggesting the formation of the electron wavepacket. The temporal evolution of electron wavapacket in N_2 was traced by precisely scanning the pump-probe delay and the the frequency components ascribed to the formation of electron wavepacket were identified [4].

These results highlight that nonlinear spectroscopic information of molecules in the short wavelength region can be obtained through the irradiation of a pair of intense APT pulses. In this progress report, we will also report on the development of multi-fragment momentum imaging method in which the momentum images of all kinds of fragment ions can be recorded simultaneously in order to extend the attosecond NFTS from diatomic to polyatomic molecules for investigating the ultrafast intramolecular electron motion in attosecond time domain.

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