Probing Ultrafast Molecular Dynamics with Intense Attosecond Pulses

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Abstract: Recent results on a new attosecond spectroscopic method called as nonlinear Fourier transform spectroscopy using attosecond pulse train and generation of multi-gigawatt isolated attosecond pulses by the infrared two-color laser field synthesis are reported.
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1. Introduction

With the progress of femtosecond laser technologies, it becomes possible to generate attosecond pulses by utilizing the coherent wavelength conversion process known as high-order harmonic generation. As a straightforward extension of femtochemistry, in which nuclear wavepacket motion is tracked in real-time, attosecond pulses enable us to capture the electron motion in atoms and molecules and the ultrafast nuclear-electron coupled motion in molecules. One of the ultimate goals of attosecond science is to perform attosecond pump and attosecond probe measurement to understand chemical reactions such as ultrafast charge migration and electron correlation with unprecedented temporal resolution of the atomic unit.

To seriously tackle the above interesting research topics, one of the most important issues is the development of high-power isolated attosecond pulses (IAPs) and/or attosecond pulse trains (APTs). IAPs and APTs are produced using high-order hamonic generation in gases. At RIKEN, high-power APTs have successfully been generated owing to research on high harmonic energy scaling using a loose-focusing geometry [1]. Using the intense ATPs, we have developed a new attosecond spectroscopic method called as nonlinear Fourier transform spectroscopy (NFTS), in which the nonlinear responses of the molecules are encoded in the energy-resolved interferometric autocorrelation traces [2, 3]. More recently, we have proposed and demonstrated a robust generation method of intense isolated attosecond pulses by employing two-color field synthesis and the energy-scaling method of high-order harmonic generation [4]. Here I report these two research topics of attosecond science at RIKEN.

2. Nonlinear Fourier transformation spectroscopy with intense attosecond pulse train

Observation and control of nuclear wavepacket and electron wavepacket are crucial to understanding ultrafast molecular dynamics. In particular, sub-10 fs high harmonics composing APT is one of the effective light sources to launch nuclear and electron wavepackets. Temporal profile of APT and the nonlinear response of molecules can be investigated by NFTS. In this study, we extended our NFTS method by introducing a velocity map imaging spectrometer with which the angular distribution of fragment ions and photoelectrons can be measured. The anisotropy of the ejection direction of the fragment ion carries the information of the symmetry of the electronic states involved in the decomposition processes. By introducing single-shot counting detection scheme, the dynamic range of the detection system is improved for extracting weak intensity variations corresponding to the molecular nonlinear responses.

Experimentally, intense APT is generated by loosely focusing the output of the femtosecond laser system to a xenon gas cell, and the resultant APT is

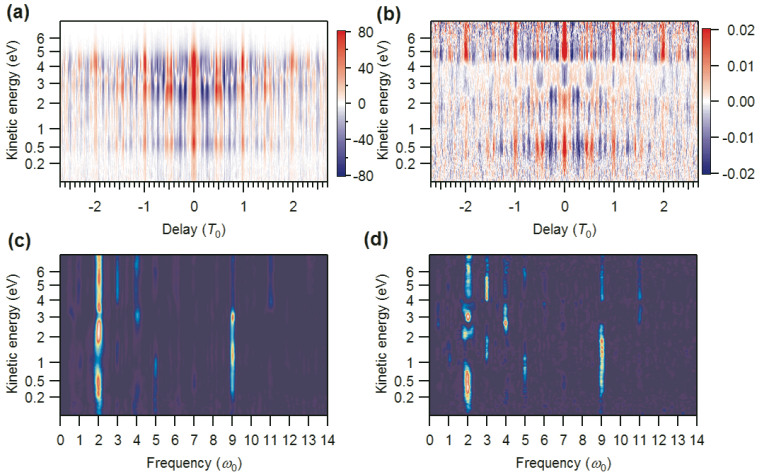

Fig. 1: (a) IAC trace $I(E, \Delta t)$ obtained from the intensity of fragment ion N^+. T_0 is the optical period of fundamental pulse. (b) IAC trace $A(E, \Delta t)$ obtained from the anisotropy of fragment ion N^+. (c) Fourier transformed frequency spectrogram of $I(E, \Delta t)$. ω_0 is the angular frequency of fundamental pulse. (d) Fourier transformed frequency spectrogram of $A(E, \Delta t)$.

propagated to a highly stable interferometer. The pair of APT is focused to a molecular beam of target molecules by a SiC concave mirror. The generated fragment ions are extracted by parallel electrodes in the velocity map imaging configuration. The ion signals are detected by a detector composed of chevron type microchannel plates and a phosphor screen. The signal of the fragment ion was selectively detected by imposing the pulse gating voltage to the detector system. The generated fragment ions (N^+) are detected with VMIS. The delay of APT is scanned (i) every 36 as from −7 fs to +7 fs for non-sequential process, and (ii) every 2 fs from −10 fs to +400 fs for sequential process.

In the short range high precision scan, the fragment ion intensity modulates as a function of the delay Δt between APT pulses as shown in Fig. 1(a). This modulation $I(E, \Delta t)$ corresponds to the interferometric autocorrelation (IAC) traces reflecting the electric waveform of APT depending on the kinetic energy (KE) of the fragment ion. The delay-dependent anisotropy of fragment ion $A(E, \Delta t)$ is plotted in Fig. 1(b). For clarifying the difference between $I(E, \Delta t)$ and $A(E, \Delta t)$, the KE-frequency spectrograms obtained by Fourier transformation of $I(E, \Delta t)$ and $A(E, \Delta t)$ are calculated as shown in Figs. 1(c) and 1(d), respectively. These spectrograms show the two-color two-photon dissociative ionization dynamics of N_2.

3. Generation of multi-gigawatt isolated attosecond pulses

We have demonstrated the generation of microjoule IAPs, which are intense enough to induce the nonlinear phenomena in atoms and molecules and perform direct characterization by autocorrelation measurement. Our generation scheme is robust and straightforward for scaling up the IAP energy, which is based on the infrared two-color laser field synthesis [5] and the energy-scaling method [1]. By carefully designing the generation configurations, we obtained IAPs with energy up to 1.3 μJ/pulse around 30 eV region, thus showing its energy enhancement from 100- to 1000-fold compared with the previous report so far. The conversion efficiency attained was improved to be 1.1 x 10^{-4} thanks to the phase matching technique. From a 500-as pulse duration determined by autocorrelation method, the peak power of this IAP was evaluated to be 2.6 GW. Furthermore, the output energy of a mid-plateau region (14 - 29 eV) having a quasi continuum spectrum by the two-color high harmonic generation attained to 10 μJ level. By utilizing this quasi-continuum spectrum, we also generated a quasi-IAP with a pulse duration of 375 as.

We are going to expand this two-color scheme into much shorter wavelength region. Since our method has the advantage that the high harmonic output yield can be linearly scaled up by increasing the high harmonic emission volume, we can estimate exactly a scaled-up configuration in the soft-X-ray region to obtain even shorter IAP durations. In the soft-X-ray region around 100 eV, we have already demonstrated a ~ 50 nJ energy per harmonic order by one-color excitation scheme in Ne gas. By straightforwardly upgrading the two-color scheme to a main pump energy of 50 mJ and adopting a focusing length of 5 m with a 5-cm-long Ne medium, we expect to achieve an isolated attosecond pulse energies grater than 0.1 μJ at 95 to 110 eV, which is almost 1000-fold higher than the energies previously reported. Our simulations indicate that IAPs can be generated with sub-300-as duration. In addition, when we switch the wavelengths of the main and supplementary fields, we can expect to further shorten the wavelength of the generated attosecond pulses to the "water window" region [6]. For this purpose, we have designed high energy femtosecond IR source based on double-chirped optical parametric amplification [7].

4. References

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