Controlling attosecond electron wave packet in a molecule

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Abstract: We control and identify the two-channel interference and the attosecond electron wave packet in polyatomic molecules by using two-dimensional high-harmonic spectra emitted from aligned molecules using the orthogonally polarized, two color laser fields.
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1. Introduction

Measuring and controlling electron wave packet motion in atoms or molecules with attosecond time-resolution have been one of the major interests in ultra-fast, attosecond science. When an intense, infrared laser pulse ($\sim 10^{14}\,\mathrm{W/cm^2}$) is irradiated to gaseous molecules, tunnel ionization will occur from the most loosely bound state (or the highest occupied molecular orbital, HOMO). If the tunnel ionization probability of HOMO is comparable with that of the second or third most loosely bound state, the hole or electron dynamics can be created in the valence state of the molecule [1]. In this case, the spectral intensity of the high-harmonics emitted from the molecule is modulated [2] or the polarization state of the harmonics changes as the valence state electron evolves in time [1].

The electron wave packet prepared by simultaneous ionizations from two states can be represented by a coherent superposition of the electron hole or Dyson orbital of each ionization channel, where the one electron is removed from the most and the second most loosely bound state. It is given by

$$\Psi(t) = a(1)\psi_1 + a(2)\psi_2 \exp(-i\varphi(t)) \qquad (1)$$

where $a(1)$ and $a(2)$ are coefficient, and $\varphi(t)$ is the phase difference between the two states. If the two states are separated by a few electron volt, the wave packet moves in the attosecond time-scale. The coefficients and $\varphi(t)$ are determined by the ratio of the tunnel ionization probabilities between them.

We have measured the dynamics of the electron wave packet prepared by this ionization process in randomly aligned C_2H_6 using the two-color laser field method [1]. However, in the randomly aligned (or unaligned) molecule, the ratio between the coefficients is fixed and cannot be controlled. On the other hand, it is difficult to identify the electron wave packet motion clearly using a *linearly polarized* probing pulse with a molecular alignment approach.

Here by combing the molecular alignment technique with the two-color field method, we demonstrate an experimental approach to control and identify uniquely the tunnel ionization ratio between the two ionization channels, thus attosecond electron wave packet. In the two-color laser field method, we combine the fundamental laser pulse with the orthogonally polarized second harmonic to generate high-harmonic emission from target molecules. The electron wave packet motion or two-ionization channel is mapped onto the two-dimensional high-harmonic spectra emitted from the molecules which are measured as a function of the phase delay between the two pulses (referred to as "two-color delay"). By aligning molecules, we select the particular ionization angle from the molecular axis. It allows us to control the amount of the contribution of each state on the total ionization probability. This can be possible because the angular distribution of the tunnel ionization probability depends on the symmetry of the molecular orbital. Using CO_2 and C_2H_6, we show how the two ionization channels or the electron wave packet motion can be controlled [3].

2. Experimental set-up

We generate 35 fs, 1 KHz, 800 nm laser pulses from Ti:Sapphire laser system (KMLabs, CEP-stabilized Dragon). We split the output beam to the pump and probe pulses. The pump pulse align the molecule non-adiabatically. Then we irradiate the orthogonally polarized, 800 nm and 400 nm laser pulses to generate high-harmonic emission as probe pulses. We focus the laser beams to a pulsed gas jet and measure the high-harmonic spectra emitted from molecules.

3. Results and discussion

Fig. 1 plots the peak intensity of the selected odd harmonics of C_2H_6 as a function of the pump-probe delay at fixed two-color delay of ~ 0.33 fs. At the 11th and 13th harmonic number, the peak intensity increases at the pump-probe delay of $0 \sim 500$ fs while at 21st harmonics the intensity decreases in this delay range. The intensity variation is due to the prompt alignment. Next, we select the pump-probe delay of -500 fs and 200 fs. Fig. 2 shows the two-dimensional high-harmonic emission spectra of C_2H_6 as a function of the two-color delay at the pump-probe delay

of (a) -500 fs and (b) 200 fs.   In both cases, as has been reported in Ref. [1], the odd and even harmonics are observed in the spectra and those intensities are modulated as a function of the two-color delay.   First we focus on Fig. 2 (a) where the molecule is unaligned. At the <15th harmonics, the even harmonic peak appears at the same phase delay from the adjacent odd harmonics, while at the >16th harmonics, the even harmonic peaks appear at the different phase delay from the adjacent odd harmonics.   Because the intensity ratio between the odd and adjacent harmonics tells us the relative polarization direction of the high-harmonic emission, the 2D map indicates that the spatial structure of the valence state electron wave function changes dynamically as the harmonic number thereby the re-collision time increases from 0.8 fs to 1.2 fs [1].   Next, we focus on Fig. 2 (b) at the pump-probe delay of 200 fs where the molecules is aligned.   In the low harmonic range, the 13th and 15th harmonic intensity increases while the 12th (not shown in this vertical scale) and 14th harmonic intensity slightly decreases compared with the unaligned spectra.   On the other hand, in the high harmonic range, the intensity of all the harmonic number decreases.   The intensity variation with alignment suggests that the contribution of HOMO-1 (the Ag state) on the valence electron wave packet is more enhanced than the HOMO (the Eg state) when the molecule is aligned.

Fig. 3 shows the two-dimensional high-harmonic spectra when we anti-align CO_2 (a) parallel and (b) perpendicular to the polarization direction of the 800 nm in the probe pulse, respectively.   The double peak structure observed at 12th is identified as the contribution of the two ionization channels: the left peak is due to the σ_u state and the right peak is due to the π_g state.   The effect of the alignment on the two-channels is clearly observed.

[1] H Niikura *et al.*, , "*Probing the spatial structure of a molecular attosecond electron wave packet*", Phys. Rev. Lett. **107,** 093004 (2011).
[2] H. Niikura *et al*., "*Mapping attosecond electron wave packet motion*", Phys. Rev. Lett. **94**, 083003 (2005).
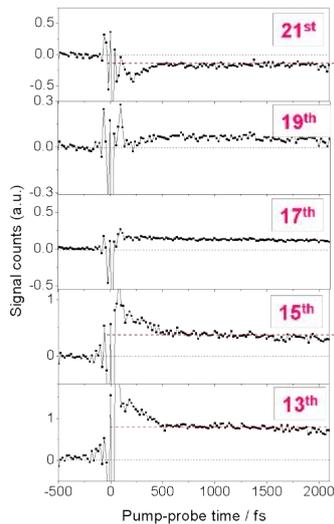[3] H. Niikura, *submitted*.

Fig. 1. The harmonic peak intensity of C_2H_6 as a function of the pump-probe delay at fixed two-color delay of ~0.33 fs.
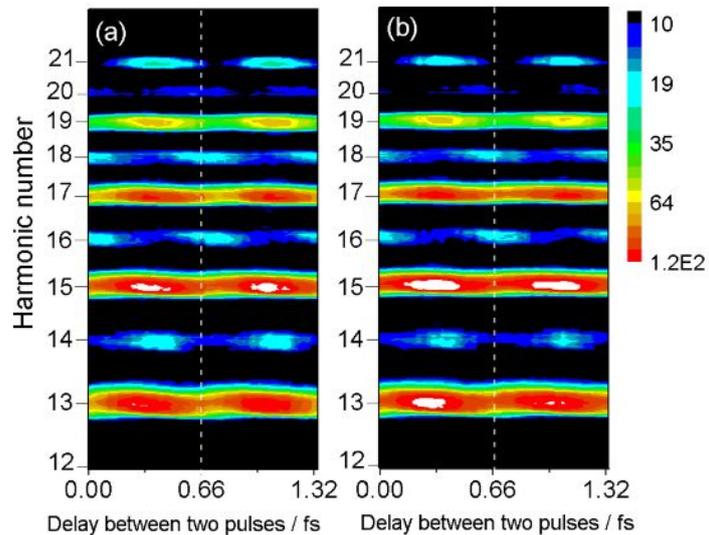
Fig. 2. The two-dimensional high-harmonic generation spectra of C_2H_6 at (a) unaligned and (b) aligned conditions.
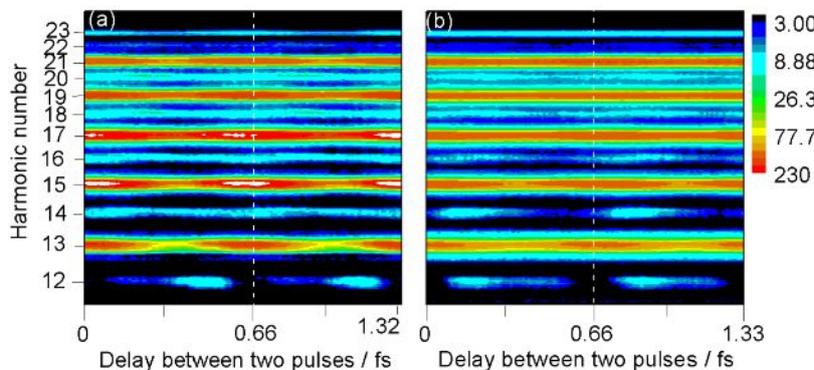

Fig. 3. The two-dimensional high-harmonic generation spectra of CO_2 at anti-aligned (a) parallel and (b) perpendicular to the main axis of the probe pulse.