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CEP dependences in single and double ionizations of methanol in intense few-cycle laser fields

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[Abstract] The carrier-envelope-phase (CEP) dependences of single and double ionization processes in methanol were investigated by detecting the fragment ions by the coincidence momentum imaging (CMI) method. It was found that CH_3^+ generated from the dissociation of CH_3OH^+ is ejected mostly opposite to the direction of the electric field, indicating that the ionization rate is larger when the electric field points from the C atom to the O atom than when it points toward the opposite direction. On the other hand, CH_3^+ generated from the Coulomb explosion of CH_3OH^{2+} exhibits the phase shift of 0.7π with respect to CH_3^+ generated from the dissociation of CH_3OH^+. This phase shift suggests that the double ionization proceeds via the re-collision of the electron ejected through the tunnel ionization.

［**Introduction]** Recent developments of ultrafast laser technologies enable us to shorten a laser pulse so that the number of optical cycles within the pulse becomes only a few. Because the temporal shape of the electric field within the pulse varies depending on the carrier-envelope phase (CEP), responses of molecules to such few-cycle laser pulses are dependent on the CEP. Indeed, it has been revealed that the ionization and dissociation processes of molecules in a few-cycle laser field are influenced by the CEP [1]. Therefore, it is expected that, through the measurements of the CEP dependences, we will be able to deepen our understanding of complex mechanisms of molecular ionization and dissociation processes in an intense laser field. However, it is difficult to determine the absolute CEP at a spatial point where atoms and molecules interact with the few-cycle laser pulses because, in most cases, the CEP monitored in a separate apparatus have a constant offset with respect to the absolute CEP at the interaction point. Recently, Fukahori et al. [2] proposed an efficient method to determine an absolute CEP through the measurements of photoelectrons ejected from Ar by the irradiation of circularly polarized few-cycle laser pulses. In the present study, by adopting this method, we investigate the absolute CEP dependences in the single and double ionization processes of methanol by monitoring the asymmetry in the ejection direction of the fragment ions.

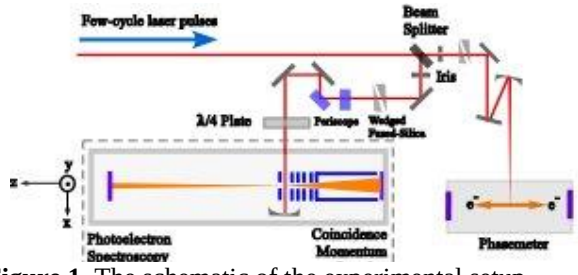

Figure 1. The schematic of the experimental setup.

[Methods] Figure 1 shows the experimental setup. Linearly polarized few-cycle laser pulses (4.5 fs, 760 nm, 5 kHz) were split into two by a beam splitter. One of the beams was introduced into a phasemeter [3] to record the CEP of each laser pulse, and the other was focused onto an effusive methanol beam in the vacuum chamber with which the CMI method are performed. The peak laser-field intensity at the laser-molecule interaction point was estimated to be 2.1×10^{14} W/cm^2, and the polarization direction of the laser pulse was parallel to the *y*-axis as shown in Fig. 1. The generated

fragment ions were accelerated by electrostatic lenses and were detected by a position sensitive detector. The three-dimensional momentum vectors of these ions were retrieved from their positions and the arrival times at the detector. The absolute CEP was calibrated by the CEP dependence of the photoelectrons generated from Ar in the circularly polarized few-cycle laser pulses.

[Results and discussion] The CEP dependences of the ejection direction of CH_3^+ produced from CH_3OH^+ ($CH_3OH^+ \rightarrow CH_3^+ + OH$) and from CH_3OH^{2+} ($CH_3OH^{2+} \rightarrow CH_3^+ + OH^+$) were investigated. The asymmetry parameter $A(\phi)$ describes the difference between the yields of CH_3^+ ejected upwards ($p_y > 0$) and downwards ($p_y < 0$) as a function of the CEP ϕ as

$$A(\phi) = \frac{N_{up}(\phi) - N_{down}(\phi)}{N_{up}(\phi) + N_{down}(\phi)}, \qquad (1)$$

where $N_{up}(\phi)$ and $N_{down}(\phi)$ denote the yields of the fragment ions ejected upwards and downwards, respectively. By a least-squares analysis, the ϕ dependence in the asymmetry parameter was fitted to a cosine function, $A(\phi) = A_0 \cos(\phi - \phi_0) + B$ where ϕ_0 denotes the CEP offset. Figure 2 shows the asymmetry parameters for the two decomposition channels producing CH_3^+. The CEP offsets were determined to be $\phi_0 = 1.13\pi$ and 1.85π for the decomposition channels from CH_3OH^+ and CH_3OH^{2+}, respectively.

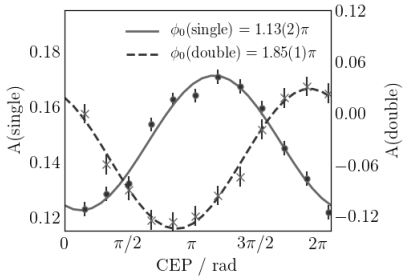

Figure 2. The CEP dependence of CH_3^+ generated from CH_3OH^+ and CH_3OH^{2+}. The black dots are the experimental data and the black lines are the best-fit asymmetry parameter, $A(\phi) = A_0\cos(\phi - \phi_0) + B$.

The CEP offsets value close to π for the decomposition channel from CH_3OH^+ shows that CH_3^+ ions are ejected preferentially toward the positive y direction at $\phi = \pi$. Figure 3(a) shows the electric field of the few-cycle laser field at $\phi = \pi$, and Fig. 3(b) shows the tunneling ionization rate w of methanol evaluated by the Ammosov-Delone-Krainov model [4]. The tunneling ionization rate becomes maxium at $t = 0$, at which the electric field is in the negative y direction, that is, the electric field is directed from the C atom to the O atom. Therefore, the tunneling ionization probability of methanol is larger when the electric field is directed from the C atom to the O atom than when the electric field direction is opposite.

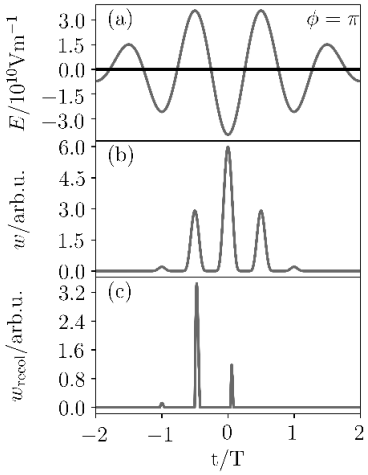

Figure 3. The results of the theoretical simulation when CEP = π (a) The electric field of the laser pulse. (b) The ionization rate of tunneling ionization. (c) The recollisional ionization rate w_{re}.

The recollisional ionization is known to contribute most to the double ionization of methanol by a few-cycle laser pulse [5]. Using the kinetic energies of a re-colliding electron obtained by solving the Newton's equation of motion, the recollisional ionization rate $w_{re}(t)$ was calculated as shown in Fig. 3(c) [1], showing that the recollsional ionization rate becomes maxium at $t = -0.47T$, where T is the period of optical cycles, at which the electric field is in the positive y direction, which is shifted by π compared with the tunneling ionization. This means that ϕ_0 of recollisional ionization is expected to be $\sim 2\pi$ ($= 0\pi$), which is in good agreement with the experimental value of $\phi_0 \sim 1.85\pi$. From the CEP dependences, we confirmed that the double ionization of methanol proceeds by the recollisional ionization.

[References]

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