Absolute carrier-envelope phase dependence in Coulomb explosion of acetonitrile induced by few-cycle intense laser pulses

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It has been revealed that the ionization and dissociation processes of molecules induced by a few-cycle laser pulse can be influenced by the changes in the carrier-envelope phase (CEP). Indeed, it was recently revealed that the dissociative ionization and hydrogen migration processes of methanol exposed to few-cycle intense laser pulses are affected sensitively by the CEP.[1] In the present study, we investigate the CEP dependences of the ejection direction of the fragments produced from two-body dissociation of acetonitrile (CH_3CN) cation and dication after the irradiation of few-cycle intense laser pulses to acetonitrile.

The acetonitrile was ionized by intense few-cycle laser pulses (6.2×10^{14} W/cm^2, 4 fs) and the three-dimensional momentum vectors of the respective fragment ions were determined by a coincidence momentum imaging spectrometer. The CEP was measured simultaneously by a phasemeter. Because the CEP obtained by the phasemeter has an offset with respect to the absolute CEP at the interaction point in the CMI chamber, the absolute CEP was calibrated based on the CEP dependence of the ejection direction of the high-energy photoelectrons (30-40 eV).[2]

The asymmetry parameters of the ejection direction of the fragment ions, P_{asym}, exhibit sinusoidal dependences on the absolute CEP, ϕ_{CEP}. The phase offset of the oscillation of P_{asym}, ϕ_0, in the CH_3^+ channel of acetonitrile cation was determined to be $\phi_0 = 0.96(5)\pi$. The CEP dependence was interpreted by the dependence of the ionization probability on the molecular orientation.[1] From the comparison with the CH_2^+ and CH^+ channels, we found a tendency that the phase offset becomes smaller as the number of migrated hydrogen atoms increases even though the extent of the decrease is as small as about 0.05π per migrated hydrogen atom. The phase offset of P_{asym} of the CH_3^+ channel and that of the CH_2^+ channel of acetonitrile dication were determined to be $\phi_0 = -0.01(3)\pi$, and $\phi_0 = 0.03(4)\pi$, respectively, both of which exhibit the phase shift of about π from the CH_3^+ channel of acetonitrile cation. The difference was interpreted as that induced by the recollision double ionization.

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