2P30 Ultrafast hydrogen migration in methanol probed by few-cycle laser pulses.

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Introduction

It has been revealed from our recent studies that, when hydrocarbon molecules are exposed to an intense laser field, ultrafast hydrogen migration proceeds, that is, hydrogen atoms within a hydrocarbon molecule migrate very rapidly in a femtosecond time scale. By using ultrashort laser pulses (38 fs) we performed pump-probe coincidence momentum imaging (CMI) measurements of the two types of two-body Coulomb explosion pathways of methanol, CH$_3$OH$^{2+} \rightarrow$ CH$_3^+$ + OH$^+$ (non-migration pathway) and CH$_3$OH$^{2+} \rightarrow$ CH$_2^+$ + OH$_2^+$ (migration pathway), and found that there were two different time scales in the ultrafast hydrogen migration processes, that is, the ultrafast hydrogen migration occurring within the laser fields (~40 fs) and that occurring after the laser-molecule interaction period (~150 fs). In the present study, in order to investigate the hydrogen migration in methanol with higher temporal resolution, we perform the pump-probe CMI measurements using few-cycle laser pulses.

Experimental Setup

The femtosecond laser pulses (0.7 mJ, 30 fs, 5 kHz, 800 nm) were focused into a hollow-core fiber filled with an Ar gas (0.5 atm) to broaden the spectral bandwidtth. The dispersion of the laser pulses was compensated by wedge plates and chirped mirrors to generate few-cycle pulses (8 fs). Pump pulses and probe pulses were produced by a Mach-Zehnder interferometer. The delay between the pump and probe pulses was modulated with a piezo-controlled translation stage in one of the two arms of the interferometer. The piezo-controlled stage was driven by a triangular wave voltage (0.2 Hz) to reduce the effect of a long term instability in the pulse duration of the few cycle pulses. The pump and probe pulses were introduced into a CMI chamber. The peak intensity of these pulses was estimated to be $4 \times 10^{13}$ W/cm$^2$.

Result and Discussion

Figures 1 show the observed momentum distributions of the fragment ions in the non-migration and migration pathway as a function of the time delay. In the momentum distribution of the non-migration pathway, two components (upper and lower strips) can be seen. The peak position of the momentum distribution of the lower strip decreases as the time delay increases. In the non-migration pathway, only a component corresponding to the lower strip can be identified. This gradual decrease in the released momentum appearing in the lower strip in both pathways can be interpreted by the gradual increase in the C-O internuclear distance after the ionization to the singly charged stage by the pump pulse.

The yield ratio of the migration pathway $\eta_{\text{mig}} = \gamma_{\text{mig}}/(\gamma_{\text{mig}} + \gamma_{\text{nonmig}})$ is found to exhibit a minimum at the delay time of 70 fs, where $\gamma_{\text{nonmig}}$ and $\gamma_{\text{mig}}$ denote the yield of coincidence events in the non-migration pathway and the migration pathway, respectively.

Reference