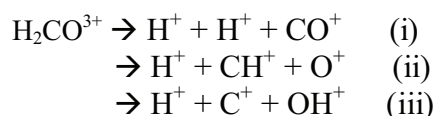**1P023**

Probing the nuclear dynamics of formaldehyde in 7 and 35 fs intense laser fields by
coincidence momentum imaging

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Momenta of fragment ions produced by molecular Coulomb explosion in ultrashort intense laser fields provide valuable information on the molecular structure just before the explosion. This "Coulomb explosion imaging" is a powerful tool to visualize ultrafast reaction dynamics, e.g., hydrogen migration, as demonstrated for the acetylene-vinylidene isomerization of $C_2H_2^{2+}$, $HCCH^{2+} \leftrightarrow CCH_2^{2+}$ [1]. Here, we report an ion coincidence momentum imaging study of Coulomb explosion of formaldehyde (H_2CO) in 7 fs and 35 fs intense laser fields (800 nm, 1.3×10^{15} W/cm^2) to understand the nuclear dynamics in intense laser fields and to seek the feasibility of applying the Coulomb explosion imaging to visualize the "roaming" hydrogen process discovered recently in the UV photodissociation of H_2CO [2].

The three-body explosion pathways identified at the present field intensity (1.3×10^{15} W/cm^2) are:

$$H_2CO^{3+} \rightarrow H^+ + H^+ + CO^+ \quad \text{(i)}$$
$$\rightarrow H^+ + CH^+ + O^+ \quad \text{(ii)}$$
$$\rightarrow H^+ + C^+ + OH^+ \quad \text{(iii)}$$

The Newton diagram of the major three-body dissociation pathway (i) in Fig.1(a) shows that the distribution of momentum angle between two protons is peaked at $\theta_{12} = 116°$, preserving the original $\angle$H-C-H bond angle ($\sim 116°$) of H_2CO in the neutral ground state, in the 7 fs intense laser fields. On the other hand, when the longer laser pulses (35 fs) were employed, a shift in the θ_{12} distribution peak ($\sim 106°$) as well as a broadening of the distribution were identified as shown in Figs. 1(b) and (d). In addition, an asymmetric energy partitioning between two H^+ ions was observed in 35 fs laser fields.
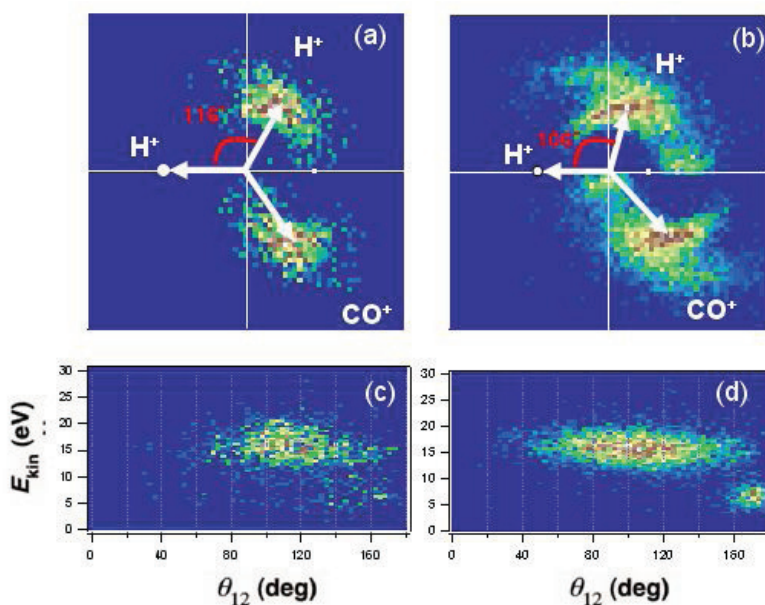

Fig.1. The Newton diagrams of $H_2CO^{3+} \rightarrow H^+ + H^+ + CO^+$ in (a) 7 fs and (b) 35 fs intense laser fields, respectively. The amplitude of the linear momentum of one of the H^+ ions is normalized to unity and the normalized vector is placed on the negative x-axis. Two dimensional maps of the momentum angle θ_{12} between $p_1(H^+)$ and $p_2(H^+)$ and the total kinetic energy release for $H_2CO^{3+} \rightarrow H^+ + H^+ + CO^+$, are shown in (c) and (d). The widths of the θ_{12} distribution at the half maximum are $\Delta\theta_{12} \sim 40°$ and $\sim 60°$ for 7 fs and 35 fs intense laser fields, respectively.

According to the theoretical calculation of the potential surface of H_2CO^{2+} [3], the $\angle$H-C-H bond angle is $\sim 60°$ at the equilibrium geometry of $H_2CO^{2+}(X\ ^1A_1)$. This suggests that the structural changes from θ_{12} (= 116°) of H_2CO to smaller $\angle$H-C-H bond angles can occur after the vertical excitation from neutral ground state in a short time scale. Moreover, the potential energy curve is repulsive along the stretching coordinate of one C–H bond (see Fig. 2). Therefore, in the 35 fs laser fields, the wavepacket launched on the dication potential will move toward the equilibrium geometry with reducing θ_{12} and elongating one of the C–H bonds prior to the formation of the tri-cation at the peak laser intensity, resulting in the smaller angle θ_{12} (=106°) and the asymmetric energy partitioning between two H^+ ions. On the other hand, the wavepacket formed in the dication state in 7 fs laser fields should be ionized without significant structural changes because the laser field intensity reaches its maximum within few optical cycles.

In conclusion, we found the geometrical structure of H_2CO significantly deforms in the 35 fs intense laser fields reflecting the dynamics in the diatonic states, while it is almost frozen during the interaction with 7 fs intense laser pulse, showing that the Coulomb explosion imaging using few-cycle intense laser pulses provides a mean to directly probe the changes of the geometrical structure of formaldehyde in chemical reaction as demonstrated for acetylene [4]. Experiments to visualize "roaming hydrogen" in the UV photodissociation of H_2CO are in process.
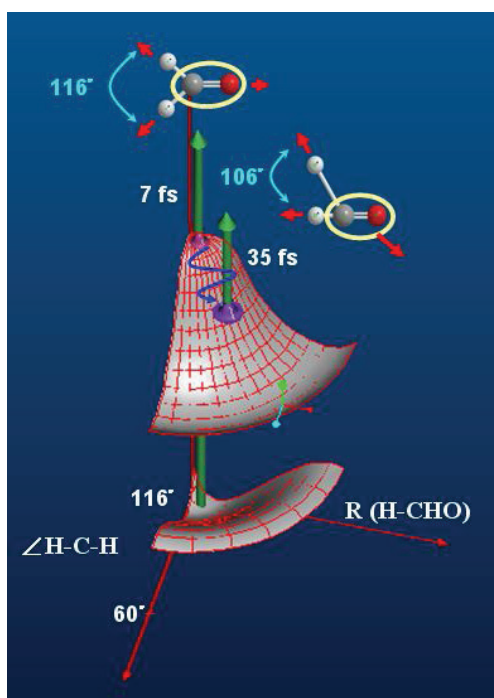

Fig. 2. Schematic potential energy surfaces of formaldehyde along one C–H bond distance R(H–CHO) and the $\angle$H-C-H bond angle.

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