Selective bond breaking of CO_2^{2+} in phase-locked two-color intense laser fields

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Synopsis Intensity dependence of selective bond breaking of CO_2^{2+} in phase-locked two-color intense laser fields has been investigated by using the three-dimensional coincidence momentum imaging. A clear intensity dependence of asymmetry parameter of the O^+ fragments detected in coincidence with CO^+ was observed in the range of 1-2×10^{14} W/cm^2, indicating that the mechanisms of selective bond breaking of CO_2^{2+} in phase-locked two-color intense laser fields involve laser-induced deformation of the potential energy surfaces and electron recollision excitation.

Molecules exposed to intense laser fields ($I \sim 10^{14}$ W/cm^2) show a variety of interesting dynamics. Since such molecular responses depend on the property of the applied laser electric fields, intense field coherent control has been demonstrated with several approaches to manipulate photodissociation dynamics at the quantum level. Two-color mixing is one of the interesting approaches, as successfully applied to the directional fragment ejection from symmetric molecules ranging from diatomic to polyatomic molecules. A previous theoretical study on CO_2^{2+} [1] predicts that the selective bond breaking in two-color intense laser fields is driven by laser-induced deformation of the potential energy surfaces. Recently, we investigated the intensity dependence of selective bond breaking of CO_2^{2+} in two-color laser fields to clarify the detail of the mechanisms [2].

The output of a Ti:Sapphire laser system (800 nm) was introduced to a β-BBO crystal to obtain 2ω pulses. The time-delay between the ω and 2ω pulses were compensated by two birefringent α-BBO crystals. The relative phase ϕ between the two pulses was locked by a feedback loop using a pair of fused silica wedges, and calibrated by separately measuring the energy distribution of the back-scattered electrons from Xe [3]. The polarization axes were paralleled to each other by a true-zero order dual-wavelength waveplate. Those pulses were focused on the molecular beam of CO_2 using a concave mirror (f = 75 mm) placed in an ultrahigh vacuum chamber. The fragment ions were accelerated to a position sensitive detector by four electrodes in a velocity map configuration. The three-dimensional momentum vector $\mathbf{p}$ of respective ions was obtained from the position (x, y) and the arrival time (t) at the detector. The momentum matching condition, $|\mathbf{p}_{CO^+} + \mathbf{p}_{O^+}| \leq 15$ a.u., was used for the coincidence detection.

Figure 1 shows the asymmetry parameters, $A(\phi) = [Y_+(\phi) - Y_-(\phi)]/[Y_+(\phi) + Y_-(\phi)]$, of O^+ fragments detected in coincidence with CO^+ at two different intensities. $Y_+(\phi)$ and $Y_-(\phi)$ are the yields of O^+ with positive and negative momenta along the laser polarization direction, respectively. At a total intensity of $I_{\omega+2\omega} = I_\omega + I_{2\omega} = 5.6 \times 10^{14}$ W/cm^2, the asymmetry has the maximum and minimum values at $\phi = 0$ and π, where the O^+ fragments are preferentially ejected to the larger field amplitude side. On the other hand, at the intensity of $I_{\omega+2\omega} = 0.76 \times 10^{14}$ W/cm^2, an increase of the amplitude from 0.04 to 0.07 and a positive shift of the phase providing the largest asymmetry are observed. These remarkable changes, observed in the range of 1-2 $\times 10^{14}$ W/cm^2, indicate that the mechanisms of the selective bond breaking of CO_2^{2+} in two-color intense laser fields involves laser-induced deformation of potential energy surfaces and electron recollision excitation.

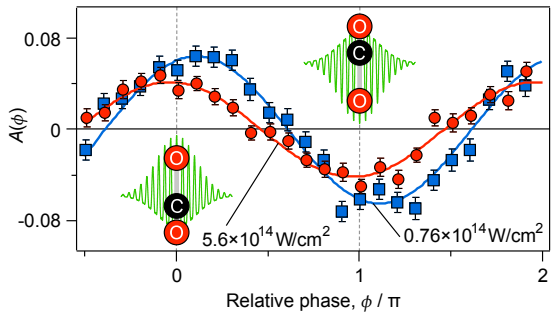

Figure 1. Asymmetry parameters of O^+ fragments at two laser field intensities, $I_{\omega+2\omega} = 5.6 \times 10^{14}$ W/cm^2 (red circle) and 0.76×10^{14} W/cm^2 (blue square).

References

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