

Dynamics of three-particle fragmentation of $(\text{CO}_2)_2^{3+}$ ions produced by intense femtosecond laser fields

P Song¹, X Wang¹, C Meng¹, Z Lv¹, D Zhang¹, Z Zhao¹, and J Yuan^{1,2*}

¹Department of Physics, National University of Defense Technology, Changsha 410073, P. R. China

²Department of Physics, Graduate School of China Academy of Engineering Physics, Beijing 100193, P. R. China

Synopsis We studied the breakup channel $(\text{CO}_2)_2^{3+} \rightarrow \text{CO}_2^+ + \text{CO}^+ + \text{O}^+$ in intense femtosecond laser fields. An analysis shows the three-body breakup through both concerted and sequential breakup channels. These two breakup channels are consistent with the instantaneous and metastable dissociation channels in the dissociation dynamics of CO_2^{2+} ion breakup into CO^+ and O^+ ions. Furthermore, we reconstructed the geometry of $(\text{CO}_2)_2$. By tracing the direction of the fragmentation ions, the O^+ was found to be produced from the C=O bond that is closer to the center of the C-C bond during direct dissociation.

Many-body fragmentation dynamics is crucial to the fundamental understanding of physical and chemical properties of polyatomic molecules. Owing to its complicated interactions of weak van der Waals and strong covalent bonds, it is a big challenge to study the geometric structure and dissociation process. With the development of experimental technologies, one of a feasible approach to explore the many-body dissociation dynamics is using the femtosecond laser Coulomb explosion imaging [1].

In this paper, we describe experiments concerning the laser-induced triple-ionization dissociation of the van der Waals complex $(\text{CO}_2)_2$ composed of two carbon dioxide molecules. Our experiment was performed on the platform of a momentum spectrometer (COLTRIMS) [2]. Dimers of the carbon dioxide molecules were generated in the supersonic expansion of CO_2 gas through a 30- μm nozzle with an 8-bar backing pressure. The linearly polarized laser pulses were centered at 790 nm with pulse duration of 25 fs and repetition rate of 10 KHz, delivered by a Ti:Sapphire multi-pass amplification system. The light intensity at the focus is estimated to be $4.5 \times 10^{14} \text{W/cm}^2$. An analysis shows that $(\text{CO}_2)_2^{3+}$ breaks up into $\text{CO}_2^+ + \text{CO}^+ + \text{O}^+$ ions through both concerted and sequential fragmentation channels. These two fragmentation channels are consistent with the instantaneous and metastable dissociation channels in the dissociation dynamics of CO_2^{2+} ion breakup into CO^+ and O^+ ions. From Coulomb explosion imaging, our results show that for the parallel sliding structure of the carbon dioxide dimer molecules, the angle between the C=O bond and the van

der Waals bond is 48° , and the intermolecular nuclear distance $R(\text{CO}_2\text{-CO}_2)$ is 4.0 Å. By tracing the direction of the fragmentation ions, the O^+ was found to be produced from the C=O bond that is closer to the centre of the C-C bond during direct dissociation. These results indicate that the dynamic characteristics of the monomer is retained and gives rise to new dynamics in the molecular cluster complexes

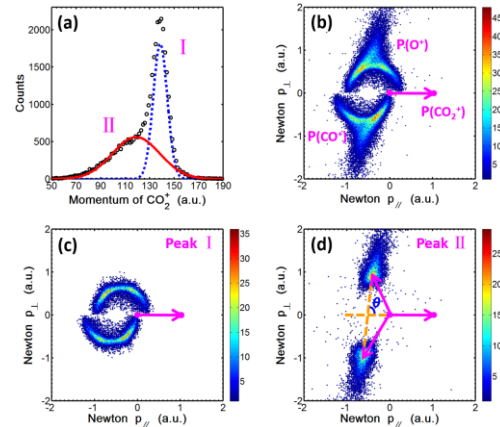


Figure 1. (a) Momentum distribution of CO_2^+ ion, black circle line is the experimental measurement data; solid red and dashed blue curves are the two Gaussian distributions of the fit. (b) Newton diagram of all events from dissociation channel $(\text{CO}_2)_2^{3+} \rightarrow \text{CO}_2^+ + \text{CO}^+ + \text{O}^+$. (c) and (d) Newton diagrams of the events corresponding to Peaks I and II, respectively. The corresponding momentum of CO_2^+ ion is (c) $125 \text{ a.u.} < |\mathbf{P}(\text{CO}_2^+)| < 155 \text{ a.u.}$, and (d) $|\mathbf{P}(\text{CO}_2^+)| < 125 \text{ a.u.}$ or $|\mathbf{P}(\text{CO}_2^+)| > 155 \text{ a.u.}$.

References

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* E-mail: jmyuan@nudt.edu.cn