Effects of laser pulse duration and intensity on Coulomb explosion of CO_2: signatures of charge-resonance enhanced ionization

Igor V. Litvinyuk[1,*], Irina Bocharova[1], Joseph Sanderson[2], Jean-Paul Brichta[3], Jean-Claude Kieffer[4] and François Légaré[4]

[1] J.R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, KS 66506, USA
[2] Department of Physics, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada
[3] Department of Physics, University of Ottawa, Ottawa, Ontario, Canada K1N 6N5
[4] Centre Énergie, Matériaux et Télécommunications, INRS, Varennes, QC, J3X 1S2, Canada

Synopsis We studied laser-induced Coulomb explosion of CO_2 by full triple-coincidence momentum resolved detection of resulting ion fragments. From the coincidence momentum data we can reconstruct molecular geometry immediately before explosion. We observe the dynamics of Coulomb explosion by comparing reconstructed CO_2 geometries for different Ti:Sapphire laser pulse durations (at the same intensity) ranging from few cycles (7 fs) to 200 fs. We conclude that for longer pulse durations (≥100 fs) Coulomb explosion proceeds through the enhanced ionization mechanism taking place at the critical O-O distance of 8 a.u., similarly to well known charge-resonance enhanced ionization (CREI) in H_2.

CREI at the critical internuclear distance is a well established mechanism for strong-field double ionization of hydrogen by sufficiently long (>20 fs) near-IR laser pulses [1, 2]. Whether a similar mechanism also operates during multiple ionization of larger molecules is still a matter of some debate. Here we present experimental evidence that multiple ionization of tri-atomic CO_2 molecules by intense femtosecond laser pulses can also proceed through enhanced ionization at a critical distance.

The experiments were performed on the 5 kHz Ti:Sapphire beamline of the Advanced Laser Light Source (ALLS) at Varennes, Quebec. A hollow-core fiber compressor was used to produce 7 fs pulses. A supersonic jet of CO_2 molecules intersected a tightly focused laser beam inside a uniform-electric-field ion spectrometer equipped with a time- and position-sensitive delay-line anode detector. All resulting ion fragments were detected and their full 3D momentum vectors were determined.

The triple coincidences of the type (O^{k+}, C^{l+}, O^{m+}) characterized by a near-zero total momentum were identified as resulting from Coulomb explosion of a single molecule into a specific (k,l,m) channel: $CO_2 \rightarrow O^{k+}+C^{l+}+O^{m+}$. For the (2,2,2) explosion channel we reconstructed the molecular structure, using an iterative procedure based on integration of the classical equations of motion for the fragments with pure Coulomb interactions.

We compared the results for various pulse durations from 7 fs to 200 fs (Figure 1). The 7 fs pulses generated the most energetic fragments and reconstructed structures were nearly identical to that of a neutral CO_2 molecule. As the laser pulses become longer the kinetic energy decreases and the O-O distance increases due to the dynamics occurring before the explosion. For 100 fs pulses the O-O distance reaches 8 a.u., and it remains the same for longer pulses.

We consider it a manifestation of the enhanced ionization process taking place in CO_2 molecular ions. However, the fragments in the (1,1,1) dissociation channel appear not to result from the enhanced ionization, pointing to the molecular tri-cation CO_2^{3+} as an intermediate state which undergoes this CREI-like process at the critical O-O separation of 8 a.u.

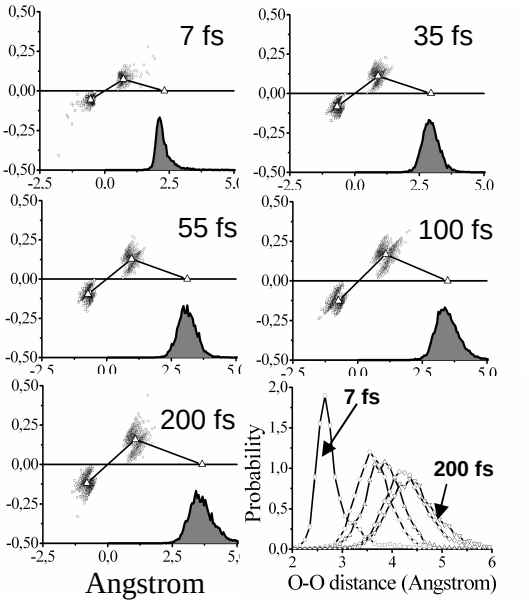

Fig. 1. Structures of CO_2 reconstructed for (2,2,2) explosion channel with different pulse durations. Note the different X- and Y-scales.

References:

1. T. Zuo and A.D. Bandrauk, Phys. Rev. A **52**, R2511 (1995).
2. T. Seideman, M.Y. Ivanov and P.B. Corkum, Phys. Rev. Lett. **75**, 2819 (1995)

* ivl@phys.ksu.edu