

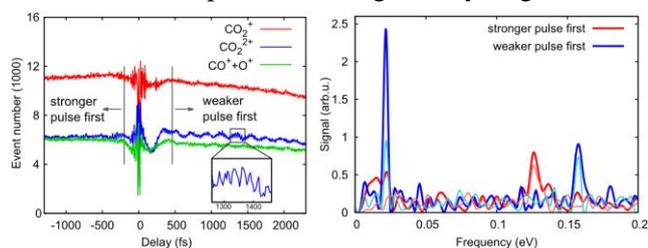
## Two-Pulse Control over Double Ionization Pathways in CO<sub>2</sub>

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**Synopsis** Using a double-pulse scheme we visualize and control molecular dynamics taking place on intermediate states populated during different sequential double ionization pathways of CO<sub>2</sub>. Exchanging the pulse-sequence can almost completely switch the pathway.

When an atom or a molecule interacts with a strong laser field multiple ionization may take place. In this process the final ionic state may be reached via different pathways, which in the case of molecules may give rise to strongly different nuclear dynamics on the intermediately populated potential energy surfaces. We report on experiments that employ a sequence of two delayed pulses for controlling and visualizing the sequential double ionization pathway in CO<sub>2</sub>. In the experiments, we measured electrons and ions in coincidence from the ionization and dissociation of CO<sub>2</sub> molecules using the COLTRIMS technique [1]. Measurements are done with two 795 nm, 25 fs laser pulses, with peak intensities of about  $1 \times 10^{14} \text{ W/cm}^2$  and  $2 \times 10^{14} \text{ W/cm}^2$ , respectively. The time delay between the two pulses was varied in steps of 4 fs using a delay stage.



**Figure 1.** (a) Measured ion signals of CO<sub>2</sub><sup>+</sup>, CO<sub>2</sub><sup>2+</sup> and CO<sup>+</sup>/O<sup>+</sup> over time delay. Positive time delay means the weaker pulse comes first. (b) Fourier-transformed spectra of the yields from (a), separately for positive and negative time-delays for CO<sub>2</sub><sup>2+</sup> (blue and red, thicker lines) and CO<sup>+</sup>/O<sup>+</sup> (orange and light-blue, thinner lines).

The measured signals of CO<sub>2</sub><sup>+</sup>, CO<sub>2</sub><sup>2+</sup> and CO<sup>+</sup>/O<sup>+</sup> are shown in Fig. 1 (a) as a function of the time delay between the two pulses. It can be clearly seen that the signals oscillate over time delay. There are at least two types of oscillations, a slow oscillation and a fast oscillation, as shown in the inset. To get insight to the oscillation pattern in CO<sub>2</sub><sup>2+</sup> and CO<sup>+</sup>/O<sup>+</sup>, we performed Fourier transforms of the signals over time delay. To avoid the region where the two pulses temporally overlap, we separated the scanned signal into left and right parts, as indicated in Fig. 1 (a). Fourier transforms are shown in Fig. 1 (b). The lower frequency peak at 0.022 eV (177 cm<sup>-1</sup>) corresponds to the slow oscillation in the signal

with a period of 190 fs. There are two higher frequency peaks at 0.126 eV (1016 cm<sup>-1</sup>) and 0.158 eV (1274 cm<sup>-1</sup>), which correspond to oscillation periods of 33fs and 26 fs, respectively. We assign the frequency peak at 0.158 eV to the symmetric stretch vibrational mode on the X-state of CO<sub>2</sub><sup>+</sup>, while the peak at 0.126 eV is assigned to dynamics on the excited A-state [2].

We found that the higher vibrational frequency peak in CO<sub>2</sub><sup>2+</sup> and CO<sup>+</sup>/O<sup>+</sup> moves from 0.158eV to 0.126 eV when we exchange the sequence of the two laser pulses. We explain this observation as follows. When the first pulse is weak, it singly ionizes the CO<sub>2</sub> molecule by removing a HOMO electron and the molecule ends at the X-state of CO<sub>2</sub><sup>+</sup>. The second strong pulse further ionizes the cation to the dication. In such a scenario, the measured peak at 0.158 eV reflects the vibrational dynamics on the X-state of CO<sub>2</sub><sup>+</sup>. However, when the strong pulse comes first, it can remove an electron from the HOMO-1 orbital and prepares CO<sub>2</sub><sup>+</sup> in its A-state. Because of its low intensity the following pulse can only further ionize CO<sub>2</sub><sup>+</sup> on the A-state, which explains why only the vibrational frequency at 0.126 eV from the A-state is present. From the measured frequency signal it can be seen that we can almost completely switch one pathway (through X-state) to the other path (through A-state) by simply exchanging the sequence of the two laser pulses. Thus, using a double-pulse scheme we could visualize and control molecular dynamics taking place on intermediately populated states during different sequential double ionization pathways of CO<sub>2</sub>. During the conference, we will also discuss the origin of the low frequency peak.

### References

- [1] X. Xie *et al* 2012 *Phys. Rev. Lett.* **109** 243001
- [2] A. Gharaibeh and D. J. Clouthier 2010 *J. Chem. Phys.* **132** 114307

