MFPADs as tool to experimentally prove O K-shell hole localization in CO_2 due to asymmetric stretching

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Synopsis Here we present an experiment on photoionization of the O 1s core-electron of CO_2. We want to test if excitation of asymmetric vibrational modes in the molecule lead to localization of the hole by measuring the vibrationally resolved MFPADs (Molecular Frame Photoelectron Angular Distributions).

The dynamical localization of the O 1s core-hole of CO_2 caused by an asymmetric stretch motion of the molecule has been predicted by Cederbaum *et al.* in 1999 [1]. Although widely accepted this has been untested by experiments until today.

Our measurements were carried out at the beamline P04 at PETRA3 (Hamburg, Germany), where a monochromatic circularly polarized photon beam interacts with the CO_2 gas jet. The absorption of a photon leads to the ejection of a photoelectron from the K-shell leaving an O 1s core-hole behind. Singly ionized CO_2^+ ($1s^{-1}$) can be left either in the vibrational ground state or in an asymmetric stretch vibrational mode. The molecule then decays to CO_2^{++} by emission of an Auger electron. The doubly charged ion dissociates into CO^+ and O^+ with a kinetic energy release (KER) of up to 11 eV. All charged fragments, namely the photoelectron, the Auger electron, and the two ions are detected in coincidence by our COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) reaction microscope [2]. For the electron spectrometer arm we made use of a retarding region to achieve sub 300meV resolution on a 20eV photoelectron. For the ion arm of our spectrometer we took advantage of an electrostatic lens in combination with time-of-flight focusing to get high momentum resolution on the ionic fragments.

As can be seen from the calculations shown in Figure 1, measured MFPADs will provide a clear fingerprint of localization or delocalization of the hole. A calculation based on the assumption of a localized hole (green and purple) predicts a highly asymmetric MFPAD while the calculation assuming a delocalized hole predicts a symmetric MFPAD at an electron energy of 20eV. Once we are able to distinguish between the vibrational states (vibrational splitting 307 meV), we can check whether the ground vibrational state shows symmetric MFPAD and the asymmetric stretch excited molecule shows an asymmetric MFPAD.

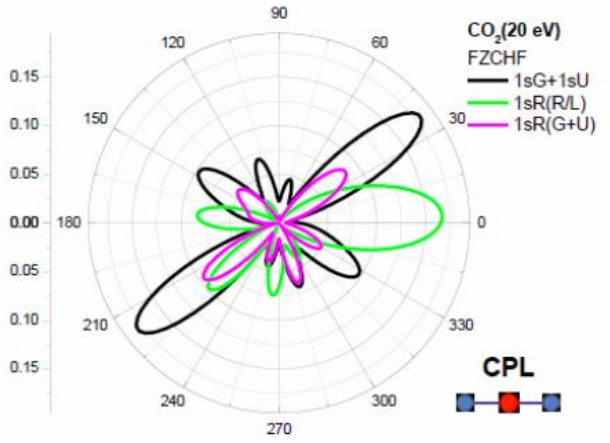

Figure 1. MFPAD for circularly polarized light (CPL). The orientation of the molecule is displayed in the bottom right corner. Under the Frozen Core Hartree-Fock (FZCHF) assumption, where G and U mean gerade and ungerade, and R and L right and left; the black line shows the calculation for a delocalized hole, while the purple and green lines show two different predictions both assuming a localized hole.

References

[1] L. S. Cederbaum *et al* 1999 *Physical Review A,* **60** 1988.
[2] R. Dörner *et al* 2000 *Physics Reports,* **330** 95

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