Towards rovibrationally resolved photochemistry in H_2O-CO_2

A.S. Bogomolov[1], S. Collignon[1], N. Moazzen-Ahmadi[2], M. Herman[3], and C. Lauzin[1]

[1]*Université catholique de Louvain, Louvain-la-Neuve, 1348, Belgium*
[2]*University of Calgary, Calgary, T2N 1N4, Canada*
[3]*Université libre de Bruxelles, Brussels, B-1050, Belgium*

Water and CO_2 are the most important greenhouse gases in our atmosphere. The solvation of CO_2 is a decisive process in the chemistry of clouds and oceans. A precise characterization of the interaction of these two molecules is thus of prime importance. In the present work, we recorded a rotationally resolved spectrum of the triple excitation of the OH stretch in the H_2O-CO_2 molecular complex. This represents a further step in our longstanding effort to better characterize the dynamics of CO_2-H_2O interactions, notably through a systematic increase in vibrational excitation [1–4]. Increasing excitation will ultimately drive the isomerization of the complex into carbonic acid H_2CO_3. The complexes were formed from an 8 cm-long pulsed slit supersonic jet and probed by the CRDS technique in the spectral range of the second OH overtone of H_2O, using the FANTASIO setup [5,6]. A home-built external cavity diode laser (ECDL), stabilized to the frequency comb, was used as the light source. The recorded spectrum was vibrationally assigned to the $(v_1,v_2,v_3) \leftarrow (v_1',v_2',v_3') = (2,0,1) \leftarrow (0,0,0)$ and (2,0,1) + intermolecular mode ← (0,0,0) where the v_1,v_2,v_3 vibrational quantum numbers of the isolated H_2O molecule are used to assign the involved states. In this talk, I will present the experimental spectrum and the improvements to the experimental setup that enabled the recording of this spectral signature. I am going to present our progress in the analysis of this spectrum using group theory and the effective Hamiltonian. Finally, perspectives on going across the isomerization barrier will be presented.

[1] C. Lauzin, A.C. Imbreckx, T. Foldes, T. Vanfleteren, N. Moazzen-Ahmadi, and M. Herman, Mol. Phys. 118, e1706776 (2020).
[2] A.S. Bogomolov, A. Roucou, R. Bejjani, M. Herman, N. Moazzen-Ahmadi, and C. Lauzin, Chem. Phys. Lett. 774, 138606 (2021).
[3] T. Gartner, C. Lauzin, A.R.W. McKellar, and N. Moazzen-Ahmadi, J. Phys. Chem. A 127, 3668 (2023).
[4] A.S. Bogomolov, S. Collignon, R. Glorieux, N. Moazzen-Ahmadi, M. Herman, C. Lauzin, Rotationally resolved triple vibrational excitation in D_2O-CO_2 van der Waals complex, in preparation (2026).
[5] M. Herman, K. Didriche, D. Hurtmans, B. Kizil, P. Macko, A. Rizopoulos, and P.V. Poucke, Mol. Phys. 105, 815 (2007).
[6] A.S. Bogomolov, R. Glorieux, M. Herman, T. Corbo, S. Collignon, B.M. Hays, D. Lederer, N. Moazzen-Ahmadi, A. Libert, B. Tomasetti, J. Fréreux, and C. Lauzin, Mol.Phys. 123, e2413417 (2025).