

Towards the quantum mechanical calculation of reaction rates

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Towards the quantum mechanical calculation of reaction rates

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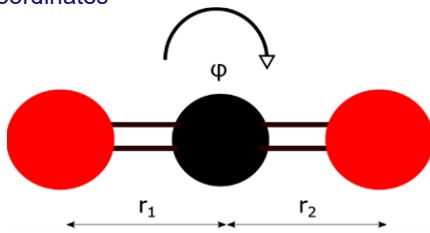


Introduction

The high CO₂ concentration in the air is driving global warming. Methods for the destruction of CO₂ molecules are investigated to limit its concentration. The most promising method uses vibrational pumping. The vibrational-vibrational reactions are poorly understood and occur in PLASIMO's CO₂ global model via empirical scaling relations. The quality of these relations is unknown. For that reason a quantum mechanical calculation of these rates is investigated. The main bottleneck is the calculation of the 11D potential.

Possible configurations

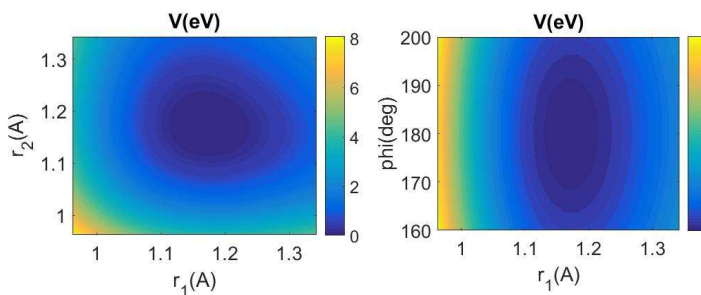
The internal structure of a CO₂ molecule can be described with 3 coordinates



An interaction between two of these CO₂ molecules therefore already contains 6 degrees of freedom. The remaining degrees of freedom include the separation between the center of mass (COM) of both molecules, the rotation along the COM of molecule 2 and the rotation of molecule 2 around molecule 1

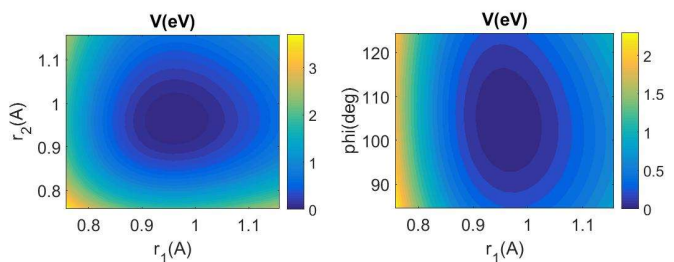
Potential surface CO₂

As a first step the potential energy surface for a single molecule is calculated. The left figure shows results for the variation of r_1 and r_2 and the right figure for r_1 and ϕ .



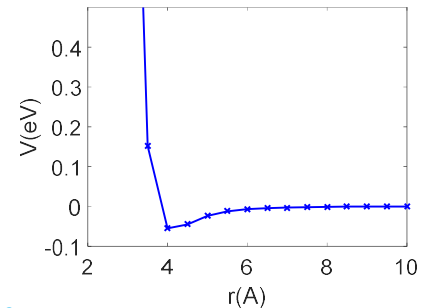
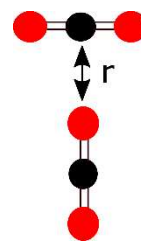
Potential surface H₂O

Since the final goal of the project is to create CH₄ the impact of H₂O on the chemistry is important as well.



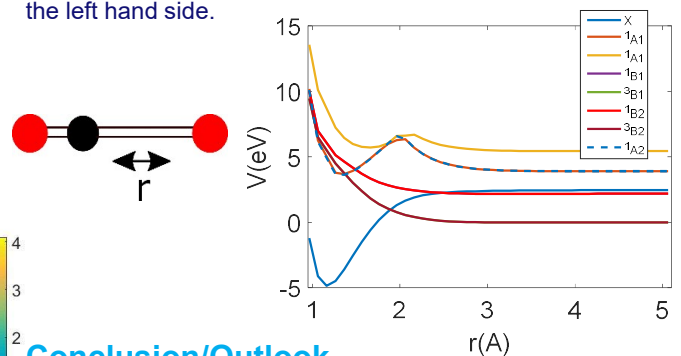
Interaction of two CO₂ molecules

Two CO₂ molecules collide as indicated in the drawing on the left hand side.



CO₂ dissociation

The potential energy curves for the CO₂ molecule are calculated for the dissociation as indicated in the figure on the left hand side.



Conclusion/Outlook

The CCSD procedure gives reliable results for the calculation of the potential energy. Further work focusses on using the optimal basis set and determining the 11D grid in such a way that all features can be captured while the calculation time remains reasonable.