



M⁺ (M=Ca, Yb) Cations Bound to Molecular Cavities: A New Strategy for Incorporating Molecular Quantum States into Quantum Information

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Introduction / Motivation

Current quantum computers utilize trapped quantum ion technology to store information in qubits (quantum bits). The trapped ion's quantum information can be transferred using laser induced coupling between qubit states. Currently only one qubit of information can be stored per trapped ion.

- This project aims to explore the vibrations and potential energy surfaces (PES) for motion of ions commonly used in quantum ion traps (e.g. Ca⁺, Yb⁺, Ba⁺) within molecular cavities of varying size. A series of nanohoops, [n]cycloparaphenylenes ([n]CPP), serve as molecular cavities containing n=5-18 phenyl rings.
- The results of this project aim to inform collaborators at SNL of the most promising ion-molecular cavity combinations to be further studied experimentally through Coulomb crystals. A long-term goal of this project is to inform the future design of higher capacity quantum computers.

Approach

The approach of this project is to utilize Gaussian software to optimize the structures and determine the binding energies and potential energy surfaces of the complexes of interest. We use Density Functional Theory (DFT) with a ω B97X-D functional and the 6-31+g(d) basis. Using these parameters, the lowest energy structures of the complexes are optimized and corresponding vibrational modes and frequencies found. The potential energy surfaces (PES) and binding energies were also found using these parameters.

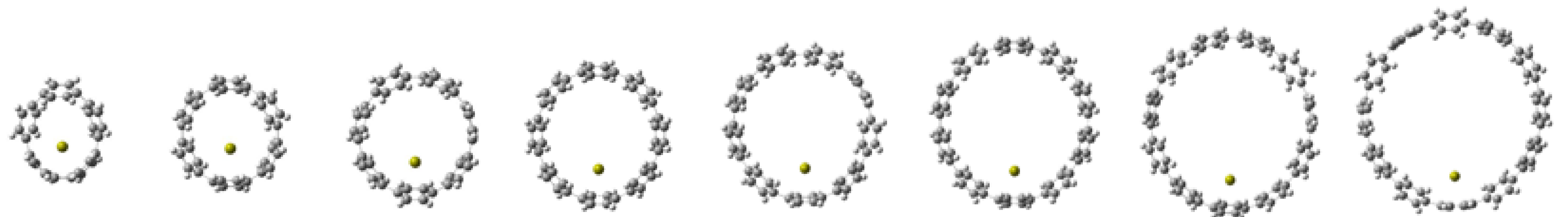


Figure 1 (left to right): Optimized nanohoop-Ca⁺ complexes of ring size [5-12]CPP. The PES included restraints to fix the ion at certain locations within the nanohoop. The PES capitalizes on the 'n'-fold symmetry of the [n]CPP cavity. Only 1/nth of the PES needed to be actively modeled, as the results were symmetric around the nanohoop per phenyl ring.

Current Status / Results

We have optimized the structures of complexes of [5-12]CPP with Ca⁺. In each ring, ([5-12]CPP) the ion gravitates towards the nanohoop wall. The nanohoop distorts in response to the ion's location. This distortion is more prevalent as ring size decreases. Binding energies have been calculated. Potential energy surface scans were conducted for [5-8]CPP-Ca⁺ systems, showing a general trend towards a 'sombbrero' shape as ring size increased.

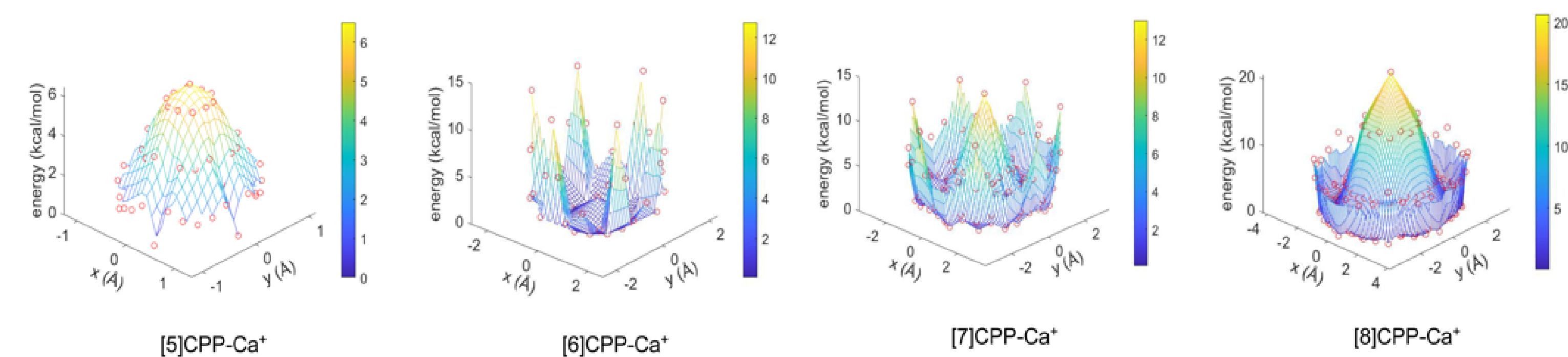


Figure 2 (left to right): Potential energy surfaces for [5-8]CPP-Ca⁺.

Optimized structure and binding energy calculations of tricyclophane are in progress to probe the molecule's viability as a molecular cavity.

Table 1: Binding Energies

[n]CPP	ΔE_{Bind} (kcal/ mol)
[5]CPP	-56.79
[6]CPP	-39.33
[7]CPP	-36.97
[8]CPP	-35.89
[9]CPP	-35.26
[10]CPP	-34.60
[11]CPP	-34.29
[12]CPP	-34.25

Challenges

Computational expenses for larger rings are high using density functional theory methods. A longer-term goal of this work is to develop a force field capable of accurately modeling the complex due to the unusual oxidation state of the ion.

Acknowledgments

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Next Steps / Future Work

- Optimize the structures of key excited electronic states of Ca⁺ used in quantum computing in order to predict the visible absorption spectrum for comparison with experimental data obtained at Sandia.
- Develop a polarizable force field for modeling ions with unusual oxidation states (Ca⁺). The polarizable force field will be used to continue the work for Ca⁺-[n]CPP (n=13-18) complexes.
- Explore the same aspects with Yb⁺ and Ba⁺ (two other common trapped ions).
- Explore tricyclophane and other cyclophanes as potential molecular cavity candidates