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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 B3LYP functional and the 6-31+g(d) basis set with the gd3bj dispersion correction. Using these parameters, the lowest energy structures of the complexes and transition state paths are optimized and corresponding vibrational modes and frequencies found.

Comparison of the optimized complex energies and the transition state energies allow us to find the potential energy barrier to movement of the Ca$^+$ between minima and consequently around the ring.

The potential energy surface scan will show a more detailed surface to ion movement in the complex.

Binding energies will be calculated for Ca+ to the nanohoop as a function of nanohoop size.

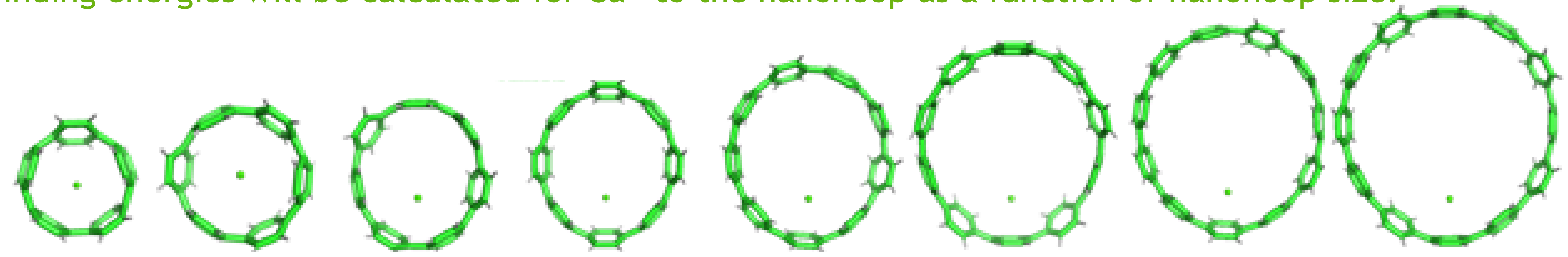

Figure 1 (left to right): Optimized nanohoop-Ca+ complexes of ring size 5cpp-12cpp.

Current Status/ Results

We have optimized the structures of complexes of [5-12]CPP with Ca$^+$. In smaller rings ([5-6]CPP) the ion prefers a position at the center of the nanohoop. In larger rings ([7-12]CPP) the ion gravitates towards the nanohoop wall. Transition states and vibrational modes for adjacent and opposite minima have been calculated for [7-9]CPP-Ca$^+$.

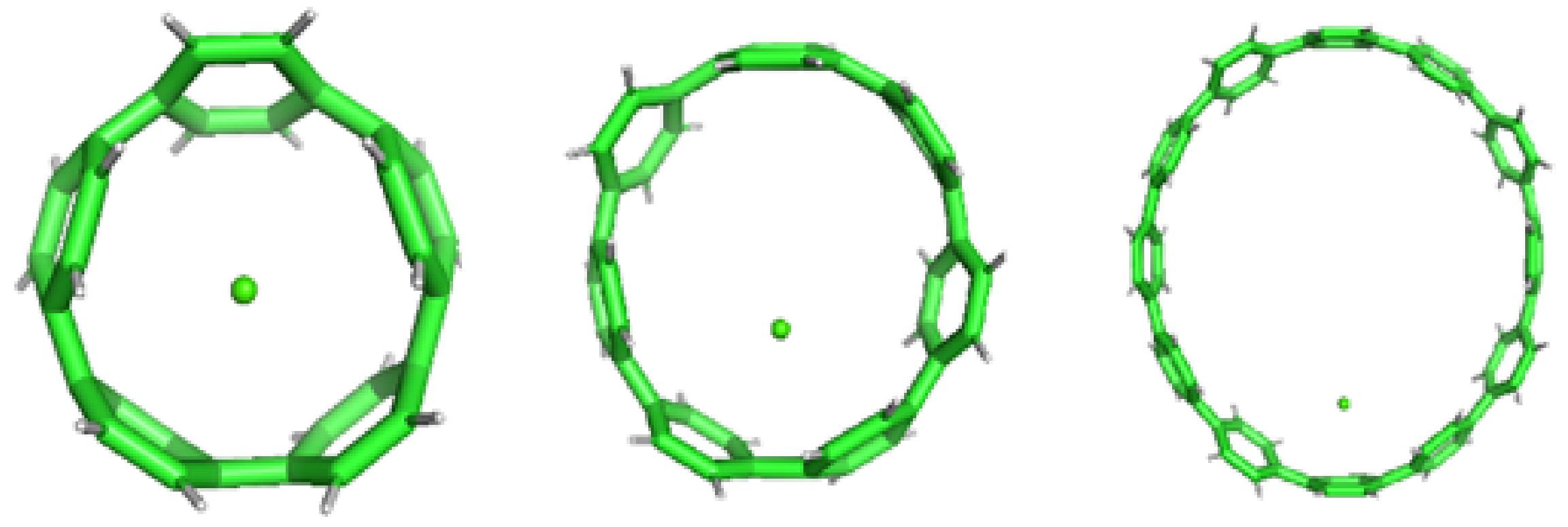

Figure 2 (left to right): Optimized ground state structures for 5, 8, and 12cpp-Ca+ complexes

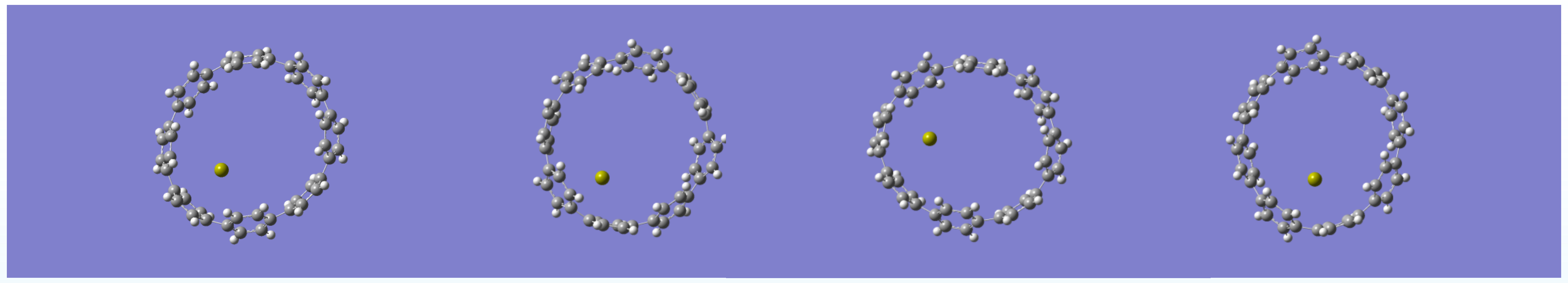

Figure 3 (left to right): 8cpp-Ca+ adjacent minima TS path and frequency, 8cpp-Ca+ opposite minima TS path and frequency

Potential energy surface scans are in progress for 5-10cpp-Ca+ which will capitalize on the 'n'-fold symmetry of the [n]CPP cavity. Only 1/nth of the PES needs to be actively modeled, as the results will be symmetric around the nanohoop per phenyl ring.

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

- Complete transition state analysis, explore relaxed potential energy surfaces and obtain accurate binding energies.
- 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^{+1}). 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).

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