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Photo/Redox-Responsive 2D-Supramolecular Assembly Involving Cucurbit[8]uril and a Star-Shaped Porphyrin Tecton

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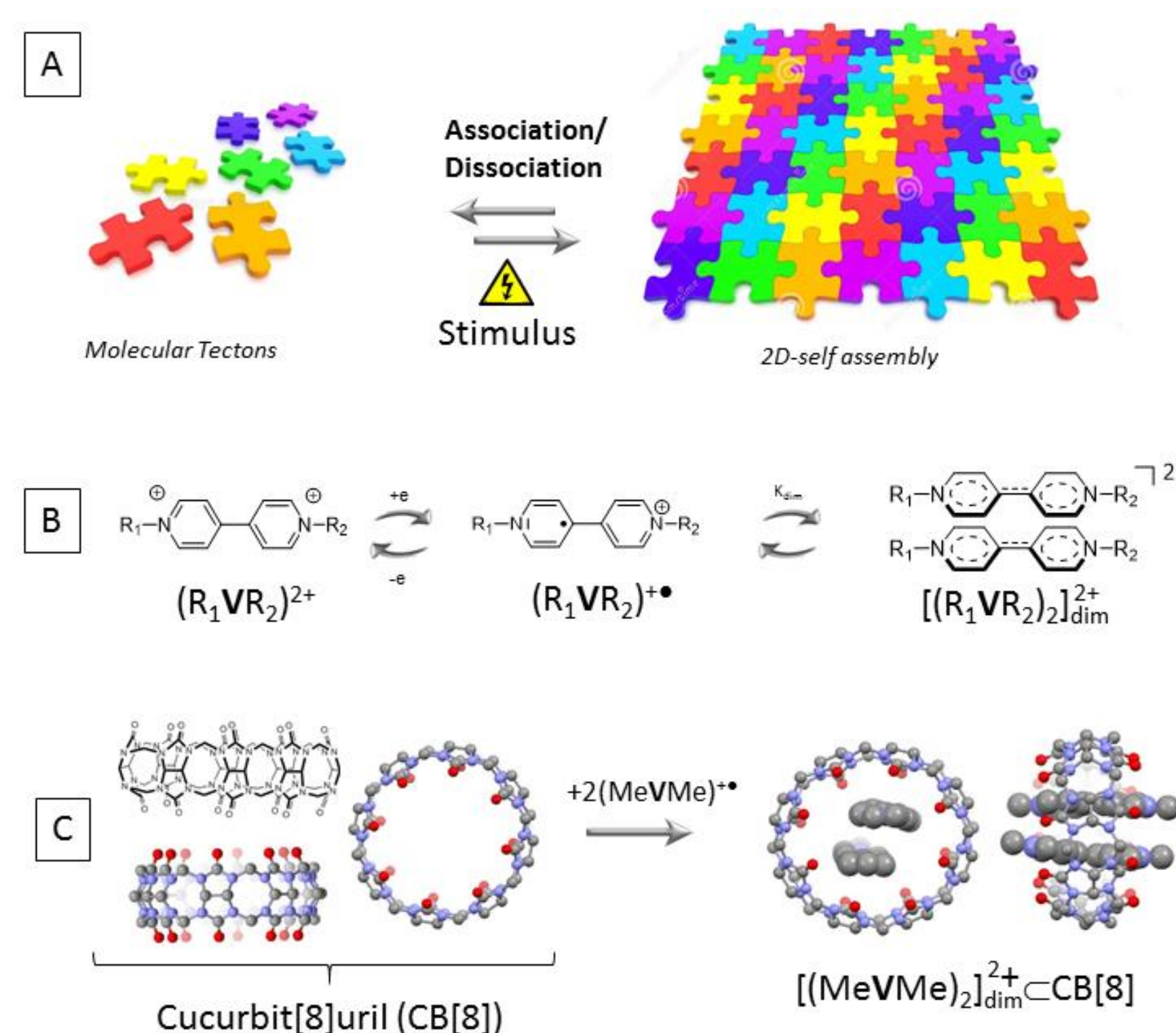
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Introduction

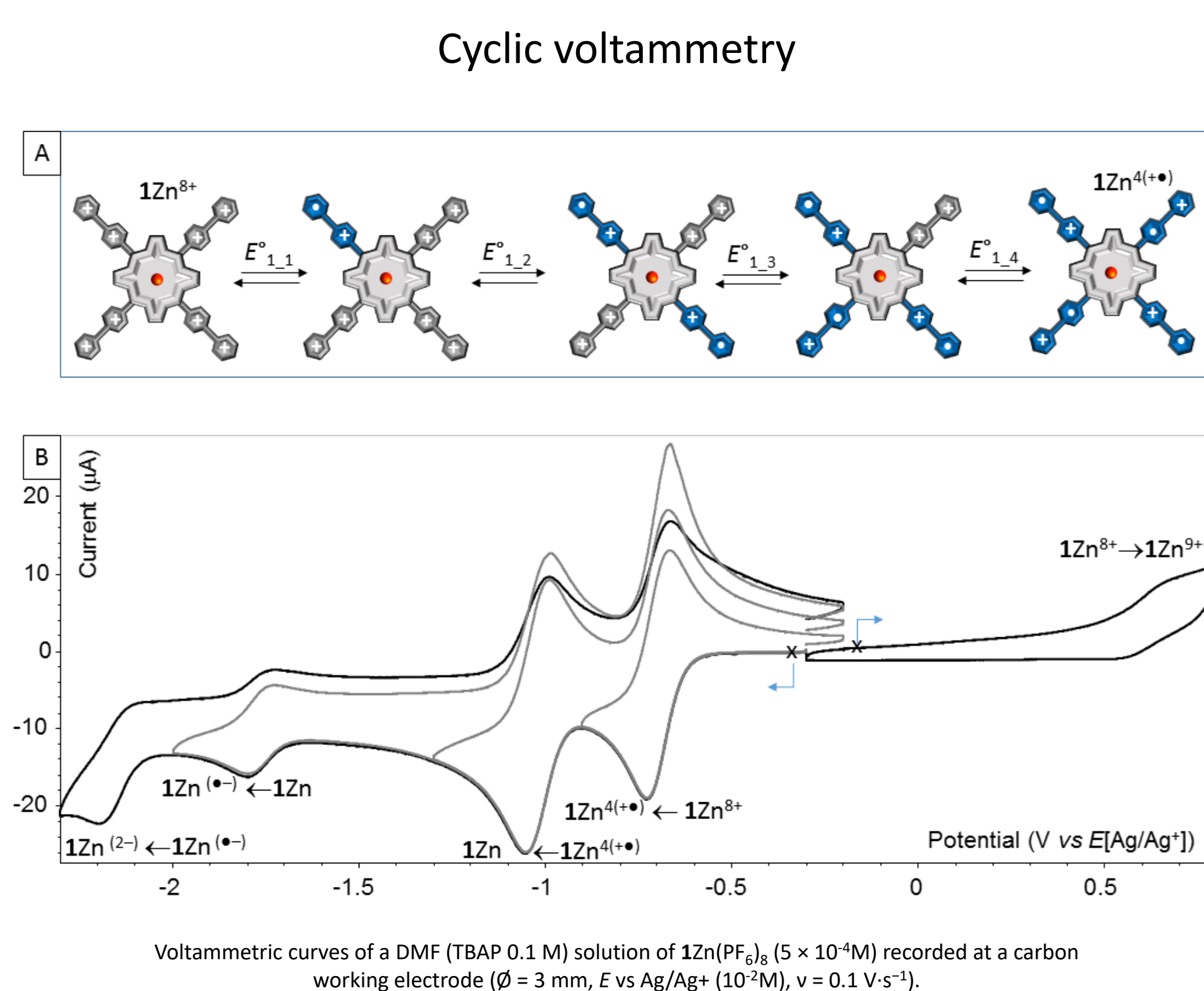
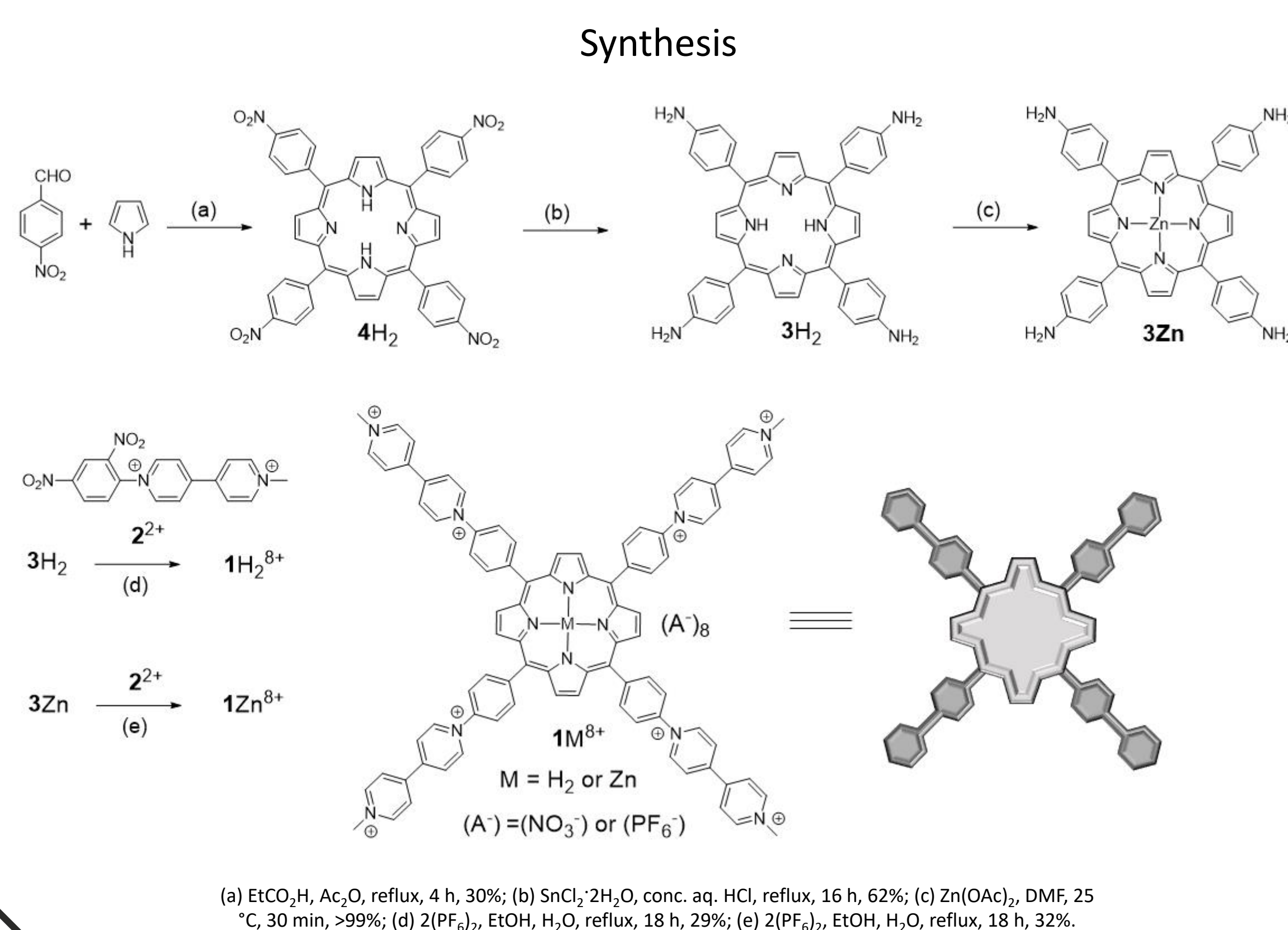
Supramolecular assemblies whose association or organization between tectons can be controlled by external stimuli have been found to be of great interest in various areas such as molecular electronics, analytical chemistry and materials science.^[1] In this context, we are interested in (supra)molecular metamorphism involving dimerization of π -conjugated radicals as driving force. Our group has been focusing over the past few years on the development of tailor-made redox-controllable molecular or supramolecular systems involving electro-generated viologen-based π -cation radicals as key responsive and/or assembling elements, using their ability to self-assemble into sandwich-like, dimeric entities called π -dimers. Such π -dimers can be observed in standard temperature and concentration ranges, when using barrel-shaped cavitands, known as cucurbit[8]urils, whose inner cavity is ideally suited to the inclusion of two viologen-based radicals.^[2]



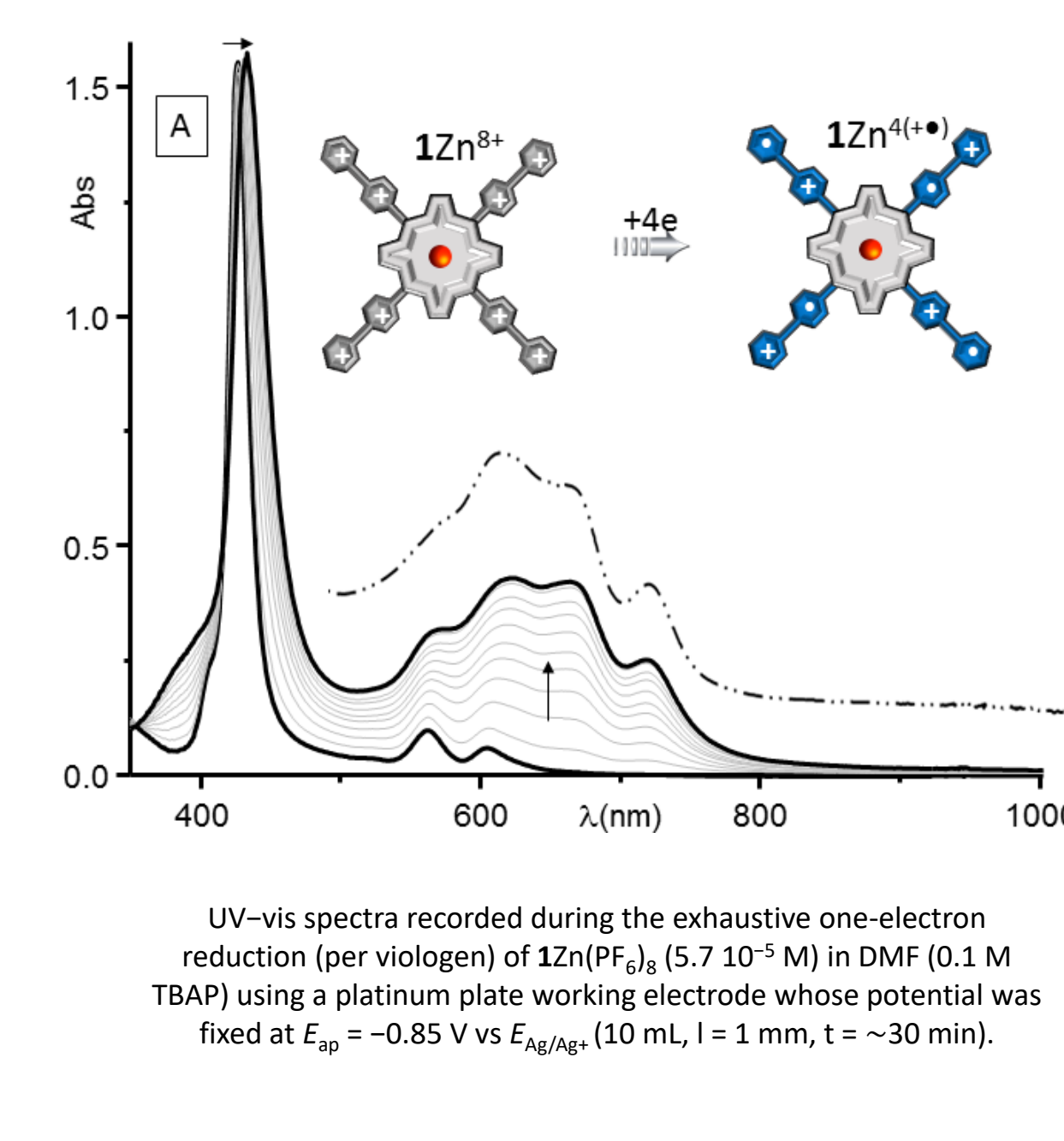
Conclusion

- Formation of discrete 4:1 pseudo-rotaxane-like caviplexes: 4 CB[7 or 8] / viologen-based star shaped porphyrin guest
- Formation of intermolecular π -dimers requires CB[8] acting as an assembling element (electrochemical, theoretical and spectroscopic analyses)
- CB[8] hosts proved useful to promote the redox-triggered formation of a 2D-supramolecular assembly in solution (electrochemical, chemical or photochemical stimulus)

Star-shaped porphyrin tecton



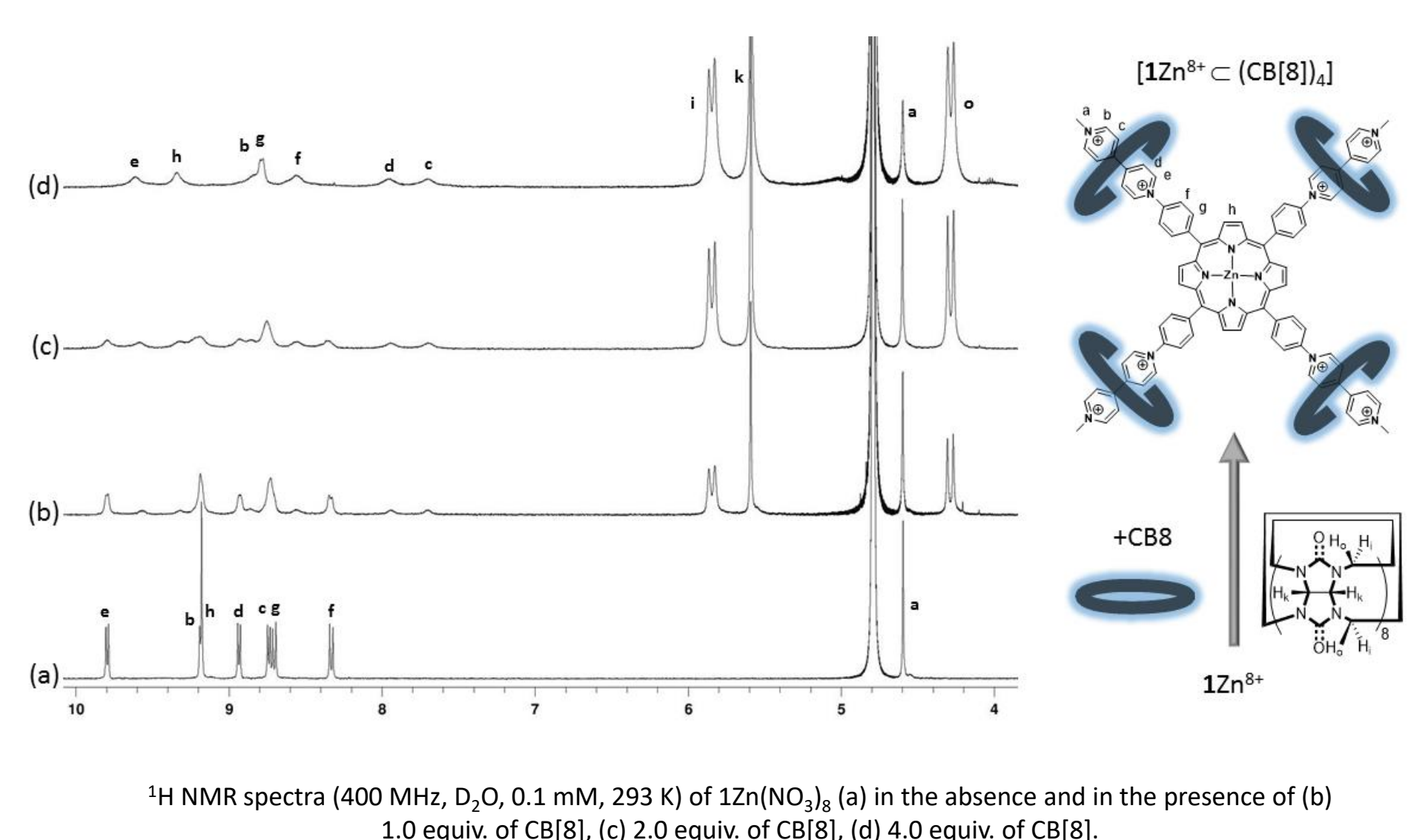
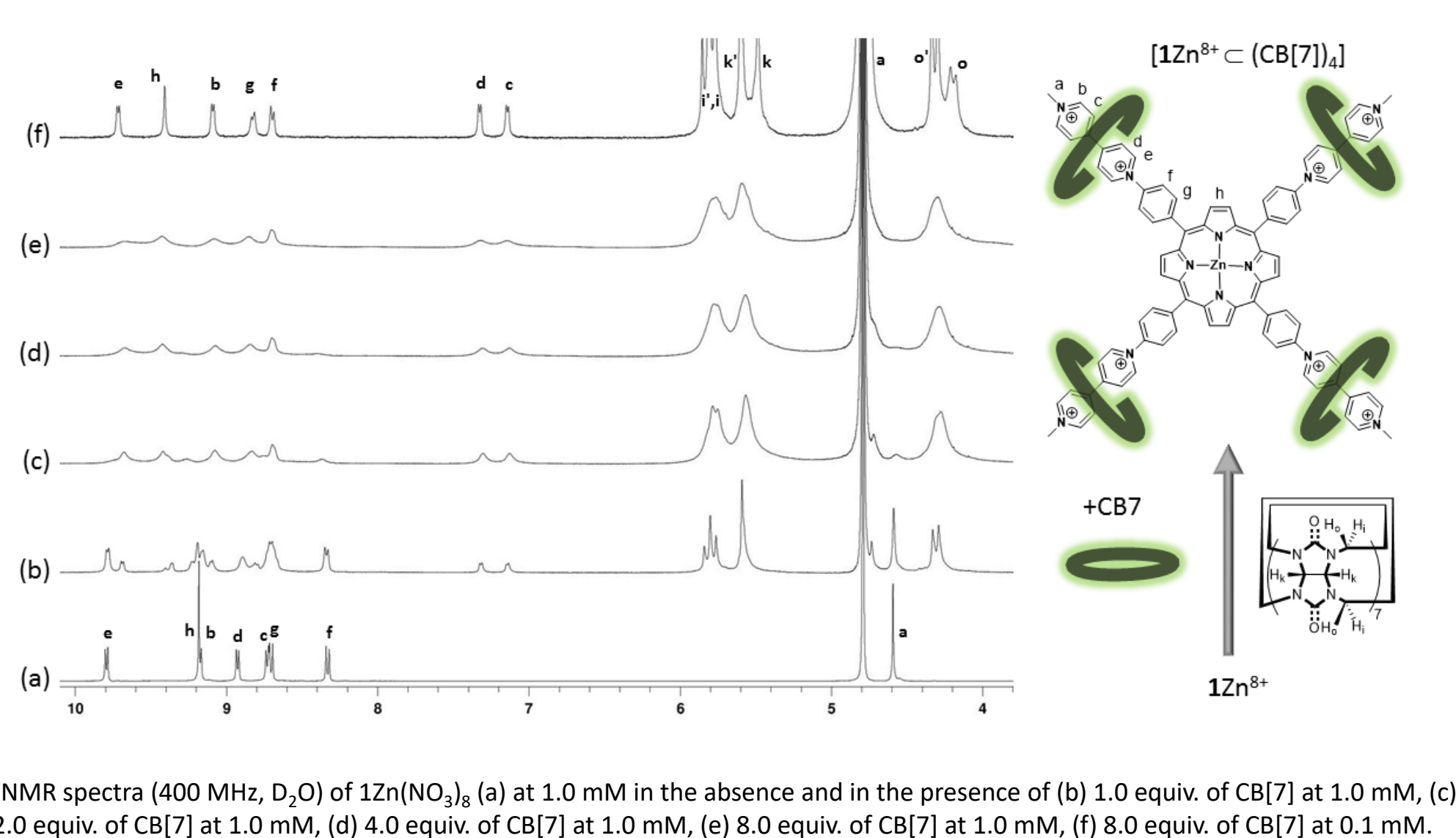
Spectro-electrochemistry



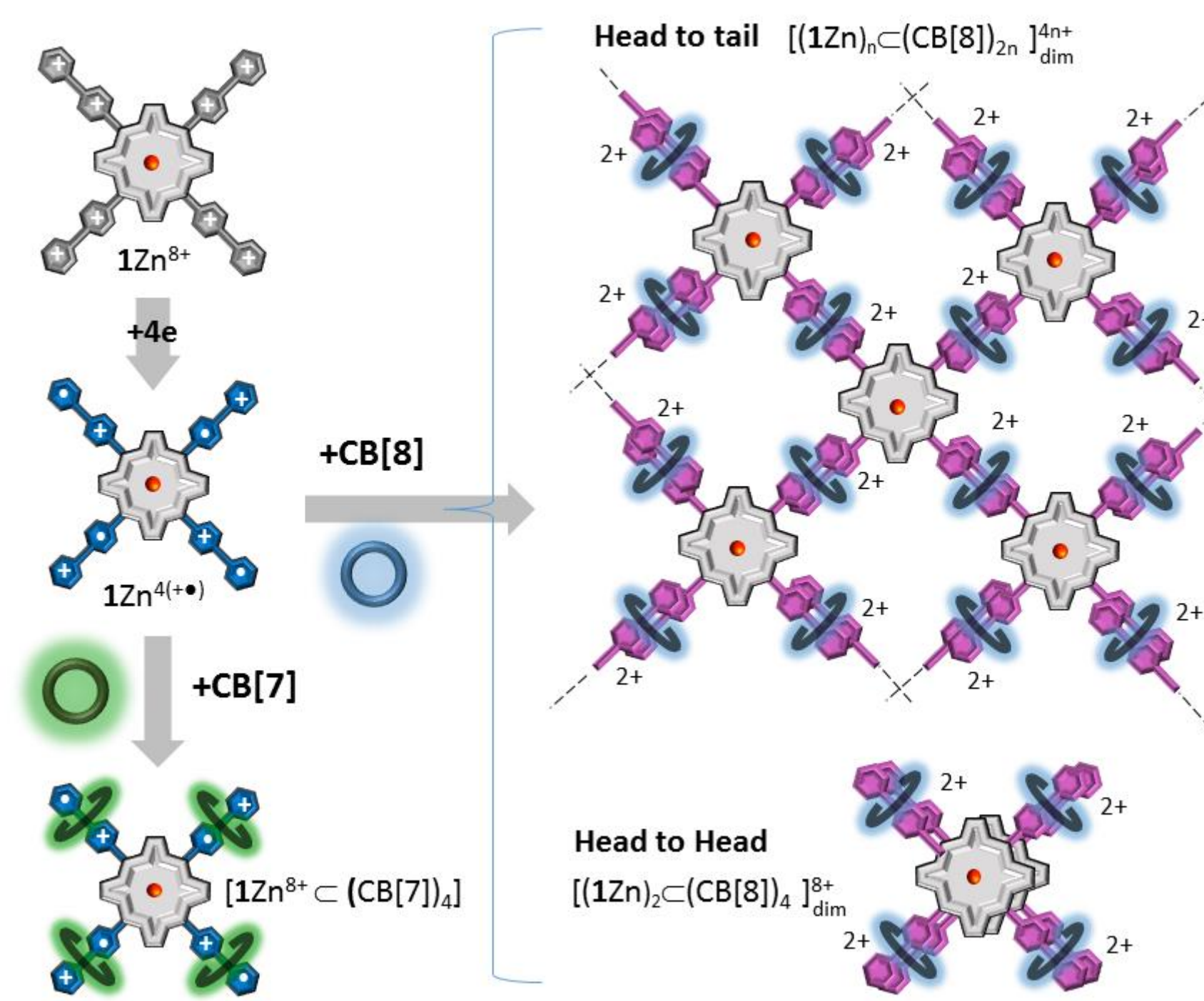
Supramolecular assembly involving cucurbituril

We have used this star-shaped porphyrin tecton to control the association/dissociation process of a 2D-supramolecular self-assembly with cucurbit[8]urils by means of electrochemical, chemical, and photochemical stimuli.^[3]

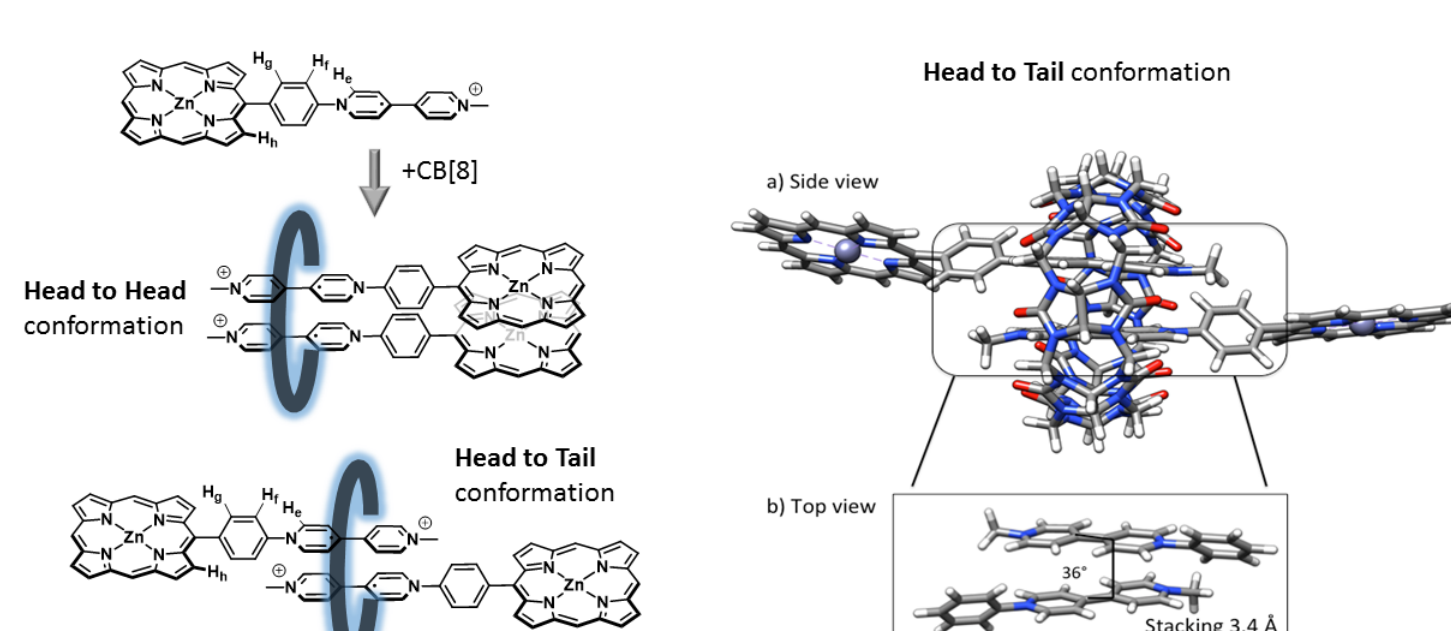
NMR Analysis of host-guest interaction of 1Zn⁸⁺ with cucurbiturils



Supramolecular self-assembly of 1Zn⁴⁺ with cucurbiturils

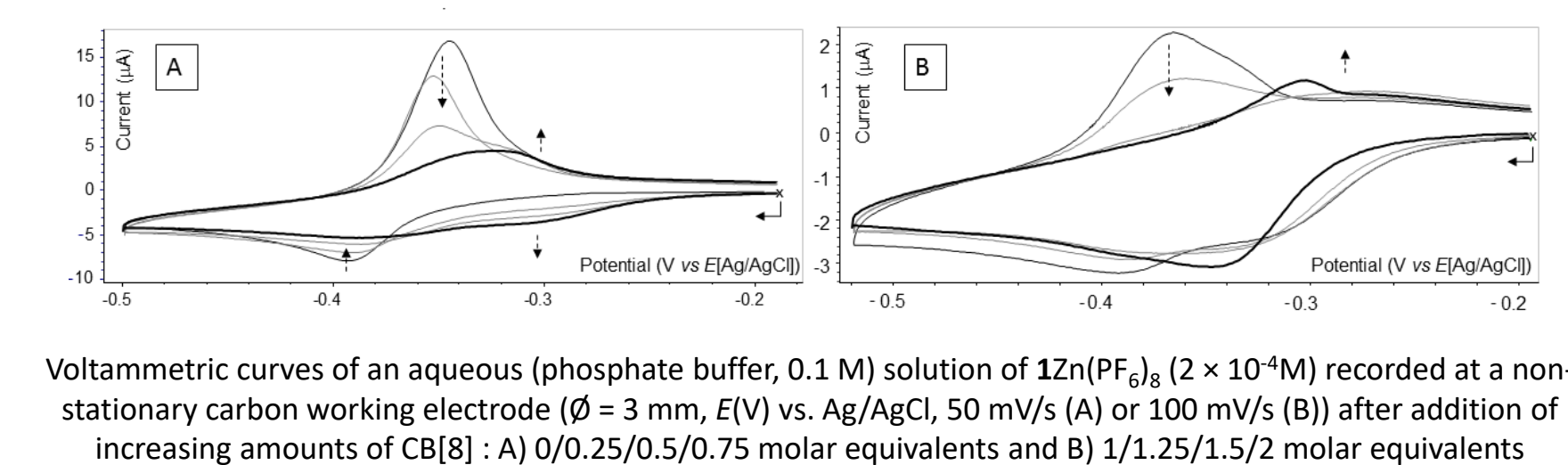


Computational analysis

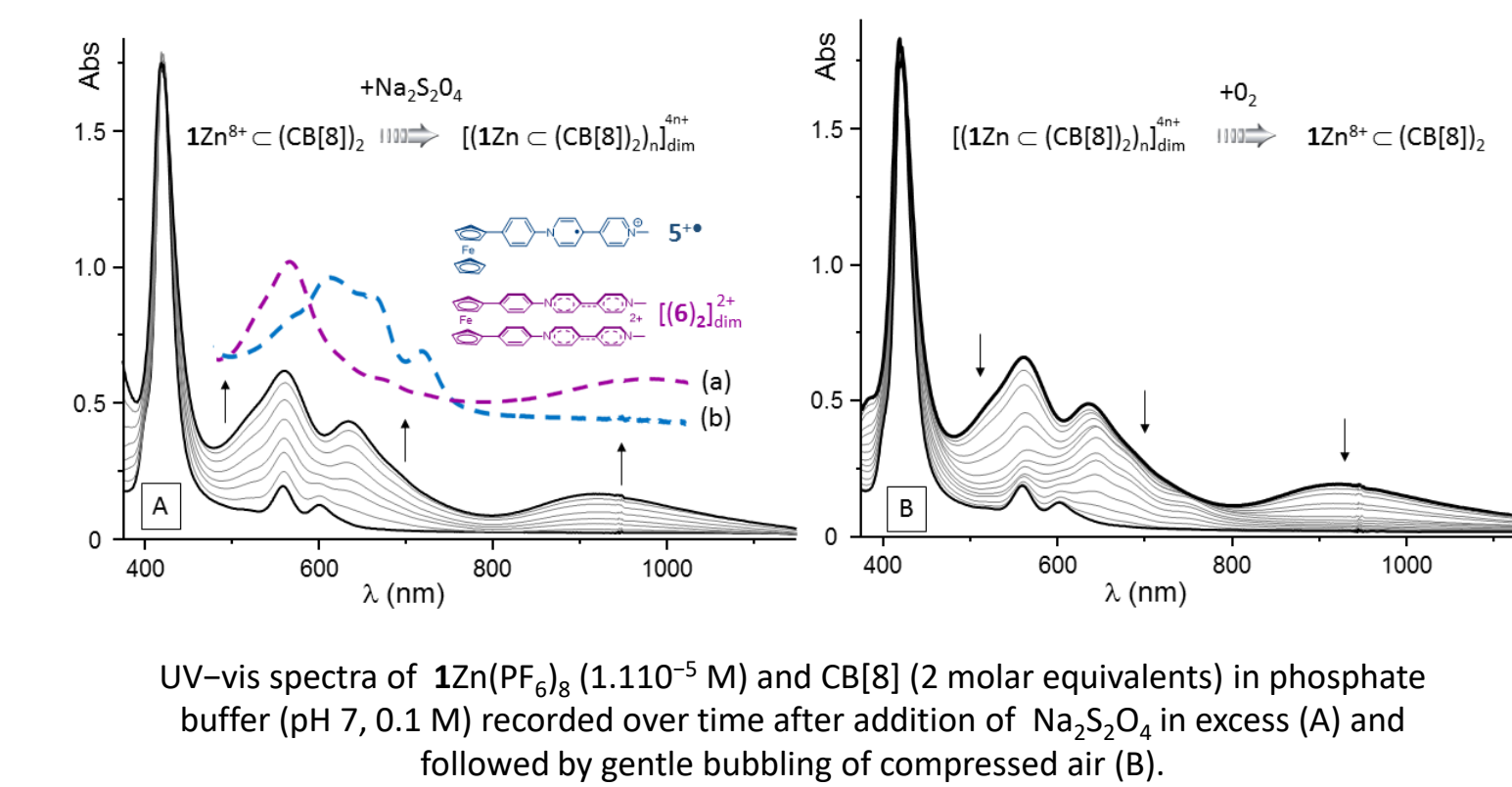


Left) Schematic representation of the face to face or head to tail conformations of the selected π -dimer model complex formed in the presence of cucurbit[8]uril (shown as a blue ring). Right) Structure of the head to tail conformer optimized at DFT/M06-2X level of theory

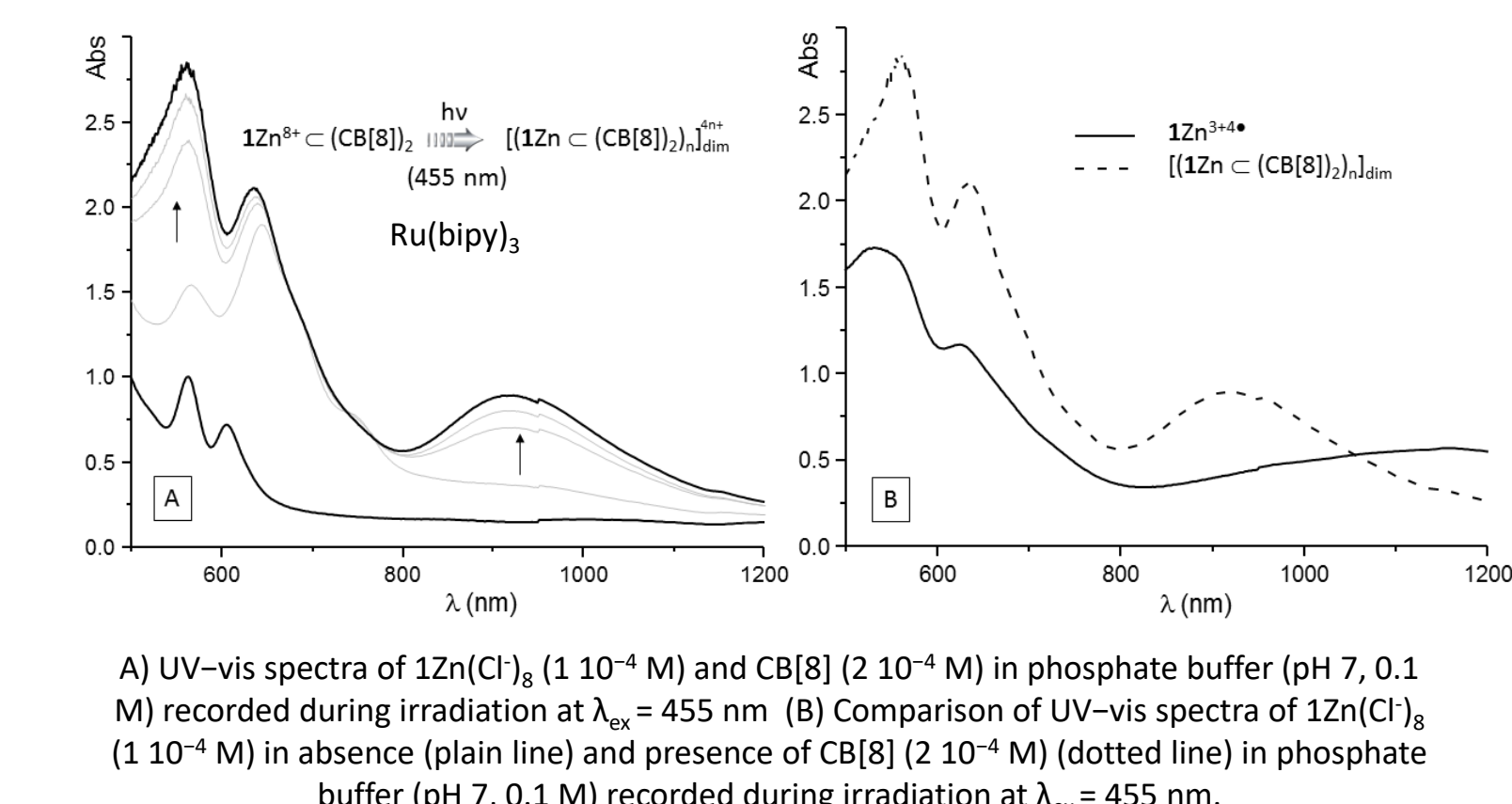
Electrochemical analysis



Chemically triggered self-assembly



Photochemically triggered self-assembly



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