Experimental and theoretical investigation of polynuclear bio-inspired copper complexes

Copper active sites play central biological roles, including electron transfer, dioxygen binding, activation and reduction, as well as denitrification processes. Enzymatic copper centers are extremely diverse in geometric and electronic structure, and range from mononuclear sites to dinuclear, trinuclear and tetranuclear clusters. The accurate description of the magnetic and spectroscopic properties of these polynuclear sites, at different stages of the reaction cycle, is of high importance to get a better understanding of the structure-function relationships and catalytic mechanism.

In this project we aim at designing bioinspired polynuclear low molecular weight copper complexes, as well as at understanding their redox, spectroscopic, electronic and magnetic properties, in relation with the corresponding enzymatic systems. The ligands and bridging atoms will be modulated to exemplify the complexity observed in Nature and their reactivity in modeling reactions will be tested. The characterization of the complexes will rely on different techniques such as electron paramagnetic resonance (EPR), UV-visible, electrochemistry etc. A special emphasis will be dedicated to the use of theoretical approaches such as Density functional theory (DFT) for the description of the properties using spin-projection methods. [1] In some cases, the magnetic interactions and associated spectroscopic behavior become highly complex so that DFT calculations can produce inconsistent results. Such limitations could be lifted by the use of advances theoretical approaches such as Density Matrix Renormalization Group (DMRG). This approach will be conducted in collaboration with a German group (Max Planck Institut, Mülheim/Ruhr, Germany). The proposed project therefore involves a combined experimental and theoretical approach. Such combination of synthesis, spectroscopy and theory aims at understanding the properties and functions of the bioinorganic sites themselves, and finally to inform the synthesis of improved and potentially functional analogues of biological multicopper sites.

The student will be hosted in the BiosCiences group at iSm2 in Marseille for a 3-year PhD contract from Aix-Marseille to begin in October 2018. There is also the possibility to teach during this 3-year contract. The BiosCiences group will provide a multidisciplinary environment and has already strong expertise in combining both experimental and theoretical approaches to study bioinspired metal complexes as well as metal-containing enzymes.[2]

References

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Keywords

copper complex, protein mimic, organic chemistry, coordination chemistry, spectroscopy, magnetism, quantum chemistry

Required skills

Background in organic synthesis and/or coordination chemistry
Knowledge in quantum chemistry and spectroscopy is welcome

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