Contribution ID : **293** Type : **Oral**

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Thursday, 19 November 2020 16:00 (60)

The Evolution of Electronic Complexity in Biology: 2p3d and 1s3p RIXS of Iron-Sulfur Clusters
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Iron sulfur proteins are ubiquitous in nature, performing essential roles in electron transfer processes, redox chemistry, regulatory sensing and catalysis. The metal active sites of these proteins range from simple single iron sites to complex eight iron clusters. Perhaps the most complex iron sulfur cluster that has been identified to date is the iron molybdenum cofactor (or FeMoco) of nitrogenase, which is capable of cleaving the strong triple bond of dinitrogen. The fundamental question that arises is how does nature evolve complexity in order to enable challenging transformations? In our view, a deeper understanding of the complex geometric and electronic structure of iron sulfur clusters requires the pursuit of novel experimental approaches for integrating their electronic structure in a detailed and quantitative fashion. To this end, we are applying both 2p3d and 1s3p resonant inelastic X-ray scattering (2p3d RIXS), in order to obtain deeper insights into the electronic structure of these important clusters. These data provide an experimental measure of the d-d transitions and allow for more detailed insights into the nature of the multiplet structure. The utility of these methods for understanding the electronic structure of nitrogenase will be highlighted. The challenges that RIXS spectroscopy presents for theoretical modeling will also be discussed.

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Session Classification : Plenary 2: Prof. Serena DeBeer, Max Planck Institute for Chemical Energy Conversion