Monday, October 15, 2018

Arnimalle 14, 14195 Berlin, Hörsaal B

SFB 1078 Protonation Dynamics in Protein Function

16:30 Johannes Messinger

17:40 Leonardo Guidoni

18:30 Finger-food buffet and beverages – you are cordially invited

Studying biological water oxidation by room-temperature serial crystallography

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Nearly 50 years ago that Bessel Kok established that biological water oxidation in photosystem II (PSII) involves a 'charge' storing unit that accumulates four oxidizing equivalents before water oxidation proceeds in a concerted reaction, thus avoiding high-energy intermediates and the formation of reactive oxygen species. We know today that each step of this five-stage reaction cycle involves the oxidation of the Mn_4CaO_5 cluster that is ligated mostly by the reaction center protein D1, and that the surrounding protein environment and water/proton networks play essential roles in activating the abundant Mn and Ca ions to catalyze the water-splitting reaction with low over potential. Although high-resolution structures of the dark-stable state (S1) of photosystem II have been obtained in recent years, we do not yet understand the crucial factors for this activation, nor the precise mechanism for water oxidation. Over the past years, several groups have worked intensively on making photosystem II amendable to room-temperature serial crystallography using femto-second X-ray laser pulses. In my presentation, I will give you a flavor of the challenges involved and present high-resolution structures of all four stable intermediates and of two transient states between the S2 and S3 states. Implications for the mechanism of water oxidation are discussed.

Ab-initio molecular dynamics simulations on water oxidation catalysis in biology and synthetic oxides

Leonardo Guidoni

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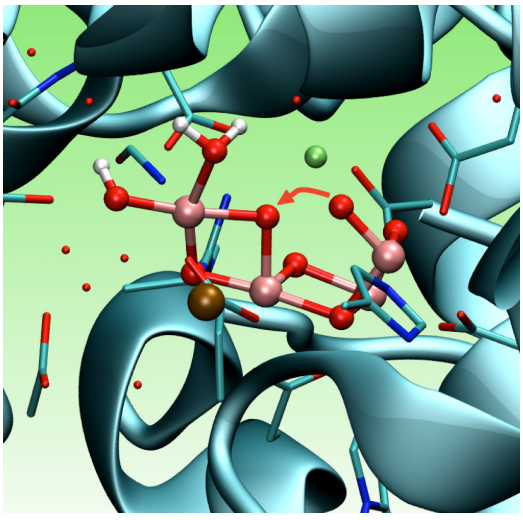

Understanding the details of the molecular mechanisms and the catalytic strategies adopted by Photosystem II to achieve the water oxidation reaction represents a major challenge for experiments and simulations. Using QM/MM dynamics and gas phase models we have built a comprehensive pathways of intermediate structures along the first steps of the Kok-Joliot cycle. Starting from the S3 state, we have investigated several possible pathways and spin surfaces that may lead to the formation of the O-O bond (see figure). The calculated structures and vibrational properties are compared with those of biomimetic compounds.