European XFEL Science Seminar

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Via Zoom

Majed Chergui

Ecole Polytechnique Fédérale de Lausanne, Lausanne Centre for Ultrafast Science, 1015 Lausanne, Switzerland

Spin cross-over dynamics and charge transfer in biology

The past decades have witnessed a revolution in sources of ultrashort pulses, from the optical to the X-ray spectral domain. This has given rise to novel approaches to track the evolution of chemical systems, with element-, spin- and structural sensitivity. In this talk, I will show the capabilities and the new insights that can be obtained from such approaches in the study of biological and molecular systems, and of materials.
We have recently investigated the initial events of the respiratory function in heme proteins. The change of the low-spin (LS) hexacoordinated heme to the high spin (HS) pentacoordinated domed form upon ligand detachment and the reverse upon ligand binding, represents the “transition state” that ultimately drives the respiratory function. The mechanism of the ligand dissociation-recombination process has been hotly debated in the past 30 years, without reaching a unified picture. We have monitored the evolution of Myoglobin-NO (MbNO) from LS to the HS state and the reverse process upon ligand rebinding to the heme, after impulsive photodissociation of the NO ligand. We monitored the evolution of the system using femtosecond (fs) Fe Kα and Kβ non-resonant X-ray emission spectroscopy (XES), which is a powerful marker of the spin state. We show that the entire ligand dissociation-recombination cycle in MbNO is a spin cross-over followed by a reverse spin cross-over process. Ferric Cytochrome c is the most important electron transfer (ET) protein in humans, whose efficiency has been associated to its ruffled heme. However, in investigating ferric Cyt c using fs X-ray absorption spectroscopy and XES, we found that the heme also undergoes doming. The latter corresponds to a much more dramatic change of redox than ruffling, and therefore likelier to modulate the ET properties of the protein.

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