



CHEMISTRY & CHEMICAL BIOLOGY COLLOQUIUM: Electronic Dynamics Underlying Molecular & Nanoplasmonic Light Harvesting Studied by X-Ray Spectroscopy

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About The Speaker:

Oliver Gessner is a Senior Scientist and the Leader of the Atomic, Molecular, and Optical Sciences (AMOS) Program at Lawrence Berkeley National Laboratory (LBNL). He pursued his undergraduate and graduate studies at the Technical University and the Free University Berlin, obtaining his PhD degree in the group of Uwe Becker at the Fritz-Haber-Institut of the Max-Planck Society in 2002. For his postdoctoral research, he moved to Canada to work with Albert Stolow at the Steacie Institute for Molecular Sciences. Gessner joined LBNL as a Divisional Fellow in 2006 and became a Senior Scientist in 2011. Since 2019, he is leading the AMOS Program at LBNL. His research group develops and applies ultrafast x-ray spectroscopy and imaging techniques to study fundamental electronic and nuclear dynamics underlying chemical transformations, and the flow of charge and energy in interfacial systems. He received an Early Career Research Program Award of the DOE Office of Science (2012), and an LBNL Director's Award for Exceptional Scientific Achievement (2014). In 2015, he was elected a Fellow of the American Physical Society.

Abstract:

The success of many emerging molecular electronics and light harvesting concepts hinges on an atomic-scale understanding of the underlying electronic dynamics. Processes evolving on spatial and temporal scales spanning orders of magnitude have to be connected in order to gain a comprehensive picture of the fundamental steps that enable molecular, interfacial, and macroscopic charge and energy transport. The presentation will focus on the potential of ultrafast X-ray spectroscopy techniques to provide a deeper understanding of photoinduced electronic dynamics in several model systems for light harvesting by heterogeneous molecular, molecule-semiconductor, and nanoplasmonic architectures.

Date:

9/11/2020

Time:

1:30 PM - 2:50 PM

Link:

Please email
snsgradstaff@ucmerced.edu
for Zoom link and passcode

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<https://ucmerced.zoom.us/j/93194925460?pwd=L1NLaDhMUzNJa1VvTGhqT3kyYkU0UT09>

Abstract (cont.):

Using femtosecond and picosecond time-resolved X-ray photoelectron spectroscopy (TRXPS) to study photoinduced charge injection in N3 dye-sensitized films of ZnO nanoparticles, the locations of intermittently trapped electrons, interfacial dipoles, as well as charge delocalization and recombination dynamics are monitored with interfacial site-specificity, resolving a long-standing controversy regarding the bottleneck for charge injection in ZnO based dye-sensitized systems.

TRXPS studies of photoinduced dynamics in copper-phthalocyanine(CuPc)-C60 heterojunctions provide a deeper understanding of the predominant energy transport and charge generation mechanisms. Contrary to common belief, fast intersystem crossing from initially excited singlet excitons to triplet excitons within the bulk of the donor domain is not a loss channel but the triplet excitons contribute to a significantly larger extent to the time-integrated interfacial charge than the initially excited interfacial singlet excitons. Ultrafast site-specific X-ray probing provides direct access to the diffusivity of the triplet excitons in the CuPc donor domain and their diffusion length. A recent TRXPS study of the CuPc-C60 heterojunction in the femtosecond domain captures the actual moment of charge generation from interfacial charge-transfer (ICT) states and provides a quantitative measure of the exciton dissociation efficiency compared to photon energy loss by exciton recombination.

In a third showcase example, photoinduced charge transfer dynamics in a nanoporous film of TiO₂ sensitized with gold nanoparticles (Au NPs) is monitored by TRXPS. Direct access to the absolute photon-to-charge conversion efficiency and subsequent electron-hole recombination dynamics in this model system for plasmon-enabled photocatalytic nano-assemblies is demonstrated with single-electron sensitivity. The results provide important boundary conditions for the design of AuNP-TiO₂ based light harvesting systems. Opportunities for extending the measurements toward studies of photoinduced chemical transformations will be discussed.

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