



SEMINARS IN CHEMICAL AND BIOMOLECULAR ENGINEERING

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Presented by

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“Effects of Molecular Environment on the Photophysics of fluorophores in Biomimetic Lights Harvesters and in Organic Light-Emitting Diodes”

The local environment of a fluorescent molecule can have large effects on its optical properties. I will discuss two particular instances where control of the interactions of a chromophore with its surroundings can be used to tune their functionality. First, I will describe our work on biomimetic light harvesters based on protein scaffolds, and then I will turn our attention to host-guest interactions of thermally-activated delayed fluorescence emitters embedded in polymer films.

Artificial light harvesters are a promising way to mimic the tight protein pockets found in natural light-harvesting complexes. With transient absorption and time-resolved fluorescence, we probed isolated chromophores covalently linked to the interior cavity of the supra-molecular assembly of a mutant tobacco mosaic virus capsid protein. The time scales over which the photoexcited chromophores are solvated, undergo conformational rearrangements, and return to the ground state are highly sensitive to their position within the cavity and are significantly slower than in a bulk aqueous solution. Tuning these relaxation processes can improve the yield of intermolecular energy transfer, increasing the collection efficiency of excitations.

Another system where the local environment has large effects on the optical properties of luminescent molecules is organic light-emitting diodes. In a new generation of emitters, dark triplet states can still result in luminescent output by a mechanism known as thermally-assisted delayed fluorescence, in which reverse intersystem crossing is followed by fluorescence decay from singlet states. Such additional processes in the excited state result in complex photophysical properties, which we have studied at the single-molecule level as a function of the host polymer. We found that the amount of heterogeneity in the fluorescent lifetime and spectral characteristics of single emitters is dominated by the static disorder of the host polymer. We are also able to locally measure singlet-triplet equilibration dynamics at room temperature using fluorescence correlation spectroscopy in small fluorophore clusters within the polymer matrix.



Extending these studies to charge-transporting polymers used in devices will lead to better control of host-guest interactions.

Rodrigo Noriega is interested in the molecular-scale determinants of the optoelectronic properties of organic fluorophores. He obtained his PhD in Applied Physics from Stanford University in 2013, working with Professor Alberto Salleo. There, he explored the connections between structural disorder and charge transport in organic semiconductors using synchrotron-based X-ray techniques, electrical measurements, and computer modeling. He then became a postdoctoral researcher at the University of California, Berkeley with Professor Naomi Ginsberg. In his postdoctoral work, he uses ultrafast laser spectroscopy to probe the effects of molecular environment on the excited state dynamics of luminescent molecules in biomimetic light harvesters and light emitting diodes.