Excitons at Organic Semiconductor Interfaces

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This lecture aims to explore key mechanistic issues in an emerging photovoltaic technology based on organic and polymeric molecules, i.e., plastic solar cells. I will present two examples from recent research in my laboratory. The first example is within the realm of conventional theories and we aim to understand a critical step in charge separation at donor/acceptor interfaces in organic photovoltaics, namely the formation and dissociation of interfacial charge transfer excitons [1]. In particular, we show the critical role of hot charge transfer excitons in setting the fundamental time limit for charge separation in organic photovoltaics [2]. The second example shows how an intriguing physical phenomenon, exciton fission in which a singlet exciton breaks up into two triplet excitons in organic semiconductor materials, may be used to build solar cells with power conversion efficiency exceeding the fundamental limit (the so-called Shockley-Queisser limit) of conventional solar cells. We show how singlet exciton fission can occur in organic semiconductors due to a many electron quantum coherent process [3-4], and how we can efficiently extract multiple electrons or energy from the singlet fission process [5-6].

[1] M. Muntwiler, Q. Yang, W. A. Tisdale, X.-Y. Zhu, “Coulomb barrier for charge separation at an organic semiconductor interface,” *Phys. Rev. Lett.* 101 (2008) 196403.
[2] A. Jailaubekov, A. P. Willard, J. Tritsch, W.-L. Chan, N. Sai, L. Kaake, R. I. Gearba, K. Leung, P. J. Rossky, X.-Y. Zhu, “Hot charge transfer excitons set the time limit for charge separation at donor/acceptor interfaces in organic photovoltaics,” *Nature Mater.* 12 (2013) 66-73.
[3] W.-L. Chan, M. Ligges, A. Jailaubekov, L. Kaake, L. Miaja-Avila, X.-Y. Zhu, “Observing the Multi-Exciton State in Singlet Fission and Ensuing Ultrafast Multi-Electron Transfer,” *Science* 334 (2011) 1541-1545.
[4] W.-L. Chan, M. Ligges, X.-Y. Zhu, “The energy barrier in singlet fission can be overcome through coherent coupling and entropic gain,” *Nature Chem.* 4 (2012) 840-845.
[5] W.-L. Chan, J. R. Tritsch, X.-Y. Zhu, “Harvesting singlet fission for solar energy conversion: one versus two electron transfer from the quantum mechanical superposition,” *J. Am. Chem. Soc.* 134 (2012) 18295–18302.
[6] J. R. Tritsch, W.-L. Chan, N. R. Monohan, X.-Y. Zhu, “Harvesting singlet fission for solar energy conversion via triplet energy transfer,” *Nature Commun*.