Ultrafast Coherent Charge Transfer in Solar Cells and Artificial Light Harvesting Systems: Toward Movies of Electronic Motion

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The efficient conversion of (sun-)light into electrical or chemical energy is one of the most fundamental and relevant challenges in current energy research. Our ability to construct artificial molecular or nanostructured devices that can harvest and exploit sunlight inevitably relies on an in-depth understanding of the elementary microscopic principles that govern the underlying light conversion processes. Generally, these processes happen on an exceedingly short femtosecond time scale, making real time studies of the light-driven dynamics particularly important. To elucidate these dynamics, we have recently combined coherent femtosecond spectroscopy and first-principles quantum dynamics simulations [1,2] and have used this approach to explore the primary photoinduced electronic charge transfer in two prototypical structures: (i) a carotene-porphyrin-fullerene triad, an elementary component for an artificial light harvesting system [2] and (ii) a polymer:fullerene blend as a model for an organic solar cell [1].

Surprisingly, our experimental and theoretical results provide strong evidence that in both systems, at room temperature, the driving mechanism of the primary step within the current generation cycle is a quantum-correlated wavelike motion of electrons and nuclei on a timescale of few tens of femtoseconds. Our results suggest that the strong coupling between electronic and vibrational degrees of freedom is of key importance for the dynamics and yield of the charge separation process. In my talk, I will present our most recent findings and their implications for the light-to-current conversion in solar cells. In an outlook, I will discuss new opportunities to probe such dynamics at a single nanostructure level.

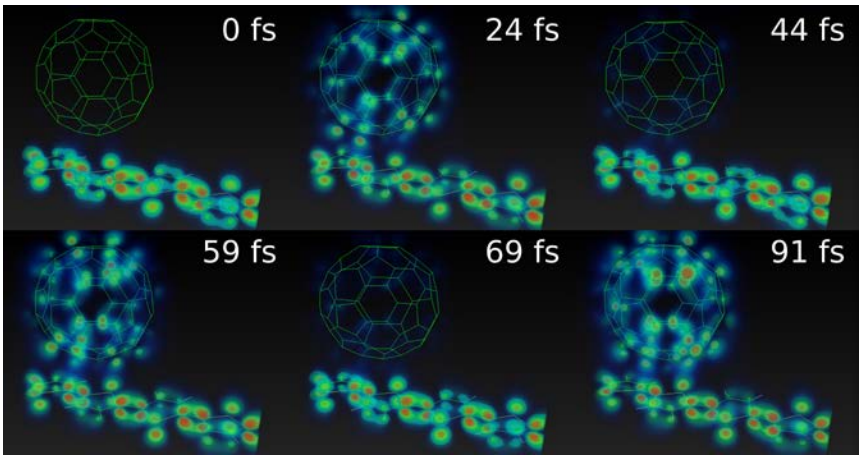

Figure: Real time simulation of the coherent charge transfer dynamics between polymer and fullerene moiety in a P3HT/PCBM thin film photovoltaic device.

[1] S. M. Falke et al., Coherent ultrafast charge transfer in an organic photovoltaic blend. Science 344, 6187 (2014).
[2] C. A. Rozzi et al. Quantum coherence controls the charge separation in a prototypical artificial light harvesting system. Nature Comm. 4, 1602 (2013).