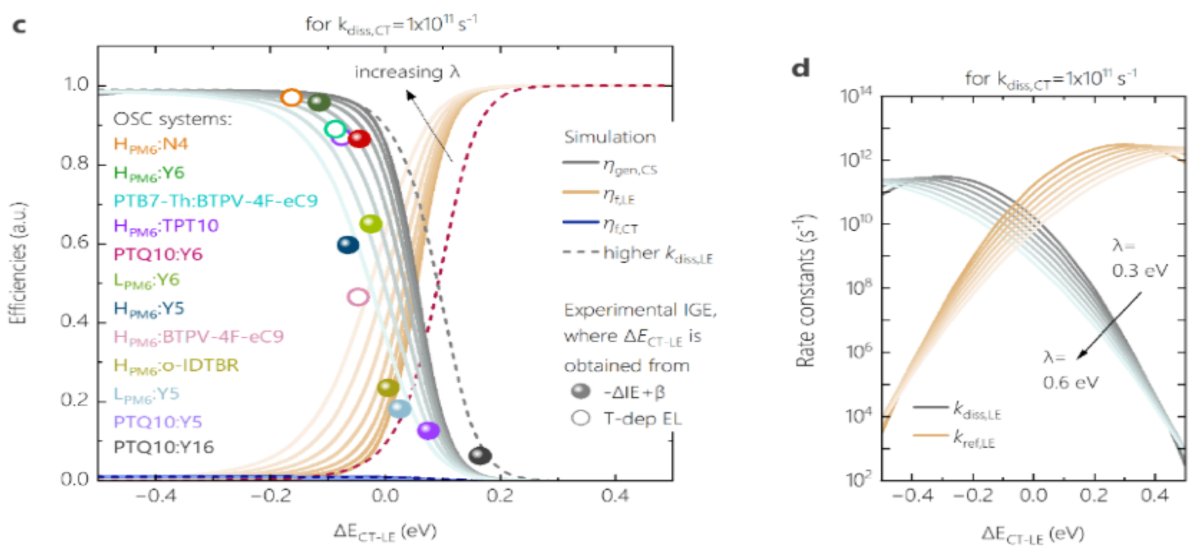
On the (Ultimate) Limits of the Efficiency of Organic Solar Cells - and How to Go Beyond

Dieter Neher

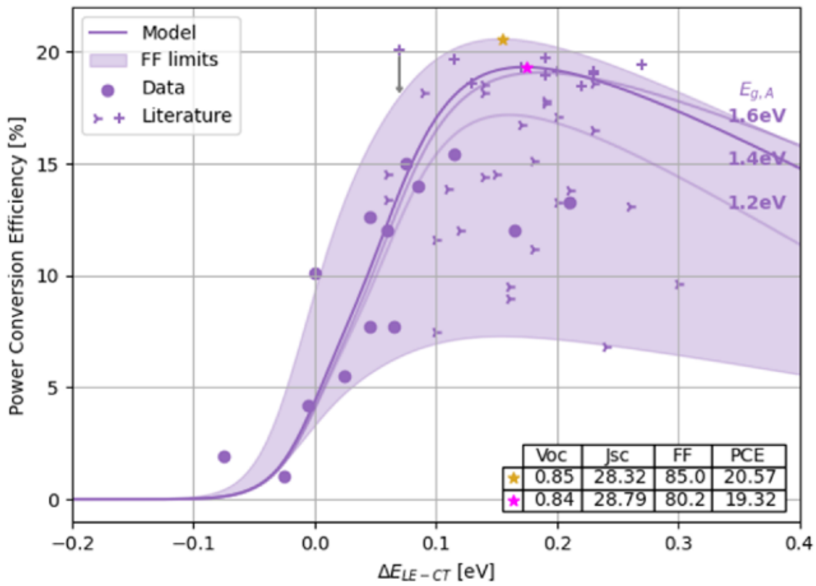
Institute of Physics and Astronomy, University of Potsdam, 14476 Potsdam, Germany

Since the advent of low bandgap non-fullerene acceptors (NFAs), the performance of organic solar cells (OSCs) has improved significantly. In combination with donor materials with complementary absorption, such NFA-based bulk heterojunction blends show efficient conversion of photons into electrons over a broad spectral range. Therefore, an important topic of current OSC research is to reduce the voltage loss by non-radiative recombination while maintaining the short-circuit current (Jsc) and the fill factor (FF) at a high level. The most popular approach to optimize the open circuit voltage of NFA-based blends is to reduce the HOMO-HOMO energy offset. Unfortunately, this is generally accompanied by a significant reduction of Jsc and FF - the reason for this being controversially discussed in the literature.[1-5]

Here we combine a wide range of methods, from spectroelectrochemistry to determine the blend energetics, to femtosecond transient absorption to study the early excitation dynamics, to bias-dependent photoluminescence spectroscopy to study the mechanisms and efficiency of free charge generation. Surprisingly, we find losses due to non-radiative CT recombination competing with charge separation to be insignificant in all blends studies. Instead, singlet exciton decay is identified as the main competing pathway for free charge generation. Our experimental data align with Marcus theory calculations supported by density functional theory simulations. Our results show that the optimal range of the energy offset for achieving optimal performance is quite narrow and lies at ca. 0.3 eV, but that this restriction is lifted in systems with a small reorganization energy for charge transfer.[6]



To explain our data as function of the HOMO offset, we set up a 5-state model which includes singlet and triplet excitons. Our results show that the optimal range of the energy offset for achieving optimal performance is quite narrow and that without additional means for efficient photon harvesting, the power conversion efficiency is limited to around 20 %. This findings aligns with recent > 20 % efficiency records which benefit from improved photon harvesting or very high fill factors.[7]



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