Adaptive Light Capture, Conversion, and Storage

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Abstract The sun is an abundant and sustainable source of energy that is vital for the on-going energy transition. For photons with energies well-above the bandgap of the absorbing material, excess energy is lost predominantly by thermalization in the form of heat. In contrast, photons with energies below the optical bandgap are not absorbed at all. Even at peak efficiencies, both thermalization and sub-bandgap losses account for over 50% of incident solar power. Therefore, single-junction solar cells are limited to a maximum performance of 33%, which is known as the detailed balance limit. It is therefore imperative to find strategies to reduce thermalization and sub-bandgap losses to achieve efficiencies beyond the detailed balance limit. Here, down- and up-conversion processes could, theoretically, increase solar-energy conversion efficiencies beyond current limitations by reaching 39% and 49%, respectively. Additionally, the integration of down- and up-shifters with the aforementioned elements will aid in controlling light throughout the solar radiation spectrum, spanning from the ultraviolet up to the infrared.

The spectral conversion enables modifying the incident solar spectrum such that a better match is obtained with the wavelength-dependent conversion efficiency of, for example, the photoactive layer of photovoltaics. We thereby demonstrate to harness down- and/or up-converting or down- and/or up-shifting of the spectrum, meaning that the energy of photons is modified at demand to either lower or higher energy. Hereby, we systematically vary the electronic coupling in molecular dimers and oligomers to tune the dynamics of all relevant down- and up-conversion steps and, in turn, deciphering not only the full mechanisms of singlet-fission (SF) and triplet-triplet-annihilation up-conversion (TTA-UC), but also all bottlenecks enroute towards the conversion targets of 200% down-converted triplets at minimum driving forces and 50% up-converted singlets at maximum anti Stokes shifts. All of our down- and up-converters will be combined with complementary absorbers to round off the optimal spectral overlap across the solar spectrum by either down- or up-shifting of the spectrum. Crucially, we achieve this not only in solution, but also in the solid state with optimized arrangement and panchromatic absorption from 300 to 1000 nm.