Organic photovoltaic (OPV) materials represent a cheaper and environmentally friendly alternative to currently used inorganic solar cells. Their usually lower power conversion efficiency (PCE) is overcompensated by their simple assembly technology and light-weight, which allows for building large-area and flexible light-harvesting devices. The weakness of OPVs, such as lower PCE and uncontrolled oxidation and reduction, are the source of fast degradation and the decrease in performance over time has become a subject of intense research in the last years. As a consequence, new and potentially better OPV materials and device architectures are developed and announced every month. Such rapid progress is directly related to a better understanding of the physical and chemical processes behind the generation of photo-current. Since experimental manipulations with a possibly large amount of organic compounds are very time consuming, optimization and preselection of the new promising organic photovoltaic compounds and their small building blocks can be achieved more effectively with the use of computer-aided simulations at the quantum level. The effective synergy between theoreticians, who indicate molecular candidates for synthesis and further investigation to experimentalists, will allow for even more rapid progress in the field of novel OPV materials.

A routine quantum chemistry approach to model the electronic structure of OPVs is Density Functional Theory (DFT) and its time-dependent variant, due to their favorable computational scaling. Unfortunately, for certain groups of molecules, DFT completely fails, giving results that are far from physical and chemical reality. A better description of electronic structures and properties of the OPV-related compounds can be achieved with more accurate and reliable wave-function methods. However, the size of the molecules used as building blocks of OPV devices often limits their application to only small model systems due to their unfavorable computational scaling. To bridge this gap and to reliably model electronic structures of large realistic OPV-related compounds, we will make use of novel, robust, and computationally inexpensive quantum chemical models that are based on the modern and efficient parametrization schemes of the wave function. This will lead to a better understanding of many OPV-related compounds, unreachable for standard quantum chemistry methods.