

Polaritons for Chemistry and Materials Science

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Exciton transport plays a crucial role in natural phenomena such as photosynthesis and in artificial devices such as organic solar cells, but is inefficient in many organic materials. We will discuss how the formation of collective polaritonic modes can dramatically enhance the efficiency of exciton transport when the molecules are strongly coupled to an electromagnetic mode [1]. This effect can be exploited to either "harvest" and direct excitations to specific positions by tuning the spatial distribution of the EM mode [2] or to extend the spatial range of the Forster energy transfer process [3]. We then show that in systems with a discrete EM mode spectrum, strong-coupling-enhanced exciton transport can proceed through "dark" modes that have no photonic component, but which nonetheless acquire a delocalized character in the strong-coupling regime [4].

In the second part, we discuss the influence of strong coupling on internal molecular structure and chemical reactions. While most models of strong coupling are based on simple two-level models, pioneering experiments have shown modifications of chemical reaction rates under strong coupling [5]. In order to address this mismatch, we have developed a first-principles model that fully takes into account both electronic and nuclear degrees of freedom [6]. We will first discuss the applicability of the Born-Oppenheimer approximation, which is challenged by the introduction of the new intermediate timescale of energy exchange between the molecule and the field. Based on these findings, we then show how photochemical

reactions such as photo-isomerization can be almost completely suppressed under strong coupling [7]. Finally, we show how polaritons can also lead to the formation of a polaritonic "supermolecule" involving the degrees of freedom of all molecules, opening a reaction path on which all involved molecules undergo a chemical transformation [8].

References

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