

Inelastic and coherent theories of resonance energy transfer for soft photovoltaic molecules

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The energy flow dynamics in systems consisting of soft macromolecules such as conjugated polymers and dendrimers are of great current interest because of their potential applications for cost effective photovoltaic devices. Understanding the energy flow dynamics in systems consisting of these molecules is a fundamental theoretical issue, which should be resolved for systematic experimental control of their optoelectronic characteristics. There are well established theories of energy transfer allowing calculation of transfer rate between simple chromophores or their aggregates. However, many of the soft photovoltaic molecules have characteristics that go beyond the perception of the established theories. Examples are *conformational fluctuations* or *vibrational dynamics* that occur in time scales comparable to that of the energy transfer, and/or the *strong resonance interaction* between neighboring excitation sites that makes the rate picture of energy transfer invalid. The present poster presents two new theoretical advances addressing these issues. The first¹ is an inelastic generalization of the Förster theory for situations where there are quantum mechanical modulation of the donor-acceptor coupling. Under the assumption that the modulations are independent of the electronic excitation of the donor and the acceptor, it is shown that a general rate expression can be found, which involves two dimensional frequency domain convolution of the donor emission lineshape, the acceptor absorption lineshape, and the spectral density of the modulation of the donor-acceptor coupling. The second² is a theory of coherent resonance energy transfer, which has been developed combining the polaron transformation and a time-local quantum master equation formulation with inhomogeneous terms which account for nonequilibrium initial preparation effects. The theory elucidates how quantum coherence and nonequilibrium effects manifest themselves in the coherent population dynamics. Numerical tests show nontrivial transient behavior due to nonequilibrium effects and the contribution of quantum coherence to the steady state donor/acceptor population. Application of these theories to the soft photovoltaic molecular systems may reveal new microscopic insights into their energy flow characteristics, which can assist experimental efforts to design more efficient photovoltaic devices.

References

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