RET in a Medium: Pair and Many-Body QED Theory

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Resonance energy transfer (RET) mediated by an inert polarizable third-body, T, [1,2] in a medium described by a complex refractive index is studied using molecular QED theory [3-5]. Donor, D, and acceptor, A, are coupled to each other, and to T, via the exchange of a single virtual polariton [6]. Fermi golden rule transition rates are evaluated for direct (D → A) and indirect (D → T → A; T → D → A; D → A → T) migration routes. A variety of pure and interference terms contribute to the total rate and their relative importance is discussed. Quantum coherence is shown to originate from the interference of different Feynman pathways that connect the initial and final states of the process. It is found that the energy conserving phase of the amplitude can enhance or suppress the RET rate. Environmentally induced decoherence is attributed to vibronic relaxation occurring in the polarizability of the mediator, and dissipation in the medium due to absorption and dispersion. The interplay of these subtle, competing effects is illustrated by calculations on a model system comprising a linear configuration of the three particles embedded in the medium [3].

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