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2014 New J. Phys. 16 105021
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Focus on quantum efficiency

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Received 17 September 2014
Accepted for publication 17 September 2014
Published 28 October 2014

Abstract

Technologies which convert light into energy, and vice versa, rely on complex, microscopic transport processes in the condensed phase, which obey the laws of quantum mechanics, but hitherto lack systematic analysis and modeling. Given our much improved understanding of multicomponent, disordered, highly structured, open quantum systems, this ‘focus on’ collection collects cutting-edge research on theoretical and experimental aspects of quantum transport in truly complex systems as defined, e.g., by the macromolecular functional complexes at the heart of photosynthesis, by organic quantum wires, or even photovoltaic devices. To what extent microscopic quantum coherence effects can (be made to) impact on macroscopic transport behavior is an equally challenging and controversial question, and this ‘focus on’ collection provides a setting for the present state of affairs, as well as for the ‘quantum opportunities’ on the horizon.

Keywords: light–energy conversion, quantum transport, open quantum systems, light harvesting networks, excitation transport, semiconductors
Modern societies are in urgent need of affordable and efficient energy technologies, and the Sun’s light is arguably the most abundant resource available. Nature is using this supply by photosynthesis with remarkable efficiency. With solar cells and light-emitting diodes, devices for an efficient conversion of light into current and vice versa are available on a large scale. In all these cases, energy conversion relies on complex, microscopic transport and recombination processes in the condensed phase, which obey the laws of quantum mechanics. However, almost exclusively, some sort of phenomenological, ‘hybrid’ quantum–classical approaches are used to describe these devices or the recombination center of the photocycle. While recombination is treated quantum mechanically, usually on the basis of Fermi’s golden rule, the interaction with a thermal bath and disorder in the system is approximated by the Bose or Fermi distribution and an inhomogeneous distribution of energy levels, respectively. This ‘focus on’ collection gathers work taking the quantum mechanical description of quantum efficiency to a new and more systematic level, including the effects of disordered systems with or without realistic coupling to a heat bath, which have hitherto been too complex for systematic (quantum) analysis and modeling.

Given our much improved understanding of multicomponent, disordered, highly structured, open quantum systems, the present ‘focus on’ collection brings together cutting-edge research on theoretical and experimental aspects of quantum transport in truly complex systems as defined by, e.g., the macromolecular functional complexes at the heart of photosynthesis [3, 6, 12, 14, 16], or semiconductor (light-emitting) diodes [5, 7]. All these examples involve transport of quantum particles or excitations as a crucial ingredient determining the efficiency of the respective system or device. Since experiments on excitonic energy transport in photosynthetic light harvesting complexes reported the existence of long coherence times [18, 19], the question of whether (or in which way) the presence of quantum coherences is related to the high transport efficiency of these complexes has been strongly debated.

As a first approach to this question, let us start out with the idealized case of an isolated—and thus fully coherent—quantum system. In [2], an ensemble of disordered networks with random coupling strengths between the various nodes is investigated as a model for coherent excitation transport. Whereas most networks within the random ensemble give rise to destructive interference on the output node—and thus rather low transport efficiency—the opposite is true for specific (and on average more symmetric, see [2]) configurations leading to constructive interference. These findings agree with the following general picture of coherent wave transport in random media [17]: coherence reduces transport on average, due to enhanced backscattering [10, 20, 21] and disorder-induced localization (e.g. Anderson localization [13, 22]), whereas, at the same time, fluctuations around the average are increased [23], thus admitting the existence of specific disorder realizations with exceptionally high transfer efficiency.

With respect to the biological systems or semiconductor devices mentioned above, this picture of coherent transport needs to be supplemented by the inevitable influence of noise induced by coupling to additional degrees of freedom such as phonons/vibrations or (e.g., for photosynthetic light harvesting units) the surrounding protein scaffold and its interplay with quantum coherent effects. In general, noise reduces coherence and thus enhances transport since, as mentioned above, most molecular configurations give rise to predominantly destructive quantum interference [24–26]. On the other hand, too much noise (e.g. dephasing noise with a very large dephasing rate) may also have a localizing effect (‘quantum Zeno effect’ [27]). Under these premises, there in general exists an optimal noise level that maximizes the transport efficiency, as established in the theory of open, disordered quantum systems [28]. When equipped with an additional switching mechanism between a nearly perfect
light-harvesting state and a quenching state, efficiency and robustness of photosynthesis may thus be ensured over a wide range of light intensities [14].

This ‘focus on’ collection contains several contributions that significantly expand our understanding of noise effects in quantum transport efficiency. In particular, various novel aspects of the intricate interplay between noise and quantum coherence in determining the efficiency of transport at the optimal level of noise are discussed in [1, 6, 9, 13]. The relevance of such an interplay is especially evident in view of the fact that, e.g. in light-harvesting complexes, electronic degrees of freedom are typically strongly coupled to vibrational modes [8]. In this case, a more accurate description of environmental effects will be obtained if the separation between ‘system’ (i.e. electronic excitations) and ‘bath’ (remaining degrees of freedom) is properly redefined, e.g., by including the most relevant vibrational modes into the system Hamiltonian [3, 29] or employing a polaron transformation that mixes system and bath degrees of freedom [9, 15]. The mechanisms by which dissipative dynamics generates entanglement may furthermore lead to a better scaling of quantum metrology error with system size [4]. Last but not least, our ‘focus on’ collection also contains new results on the modeling of non-Markovian [11] or site-correlated [12] baths, which provide an additional degree of freedom for the optimisation of transport efficiencies.

References


7 [1–16] refer to the present ‘focus on’ collection
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