Stationary quantum coherence and transport in disordered networks

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Abstract. We examine the excitation transport across quantum networks that are continuously driven by a constant and incoherent source. In particular we investigate the coherence properties of incoherently driven networks by employing recent tools from entanglement theory that enable a rigorous interpretation of coherence in the site basis. With these tools at hand we identify coherent delocalization of excitations over several sites to be a crucial prerequisite for highly efficient transport across networks driven by an incoherent source. These results are set into context with the latest discussion of the occurrence of coherence in molecular complexes that are driven by incoherent sun light.

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1. Introduction

Coherent transport across disordered networks is a process that has gained a lot of interest over the last years. Whereas the transport process is a general and by itself rather abstract problem, it applies to various practical questions as for example excitation dynamics in Rydberg gases [1], efficient charge transport in organic solar cells [2] or exciton dynamics in light harvesting complexes. The latter system mediates the transport of excitations between an antenna complex that absorbs sun light and a reaction center in which the excitation is converted into chemical energy.

Recently observed beatings in spectroscopic data have been interpreted as signatures of long-lived quantum coherence [3–6]. Coherently delocalized excitations enable the interference of several path-alternatives across the network. If this interference is constructive for the output site, this leads to enhanced transport. Consequently, quantum coherence is actively discussed as a potential explanation for the efficiency of transport in such disordered networks [7–12]. The potential existence of quantum coherence at physiological temperatures and rather strong dephasing [13, 14] has also raised hope that similarly efficient transport properties based on quantum interference can be achieved in man-made devices [15, 16].

Independently of the specific physical properties of the network, each transport process is initiated by injecting an excitation into the system which can be done in different ways. A common experimental technique which is in particular used in spectroscopy experiments is to induce an excitation through the application of an ultra-short and highly coherent light pulse. As the single excitations subsequently propagate from the source to the drain, this scenario is referred to as the transient scenario [8, 17].

Recent objections question whether observations and explanations made for the transient scenario based on a coherent excitation process also apply to processes that take place under constant and incoherent driving [18–20]. Instead of an application of a pulsed and coherent light source, systems like light harvesting complexes or artificial devices designed to utilize sun light are rather exposed to an incoherent and stationary source of excitation that drives the networks into a stationary state. As this state is characterized by a constant flux of excitations from the source to the drain, this scenario is referred to as the stationary state approach [8, 17].
and its relation to the transient scenario as well as the existence and role of quantum coherence in the steady states is intensively debated [17–22].

We want to pick up this discussion of excitation transport and examine the differences between the transient and the stationary state approach for fully coupled random networks by comparing the transport efficiencies of these two scenarios. To this end, we examine how coherent delocalization of excitations relates to the transport efficiency in an incoherently and continuously driven system in order to estimate the effect of constructive interference on the transport efficiency in the stationary state scenario. Since the number of path-alternatives that interfere constructively is limited by the number of sites over which the excitation is coherently delocalized, we expect this number to characterize the potential benefit that coherence can have for the transport process. To substantiate this, we employ recent tools from entanglement theory [23] which enable a rigorous characterization of the number of sites over which an excitation is coherently delocalized.

2. Description of the model

For the comparison of the transient and the stationary state approach we consider a fully connected network of \( N \) two-level systems referred to as sites all of which have the same on-site energy. This restriction is of rather technical nature but does not fundamentally affect the relations that are under examination, i.e. the following results are robust against energetic disorder provided the width of the energy distribution is small as compared to the average intersite coupling strength. Variations of the energy levels merely change the effective coupling between the various sites which can essentially be compensated by adjusting the network geometry such that the Hamiltonian for the system can be simplified to [24]

\[
H = \sum_{i,j=1 \atop i \neq j}^{N} \frac{\Xi}{|\vec{r}_i - \vec{r}_j|^3} \sigma_i^+ \sigma_j^-.
\] (1)

The operators \( \sigma_i^+ \) and \( \sigma_i^- \) are the raising and lowering operator on site \( i \), and the interaction decays cubically with the distance between site \( i \) and \( j \) in accordance with a dipole–dipole interaction. We assume the excitations to be induced inside some external unit (e.g. an antenna) from which they are injected into the network via an input site [24] which we label site 1. From there, the excitation is supposed to propagate to site \( N \) which together with site 1 defines the poles of a sphere, and a random arrangement of the other sites within this sphere defines one realization of a network [10]. This model is scale invariant, i.e. increasing the size of the sphere or the interaction constant \( \Xi \) can be compensated completely through a proper re-scaling of time. We can therefore specify all lengths in terms of multiples of \( |\vec{r}_1 - \vec{r}_N| \) and introduce a scaled time \( t = \Xi / |\vec{r}_1 - \vec{r}_N|^3 t_r \), where \( t_r \) is the actual time\(^2\).

In the stationary state approach the full dynamics including coherent and incoherent contributions is modeled by a phenomenological master equation. Non-Markovian features can be incorporated in this framework through the use of time-dependent coupling constants. In the stationary state, however, also these become time-independent [25], so that the present framework with time-independent rates does not necessarily imply a limitation to Markovian

\(^2\) For typical light harvesting complexes \( t = 1 \) corresponds to \( t_r \) of the order of \( 10^{-13} \) s [24].
dynamics. More explicitly, the master equation reads

$$\dot{\rho} = -i [H, \rho] + \mathcal{L}_{\text{in}} (\rho) + \mathcal{L}_{\text{rec}} (\rho) + \mathcal{L}_{\text{out}} (\rho) + \mathcal{L}_{\text{deph}} (\rho).$$

(2)

The operators $\mathcal{L}_{\text{in}}$ and $\mathcal{L}_{\text{out}}$ describe the coupling to the external antenna which itself interacts with the incoherent light field and to the sink that extracts excitations from the network, respectively. $\mathcal{L}_{\text{deph}}$ incorporates the dephasing induced by an environment, whereas $\mathcal{L}_{\text{rec}}$ implements recombination, i.e. the loss of an excitation due to a finite lifetime of a site’s excited state.

Incoherent feed-in of excitations is modeled by

$$\mathcal{L}_{\text{in}} (\rho) = \gamma_{\text{in}} \left( \sigma_i^- \rho \sigma_i^+ - \frac{1}{2} \left\{ \sigma_i^+ \sigma_i^-, \rho \right\} + \sigma_i^+ \rho \sigma_i^- - \frac{1}{2} \left\{ \sigma_i^- \sigma_i^+, \rho \right\} \right),$$

(3)

where $\gamma_{\text{in}}$ is the rate of absorption from and emission to the incoherent source as typically realized by an antenna that is in turn coupled to the light field [8, 14, 26]. In addition to re-emission from the first site into the heat bath all sites can also loose their excitation through recombination as induced by

$$\mathcal{L}_{\text{rec}} (\rho) = \gamma_{\text{rec}} \sum_i \left( \sigma_i^- \rho \sigma_i^+ - \frac{1}{2} \left\{ \sigma_i^+ \sigma_i^-, \rho \right\} \right),$$

(4)

and the coupling to the sink that is described by

$$\mathcal{L}_{\text{out}} (\rho) = \gamma_{\text{out}} \left( \sigma_N^- \rho \sigma_N^+ - \frac{1}{2} \left\{ \sigma_N^+ \sigma_N^-, \rho \right\} \right).$$

(5)

The sink can only withdraw excitation but not feed them back into the network. Additionally to these dissipative effects of source, sink and recombination the decay of the inter-site coherences is governed by the dephasing operator

$$\mathcal{L}_{\text{deph}} (\rho) = \gamma_{\text{deph}} \sum_i \left( \sigma_i^- \rho \sigma_i^+ - \frac{1}{2} \left\{ \sigma_i^+ \sigma_i^-, \rho \right\} \right),$$

(6)

whose prefactor’s inverse $\gamma_{\text{deph}}^{-1}$ defines an upper limit for the maximal coherence time.

The sink rate $\gamma_{\text{out}}$ is chosen to be $\gamma_{\text{out}} = 20$ as used before in theoretical studies [27] and in agreement with the experimentally determined interaction timescales for light harvesting complexes [28]. We assume to have at most one excitation in the network at the same time as we are interested in systems that provide fast transport and where the inverse of the propagation time is smaller than the absorption rate. To incorporate the absence of double-excitations in our model we will thus choose $\gamma_{\text{in}} = 2 \times 10^{-4}$ since we found that for this choice probabilities for double-excitations are deemed negligible such that we can restrict the following discussion to the zero- and one-excitation subspace. It should, however, be mentioned that moderate variations of $\gamma_{\text{out}}$ or $\gamma_{\text{in}}$ do not lead to qualitatively different results as long as $\gamma_{\text{in}} \ll \gamma_{\text{out}}$ holds.

### 3. Transport efficiency in the transient and the stationary scenario

The examination of the transport process requires a quantification of a given network’s transport efficiency. Such is obtained by introducing two efficiency functions one of which measures the probability for rapid excitation transport in the transient case whereas the other one estimates the steady excitation flux to the sink in the stationary state scenario. Applying these functions to an ensemble of randomly generated networks enables a comparison of those two approaches.
3.1. Transient and stationary efficiency functions

In the case of transient dynamics the actual process of extracting the excitation from the network is often not described explicitly, but efficiency is defined in terms of the probability of the excitation to reach site \(N\) [10]. A suitable definition is a time-weighted average probability

\[
E_t = b_t(T) \int_0^\infty dt \varrho(t) \sigma^+_N \sigma^-_N e^{-T} t,
\]

where the choice of the time constant \(T\) permits to gauge the importance attributed to fast transport. The prefactor \(b_t(T) = T^{-1}\) is chosen such that \(E_t = 1\) is obtained for a hypothetical optimal system that instantly propagates the excitation to the output without any excitation loss, i.e. \(\varrho(t) = |N\rangle\langle N|\) for all times \(t\).

To achieve a meaningful comparison between transient dynamics and steady state properties we need to ensure comparable time-windows for the excitation to propagate from the input to the output site. Whereas for the transient case this time window is defined by \(T\) we can use the recombination rate \(\gamma_{\text{rec}}\) to provide a limitation on the propagation time in the stationary state approach. We will therefore choose the inverse of the recombination constant \(\gamma_{\text{rec}}^{-1}\) in the stationary scenario to approximate the timescale \(T\) considered in the transient case.

Whereas for the transient dynamics efficiency can be defined in terms of the probability for an excitation to reach site \(N\), in the steady state the flux of excitations to the sink is the figure of merit [8, 26]. To identify that sink flux we consider the temporal change of the number of excitations in the network by obtaining the expectation value of the number operator \(\hat{N} = \sum_i \sigma^+_i \sigma^-_i\) in the stationary state as

\[
\begin{align*}
\text{tr} \hat{N} \dot{\varrho} &= -i \text{tr} \hat{N}[H, \varrho] + \text{tr} \hat{N} \mathcal{L}_{\text{in}}(\varrho) + \text{tr} \hat{N} \mathcal{L}_{\text{rec}}(\varrho) \\
&= \hat{J}_{\text{in}} + \hat{J}_{\text{rec}} + \hat{J}_{\text{deph}} + \hat{J}_{\text{out}} = 0.
\end{align*}
\]

One identifies three non-vanishing quantities \(\hat{J}_{\text{in}}, \hat{J}_{\text{rec}}\) and \(\hat{J}_{\text{out}}\) which describe the incoming flux, the recombination loss and the sink flux, respectively and whose signs have been chosen such that all three quantities are non-negative. With the specific form of \(\mathcal{L}_{\text{in}}\) defined above in equation (3) the incoming flux can be evaluated to

\[
\hat{J}_{\text{in}} = \gamma_{\text{in}} (1 - 2 \text{tr} \varrho \sigma^+_1 \sigma^-_1),
\]

while the sink flux can be written as

\[
\hat{J}_{\text{out}} = \gamma_{\text{out}} \text{tr} \varrho \sigma^+_N \sigma^-_N,
\]

where we used the expression for \(\mathcal{L}_{\text{out}}\) as specified in equation (5). As all the three quantities introduced in equation (8) are non-negative, we find the sink flux to be bounded by the incoming excitation flux which, in turn, is bounded by the injection rate \(\gamma_{\text{in}}\) according to equation (9), i.e. \(\hat{J}_{\text{out}} \leq \hat{J}_{\text{in}} \leq \gamma_{\text{in}}\). We therefore normalize the sink flux \(\hat{J}_{\text{out}}\) with respect to the injection rate, which yields the stationary transport efficiency

\[
E_s = \frac{\gamma_{\text{out}}}{\gamma_{\text{in}}} \text{tr} \varrho \sigma^+_N \sigma^-_N.
\]

This quantity will be used in the following for estimating the transport performance in the stationary state.
3.2. Comparison of the transport efficiency in the transient and the stationary state scenario

As we have introduced efficiency quantifiers for both of the considered transport scenarios, we can now compare the efficiency in the stationary state without explicit dephasing according to equation (11) with the efficiency in the transient case given by equation (7) for randomly generated networks. Figure 1(a) depicts $E_t$ as a function of $E_s$ for $10^5$ random systems with $N = 7$ sites. A sufficiently small choice of the transient excitation lifetime $T = \frac{1}{40} \tau_{\text{direct}} = \frac{\pi}{80}$ sets the focus on short-time dynamics, i.e. we only identify those networks as efficient in which the excitation can reach the output significantly faster than allowed by the direct interaction between input and output site that takes place on the timescale $\tau_{\text{direct}} = \frac{\pi}{2}$. The excitation lifetime in the stationary state is governed by the inverse of the recombination rate $\gamma_{\text{rec}}^{-1} = \frac{1}{50}$. As it can be seen in figure 1(a), $E_t$ and $E_s$ are highly correlated, i.e. the efficiency in the transient scenario allows to infer about the efficiency in the stationary case and vice versa. The correlations are not perfect, i.e. $E_t$ is not a function of $E_s$ alone, but it can be quantified by the correlation coefficient

$$\kappa (E_t, E_s) = \frac{\langle (E_t - \langle E_t \rangle) (E_s - \langle E_s \rangle) \rangle}{\sigma_{E_t} \sigma_{E_s}},$$  

(12)

where $\sigma_X = \sqrt{\langle X^2 \rangle - \langle X \rangle^2}$ is the standard deviation and $\langle X \rangle$ stands for the average over the ensemble of random networks. A correlation coefficient of $\kappa = \pm 1$ indicates that the value of one quantity determines the value of the other exactly, whereas $\kappa = 0$ signifies that the knowledge of one does not provide any information about the other. For the data displayed in figure 1(a) we obtain a correlation of $\kappa \approx 0.97$, i.e. a close-to-maximal value what substantiates that observations made for the transient approach transfer to the stationary scenario essentially perfectly and vice versa.
One might, however, expect that this compatibility of the two transport scenarios relies on the similarity of the transport timescales, i.e. it does not apply anymore if $\gamma_{\text{rec}} T$ differs substantially from unity. To test that, we plot $\kappa$ as a function of the recombination rate’s inverse $\gamma_{\text{rec}}^{-1}$ for different choices of $T$ in figure 1(b). The case $T = \frac{1}{40} T_{\text{direct}}$ corresponding to figure 1(a) is depicted in red (dashed) and a maximum of the correlations for $\gamma_{\text{rec}} \approx 0.05$ is clearly discernible. The maximum, however, is rather broad, and strong correlations with $\kappa > 0.9$ are obtained for a wide range of excitation lifetimes in the stationary scenario $0.03 \lesssim \gamma_{\text{rec}}^{-1} \lesssim 0.1$. That is, the transfer of observations between stationary and transient approach does not require precise knowledge of parameters like the recombination rate, but a rough estimate is sufficient for qualitative assessments.

A variation of the transient lifetime $T$ confirms that optimal correlation is, however, obtained if $T$ and $\gamma_{\text{rec}}^{-1}$ define comparable timescales. Figure 1(b) depicts that the maximum of $\kappa$ is shifted to larger values of $\gamma_{\text{rec}}^{-1}$ as $T$ is increased what clearly underlines the correspondence between these two timescales. The correlation, however, gets less significant for longer excitation lifetimes that invoke a consideration of networks with less rapid dynamics. Furthermore, optimal correlations are always obtained for $\gamma_{\text{rec}} T < 1$, i.e. for the case in which the excitation is given a shorter time window in the transient case to reach the output site than in the stationary state scenario. To appreciate this difference one has to take into account the presence of the sink in the stationary state approach which additionally shortens the excitation lifetime. Whereas in the purely coherent case the excitation loss (and therefore also the finite excitation lifetime) is only determined by the value of $T$, the stationary state’s excitation lifetime is affected by both recombination and sink drainage. The choice of the sink rate $\gamma_{\text{out}}$ does fundamentally affect the period for which an excitation is able to stay in the network as well as the maximally obtainable transport efficiency. In the transient approach, however, there is no comparable analogue to the sink, what makes these two concepts differ systematically.

Despite these differences, the correlations between $E_t$ and $E_s$ as well as the correspondence of $\gamma_{\text{rec}}^{-1}$ and $T$ suggest a major agreement of the transient and the stationary state approach. Efficiency thus does not primarily depend on the injection mechanism but is a rather universal feature, i.e. a given spacial arrangement shows a similar transport performance under different feed-in scenarios.

4. Transport efficiency and quantum coherence

For the transient scenario it is widely accepted that quantum coherence is a crucial prerequisite for efficient transport across molecular networks [3, 10, 11]. Given the agreement of the transient and the stationary state scenario in terms of efficiency one might raise the question of whether the similarities go beyond the mere transport performance and also hold for the occurrence and role of coherence, i.e. whether coherence in continuously, incoherently driven networks does play the same crucial role for the transport process as assumed for the transient case.

The role of coherence for excitation transport is readily illustrated by the double-slit experiment in which a coherent superposition of two path-alternatives gives rise to an interference pattern of alternating regions of enhanced and reduced arrival probabilities. Increasing the number of coherent path-alternatives through an increasing number of slits changes the interference pattern such that it increases the contrast, i.e. the differences in arrival probabilities between spots with constructive and spots with destructive interference. Similar consequences also apply to an excitation that can take several path-alternatives in order...
to propagate from the input to the exit. If two of these path-alternatives are in a coherent superposition such that it features constructive interference for the output site then this yields a high arrival probability at the output which, in turn, results in an enhancement in transport efficiency. In analogy the multi-slit experiment this enhancement can even be higher if there is a constructive interference of not only two but three or more path-alternatives. The more different paths are taken coherently, the more potential benefit this can have for the transport efficiency.

A coherent superposition of different path-alternatives, however, requires a coherent delocalization of the excitation over various sites. The number of sites over which an excitation is coherently delocalized and which we also refer to as the extent of coherent delocalization is strongly correlated to the number of paths that are in a coherent superposition and can thus be expected to also correlate to the optimal transport efficiency.

4.1. Characterizing coherence in the stationary state

As discussed before, we can restrict the following discussion to the zero- and one-excitation subspace as due to the choice of coupling constants the state amplitudes with more than one excitation are negligible. Since all terms in the equations of motion that induce transport of excitation (i.e. the Hamiltonian defined in equation (1)) conserve the number of excitations, the system ground state does not take part in the actual transport so that we only need to consider that part of the density matrix that describes a single excitation.

The objective is to characterize to what extent the excitation is coherently delocalized [29]. For that purpose, we project the density matrix onto the single-excitation subspace and apply a renormalization such that we obtain

$$\tilde{\rho} = \frac{P \rho P}{\text{tr} \rho P} \quad \text{with} \quad P = \sum_{i=1}^{N} |i\rangle\langle i|,$$

where $|i\rangle$ denotes the state of the $i$th site excited and all other sites in the ground state. Since we expect the benefits and disadvantages of quantum coherence to result from constructive and destructive interference of different path-alternatives across the network which, in turn, requires a coherent delocalization of an excitation, we will characterize quantum coherence in terms of the number of sites over which an excitation is coherently delocalized. An excitation in a pure state is coherently delocalized over $K$ sites if the state vector

$$|\Psi_{KN}\rangle = \sum_{i} \psi_{i} |i\rangle$$

contains $K$ finite amplitudes $\psi_{i}$. Because of the coupling to incoherent reservoirs we, however, always face mixed states here for which we have to generalize the concept of $K$-site coherence. The formal generalization is fairly straightforward: a mixed state $\tilde{\rho}_{KN}$ is considered to feature $K$-site coherence if it cannot be described as an ensemble of pure states without at least one state-vector with at least $K$-site coherence, i.e.

$$\tilde{\rho}_{KN} \neq \sum_{j<K} \sum_{i} p_{ij} |\Psi_{jN}^{(i)}\rangle \langle \Psi_{jN}^{(i)}|.$$

Rigorously identifying $K$-body coherence, on the other hand, is typically rather cumbersome, but given the formal similarity between $K$-site coherence and $K$-body entanglement in the
one-excitation subspace [30, 31], efficient practical tools can be imported from entanglement theory [23]. We will employ in the following the functions:

\[
\tau_{KN}(\rho) = \max_{\langle \psi \rangle} b_{KN} \left( |\langle \Phi_1| \rho \rangle| \Phi_2 \rangle \right) - a_{KN} \sum_{i=1}^{N} \sqrt{|\langle \Phi_1^{(i)}| \rho \rangle| \langle \Phi_2^{(i)}| \rho \rangle| \langle \Phi_2^{(i)}| \rho \rangle| \langle \Phi_2^{(i)}| \rho \rangle|}
\]

(16)

with \(N\)-body product state vectors \(|\Phi_1\rangle, |\Phi_2\rangle, |\Phi_1^{(i)}\rangle, |\Phi_2^{(i)}\rangle\) defined in terms of pairs of orthogonal states (i.e. \(\langle \phi_j \mid \phi^+_j \rangle = 0\)) as

\[
|\Phi_1\rangle = \otimes_{j=1}^{N} |\phi_j\rangle, \quad |\Phi_1^{(i)}\rangle = \otimes_{j=1}^{i-1} |\phi_j\rangle \otimes |\phi^+_j\rangle \otimes_{j=i+1}^{N} |\phi_j\rangle,
\]

\[
|\Phi_2\rangle = \otimes_{j=1}^{N} |\phi^+_j\rangle \quad \text{and} \quad |\Phi_2^{(i)}\rangle = \otimes_{j=1}^{i-1} |\phi^+_j\rangle \otimes |\phi_i\rangle \otimes_{j=i+1}^{N} |\phi^+_j\rangle.
\]

(17)

With the prefactor \(a_{KN}\) defined as

\[
a_{KN} = 1/(N - K + 1) \quad \text{for} \quad K \neq 2, \quad a_{KN} = 1/N \quad \text{for} \quad K = 2,
\]

\(\tau_{KN}\) is non-positive for all \(N\)-body quantum states with a single excitation that do not have at least \(K\)-site quantum coherence. The normalization constant \(b_{KN}\) is chosen such that \(\tau_{KN}\) adopts the value of unity for the state \(|W_{KN}\rangle = \sum_{i=1}^{K} |i\rangle/\sqrt{K}\), i.e. the state of a system with \(N\) sites and an excitation that is perfectly coherently delocalized over \(K\) sites.

Although not strictly necessary for identification of \(K\)-site coherence, we always perform a numerical optimization over the state-vectors \(|\phi_j\rangle\). This can be done by means of standard unconstraint nonlinear optimization routines which are part of all recent numerical computation environments and computer algebra packages. Most standard routines are well suited for finding the global maximum for cases in which the value \(a_{KN}\) in equation (16) is small. A good choice is \(a_{KN} = 1/N\), i.e. the detection of two-site coherence is usually very reliable. Larger values of \(a_{KN}\), however, increase the probability of being trapped in a local maximum. To avoid local maxima we thus apply a two-fold approach. First, we choose a small value for \(a_{KN}\) for which we determine the optimal \(|\phi_j\rangle\) by means of standard optimization routines. We then slightly increase the value for \(a_{KN}\) and repeat the numerical optimization with the starting point defined by the vector \(|\phi_j\rangle\) which has been obtained by the previous optimization. These two steps of determining the optimal choice of \(|\phi_j\rangle\) which serves as the new starting point followed by an increase of \(a_{KN}\) are alternately applied until the desired value of \(a_{KN}\) which is necessary to identify \(K\)-site coherence is reached. This procedure was found to be the most reliable way to reproducibly obtain \(\tau_{KN}\) also for large values of \(K\).

4.2. Coherent excitation transport under incoherent driving

With these tools at hand, we can now strive for the characterization of coherence properties and their examination with respect to the stationary transport efficiencies. That is, we would like to verify if coherent delocalization of the excitation is necessary for fast, efficient transport, or whether the identification of this precondition made in the transient regime [10] is no longer possible due to the permanent de-cohering impact of the coupling to heat baths.

There are two extreme regimes, in which the recombination rate \(\gamma_{rec}\) is dominant and negligible, respectively as compared to the inter-site coupling strengths. In the former case,
where there is no path for the excitation to reach the output faster than the limit set by \( \gamma_{\text{rec}} \), the efficiencies are close to zero. In the latter case, where \( \gamma_{\text{rec}} \) is small compared to the average intersite coupling, the excitation will leave through the sink almost with certainty as the probability of a backflow to the source is low and all networks will be characterized efficient independently of the question whether transport is coherent or not.

We will therefore focus in the following on the intermediate regime where the value of \( \gamma_{\text{rec}} \) is of the same order of magnitude as the typical site–site coupling, i.e. large enough to permit the identification of fast transport, but still small enough to enable a significant sink flux. We will investigate the coherence properties of all networks with similar currents, that is we introduce a binning of the \( E_s \)-axis with windows of width \( \Delta E_s \) centered around \( E_s \). We then consider the average \( K \)-site coherence

\[
\bar{\tau}_{K,N}(E_s) = \langle \tau_{K,N}(\tilde{\rho}) \rangle_{E_s(\tilde{\rho}) \in w_{E_s}}
\]

with the average taken over all networks with a current within the window \( w_{E_s} = [E_s - \Delta E_s, E_s + \Delta E_s] \), and the width

\[
\sigma_{K,N}(E_s) = \sqrt{\langle (\tau_{K,N}(\tilde{\rho}) - \bar{\tau}_{K,N})^2 \rangle_{E_s(\tilde{\rho}) \in w_{E_s}}}
\]

of the distribution of \( \tau_{K,N} \) within a bin.

For the absence of dephasing (i.e. for \( \gamma_{\text{deph}} = 0 \)) figures 2(a) and (b) depict the average two- and three-site coherence defined in equation (19) for the recombination rate \( \gamma_{\text{rec}} = 20 \). Since this choice of \( \gamma_{\text{rec}} \) is of the same order of magnitude as the typical interaction strength between two sites, this amounts to rather short-time dynamics. The standard deviation \( \sigma_{K,N} \) given in equation (20) is depicted by a bordered area centered around the average value. Additionally to \( \sigma_{K,N} \) caused by the variance of the coherence properties of different random networks in the

Figure 2. Expectation value \( \bar{\tau}_{K,N}(W_{K'}) \) (dark green) and standard deviation \( \sigma_{K,N}(W_{K'}) \) (light green) of the coherence \( \tau_{K,N}(W_{K'}) \) of \( K = 2 \) (a) and 3 (b) sites as a function of the stationary transport efficiency \( E_s \) for randomly arranged dephasing-free networks (\( \gamma_{\text{deph}} = 0 \)). The error bars show the statistical error on the sample mean \( \bar{\tau}_{K,N} \) and on the standard deviation \( \sigma_{K,N} \). The dashed lines represent the coherences \( \tau_{K,N}(W_{K'}) \) for pure \( W_{K'} \)-states and indicate the maximal value of \( \tau_{K,N} \) that can be obtained without \( (K' + 1) \)-site coherent contribution. Whereas rather strong two-site coherence can be found for almost all systems the most efficient networks excel by a significant extent of three-site coherence.
same efficiency interval we have to account for a statistical error \( S \) on the sample mean \( \bar{\tau}_{KN} \) as well as on \( \sigma_{KN}^2 \) due to the finite sample size. These errors are indicated by error bars and can be estimated by \( S(\bar{\tau}_{KN}) = \sigma_{KN}/\sqrt{n} \) for the average value and \( S(\sigma_{KN}) = \Sigma(\sigma_{KN}^2)/(2\sqrt{\sigma_{KN}^2}) \) for the standard deviation where \( n \) is the number of networks per bin and

\[
\Sigma^2(\sigma_{KN}^2) = \frac{1}{n} \left( \mu_4 - \frac{n-3}{n-1} \sigma_{KN}^4 \right)
\]

is the variance of the sample variance \( \sigma_{KN}^2 \) [32] with \( \mu_4 = \langle (\bar{\tau}_{KN} - \tau_{KN}(\bar{\omega}))^4 \rangle_{E_i(\bar{\omega}) \in \mathcal{E}_N} \) being the fourth moment about the mean. One finds these statistical error \( S \) to increase for higher efficiencies as efficient networks are less likely to be randomly sampled as compared to rather inefficient configurations [10]. Whereas sample sizes of roughly \( 2 \times 10^3 \) networks per bin are a sound foundation for obtaining reliable expectation values, the bin with the highest efficiency contains less than a hundred networks even though the bin size has been increased.

Despite this statistical error, figure 2 shows a strong correlation between exciton current and coherence, i.e. networks that feature maximal transport also show substantial coherence. Two-site coherence can be detected in almost all networks independently of the excitation flux but gains significance as efficiency increases. As the value of \( \tau_{2\gamma}(W_2) \) obtained for a pure \( W_2 \)-state is significantly exceeded already for fairly low efficiencies (see figure 2(a)) one can expect a relevant contribution of three-site coherence, which is confirmed by scrutinizing \( \bar{\tau}_{3\gamma} \) in figure 2(b). The enhancement of efficiency thus requires a substantial extent of coherent delocalization.

In contrast to the case of \( \tau_{2\gamma} \), however, significant three-site coherence cannot be identified in every network. In the lowest quarter of the efficiency spectrum, i.e. for \( E_s \leq 0.05 \), values of \( \bar{\tau}_{3\gamma} \geq 0.5 \) can for example only be found for 46% of all networks. In the most efficient regime \( E_s \geq 0.15 \), this is the case for more than 98% of all systems. This makes three-site coherence a crucial prerequisite for efficient transport.

Coherent delocalization over more than three sites is hardly detectable. As this does not change for larger networks of \( N = 9 \) or 12, we consider this not to be a finite-size effect due to the limited number of sites in the first instance but a consequence of the short time-window provided for establishing coherence which is governed by the maximal excitation lifetime \( \gamma_{rec}^{-1} \). Positive values for \( \bar{\tau}_{4\gamma} \) can still be found sporadically but are negligible as compared to \( \tau_{4\gamma}(W_4) = 1 \) and the average value \( \bar{\tau}_{4\gamma} \) is smaller than the standard deviation for all efficiencies what prevents any statistical significance. Considering results obtained in a study of the purely coherent transient case [10] we must, however, question whether stronger four-site coherence would indeed yield more efficient transport. While an examination of the coherent case revealed \( K \)-site coherence with \( K = 2 \) and 3 to be strictly required for high transport efficiency, this relation is weakened drastically for \( K \geq 4 \). This is reasonable as coherent delocalization as a requirement for constructive interference competes with localization on the output site in order to obtain optimal efficiency. The transport properties for two- and three-site coherent states in the transient scenario do therefore perfectly agree with the results shown here and underline the similarity between coherently and incoherently induced transport.

Despite the dephasing impact of the reservoirs and the recombination, networks of suitable geometry have been shown to induce a sufficient amount of coherence which has been identified as a prerequisite for optimal transport. To test if the correlation between coherence and efficiency persists under the application of additional dephasing, we modify the situation discussed before by changing the dephasing rate to \( \gamma_{deph} = 10 \). The coherence time \( \gamma_{deph}^{-1} = 0.1 \)
Figure 3. Expectation value $\bar{\tau}_{K7}$ (dark blue) and standard deviation $\sigma_{K7}$ (light blue) of the coherence $\tau_{K7}$ of $K = 2$ (a) and $3$ (b) sites as a function of the stationary transport efficiency $E_s$ for randomly arranged networks exposed to a dephasing of $\gamma_{\text{deph}} = 10$. The error bars show the statistical error on the sample mean $\bar{\tau}_{K7}$ and on the standard deviation $\sigma_{K7}$. Comparing the results with the dephasing-free scenario depicted in figure 2 (shown here dashed and in green) one finds the correlation between coherence and efficiency to be robust under dephasing. Despite the decrease in efficiency of formerly optimal networks the most efficient systems still employ a significant extent of coherence which thus can be deemed required for high efficiencies.

is now comparable to the excitation lifetime $\gamma_{\text{rec}}^{-1} = 0.05$ that defines the time window relevant for the system dynamics. A comparison of the transport and coherence properties in the dephasing-free approach depicted in figure 2 and the case of additional dephasing assumed for figure 3 suggests that the incorporation of additional noise does not qualitatively change the relation between coherence and transport efficiency. Whereas a finite value of $\gamma_{\text{deph}}$ leads to reduced coherence, the correlation between $\bar{\tau}_{K7}$ and transport efficiency stays rather unaffected. This is in perfect agreement with the decrease of the maximal efficiency when comparing figures 2 and 3, which shows that optimal networks loose efficiency when they are exposed to dephasing. Despite the additional noise, suitable systems are, however, capable to successfully employ interference as long as the system can accumulate a sufficient amount of coherence.

5. Conclusion

Based on a comparison of transport efficiencies we found strong similarities between the coherently induced transient and the incoherently induced stationary excitation transport suggesting that the underlying transport mechanisms of these two scenarios are rather equivalent. As quantum coherence is capable to enhance the transport efficiency in the transient scenario, this similarity raises the question whether quantum coherence in the site basis can also be maintained in the stationary state that is obtained under constant and incoherent driving and how it relates to the transport efficiency.

An application of recent tools from entanglement theory [23] on a sample of incoherently driven random networks enabled us to identify a strong correlation between coherence and transport efficiency and provides a clear interpretation of the considered concept of coherence.
which in our context always refers to a coherent delocalization of an excitation over various sites. For the first time, we could quantify the extent of this delocalization, i.e. the number of the sites that are involved, and relate it to the stationary transport efficiency. Despite the decohering effects of the source and the drain, quantum coherence can be established in the stationary state and is even required to obtain optimal transport. Furthermore, we could show that this correlation between coherence and transport efficiency is robust against additional dephasing as long as the coherence time does not exceed the transport time.

With regard to light-harvesting complexes or artificial systems engineered to turn light into electrical energy our results suggest in particular that the transport process induced by coherent laser pulses permits to draw conclusions on the propagation under constant and incoherent driving by natural sun light. That is, suitable molecular networks are capable to exploit quantum coherent delocalization of excitations for the purpose of efficient excitation transport even if they are fed by an antenna that is coupled to a thermal light source rather than a coherent light source. Whether a network employs quantum coherent effects to enhance the transport efficiency thus does not depend on the driving mechanism in the first instance but is rather a question of the network geometry and the coherence time. Provided the coherence time is at least of the same order as the transport time, quantum coherence can be maintained also in the stationary state. Under the premise that two-dimensional spectroscopic measurements can indeed verify electronic coherence times as typically assumed [33, 34], but also not unambiguously agreed upon [35], our results suggest that such measurements can be used to qualitatively estimate quantum coherence also in the stationary states.

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