Survival Probabilities in Coherent Exciton Transfer with Trapping

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In the quest for signatures of coherent transport we consider exciton trapping in the continuous-time quantum walk framework. The survival probability displays different decay domains, related to distinct regions of the spectrum of the Hamiltonian. For linear systems and at intermediate times the decay obeys a power-law, in contrast to the corresponding exponential decay found in incoherent continuous-time random walk situations.

To differentiate between the coherent and incoherent mechanisms, we present an experimental protocol based on the theoretical side, such investigations are of long standing; see, e.g., [1]. Here, tight-binding models, which model coherent exciton transfer, are closely related to the quantum walks (QW). As their classical random walk (RW) counterpart, QW appear in two variants: discrete-time QW [2] and continuous-time QW (CTQW) [3]. Experimental implementations have been in microwave cavities [4], ground state atoms [5] or Rydberg atoms [6] in optical lattices, or the orbital angular momentum of photons [7].

An appropriate means to monitor transport is to follow the decay of the excitation due to trapping. The long time decay of chains with traps is a well studied problem for classical systems [8, 9]: for an ensemble of chains of different length with traps at both ends the averaged exciton survival probability has a stretched exponential form exp(−b(−λt)), with λ = 1/3 (see, e.g., [9]). In contrast, quantum mechanical tight-binding models lead to λ = 1/4 [10, 11]. However, up to now only little is known about the decay of the quantum mechanical survival probability at experimentally relevant intermediate times.

Here we evaluate and compare the intermediate-time decays due to trapping for both RW and QW situations by employing the similarity of the CTRW and the CTQW formalisms. Without traps, the coherent dynamics of excitons on a graph of connected nodes is modeled by the CTQW, which is obtained by identifying the Hamiltonian H₀ of the system with the CTRW transfer matrix T₀, i.e., H₀ = −T₀; see e.g. [3, 12] (we will set ℏ = 1 in the following). For undirected graphs, T₀ is related to the connectivity matrix A₀ of the graph by T₀ = −A₀, where (for simplicity) all transmission rates are taken to be equal. Thus, in the following we take H₀ = A₀. The matrix A₀ has non-diagonal elements 0 kj the values −1 if nodes k and j of the graph are connected by a bond and 0 otherwise. The diagonal elements 0 kj of A₀ equal the number of bonds f j which exit from node j. By fixing the coupling strength between two connected nodes to |H0 kj | = 1, the time scale is given in units of [ℏ/H0 kj]. For the Rydberg gases considered in the following, the coupling strength is roughly H0 kj/ℏ ≳ 1 MHz, i.e., the time unit for transfer between two nodes is of the order of a few hundred nanoseconds.

The states |j⟩ associated with excitons localized at the nodes j (j = 1, . . . , N) form a complete, orthonormal basis set (COBS) of the whole accessible Hilbert space, i.e., ⟨k|j⟩ = δkj and ∑k |k⟩⟨k| = 1. In general, the time evolution of a state |j⟩ starting at time t₀ = 0 is given by |j⟩ = |j⟩ exp(−iH₀t)/ηj; hence the transition amplitudes and the probabilities read αkj(t) = ⟨k|exp(−iH₀t)/ηj|j⟩ and πkj(t) = |αkj(t)|², respectively. In the corresponding classical CTRW case the transition probabilities follow from a master equation as pkj(t) = |k⟩ exp(T₀t)/ηj |j⟩ [3, 13].

Consider now that out of the N nodes M are traps with M ≤ N; we denote them by m, so that m ∈ M, with M ⊂ {1, . . . , N}. We incorporate trapping into the CTQW formalism phenomenologically by following an approach based on time dependent perturbation theory [10, 11, 13]. The new Hamiltonian is H = H₀ + ιΓ, where the trapping operator ιΓ has the trap nodes m purely imaginary diagonal elements ιΓmm, which we assume to be equal for all m (Γmm ≡ Γ > 0), and is zero otherwise. As a result, H is non-hermitian and has N complex eigenvalues, ζl = ℏl − γm (l = 1, . . . , N). In general, H has N left and N right eigenstates |Φl⟩ and ⟨Φl|, respectively. For most physically interesting cases the eigenstates can be taken as biorthonormal, ⟨Φl|Φl'⟩ = δll', and complete, ∑l=1N |Φl⟩⟨Φl| = 1; see, e.g., Ref. [14]. Moreover, we have |Φl⟩* = ⟨Φl|k). Thus, the transition amplitudes can be calculated as αkj(t) = ∑l=1N exp[−γl t]|Φl⟩⟨Φl|Φl|k⟩; here the imaginary parts γl of ζl determine the temporal decay of πkj(t) = |αkj(t)|².

In an ideal experiment one would excite exactly one node, say j /∈ M, and read out the outcome πkj(t), i.e., the probability to be at node k /∈ M at time t. However, it is easier to keep track of the total outcome at all nodes k /∈ M, namely, ∑k/∈M πkj(t). Since the states |k⟩ form a COBS we have...
depend on the mutual distance between the nodes as \( R \). The trapping of the exciton occurs at the ends of the chain. The dipolar interactions between Rydberg atoms depend on the mutual distance \( R \) between the nodes as \( R^{-3} \).

Now, CTRW over a chain of regularly arranged sites lead both for nearest-neighbor steps and for step distributions depending on \( R \) as \( R^{-\gamma} \), with \( \gamma > 3 \), to a standard diffusive behavior and, therefore, belong to the same universality class, see e.g. [16]. The reason is that in one dimension for \( \gamma > 3 \) the first two moments, \( \langle R \rangle \) and \( \langle R^2 \rangle \), are finite. Thus, although the quantitative results will differ, the qualitative behavior is similar. Hence, we focus on a nearest-neighbor tight-binding model and consider a chain of length \( N \) with two traps \((M = 2)\) located at its ends \((m = 1 \text{ and } m = N)\), [24]. The CTQW Hamiltonian thus reads

\[
H = \sum_{n=1}^{N} \left( 2|n\rangle\langle n| - |n-1\rangle\langle n| - |n+1\rangle\langle n| \right)
+ i\Gamma \sum_{m=1,N} |m\rangle\langle m|.
\]

For CTRW we include trapping in a formally similar fashion as for the CTQW. Here, however, the classical transfer matrix \( T_0 \) is modified by the trapping matrix \( \Gamma \), such that the new transfer matrix is \( T = T_0 - \Gamma \), [15]. For a single linear system with traps at each end, the mean survival probability \( P_M(t) = -N/M \sum_{j \notin M} \sum_{k \notin M} \pi_{kj}(t) \) decays exponentially at intermediate and at long times [18]. As we proceed to show, the decays of \( \Pi_M(t) \) and \( P_M(t) \) are very different, thus allowing to distinguish experimentally whether the exciton transfer is coherent or not.

For long \( t \) and small \( M/N \), Eq. (2) simplifies considerably: At long \( t \) the oscillating term on the right hand side drops out and for small \( M/N \) we have \( 2 \sum_{m \in M} |\Phi_t| m \rangle \langle m | \Phi_t| \ll 1 \). Thus, \( \Pi_M(t) \) is mainly a sum of exponentially decaying terms:

\[
\Pi_M(t) \approx \frac{1}{N-M} \sum_{i=1}^{N} \exp[-2\gamma t].
\]

Asymptotically, Eq. (3) is dominated by the \( \gamma_l \) values closest to zero. If the smallest one, \( \gamma_{min} \), is well separated from the other values, one is led for \( t > 1/\gamma_{min} \) to the exponential decay found in earlier works, \( \Pi_M(t) = \exp(-2\gamma_{min} t) \) [11].

Such long times are not of much experimental relevance (see also below), since most measurements highlight shorter times, in which many \( \gamma_l \) contribute. In the corresponding energy range the \( \gamma_l \) often scale, as we show in the following, so that in a large range \( \gamma_l \sim a \mu \). The prefactor \( a \) depends only on \( \Gamma \) and \( N \) [11]. For densely distributed \( \gamma_l \) and at intermediate times one has, from Eq. (3),

\[
\Pi_M(t) \approx \int dx \ e^{-2at x} = \int dy \ \frac{e^{-y \mu}}{(2at)^{1/\mu}} \sim t^{-1/\mu}.
\]

The envisaged experimental setup consists of clouds of ultra-cold Rydberg atoms assembled in a chain over which an exciton migrates; the trapping of the exciton occurs at the ends of the chain. The dipolar interactions between Rydberg atoms depend on the mutual distance \( R \) between the nodes as \( R^{-3} \).
and in the intermediate time domain

\[ N \] scales (lower three curves). Indicated are the fits to \( \Pi_M(t) \) (long
dashed lines) in the intermediate (upper red) and the long (lower
blue) time regime.

FIG. 2: (Color online) Temporal decay of \( \Pi_M(t) \) (solid black lines)
and \( P_M(t) \) (short dashed green lines) for \( N = 100 \) and \( \Gamma = 1 \)
in double logarithmic scales (upper three curves) and in logarithmic
scales (lower three curves). Indicated are the fits to \( \Pi_M(t) \) (long
dashed lines) in the intermediate (upper red) and the long (lower
blue) time regime.

We now turn to the parameter dependences of \( \Pi_M(t) \). Figure 3 displays
the dependence of \( \Pi_M(t) \) on \( N \). We note that the scaling regime, where \( \Pi_M(t) \sim t^{-1/\mu} \), gets larger with increasing \( N \). The cross-over to this scaling region from the
domain of short times occurs around \( t \approx N/2 \). For larger \( N \) and in the intermediate time domain \( \Pi_M(t) \) scales nicely with
\( N \). In this case, the power-law approximation [Eq. (3)] holds and by rescaling \( t \) to \( l/N \) we get from Eq. (3) that

\[
\Pi_M(t) \sim \sum_\ell e^{-2N^{-3\mu}t} = \sum_\ell \exp\left[ -2(l/N)^\mu N^{-(3-\mu)t} \right],
\]

where we used that \( a \sim N^{-3} \) for a linear system \([11]\). Thus
when rescaling \( l \) to \( l/N \), time has to be rescaled by the factor \( N^{-3(3-\mu)} \). Indeed, all curves where a power-law behavior can
be justified fall on a master curve; see the inset in Fig. 3.

FIG. 3: (Color online) \( N \)-dependence of \( \Pi_M(t) \) for \( \Gamma = 1 \); \( N \)
increases in steps of 10 from 20 (blue line) to 100 (green line). The
inset shows \( \Pi_M(t) \) versus the rescaled time \( t/N^{3-\mu} \).

The temporal decay does not only depend on \( N \) but also on \( \Gamma \). Figure 4 shows \( \Pi_M(t) \) for \( N = 50 \) and different \( \Gamma \). For
values \( \Gamma \gg 1 \) (green lines) and \( \Gamma \ll 1 \) (black lines) the curves
shift to longer times. Values of \( \Gamma \) close to 1 (blue lines) lead
to the quickest decay. Note that these values are of the same
order as the coupling strength between the non trap nodes,
\( H_{j,j\neq 1} = -1 \).

An experimental implementation of the described system
has to meet several criteria. A single node must represent
a well-defined two-level system to ensure coherent energy
transfer while at the same time a mechanism is needed to
trap an exciton with a controllable trapping efficiency.
Furthermore, the chain must be static with negligible motion and
should allow for spatially selective excitation and detection of
the exciton. These demands rule out many possible candidates
for an experimental realization of CTQW. A frozen Rydberg
gas [17] can meet all of the above demands by combining
the rich internal structure of highly excited atoms with the
full quantum control over the external degrees of freedom that
is available in up-to-date experiments with ultracold atoms.
The internal structure of Rydberg atoms provides both decoupled
two-level subsystems and tunable traps, while the pronounced
Stark shift allows to selectively address single sites in a
chain when an electric field gradient is applied. At the same
time, experimentally accessible temperatures below 1 \( \mu \)K en-
sure that the thermal motion is negligible.

Our scheme starts from a cloud of laser-cooled ground state
atoms prepared in a chain of optical dipole traps [18]. Each
site represents one node with distances between sites of 5 to
20 \( \mu \)m. For an experimentally achievable extension of 1 mm
this translates into approximately 100 nodes. All nodes are ex-
cited to Rydberg states exploiting the dipole blockade mecha-
nism to ensure a single Rydberg excitation per node [19]
which avoids many-body effects [20]. A two-level system is
realized by a transition between energetically isolated states,
i.e., by low-angular-momentum states which exhibit a large
quantum defect, e.g., \( nS \rightleftharpoons n'P \). A number of ex-
periments has revealed the coherent character of this process [20]. By contrast to low-\( \ell \) states, states with angular
momenta \( \ell \geq 3 \) have no quantum defect and are degenerate.
This allows to construct an exciton trap with the transitions
\( n'D \rightleftharpoons n''F \rightleftharpoons n''\ell (\ell \geq 3) \), where the first transition is the
dipole transition providing the coupling to neighboring nodes
[23] while the second transition, driven by a radio-frequency

![Graph](https://via.placeholder.com/150)

FIG. 4: (Color online) \( \Gamma \)-dependence of \( \Pi_M(t) \) for intermediate \( t \)
and \( N = 50 \).
(rf) field, represents the trap and decouples this site from the energy transfer, as the large degeneracy of the high-\(\ell\) field, ensures an efficient suppression of the coupling back to the \(11\) state [26]. By changing the strength of the driving rf field, the trapping efficiency can be tuned. The population of the \(\ell\) state is directly proportional to \(1 - M(\ell, 1)\), and can be determined by state selective field ionization [21]. In an experiment the central nodes would be prepared in the S state and the trap nodes in the D state. A single S node is swapped to P through a microwave transition in an electric field gradient which makes the resonance S\(\rightarrow\)P position sensitive. This is equivalent to exciting a single exciton. The energy transport is started by removing the field gradient making the transition energy the same for all nodes.

There are two important decoherence mechanisms which are given by the spontaneous decay of the involved Rydberg states and by the atomic motion. Exemplarily, for the 71S and 61D states of rubidium and a distance of 20\(\mu\)m between nodes we calculate a transfer time of \(\tau = 145\) ns between two neighboring sites, radiative lifetimes including black-body radiation of \(\geq 100\) \(\mu\)s and residual thermal motion that leads to a change of the interatomic distance of 1.4 \(\mu\)m per 100 \(\mu\)s at a temperature of 1 \(\mu\)K. Another source of decoherence is the interaction-induced motion \([23]\). We can model this motion quantitatively \([23]\) and calculate negligible changes of the interatomic distances of less than 0.2 \(\mu\)m per 100 \(\mu\)s. This means that both the chain and the elementary atomic system sustain coherence over timescales on the order of several ten \(\mu\)s and longer.

In conclusion, we have identified different time domains in the CTQW exciton decay in the presence of traps, domains which are directly related to the complex spectrum of the system’s Hamiltonian. The CTQW average survival probability \(\Pi_M(\ell)\) for an exciton to stay inside a linear system of \(N\) nodes with traps at each end can clearly be distinguished from its classical CTRW counterpart, \(P_M(\ell)\). Finally, we proposed an experimental test for coherence on the basis of ultra-cold Rydberg atoms.

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[24] All numerical results were obtained by using FORTRAN’s LAPACK routines for diagonalizing non-hermitian matrices.
[25] In order to ensure the right coupling strength to neighboring nodes both the energy difference and the transition dipole moments of the processes \(nS \rightarrow nP\) and \(nD \rightarrow n''F\), \(\Delta E_S/P = \Delta E_D/B = h\ 1.0\) GHz and radial transition matrix elements of 5200 au and 4800 au, respectively.
[26] Note that the rf frequency is detuned for any transitions in the other nodes as those involve different atomic states.