Discrete-time quantum simulation of non-Markovian behaviour of transport dynamics in FMO complex

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The Fenna-Mathews-Olson (FMO) complex present in green sulphur bacteria are known to mediate the transfer of excitation energy between light-harvesting chlorosomes and membrane-embedded bacterial reaction centres. Due to high efficiency of such transport process, it is an extensively studied pigment-protein complex system with the eventual aim of modelling and engineering similar dynamics in other systems and use it for real time application. Some studies have attributed the enhancement of transport efficiency to wave-like behaviour and non-Markovian quantum jumps resulting in long lived and revival of quantum coherence, respectively. Since dynamics in these systems resides in quantum-classical regime, quantum simulation of such dynamics will help in exploring the subtle role of quantum features in enhancing the transport efficiency which has remained unsettled. Discrete quantum simulation of the FMO complex dynamics can help in efficient engineering of the heat bath and controlling the environment with the system. In this work, using the discrete quantum simulation framework we quantify and show that the non-Markovian memory effects are present in specific site-pairs when internal structures and environmental effects are in favour of faster transport. As a consequence, our study lean towards the connection between non-Markovianity in the quantum jumps with the enhancement of transport efficiency.

I. INTRODUCTION

Photosynthesis is one of the primordial process in nature that provides energy for sustaining life. Dynamics of photosynthetic organisms, which have been successfully harvesting and transferring solar energy for several billion years are far more efficient than any man-made devices known till date\cite{1–4}. Decades of studies have attributed the remarkable efficiency of the photosynthesis process to light-harvesting antenna complexes which are funneling the excitation energy from captured photons to a reaction center where it is converted to the chemical energy. Photosynthetic complexes have differences based on their living conditions and habitats, but all follow the same procedure: they absorb solar energy (photons) in the form of electronic excitation by an antenna, and then this excitation is transported to a reaction center where charge separation transforms it into a more stable form of energy\cite{5, 6}. The most simple and well-studied examples of such a light harvesting system are found in green sulfur bacteria, which is an organism that depends only on sunlight as a source of energy. Its antenna is quite large and made out of chlorosomes which allows them to thrive in extremely low light conditions. A special kind of structured complex can be found in them, called the Fenna-Mathews-Olson (FMO) complex. These complexes connects the antenna to the reaction centre\cite{7, 8}. They are small in size, water-soluble, and, most importantly, they transports excitation from the antenna to the reaction centre with more than 90\% efficiency. Given that these excitons are short-lived (lifetime is less than a nanosecond), the efficiency of the transport process has motivated and encouraged intense research in the direction of understanding and modelling environment assisted energy transport\cite{9–12}.

Mathematical modelling to mimic the observed dynamics is one of the most effective way to understand the dynamics in these photosynthetic complexes. In the year 2007, a new model was presented with the evidence for wave-like behaviour and presence of quantum coherence during these exciton transport\cite{13}. Photosynthesis usually occurs at ambient temperature, and the presence of quantum coherence at such temperature for 300 – 500 fs led to numerous research in the direction of exploring environment assisted quantum transport to find further evidences for presence of quantum coherence for high transport efficiency\cite{14}. Numerous study on FMO complex has outlined the structural details of the FMO complex and that has been used to study dynamics in quantum-mechanical framework\cite{15}. In the widely accepted form, the FMO complex is a trimer formed by three identical monomers that each bind seven Bacteriochlorophyll-a (BChla) molecules, since monomers function independently, without loss of generality, studies are restricted to a single monomer. There are seven sites in the FMO complex, and it is assumed that there is at most one exciton in the complex at any time. Considering one exciton at any time is a reasonable assumption because these bacteria usually receive very less sunlight. The initial excitation occurs at site 1 or 6 and is transported to the sink at sites 3 and 4, the two dominant pathways for exciton transfer are (1 → 2 → 3) and (6 → (5, 7) → 4 → 3).
In the wave-like description of dynamics in FMO complex which has put forward the contribution of quantum coherence for better efficiency, the wave function of the excitation enters a superposition state of multiple combined pigments instead of the excitation transferring from pigment to pigment sequentially. It helps the exciton to traverse several paths at once and find the shortest route from the antenna to the reaction center [17]. A more rigorous and generic theoretical treatment on the FMO complex in quantum mechanical framework has been presented in two seminal works by Plenio & Huelga [18] and Mohseni et al. [19], respectively. There, it has been shown that only coherence dynamics cannot transport exciton with such high efficiency, and the interplay between coherent dynamics and environmental induced noise transports exciton with such high efficiency. For these studies, the FMO complex and its surrounding environment have been modeled into an open quantum system set-up, and quantum master equations have been used to solve these problems [20, 21].

Studies have also shown that the coherence might be helpful in transport dynamics, but it does not contribute to the high efficiency of this exciton energy transport dynamics [22]. An other recent study argue that the contribution from quantum coherence is minute at best in photosynthesis transport process [23]. Other than quantum coherence, quantum features like entanglement and quantum memory effects (non-Markovianity) have been explored additionally to find their impacts on this highly efficient transport process. The presence of bipartite entanglement in the FMO complex has been detected [16] and it has been tried to understand the importance it has in the dynamics of the FMO complex with its usefulness in quantum technologies. Studies have also described the possibility that the non-Markovianity might be helpful in this type of transport process [24]. The role of quantum features and quantum advantage in photosynthesis continues to remain unsettled paving way for further investigations.

In addition to all these previously used techniques to study FOM complex, studies have focused to quantum simulation approach. In quantum simulation approach, one can engineer the dynamics and control the parameters in the dynamics that could effectively lead to the observed phenomena and help in understanding the complex properties in FMO system. With noisy intermediate-scale quantum (NISQ) devices [25] being available, FMO complex systems which are defined on 7-site system can be simulated on a smaller number of qubit, NISQ devices. Quantum simulators are available in two different formats: analog quantum simulators and digital quantum simulators. Analog quantum simulators use continuous-time evolution equations, like the Lindblad master equation. Digital quantum simulations use universal quantum gates to simulate the discrete-time evolution equation, thus it provides flexibility and universality over analog quantum simulation [26, 27]. Analog quantum simulations of the FMO complex have been reported using ultracold atoms [28], superconducting circuits [29, 30] and NMR quantum computer [31, 32]. Mahdian et al. presented a setup for digital simulation of the FMO complex, but their study does not include the interplay of coherent dynamics and quantum jumps [33].

Recently, a framework for digital quantum simulation for FMO complex has been presented which includes the interplay of coherent dynamics and quantum jumps [34]. Using the controllable parameters in the digital quantum simulation framework using interplay of coherent dynamics and quantum jumps, one can quantify non-trivial quantum features in the dynamics and have a better understanding of the high-efficient exciton transport process of FMO complex.

In this work, using the discrete-time quantum simulation model we study the dynamics in site pairs of FMO complex and quantify the non-trivial quantum features which may aid this high efficient transport. For simulating site-pairs in digital simulation method, we have prepared a theoretical framework by taking conceptual understanding from the previous studies. We have simulated different site-pairs of FMO complex and used trace distance and BLP measure to identify and quantify, respectively the non-Markovian memory effects in the dynamics. The simulation results shows the presence of non-Markovian memory effects in some specific site pairs. This memory effect is controlled by the structural features of the FMO complex (site couplings and site energy differences) and environmental influence. This results signifies that digital simulation method can be useful for studying finer details of photosynthetic complex. Our results match with previous theoretical findings and show that non-Markovian memory effects is present in those conditions where internal structures and environmental effects are in favour of faster transport.

This article is organised as follows, in Sec. II we present the model with open quantum system framework to model the system-environment coupling, study site-pair dynamics and method to quantify the non-Markovian behavior in the dynamics. In Sec. III we presents the results for different pairs of sites in the FMO complex and analyze non-Markovian behaviour in the dynamics and in Sec. IV we conclude with the summary of our observations.

II. MODEL AND METHODS

The FMO complex serves as a transport channel in green sulphur bacteria which transports the exciton energy from site 1 or site 6 to site 3. The dynamics of FMO complex systems is championed by quantum features and can be modeled into open quantum system dynamics. Thus, its quantum and dissipative effects can be captured using a combination of Kraus operators and unitary quantum evolution. By using those discrete time evolution operators the exciton energy transport in 7-site
FMO complex can be effectively and efficiently simulated using digital quantum simulation method [34]. Thus, in this work we use the previously proposed open quantum system framework of energy transfer in 7 site FMO complex to prepare simulation framework for site-pairs (2-site system). In Fig. 1, the schematic structure of FMO complex is shown where each sphere represents a BChl site. Initial excitation occurs at site 1 or 6 and it is transported to sink sites 3 and 4 using sites 2, 5, and 7 as pathways in different possible configurations.

**FIG. 1: A schematic structure of 7-site FMO complex where each sphere represents a BChl a site. Site 1 and 6 represent the initial site where excitation occurs. Site 3 and 4 represent the sink site to which excitation gets transported through intermediate sites, 2, 5, and 7.**

A. Framework for simulating 2-sites system

At the time of working with a specific site pair, we can treat the specific site pair with other sites as the system and the protein environment as the bath, or the specific site pair as the system and the protein environment with other sites as the bath. In the first case, the system will be represented by 3 qubits, and in the second case, the system will be represented by 1 qubit [as for representing \(2^n - 1\) sites, we need a minimum of \(n\) qubits]. If we treat the bath with minimum one qubit, then in the first case we have to deal with 4 qubits or \(2^4\) dimensional space, and in the second case we have to deal with 2 qubits or \(2^2\) dimensional space. The second option has been chosen here to allow for less computational complexity.

For simulating site-pairs of FMO complex in digital quantum simulation method, we need to first initialize the state, follow it up with state evolution by taking all environmental effect into consideration and final state is measured. Before initializing the state, we will demonstrate this problem using an illustration. In Fig. 2, the two level system is shown where the two levels are denoted by \(|0\rangle\) and \(|1\rangle\). The two sites of the FMO complex are mapped into these two levels. The bath induces a jump between these two sites. Here, the jump probability is denoted by \(\gamma_{i\rightarrow j}\) where \(i\) and \(j\) are two sites. The initial system-environment state can be represented by

\[
|\psi(0)\rangle = |i_S\rangle \otimes |0_E\rangle.
\]  

(1)

At the time of evolution, we need to capture the dissipative environmental effects as well as its internal unitary dynamics. The dissipative effects can be represented in form of environment-induced quantum jump which is implemented as

\[
|i_S\rangle |0_E\rangle \mapsto \sqrt{1-\gamma_{i\rightarrow j}} |i_S\rangle |0_E\rangle + \sqrt{\gamma_{i\rightarrow j}} |j_S\rangle |1_E\rangle.
\]  

(2)

It can be implemented using a quantum channel (in matrix form) rather than in the form of Kraus operators. The matrix form can be written as

\[ M = M_1C, \]

(3)

where \(M_1\) and \(C\) are both matrices given by,

\[
M_1 = \begin{bmatrix}
\sqrt{1-\gamma_{i\rightarrow j}} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 \\
\end{bmatrix}
\]  

(4)

\[
C = \begin{bmatrix}
1 & 0 & 0 & 0 \\
0 & 0 & 0 & 1 \\
0 & 0 & 1 & 0 \\
0 & 1 & 0 & 0 \\
\end{bmatrix}
\]  

(5)

Here, our main goal is to focus on the dynamics between the two sites of FMO, so the coherent dynamics between two sites is implemented by,

\[ U \equiv U_S \otimes I, \]

(6)

where

\[
U_s = e^{-i\frac{\text{H}t}{\hbar}},
\]

(7)

**FIG. 2: A schematic illustration of two level system which is used to represent two sites where bath induces jump between the sites with probability \(\gamma_{i\rightarrow j}\).**


and

\[
H_c = \begin{bmatrix}
H_{11} & H_{12} \\
H_{21} & H_{22}
\end{bmatrix}.
\] (8)

The \( \gamma \) values of Eq. 2 and the Hamiltonian \( H_C \) of Eq. 8 have been taken from ref. [35], [36] and [37]. The combined system and environment state is evolved as per Eq. 3 and Eq. 6 and we trace out environment at each step to see whether the reduced dynamics of system has any non-Markovian effects.

### B. Trace distance and BLP measure

The trace norm of a trace class operator \( A \) is defined by \( ||A|| = tr|A| \), where the modulus of the operator is given by \( |A| = \sqrt{A^\dagger A} \). If \( A \) is self-adjoint, the trace norm can be expressed as the sum of the moduli of the eigenvalues \( a_i \) of \( A \) counting multiplicities,

\[
||A|| = \sum_i |a_i|.
\]

This norm leads to a natural measure for the distance between two quantum states \( \rho_1 \) and \( \rho_2 \) known as trace distance,

\[
D(\rho_1(t), \rho_2(t)) = \frac{1}{2} ||\rho_1(t) - \rho_2(t)||. \tag{9}
\]

To quantify the non-Markovianity of the reduced dynamics of the system, we can use the Breuer-Laine-Piilo (BLP) measure. The BLP measure is defined as follows

\[
N = \int_{\sigma > 0} \sigma(t, \rho_{1,2}(0)) dt, \tag{10}
\]

where

\[
\sigma(t, \rho_{1,2}(0)) = \frac{d}{dt} D(\rho_1(t), \rho_2(t)). \tag{11}
\]

According to this definition, the non-Markovian process will have a positive finite value for \( N \) where the value of the Markovian process of \( N \) will always be 0 [38–40]. We use trace distance to detect the presence of non-Markovianity in the dynamics and later we quantify the non-Markovianity using the BLP measure. Eq. 2 is an implementation of a quantum jump, which happens due to the effect of environment on the system, and Eq. 6 is an implementation of coherent dynamics which is due to couplings between different sites.

### C. Framework for tunable bath couplings

FMO dynamics depends on environmental assistance and in this framework, the environmental effects are implemented using quantum jumps. Thus, for variable system-bath coupling, quantum jump rates will be affected by the coupling constant. Now in this scenario, the environment-induced quantum jump is implemented as follows:

\[
|i\rangle_S 0 |E \rightarrow \chi \sqrt{1 - \gamma_{i \rightarrow j}} |i\rangle_S |0\rangle_E + \chi \sqrt{\gamma_{i \rightarrow j}} |j\rangle_S |1\rangle_E, \tag{12}
\]

where \( \chi \in [0,1] \). For this scenario, the modified matrix \( M_1 \) \( (M_1') \) will take the form,

\[
M_1' = \begin{bmatrix}
\chi \sqrt{1 - \gamma_{i \rightarrow j}} & 0 & 0 & 0 \\
\chi \sqrt{\gamma_{i \rightarrow j}} & 0 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1
\end{bmatrix}. \tag{13}
\]

### III. RESULTS AND DISCUSSIONS

Each site pairs of FMO complex have different features which facilitate the exciton energy transfer through specific paths. Thus, here our primary goal is to check whether non-Markovianity or memory effects have any influence on the exciton energy transfer dynamics of the FMO complex. For doing that some selected site pairs are simulated using the framework presented in Sec. II. In addition to that, trace distance and BLP measure are calculated to detect the presence of non-Markovianity and quantify non-Markovianity, respectively in the dynamics. Fig. 3 presents the numerically calculated change in trace distance with time (fs) for sites 1 and 2. The trace distance as function of time does not decrease monotonically as we can see a small increase a few instances for some \( t \geq 0 \). This denotes the presence of non-Markovianity in its dynamics. This result agrees with the result from ref. [24] that showed strongly coupled sites 1 and 2 have signature of non-Markovianity in the dynamics. Similarly, we have calculated the relation between trace distance and time for other site pairs to get information about non-Markovian influence in dynamics.
FIG. 4: Trace distance as function of time for site-pairs 1 - 2 and 3 - 5. For the directly connected sites (1-2) we can see small oscillation in trace distance whereas a monotonic decrease is seen for pair which has no direct connection.

FIG. 5: Trace distance as function of time for site-pairs 3 - 4 and 2 - 5. For the directly connected sites (3-4) we can see small oscillation in trace distance whereas a monotonic decrease is seen for pair which has no direct connection.

FIG. 6: Trace distance as function of time for site-pairs 4 - 7 and 3 - 6. For the directly connected sites (4-7) we can see small oscillation in trace distance whereas a monotonic decrease is seen for pair which has no direct connection.

At the time of exciton energy transfer, it is transferred from site 1 or 6 to site 3, mainly through two dominant transfer pathways, which are $1 \rightarrow 2 \rightarrow 3$ and $6 \rightarrow (5,7) \rightarrow 4 \rightarrow 3$ as shown in Fig. 1. Thus, we have chosen 3 site pairs that are in the dominant transfer pathways (directly connected pairs) and other 3 site pairs that are not in the dominant transfer pathways to find the difference in the dynamics present in them. In Fig. 4, Fig. 5 and Fig. 6 the blue line curves with triangular symbol are for those site pairs present in the dominant transfer pathways and the green line curves are for those site pairs which are not present in dominant transfer pathways. We see that from Fig. 4, Fig. 5 and Fig. 6, site pairs from dominant transfer paths show small oscillations in trace distance with time, that is, $D(\Phi(\rho_1), \Phi(\rho_2))$ is increasing for some $t \geq 0$, which means that information is coming back into the system from environment. This indicates that those pair of sites have non-Markovianity in their dynamics. The presence of non-Markovianity or memory effects in the dominant transport pathways of the FMO complex is an interesting result that is obtained here by simply considering site pairs in discrete-simulation framework and it clearly concur with previous theoretical understandings [24]. It can be understood that the site pairs present in the dominant transfer pathways have site coupling much higher than the others. Site coupling plays a very important role in exciton energy transfer because on the basis of site couplings the exciton de-localizes in between sites, which gives an exponential speedup to this transport. From this result, we can say that the non-Markovianity which is present only in some specific site pairs depends on the site coupling, an internal feature of the system.

We have explicitly quantified the presence of non-Markovianity for those site pairs with dominant couplings. For weakly coupled pairs, the trace distance decreases monotonically with time, thus the BLP measure is zero for such pairs according to Eq. 10. Fig. 7 shows the change of the BLP measure with time for all those sites where the trace distances do not decrease monotonically. The trace distance detects the presence of non-Markovianity and the BLP measure quantifies that the non-Markovianity present. Fig. 6 shows that in the case of site-pairs 4-7, the non-Markovian oscillations stay longer than for site-pairs 1-2 and 3-4. As a result, in Fig. 7 we can see that the BLP measure grows with time and also has a higher value. In the other two cases, the BLP measure has a lower value and almost saturates after a certain time.

This result is consistent with other theoretical findings [24] and it can be understood from the fact that non-Markovianity decreases with increasing site energy difference. Site pair 4-7 has much lesser site energy difference than site pairs 1-2 and 4-7, as a result site pair 4-7 has higher non-Markovianity. If we look into the FMO complex, physical understanding is possible for decreas-
FIG. 7: BLP measure as function of time for different site pairs with dominant pathways. The BLP measure quantifies the non-Markovianity and we can see that the non-Markovian oscillation stay for long for site pair 4-7 compared to other pairs.

FIG. 8: Trace distance as function of time for site pair 1-2 with tunable system-bath couplings. With increase in coupling strength we see an increase in non-Markovian oscillation.

Fig. 8, Fig. 9 and Fig. 10 show the trace distance as function of time with different system-bath coupling strength for different site-pairs, 1-2, 3-4, and 4-7, respectively. The values are calculated for four different coupling constants ranging from 1 to 0.4. One common conclusion can be drawn from all these plots is that the weakly coupled environment leads to less non-Markovian oscillation than the strongly coupled one. In case of strong system-bath couplings, environment is having more influence on the system, thus the lost information from system is getting stored in the environment for some time and again coming back into the system after some time. Thus, we observe more non-Markovian oscillations in case of strong system-bath couplings.

Next, we have quantified non-Markovianity using BLP measure for these site pairs in the case of coupling constant 1 (high level coupling) and 0.4 (low level coupling). Fig. 11, Fig. 12 and Fig. 13 shows the BLP measure as function of time plot for the site pairs 1-2, 3-4, and 4-7, respectively. From the observations we can conclude that in case of low level system-bath coupling (coupling constant=0.4), BLP measure is zero which denotes no information back-flow into system indicating that it is a Markovian dynamics, where in case of high level system-bath coupling (coupling constant=1.0) BLP measure gives non-zero value which denotes information back-flow into the system from environment indicating the existence of non-Markovianity in dynamics. Thus, here we can see that by using variable system-bath cou-
plings, it is possible to tune and interchange between non-Markovian and Markovian dynamics. One more important point to note here is that that system-bath couplings depend on temperature, thus by tuning temperature it is possible to interchange between Markovian and non-Markovian dynamics. The site-pairs of the FMO complex were simulated using discrete-time quantum simulation approach to get detail information about their dynamics and the presence of non-Markovianity has been detected using trace distance and the BLP measure method. From our results, it can be established that the presence of non-Markovianity in some specific site pairs depends on the internal structure of the FMO complex and the influence of the environment.

IV. CONCLUSIONS

We have presented a theoretical framework for digital quantum simulation of site-pair dynamics of FMO complex. The site-pair description along with tunability of system-environment coupling presented in this work has paved way to engineer the dynamics and explore the presence of quantum features across variable coupling parameters. We have used trace distance to establish the presence of non-Markovian memory effects and BLP measure to quantify the memory effect between site pairs with variable system-environment coupling strength. This memory effect is controlled by structural features of the FMO complex (site couplings and site energy differences) and environmental influence. Our results using discrete-time approach match with previous theoretical findings using continuous-time approach and show that non-Markovian memory effects is present in those conditions where internal structures and environmental effects are in favour of faster transport. Using BLP measure we could also show that the memory effect is zero even for dominant pathways when the system-environment coupling is weak. Therefore, critical coupling strength is important to see memory effect even for the prominent pathways.

We may conclude from these results that the non-Markovian memory effects facilitate energy transfer by giving special importance to certain site pairs that help the exciton to travel through a certain direction from antenna to sink. This study also sheds light on the fact that memory effects can help in noise-assisted transport dynamics, which can be useful for real-life applications. This study shows non-Markovianity is dependent on the system structure and environment couplings, which is an important information if one wants to get information back-flow in such noise-assisted transport dynamics. As discrete (digital) quantum simulation provides universal- ity, the same kind of framework can be applied to other photosynthesis complexes as well and can be simulated using quantum circuits. Here, the digital simulation scheme was able to capture memory effects; thus, we may say that in some other situations, digital simulation can be helpful in capturing such finer details in dynamics.

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