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Generalized master equation with non-Markovian multichromophoric Förster resonance energy transfer for modular exciton densities

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A generalized master equation (GME) governing quantum evolution of modular exciton density (MED) is derived for large scale light harvesting systems composed of weakly interacting modules of multiple chromophores. The GME-MED offers a practical framework to incorporate real time coherent quantum dynamics calculations of small length scales into dynamics over large length scales, and also provides a non-Markovian generalization and rigorous derivation of the Pauli master equation employing multichromophoric Förster resonance energy transfer (MC-FRET) rates. A test of the GME-MED for four sites of the Fenna-Matthews-Olson complex demonstrates how coherent dynamics of excitonic populations over coupled chromophores can be accurately described by transitions between subgroups (modules) of delocalized excitons. Application of the GME-MED to the exciton dynamics between a pair of light harvesting 2 (LH2) complexes in purple bacteria demonstrates its promise as a computationally efficient tool to investigate large scale exciton dynamics in complex environments.

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Most photosynthetic units of bacteria and higher plants have modular structures where the entire systems are composed of smaller subunits, or “modules” of protein-chromophore complexes[1, 2]. While the nature of interactions and quantum dynamics within each module varies, the inter-module interactions in such systems are generally weak. A striking characteristic is that excitons can migrate through those weak links and find their destinations with near unit efficiency within picoseconds. How can this be accomplished despite significant disorder and fluctuations? What are the general conditions ensuring such high efficiency of natural systems? Recent theoretical studies provide some clues[3–7], but the answers for the above fundamental questions are far from being settled. To this end, simulation of exciton dynamics over larger length and longer time scales including realistic effects of disorder/fluctuations is needed. However, accurate quantum dynamical calculations typically apply to small (∼ 7 chromophores) [8, 9] or medium range systems having up to ∼ 30 chromophores [10–13], with the latter already requiring massive computational resources. Thus, application of such approach to simulation of complexes with hundreds of chromophores (e.g., photosystem II) also while averaging over sufficiently large ensemble of disorder is impractical at present. Instead, the Pauli master equation (PME) is frequently used for such calculations[14–20], but without clear derivation of its kernels from quantum dynamical principles. As such, it is difficult to establish connection between the observed phenomenology and the key microscopic features.

In this work, we derive a generalized master equation (GME) for the time evolution of the exciton density coarse-grained over a module, i.e., the modular exciton density (MED). The resulting GME-MED complements a recent analysis of coherence propagation between weakly coupled multichromophore units[6], clarifies assumptions underlying the use of multichromophoric Förster resonance energy transfer (MC-FRET) rate[3, 21, 22] in a PME, and provides its non-Markovian generalization. We also demonstrate that the GME-MED serves as a practical means to incorporate high level intra-module quantum calculations into energy transfer simulation over significantly longer length scales.

Consider a total Hamiltonian given by $H = H_0 + H_c$, where $H_0$ represents noninteracting modules of excitons plus their environmental degrees of freedom and $H_c$ the

FIG. 1: Schematic of a modular system. Arrows represent transition dipoles of each chromophore, dotted lines their electronic couplings, and wavy lines exciton-bath couplings. The grey region represents MED.
couplings between different modules. Each module is denoted as \( n \) or \( m \), and a chromophore in the \( n \)th module is denoted as \( j_n \), \( k_n \), etc. Thus,

\[
H_0 = \sum_n H_n = \sum_n \left( H_n^e + \sum_{\ell=j_n} B_{\ell j_n} |\ell_n\rangle \langle j_n| + H_n^g \right), \tag{1}
\]

with \( H_n^e \) the single exciton Hamiltonian of the \( n \)th module, \(|\ell_n\rangle\) the site excitation state of the \( \ell \)th chromophore in the \( n \)th module, \( B_{\ell j_n} \) the bath operator coupled to the excitonic term \(|\ell_n\rangle\langle j_n|\), and \( H_n^g \) the bath Hamiltonian (the Hamiltonian in the ground electronic state) of the \( n \)th module. The inter-module coupling Hamiltonian has the form:

\[
H_c = \sum_{n,m} \sum_{j_n k_m} J_{j_n k_m} |j_n\rangle \langle k_m|, \tag{2}
\]

where \( J_{j_n k_m} \) is assumed to be real and symmetric. By definition, \( J_{j_n k_m} = 0 \) for \( n \neq m \). For generality, we assume that \( H_n^c, B_{\ell j_n}, \) and \( H_c \) are time dependent (although not shown explicitly) but that \( H_n^g \) is time independent. Figure 1 provides a schematic modular structure.

We denote the time evolution operator for the interaction free Hamiltonian \( H_0 \) as \( U_0(t, t') = \exp_{(+)} \{-i \int_t^{t'} d\tau H_0(\tau)/\hbar\} = \prod_n U_n(t, t') \), where the subscript \( (+) \) is implied chronologically. Assuming that the exciton is created at time \( t = 0 \), we shall abbreviate \( U_0(t, 0) = U_n(t, 0) = U_0(t) \) and \( U_n(t) \) the total density operator is denoted as \( \rho(t) \). In the interaction picture with respect to \( H_0 \), \( \rho(t) = U_0^\dagger(t) \rho U_0(t) \) evolves according to the equation

\[
\frac{\partial}{\partial t} \rho(t) = -i[H_c, \rho(t)] = -i\{H_c, \rho(t)\}/\hbar = -i\mathcal{L}_{c,j}(t),
\]

where \( \mathcal{L}_{c,j}(t) = U_0^\dagger(t) H_c U_0(t) \), the second equality defines \( \mathcal{L}_{c,j}(t) \). The ground state time evolution operator of the \( n \)th module is denoted as \( \rho_n(t) = \exp[-iH_n^g t/\hbar] \). Since \(|\ell_n\rangle\) represents the state where only the \( \ell \)th chromophore in the \( n \)th module is excited while all other modules are in the ground electronic state, \(|\ell_n\rangle|U_0(t)\rangle = \left( \prod_{n \neq \ell} U_0^\dagger(t) \right) |\ell_n\rangle|U_0(t)\rangle \). Thus,

\[
H_{c,j}(t) = \sum_{n,m} \sum_{j_n k_m} J_{j_n k_m} T_{j_n k_m}(t) = \sum_{n,m} \mathcal{F}_{nm}(t), \tag{3}
\]

where \( T_{j_n k_m}(t) = U_0^\dagger(t) U_0^\dagger(t) |j_n\rangle \langle k_m|U_0^\dagger(t) U_0(t) \) and the second equality defines \( \mathcal{F}_{nm}(t) \). By definition, \( \mathcal{F}_{nm}(t) \) vanishes for \( n = m \). We denote the identity operator in the single exciton space of each module as \( I_n = \sum_{j_n} |j_n\rangle \langle j_n|\) and that in the total single exciton space as \( I = \sum_n I_n \). The equilibrium bath canonical density operator of the \( n \)th module in the ground electronic state is \( \rho_0 = e^{-\beta H_n^g}/\text{Tr}_b \{ e^{-\beta H_n^g} \} \).

The key idea in deriving the GME-MED is to introduce the following modular projection super-operator \( \mathcal{P} \):

\[
\mathcal{P}(\cdot) = \sum_{n} \rho_{nc_n} \mathcal{Tr}_{bC_n} \{ 1_n(\cdot)1_n \}, \tag{4}
\]

where \( \{ \cdot \} \) represents an arbitrary operator, \( \rho_{nc_n} = \prod_{n \neq \ell} \rho_0 \), and \( \mathcal{Tr}_{bC_n} \) represents the trace over all baths except for those associated with the \( n \)th module. The super-operator \( \mathcal{P} \) projects the total density operator into an independent sum of blocks, each representing a module, and satisfies the required condition of \( \mathcal{P}^2 = \mathcal{P} \). It also satisfies the identity of \( \mathcal{P} \mathcal{L}_{c,j}(t) \mathcal{P} = 0 \) (see Supplemental Material (SM)). We assume a simple initial condition at time \( t = 0 \) with no intermodule quantum coherence because the likelihood of such a coherent state under natural condition is very small, so that \( (1 - \mathcal{P}) \rho_0(0) = 0 \). Then, from the well-known formal solution for \( \mathcal{P} \rho(t) \) (see SM), it is easy to derive the following time evolution equation for the total \( n \)th module density operator, \( \rho_{n,t}(t) = \mathcal{Tr}_{bC_n} \{ 1_n \rho_0(t) 1_n \} \):

\[
\frac{\partial}{\partial t} \rho_{n,t}(t) = -\sum_{m} \int_0^t d\tau \mathcal{L}_{c,j}(t) \mathcal{L}_{c,j}(\tau) \rho_{nc_m} \rho_{n,m,t}(\tau) \}, 1_n, \tag{5}
\]

which is formally exact but not amenable for practical solution yet. Under the assumption that the inter-module coupling \( H_c \) is small compared to \( H_0 \), the approximation \( e^{-i\int_0^t d\tau' (1 - \mathcal{P}) \mathcal{L}_{c,j}(\tau')} \approx 1 \) can be made in Eq. (5). This results in the following 2nd order approximation with respect to \( H_c \):

\[
\frac{\partial}{\partial t} \rho_{n,t}(t) = -\sum_{m} \int_0^t d\tau \mathcal{L}_{c,j}(t) \mathcal{L}_{c,j}(\tau) \rho_{nc_m} \rho_{n,m,t}(\tau) \}, 1_n, \tag{6}
\]

The above equation provides a complete prescription to incorporate full quantum dynamics calculations for each module (made using, e.g., the methods of [23, 24]) into a consistent description of the dynamics across all coupled modules. Note that the only assumption invoked here is the smallness of \( H_c \) compared to \( H_0 \). This assumption can be used as long as a natural division into weakly coupled modules exists. This can be justified in most light harvesting super complexes, with the possible exception of the chlorosome of the green sulfur bacteria[25], which appears to have an extended network of coupled chromophores. However, even in this case, the excitons have finite coherence lengths due to disorder or exciton-phonon couplings and application of our formalism may be feasible in this case by appropriately choosing modules comparable to the coherence sizes of excitons [20].

When the main focus is on the exciton states, the equation for the reduced system density operator, \( \sigma_{n,t}(t) = \mathcal{Tr}_{bn} \{ \rho_{n,t}(t) \} \), can be obtained by tracing Eq. (6) over the bath of the \( n \)th module and employing the explicit
expression for $H_{c,t}(t)$ of Eq. (3). The result becomes

$$\frac{\partial}{\partial t}\sigma_{n,t}(t) = -\frac{1}{\hbar^2} \sum_{m\neq n} \int_0^t d\tau \left( \text{Tr}_b \{ F_{nm}(t) \rho_{n,\tau}(t) \rho_{bC_n} \} + \text{Tr}_b \{ \rho_{bC_n} \rho_{n,\tau}(t) \rho_{C_m} F_{mn}(t) \} - \text{Tr}_b \{ F_{nm}(t) \rho_{bC_m} \rho_{n,\tau}(t) \rho_{C_m} F_{mn}(t) \} - \text{Tr}_b \{ F_{nm}(t) \rho_{bC_m} \rho_{n,\tau}(t) \rho_{C_m} F_{mn}(t) \} \right), \quad (7)$$

where the fact that $\text{Tr}_b \rho_{bC_n} = \rho_{bC_n}$ has been used.

The integrands of Eq. (7) can be related to lineshape operators of each module. For this, we introduce the following exciton space operators:

$$\mathcal{I}_n(t,\tau) = \text{Tr}_b \{ U_n(t,\tau) 1_n \rho_{bC_n} U_{n}^\dagger(t,\tau) \}, \quad (8)$$

$$\mathcal{E}_n(t,\tau;\rho_n) = \text{Tr}_b \{ U_{n}^\dagger(t,\tau) \rho_n U_n(t,\tau) \}, \quad (9)$$

where $\rho_n(\tau) = U_n(\tau) \rho_n U_n^\dagger(\tau)$. The Fourier transform of $\mathcal{I}_n(t,\tau)$ with respect to $t-\tau$ produces the absorption lineshape when contracted with transition dipole vectors. A similar procedure with $\mathcal{E}_n(t,\tau;\rho_n)$ leads to the time dependent emission lineshape depending on $\rho_n(\tau)$ as its initial condition. For complete representation of all the integrands of Eq. (7), we need to define additional operators with specific coherence information built in as follows:

$$\mathcal{I}_{n,j_n^m,j_n^m'}(t,\tau) = \text{Tr}_b \{ U_n(t,\tau) 1_{n,j_n^m,j_n^m'} U_{n}^\dagger(t,\tau) \rho_{bC_n} \}, \quad (10)$$

$$\mathcal{E}_{n,j_n^m,j_n^m'}(t,\tau;\rho_n) = \text{Tr}_b \{ U_{n}^\dagger(t,\tau) \rho_n U_n(t,\tau) \}, \quad (11)$$

Then, it is possible to show (see SM) that Eq. (7) is equivalent to the following time evolution equation:

$$\frac{\partial}{\partial t} \langle j_n^m|\sigma_{n,t}(t)|j_n^m'\rangle = -\frac{1}{\hbar^2} \sum_{m\neq n} \sum_{j_n^m,k_n^m} \sum_{j_n^m',k_n^m'} J_{j_n^m,k_n^m} J_{j_n^m',k_n^m'}$$

$$\times \int_0^t d\tau \left\{ \langle k_n^m|\mathcal{I}_m(t,\tau)|k_n^m'\rangle |j_n^m'|\mathcal{E}_{n,j_n^m'j_n^m'}(t,\tau;\rho_n)\rangle \langle j_n^m'| \right\}$$

$$+ \langle k_n^m|\mathcal{I}_m(t,\tau)|j_n^m\rangle |j_n^m'|\mathcal{E}_{n,j_n^m'j_n^m'}(t,\tau;\rho_n)\rangle \langle j_n^m'| \right\}$$

$$- \langle j_n^m|\mathcal{I}_m(t,\tau)|j_n^m'\rangle |k_n^m'|\mathcal{E}_{n,j_n^m'j_n^m'}(t,\tau;\rho_n)\rangle \langle k_n^m' | \right\}$$

$$- \langle j_n^m|\mathcal{I}_m(t,\tau)|j_n^m'\rangle |k_n^m'|\mathcal{E}_{n,j_n^m'j_n^m'}(t,\tau;\rho_n)\rangle \langle k_n^m' | \right\}. \quad (12)$$

The GME for the MED, $p_n(t) = \sum_{j_n^m} \langle j_n^m|\sigma_{n,t}(t)|j_n^m\rangle$, can be obtained by summing the diagonal components of Eq. (12) and utilizing the fact that $\mathcal{I}_n(t,\tau) = \sum_{j_n^m} \mathcal{I}_{n,j_n^m,j_n^m}(t,\tau)$ and $\mathcal{E}_n(t,\tau;\rho_n) = \sum_{j_n^m} \mathcal{E}_{n,j_n^m,j_n^m}(t,\tau;\rho_n)$, yielding

$$\frac{\partial}{\partial t} p_n(t) = -\frac{1}{\hbar^2} \sum_{m\neq n} \sum_{j_n^m,k_n^m} \sum_{j_n^m',k_n^m'} J_{j_n^m,k_n^m} J_{j_n^m',k_n^m'}$$

$$\times 2\text{Re} \int_0^t d\tau \left\{ \langle k_n^m|\mathcal{I}_m(t,\tau)|k_n^m'\rangle |j_n^m'|\mathcal{E}_{n,j_n^m'j_n^m'}(t,\tau;\rho_n)\rangle \langle j_n^m'| \right\}$$

$$- \langle j_n^m|\mathcal{I}_m(t,\tau)|j_n^m'\rangle |k_n^m'|\mathcal{E}_{n,j_n^m'j_n^m'}(t,\tau;\rho_n)\rangle \langle k_n^m' | \right\}. \quad (13)$$

Higher order versions of this equation can be obtained from Eq. (5) by following similar procedures including higher than second order terms[27, 28, 40]. In the limit where each module consists of a single chromophore, the GME-MED reduces to the GME for localized excitons[29, 30].

Equation (13) is the main formal result of the present letter, but its solution requires full knowledge of the total density operator of each module, due to the dependence of $\mathcal{E}_n(t,\tau;\rho_n)$ on $\rho_n(\tau)$. We now describe a generic approximation removing such dependence, which is implicit in applications employing MC-FRET rates in the PME[14, 18, 20] and is believed to be appropriate for many natural photosynthetic systems. To simplify the argument, we shall assume that all $H_n$ are time independent. Then, $U_n(t,\tau) = U_n(t-\tau)$ and $\mathcal{I}_n(t,\tau) = \mathcal{I}_n(t-\tau,0) \equiv \mathcal{I}_n(t-\tau)$. If the dynamics driving intra-module detailed balance are fast compared to the inter-module population dynamics, one may invoke the following steady state approximation: $\rho_n(\tau) \approx \rho_n^s \rho_n(\tau)$, where $\rho_n^s = e^{-\beta H_n}/\text{Tr}_b \{ e^{-\beta H_n} \}$. This does not imply complete time scale separation between intra-module and inter-module dynamics, and takes the full effect of exciton-bath entanglement into consideration through $\rho_n^s$. With this approximation, $\mathcal{E}_n(t,\tau) \approx \mathcal{E}_n^s(t-\tau) \rho_n(\tau)$, where $\mathcal{E}_n^s(t) = \text{Tr}_b \{ U_{n}^\dagger(t) \rho_n U_n(t) \}$. Equation (13) then reduces to the following closed-form expression:

$$\frac{\partial}{\partial t} p_n(t) = \sum_{m\neq n} \int_0^t d\tau \{ \mathcal{K}_{n\rightarrow m}(t-\tau) p_m(\tau) - \mathcal{K}_{m\rightarrow n}(t-\tau) p_n(\tau) \} \quad (14)$$

where

$$\mathcal{K}_{n\rightarrow m}(t) = \frac{2}{\hbar^2} \text{Re} \sum_{j_n^m,k_n^m} \sum_{j_n^m',k_n^m'} J_{j_n^m,k_n^m} J_{j_n^m',k_n^m'}$$

$$\times \langle k_n^m|\mathcal{I}_m(t)|k_n^m'\rangle \langle j_n^m'|\mathcal{E}_n^s(t)|j_n^m\rangle \quad (15)$$

The GME-MED of Eq. (14) can now be solved employing the pre-determined kernels of Eq. (15). Alternatively, a time-local version of Eq. (14) can also be derived following a similar step but utilizing the cumulant expansion approach[31]. This results in

$$\frac{\partial}{\partial t} p_n(t) = \sum_{m\neq n} \{ W_{n\rightarrow m}(t)p_m(t) - W_{m\rightarrow n}(t)p_n(t) \} \quad (16)$$

where $W_{n\rightarrow m}(t) = \int_0^t d\tau \mathcal{K}_{n\rightarrow m}(\tau)$. In the Markovian limit where all the intra-module exciton dynamics are much faster than the inter-module dynamics, both Eqs. (14) and (16) become equivalent and reduce to the PME with the following MC-FRET rate[22]:

$$W_{n\rightarrow m}(\infty) = \sum_{j_n^m,k_n^m} \sum_{j_n^m',k_n^m'} J_{j_n^m,k_n^m} J_{j_n^m',k_n^m'}$$

$$\times \int d\omega \mathcal{E}_n^s(j_n^m)(\omega) I_{m,k_n^m}(\omega). \quad (17)$$
where $f_m^{k_m'}(\omega) = \int_{-\infty}^{\infty} dt \ e^{i\omega t} \langle k_m(t)|k_m'\rangle$ and $E_m^{n,j}\omega^{n,j}(\omega) = 2 \ Re \int_{-\infty}^{\infty} dt \ e^{-i\omega t} \langle j_n'|E_n^{\omega}(t)|j_n\rangle$. This analysis clarifies the assumptions involved in the use of MC-FRET in PME (see also Ref. 32), while also providing a non-Markovian generalization of that approach.

As a simple demonstration of accuracy, we consider a system consisting of bacteriochlorophylls (BChls) 1-4 in the Fenna-Matthews-Olson (FMO) complex and its protein bath, using parameters adopted from previous works[6, 8] and modeling them as a two-module system (Fig. 2 (a)). The exciton Hamiltonian of each module is given by $H_n^B = E_1 |1_n\rangle\langle 1_n| + E_2 |2_n\rangle\langle 2_n| + \Delta_n (|1_n\rangle\langle 2_n| + |2_n\rangle\langle 1_n|)$, for $n = 1, 2$. Any type of spectral density for the bath can be used for the GME-MED. However, in order to make comparison with the hierarchical equation of motion (HEOM) approach[33], which is limited to the Ohmic-Drude spectral density[6, 8], we use the same spectral density, assuming the usual site-local reorganization energy of $\lambda = 35$ cm$^{-1}$ and Drude cutoff at $\hbar\omega_c = 106$ cm$^{-1}$. The resulting modular excitonic densities calculated for two different initial conditions, one starting from $|1_1\rangle$ and the other starting from $|2_1\rangle$ are shown in Fig. 2 as blue and red dashed lines, respectively, at $T = 150$ and 300 K. Although the exciton population at each BChl is sensitive to the initial condition and exhibits strongly coherent behavior (see insets), the dynamics of modular exciton density is monotonic and much less sensitive to the initial condition.

Employing Eq. (16), the time dependent MED was then calculated with two approximations for Eq. (15). The first neglects the off-diagonal elements of exciton-bath couplings in the exciton basis, while including all the diagonal exciton-vibrational terms (GME-MED-1). The second employs a second order time-local quantum master equation approach neglecting initial exciton-bath coupling (GME-MED-2). Detailed expressions are provided in the SM. The results in Fig. 2(b) show excellent agreement of GME-MED-1 with the corresponding HEOM populations, both in the initial times and the steady state limits. In contrast, the GME-MED-2 results are much less accurate.

The excellent agreement of GME-MED-1 with HEOM at both low and room temperatures suggests that non-equilibrium effects, inter-module non-adiabatic couplings, and quantum coherence, none of which are fully accounted for here, have minor effects. Furthermore, comparison with the results in the Markovian limit (not shown) confirms that non-Markovian effects are also not significant. On the other hand, the relatively poor performance of GME-MED-2 demonstrates the importance of an accurate description of the exciton-bath coupling, some of which can be improved through non-perturbative treatment of the initial condition through analytic continuation of time [32]. These results illustrate the importance of using sufficiently accurate lineshape expressions for PME calculations[14, 17–20] to attain reliable accuracy.

As a further example demonstrating the capability of GME-MED in simulating large scale systems comparable to those studied recently by phenomenological approaches[20, 34, 35], we have calculated the exciton population dynamics between a pair of B850 rings in adjacent LH2 light harvesting complexes of a purple bacterium, Rps. Acidophila. Each ring constitutes a module...
containing 18 bacteriochlorophyll molecules, for a total of 36 chromophores. Each B850 ring is described by a well tested exciton Hamiltonian and spectral density[3]. The inter-B850 BCHl electronic couplings are given by transition dipole interactions, which is an accurate description at the typical distances in *Rps. Acidiphila* (≥ 2.5 nm). Figure 3(a) shows the time dependence of exciton population in the donor B850 ring at three different temperatures calculated by GME-MED-1 (black solid lines) and by its Markovian limit, PME with MC-FRET rates (red dotted lines). At short times we see significant differences between the GME-MED and MC-FRET curves, which reflect the influence of the non-Markovian bath dynamics in the GME-MED description compared to the Markovian dynamics assumed in MC-FRET theory. Figure 3(b) shows the temperature dependence of the effective forward rate for GME-MED-1, $k_{eff} = P_A(\infty)/\tau_{tr}$, where $\tau_{tr}$ satisfies the condition of $\ln(P_D(\tau_{tr}) - P_A(\tau_{tr})P_D(\infty)/P_A(\infty)) = -1$ (see SM), and the Markovian MC-FRET rate, $W_{D \to A}(\infty)$. We note that the effective rate reflecting the non-Markovian effect is significantly smaller than the Markovian rate at low temperatures, but approaches it at room temperatures and above. GME-MED-1 is much more efficient than the HEOM approach[10] (each calculation takes only a few seconds on a typical desktop computer), and makes it possible to simulate excitonic energy transfer between e.g., larger aggregates of LH2 with incorporation of energetic disorder, a key feature to explain experimental results[36, 37] and to understand the robustness of energy transfer dynamics in light harvesting systems over a broad range of temperature.

In summary, we have presented a general derivation of a generalized master equation describing the quantum evolution of exciton density in modular materials, GME-MED, which provides a rigorous derivation of the PME with MC-FRET rates and offers its non-Markovian generalization. As a proof of principle demonstration, we showed that this approach allows accurate description of population dynamics between modules in sub-complexes of FMO within which significant electronic coherence exists. As a further demonstration of its applicability to large scale systems, we provided results for exciton energy transfer between B850 units of LH2 complexes that revealed the significance of non-Markovian effects over a range of temperatures. Taken together, these demonstrations support the GME-MED approach as a novel route to calculation of long range transfer of excitonic energy between modules within which electronic coherence contributes.

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[38] See Supplemental Material [url], which includes Refs. [39-43].