Energy transfer between two aggregates in light-harvesting complexes*

Wang Xiao-Lian(汪小莲), Li Hong-Rong(李宏荣)†, Zhang Pei(张 沛), and Li Fu-Li(李福利)

Department of Applied Physics, Xian Jiaotong University, Xian 710049, *China*

(Received 27 April 2013; revised manuscript received 27 May 2013)

Energy transfer processes between two aggregates in a coupled chromophoric-pigment (protein) system are studied via the standard master equation approach. Each pigment of the two aggregates is modeled as a two-level system. The excitation energy is assumed to be transferred from the donor aggregate to the acceptor aggregate. The model can be used to theoretically simulate many aspects of light-harvesting complexes (LHCs). By applying the real bio-parameters of photosynthesis, we numerically investigate the efficiency of energy transfer (EET) between the two aggregates in terms of some factors, e.g., the initial coherence of the donor aggregate, the coupling strengthes between the two aggregates and between different pigments, and the effects of noise from the environment. Our results provide evidence for that the actual numbers of pigments in the chromophoric rings of LHCs should be the optimum parameters for a high EET. We also give a detailed analysis of the effects of noise on the EET.

Keywords: light-harvesting complexes, energy transfer, decoherent effects

PACS: 71.35.–y, 71.23.An **DOI:** 10.1088/1674-1056/22/11/117102

1. Introduction

The energy crisis is one of the most important issues in the 21st century. The world's current energy requirements are far from being satisfied and, as a clean and almost permanent energy source, photosynthesis has been studied for a long time. Because of developments in science and technology, we can now determine the operational mechanisms of photosynthesis on the scale of biological organisms and the structures of light-harvesting complexes (LHCs) in plants or other light-harvesting bacteria.[1] In most light-harvesting processes, light is first captured by solar antennae in photosynthetic bacteria or plants; the absorbed energy is then transferred to the reaction center (RC) where the energy is converted into chemical energy through electron-transfer reactions.[2] There are two types of LHCs in most purple bacteria, a ring-shaped light-harvesting complex I (LHI) and the multiple copies of a similar complex, light-harvesting complex II (LHII). The LHI directly surrounds the RC, while the LHII does not contain the RC. Both LHI and LHII are trans-membrane proteins. In some bacteria and higher plants, the efficiency of energy transfer (EET) is found to be higher than 99%.[1] Recent experimental observations using electronic spectroscopy technology demonstrated that quantum coherence is involved in the processes of excitation energy transfer in LHCs and Fenna–Matthews–Olson (FMO) complexes.[3–7] Revealing the physical mechanism of such processes is not only necessary for scientific exploration but can also help us understand the secrets of photosynthesis and eventually create effective artificial photosynthetic devices.

On the molecular scale, LHCs are multi-site quantum systems, and the associated processes can be understood through quantum dynamics theory. When we address the problems of energy transfer in LHCs, there are many factors that may affect the EET. First, as shown in recent experimental reports,[3–7] the excitation energy transfer in photosynthetic systems takes a wavelike form. As a result, quantum coherence exists in the energy transfer processes and may affect the EET in LHCs. A long-lived quantum coherence survives in the FMO complex at physiological temperature long enough to impact the energy transfer.[8,9] Second, because the wave packet of the photons captured by the antenna pigments is always larger than or comparable to the scale of multi-chromophoric molecules,[1,2] the initial excitation always takes place coherently among the antenna pigments; thus, the EET from the antenna to the RC depends intimately on the quantum superposition properties of the initial states.[10] Third, as open quantum systems, LHCs are unavoidably affected by decoherent processes of the environment, and those processes are believed to always deteriorate the EET. In a series of recent works,[11–15] by considering the interplay between the coherent dynamics of the LHCs and the incoherent actions of the environment, the authors surprisingly found that the dephasing processes will lead to the enhancement of the EET under some conditions. Beyond the normal weak system–bath coupling, Nazir *et al.* investigated the processes of energy transfer and demonstrated a crossover from coherent to incoherent transfer for resonant donor–acceptor pairs with increasing temperature.[16,17] The

*Project supported by the National Natural Science Foundation of China (Grant No. 11174233) and the National Basic Research Program of China (Grant No. 2011CB311807).

†Corresponding author. E-mail: hrli@mail.xjtu.edu.cn

© 2013 Chinese Physical Society and IOP Publishing Ltd

fourth factor that affects the EET is the structures of the LHCs, such as the number of pigments in the chromophoric proteins. For example, there are 32 molecules in the chromophoric ring of LHI; LHII has a double-layered ring structure, the top ring contains 16 (or 18) BChl molecules and the bottom ring contains 8 (or 9) BChl molecules.[1,2] We may understand these numbers to be the natural choice, but why? Can we explain the structures of LHCs, e.g., the numbers of natural BChl molecules to be the optimal parameters for high EET? There is also much interest in showing the genius of quantum phenomena that exist in LHCs, e.g., quantum entanglement in multi-chromophoric light-harvesting complexes has been studied.[18–21] Many other important studies have also been reported recently.[22–29]

As one type of the most complicated biological systems, LHCs optimally implement solar energy conversion. LHCs adapt to all types of environmental conditions, and the coupling properties among the different components of the LHCs satisfy all the requirements of nature.[30] Therefore, the EET in LHCs is in fact the result of competition among the mechanism of the LHCs, environmental noise, and other factors. In spite of the above experimental and theoretical studies, the underlying mechanism in which factors under certain noise conditions support high-efficiency transfer has yet to be precisely identified. Especially in a substantial process, quantum dynamics in LHCs are always accompanied by simultaneous dephasing and dissipation. Therefore, it is necessary to determine if there are any optimal ratios between the dephasing rate and the dissipation rate to enhance the EET. Motivated by the above considerations, we will study the scenarios by proposing a theoretical model and derive the transfer capacities.

In our model, light is first captured by the donor pigments and the excitation is created among them simultaneously. The excitation energy is then transferred to the acceptor aggregate under the influences of the environment. Every pigment in the donor aggregate and the acceptor aggregate is defined as a two-level system; we thus treat each pigment as a quasi spin. Using a master equation in the Lindblad form,[31] we calculate the EET for different initial states and different environmental factors. Our results thoroughly explain how the structures of LHCs, the environmental dephasing, and the dissipation affect the EET.

This paper is organised as follows. In Section 2, a theoretical model is developed to simulate the LHCs, and a simple analytical discussion is presented. In Section 3, numerical results are shown. Finally, conclusions and final remarks are presented.

2. Theoretical model and master equation

We consider a coupled chromophoric-pigment (protein) system in the single-excitation space. The system is composed of a donor aggregate with m pigments and an acceptor aggregate with n pigments. Each pigment is modeled as a two-level system with excited state $|e_i\rangle$ and ground state $|g_i\rangle$. A diagram of the theoretical model is shown in Fig. 1. In our model, the excitation energy is assumed to be transferred from the donor aggregate to the acceptor aggregate; the present setup can be used to describe or simulate many aspects in the LHCs, e.g., the coupling between different chromophoric rings in LHII and the coupling between rings from LHII and LHI.

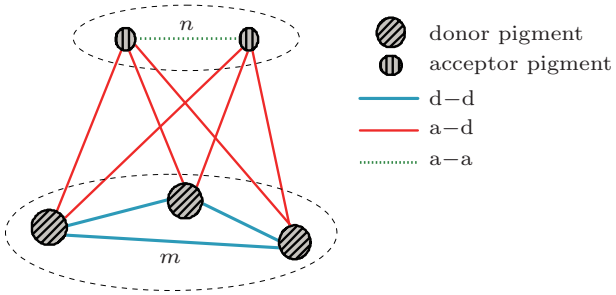


Fig. 1. (color online) Diagram of the theoretical model. The system includes two chromophoric aggregates, the donor aggregate has m pigments, and the acceptor aggregate has n pigments. Each pigment is modeled as a two-level system, and the two different pigments are coupled through dipole–dipole interactions. We use d–d, a–d, and a–a to represent the couplings between two different pigments in the donor aggregate, one pigment in the donor aggregate and one pigment in the acceptor aggregate, and two pigments in the acceptor aggregate, respectively.

The Hamiltonian of the system is

$$H = H_0 + H_{\rm I}, \tag{1}$$

where the site-dependent free Hamiltonian is

$$H_0 = \sum_{i=1}^{m+n} \varepsilon_i \sigma_i^+ \sigma_i^-, \tag{2}$$

and ε_i is the energy difference between the two states of pigment i. We assume that all pigments in the donor aggregate have the same energy difference ε_1, and all of the acceptor pigments have the same energy difference ε_2, i.e.,

$$\varepsilon_i = \begin{cases} \varepsilon_1, & i \le m, \\ \varepsilon_2, & m+1 \le i \le m+n. \end{cases} \tag{3}$$

The Hamiltonian that describes the coupling between every two different pigments is

$$\begin{aligned} H_{\rm I} = & \sum_{i=1}^{m} \sum_{c=m+1}^{m+n} \gamma_{ic} (\sigma_i^+ \sigma_c^- + \sigma_c^+ \sigma_i^-) \\ & + \sum_{i=1}^{m} \sum_{j>i}^{m} J_{ij} (\sigma_i^+ \sigma_j^- + \sigma_j^+ \sigma_i^-) \\ & + \sum_{c=m+1}^{m+n} \sum_{r>c}^{m+n} g_{cr} (\sigma_c^+ \sigma_r^- + \sigma_r^+ \sigma_c^-), \end{aligned} \tag{4}$$

where $\sigma_i^+ = |e_i\rangle \langle g_i|$ and $\sigma_i^- = |g_i\rangle \langle e_i|$ are the raising and lowering operators for site i, respectively, γ_{ic} denotes the coupling strength between the i-th donor pigment and the c-th acceptor pigment, J_{ij} denotes the coupling strength between the i-th

and the j-th pigments of the donor aggregate, and g_{cr} denotes the coupling strength between the c-th and the r-th pigments of the acceptor aggregate. The system is unavoidably affected by the noise of the environment, which can be described with a Markovian master equation[31]

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -\mathrm{i}[H,\rho] + L_{\mathrm{deph}}(\rho) + L_{\mathrm{diss}}(\rho), \tag{5}$$

where the Lindblad super-operator

$$L_{\mathrm{deph}}(\rho) = \sum_{i=1}^{m+n} \Gamma_i'[-\{\sigma_i^+\sigma_i^-,\ \rho\} + 2\sigma_i^+\sigma_i^-\rho\sigma_i^+\sigma_i^-]$$

denotes the dephasing process that destroys the phase coherence in any superposition state of the system, and

$$L_{\mathrm{diss}}(\rho) = \sum_{i=1}^{m+n} \Gamma_i[-\{\sigma_i^+\sigma_i^-,\rho\} + 2\sigma_i^-\rho\sigma_i^+]$$

denotes the dissipative process that dissipates the excitation energy to the environment. We assume that all pigments in the donor (or in the acceptor) aggregate have the same dephasing and dissipation rates, i.e.,

$$\Gamma_i = \begin{cases} \Gamma, & i \le m, \\ \kappa, & m+1 \le i \le m+n, \end{cases} \tag{6}$$

$$\Gamma_i' = \begin{cases} \Gamma', & i \le m, \\ \kappa', & m+1 \le i \le m+n. \end{cases} \tag{7}$$

In the single-excitation space, we choose the basis of state to be $S = \{|0\rangle, |i\rangle\}$, where $|0\rangle$ denotes that all the pigments are in their ground states, and $|i\rangle$ denotes that the i-th pigment is in its excited state and all the other pigments are in their ground states. In this basis and based on the master equation, we can obtain the Bloch equation[31] of the system by calculating all the transition momenta $\rho_{ij} = \langle i|\rho|j\rangle$. We suppose that the excitation first locates at the donor aggregate and then is transferred to the acceptor aggregate, the EET is defined by

$$\eta = 2\kappa \int_0^\infty \sum_{c=m+1}^{m+n} \rho_{cc}\,\mathrm{d}t. \tag{8}$$

The so-defined EET is in fact associated with the total population of the acceptor aggregate. To obtain the dynamics of η, we only need to calculate the sum of the population of all the acceptor pigments in their excited states. We divide the entire transition momentum into several parts. If we use indexes $i,j,k \in [1,m]$ and $c,g,r \in [m+1, m+n]$, then the new variables denoting different parts of the transition momentum are defined by

$$\begin{aligned} j_1 &= \sum_{i=1}^{m}\sum_{j=1}^{m}\rho_{ij}, \quad j_2 - ij_3 = \sum_{i=1}^{m}\sum_{c=m+1}^{m+n}\rho_{ic}, \\ j_4 &= \sum_{c=m+1}^{m+n}\sum_{r=m+1}^{m+n}\rho_{cr}, \quad j_5 = \sum_{i=1}^{m}\rho_{ii},\ j_6 = \sum_{c=m+1}^{m+n}\rho_{cc}, \end{aligned} \tag{9}$$

where j_2 and j_3 are both real variables. From master equation (5), we obtain the following dynamic equations:

$$\begin{aligned} \dot{j}_1 &= -2(\Gamma+\Gamma')j_1 + 2m\alpha_0 j_3 + 2\Gamma' j_5, \\ \dot{j}_2 &= -(\Gamma+\Gamma'+\kappa+\kappa')j_2 - (\varepsilon_1 - \varepsilon_2 + \beta - \gamma)j_3, \\ \dot{j}_3 &= -n\alpha_0 j_1 + (\varepsilon_1 - \varepsilon_2 + \beta - \gamma)j_2 \\ &\quad -(\Gamma+\Gamma'+\kappa+\kappa')j_3 + m\alpha_0 j_4, \\ \dot{j}_4 &= -2n\alpha_0 j_3 - 2(\kappa+\kappa')j_4 + 2\kappa' j_6, \\ \dot{j}_5 &= 2\alpha_0 j_3 - 2\Gamma j_5, \\ \dot{j}_6 &= -2\alpha_0 j_3 - 2\kappa j_6, \end{aligned} \tag{10}$$

$$\dot{\rho}_{00} = 2\Gamma j_5 + 2\kappa j_6. \tag{11}$$

In deducing the above equations, we have assumed that

$$\begin{aligned} &\gamma_{ic} = \alpha_0 \left(\sum_{i=1}^{m}\gamma_{ic} = m\alpha_0 = \alpha\right), \\ &\sum_{i\ne k}^{m} J_{ik} = \beta, \quad \sum_{r\ne c}^{m+n} g_{cr} = \gamma, \end{aligned} \tag{12}$$

where α_0, β, and γ are constants, and the assumption $\gamma_{ic} = \alpha_0$ requires that the distance between the two aggregates is larger than the spatial scale of each single system. If we let $v = 2\kappa j_6$, then $\eta = \int_0^\infty v\,\mathrm{d}t$. By obtaining the Laplace transformation of Eq. (10) under the initial conditions of the states $j_1(0) = A(0), j_2(0) = j_3(0) = j_4(0) = j_6(0) = 0, j_5(0) = 1$, we find

$$\begin{aligned} \bar{v} = \frac{1}{\Delta}\frac{4n\kappa\alpha_0^2}{p+2\kappa}\Bigg[&\frac{A(0)}{p+2\Gamma+2\Gamma'} \\ &+ \frac{2\Gamma'}{(p+2\Gamma)(p+2\Gamma+2\Gamma')}\Bigg], \end{aligned} \tag{13}$$

where $\bar{v}$ represents the Laplace transform of v.

Equation (13) is not the final form for η, however we can find some useful information from it. We observe that $\bar{v}$ includes two terms; the first term is proportional to $A(0) = j_1(0) = \sum_{i,j=1}^{m}\rho_{ij}(0) = \sum_{i=1}^{m}\rho_{ii}(0) + \sum_{i\ne j}^{m}\rho_{ij}(0) = j_5(0) + \sum_{i\ne j}^{m}\rho_{ij}(0)$. Because we have already set $j_5(0) = 1$, $A(0)$ corresponds to the initial coherence of the donor aggregate in fact. The second term is a function of Γ', i.e., the term is a function of the dephasing rate of the pigments in the donor aggregate. The first term means that the EET depends on the initial coherence of the donor aggregate.[10–14] The second term tells us that the dephasing processes of the donor aggregate play nontrivial roles in the energy transfer, and these roles can be understood more clearly when the first term vanishes. In fact, if the initial state of the donor aggregate has the following form:

$$|\Psi(0)\rangle = \frac{1}{\sqrt{m}}\sum_{i=1}^{m}\mathrm{e}^{\mathrm{i}\theta_i}|i\rangle, \quad \theta_i = \frac{2\pi}{m}i, \tag{14}$$

then $A(0) = 0$. In this case, although the initial coherence of the donor aggregate does not vanish, the excitation is trapped among the donor pigments if there is no dephasing due to the

environment. When $\Gamma' \neq 0$, one easily finds that the dephasing processes of the donor aggregate will destroy the trapping conditions and thus enhance the EET; similar results were also shown in other studies.[11–13,32] Because the dephasing rate is a function of temperature,[31] an increase in temperature will increase the dephasing rate and is thus beneficial to the excitation transfer. However, an increase in temperature also normally increases the dissipation rate of the system; this rate is always harmful to the excitation transfer, so an optimal temperature exists for the excitation transfer.[12]

Performing an inverse Laplace transformation on Eq. (13), we obtain

$$v(t) = 4\kappa n\alpha_0^2[2\Gamma'\omega_1(t) + A(0)\omega_2(t)]. \tag{15}$$

Correspondingly, we can divide the EET into two parts

$$\eta = \eta_1 + \eta_2, \tag{16}$$

where

$$\begin{aligned} \eta_1 &= 4n\kappa\alpha_0^2 \int_0^\infty 2\Gamma'\omega_1(t)\,\mathrm{d}t, \\ \eta_2 &= 4n\kappa\alpha_0^2 \int_0^\infty A(0)\omega_2(t)\,\mathrm{d}t. \end{aligned} \tag{17}$$

Clearly, η_1 is proportional to the dephasing rate Γ', and η_2 depends on $A(0)$. From Eq. (13), we find

$$\bar{\omega}_1(p) = \bar{\omega}_2(p)\frac{1}{p+2\Gamma}. \tag{18}$$

So, we have

$$\omega_1(t) = \mathrm{e}^{-2\Gamma t}\int_0^t \omega_2(\tau)\mathrm{e}^{2\Gamma\tau}\,\mathrm{d}\tau. \tag{19}$$

Then, we obtain

$$\frac{\eta_1}{\eta_2} = \frac{1}{A(0)}\frac{\Gamma'}{\Gamma}. \tag{20}$$

Equation (20) is a comparative relation between the two parts of the EET defined in Eq. (17). From Eq. (20), we can preliminarily learn the key factors that affect the EET in systems similar to our model. For example, if we suppose $\Gamma'/\Gamma =$ constant, increasing the initial coherence of the donor aggregate will make η_1/η_2 decrese and thus enhance the EET, provided that η_1 does not change. Especially for artificial photosynthetic units, this equation is the basic equation for the realization of an effective design.

3. Numerical results

In the above section, from Eq. (13), we find that the EET is in fact affected by the structural parameters of the two aggregates, the initial coherence of the donor aggregate, and the dephasing and dissipation processes of the system. In the following, based on Eq. (10), we will show some numerical results that reveal in detail the functionality of each factor.

For the excitation transfer processes in LHCs, there are many potential pathways for the absorption of photons and for the subsequent transfer of excitations to the RC. One path originates with the excitation of the bottom ring of LHII by 800-nm photons, and the excitations are transferred to the RC[33] through B800 (LHII, the bottom ring) → B850 (LHII, the top ring) → B850 (LHII) → LHI → RC. It was revealed[33] that the excitation transfer from B800 → B850 proceeds within approximately 700 fs and is associated with a transition rate greater than $\kappa = 1.429 \times 10^{12}$ Hz. For B850 → LHI, the time constant is 3–5 ps, which corresponds to $\kappa = 2.0 \times 10^{11}$–$3.33 \times 10^{11}$ Hz. The excitation transfer of LHI → RC in the final step requires approximately 35 ps, which corresponds to $\kappa \approx 2.86 \times 10^{10}$ Hz.

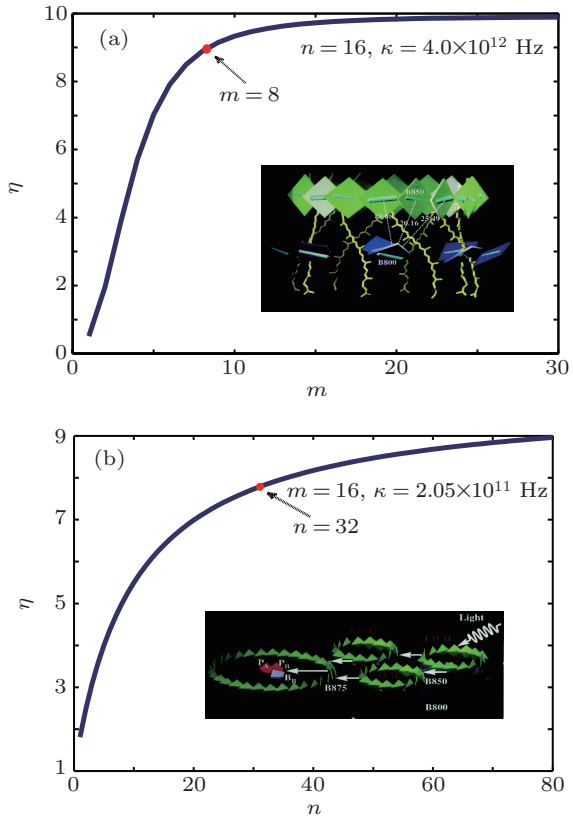


Fig. 2. (color online) (a) The η as a function of the number of donor pigments m for 16 acceptor pigments; (b) η as a function of the number of acceptor pigments n for 16 donor pigments. The two insets are extracted from Ref. [33], $A(0) = 10$.

To reveal how the numbers m and n affect the EET in the above sequential steps, we show numerical results in Fig. 2. We choose $\kappa = 4.0 \times 10^{12}$ Hz in Fig. 2(a), with which we imitate the process from B800 → B850. In Fig. 2(b), we set $\kappa = 2.05 \times 10^{11}$ Hz to imitate the process of B850 → LHI. The other associated parameters are chosen to be $\varepsilon_1 - \varepsilon_2 = 4.89 \times 10^{12}$ Hz, $\alpha_0 = 2.413 \times 10^{11}$ Hz, $\beta = 3.549 \times 10^{13}$, and

$\Gamma = 1.0 \times 10^9$ Hz; all of the parameters are chosen to imitate the real LHCs[10–15] and will be used in all of the following figures. In Fig. 2(a), we set $n = 16$, and plot η as a function of the number of the donor pigments m. We find that η increases with increasing m and approaches a saturation, and the saturation phenomenon begins at near $m = 8$. The results show that our model can be used to imitate the actual excitation transfer process from B800 $\rightarrow$ B850 in LHII (shown in the inset[33]), the numbers of pigments in the actual double-layered rings of LHII, $m = 8$ (or 9) in the bottom ring and $n = 16$ (or 18) in the top ring, are the most economical parameters for LHII to obtain a high EET. Are these numbers only a coincidence or are they the result of natural selection? In Fig. 2(b), η is plotted as a function of the number of acceptor pigments n with $m = 16$, we also find similar results that approximatively imitate the excitation transfer for the B850 $\rightarrow$ LHI step (shown in the inset[33]). Certainly, these results are not accurate; more strict proofs are needed to obtain more complete conclusions. However, we at least provide an evidence for the conclusion that the numbers of pigments (proteins) of actual LHC molecules are the optimum parameters for obtaining a high EET.

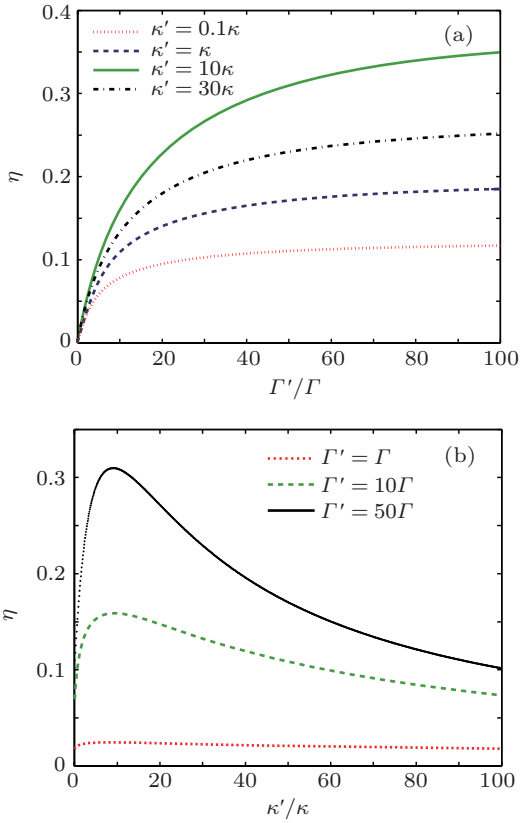


Fig. 3. (color online) (a) The η as a function of ratio Γ'/Γ; (b) η as a function of ratio κ'/κ, where $A(0) = 0$.

In the following, we will show how the dephasing and dissipation rates affect the EET. Without loss of generality, we set $m = 32$ and $n = 1$ in all the following numerical calculations, which simply model the energy transfer from LH1 to RC. First, we suppose $A(0) = 0$. In Fig. 3(a), η is plotted as a function of ratio Γ'/Γ for different κ'/κ. Two features can be observed in the figure. One feature is that η increases and approaches a saturation as Γ'/Γ increases, i.e., as the dephasing rate of the donor increases. Another feature is the existence of an optimum κ'/κ, and we show this more clearly in Fig. 3(b). For the given Γ'/Γ, we find that the optimum value is approximately $\kappa'/\kappa = 10$, which provides an optimum ratio between the dephasing rate of the acceptor and the dissipation rate under nearly realistic conditions.

Secondly, we consider the cases with nonzero $A(0)$. We set $A(0) = 10$ and find from Fig. 4(a) that all the values of η for different Γ'/Γ have almost the same optimum area at approximately $\kappa'/\kappa \approx 10$; this feature is similar to that for $A(0) = 0$. There are three different areas, A, B, and C, in the figure; η increases as Γ'/Γ increases in area B, and the opposite results are found in areas A and C. The results are more clearly revealed in Fig. 4(b). It is obvious that area B is more interesting than the other two areas because in area B, η increases as the dephasing rate of the donor aggregate increases. Can we amplify this area? The anticipated results are shown in Figs. 5(a) and 5(b) for $A(0) = 5$. More numerical calculations (not shown here) show that area B is larger for smaller values of $A(0)$ and area B tends toward infinity if $A(0) \rightarrow 0$ (see Fig. 3(a)). From Figs. 4 and 5, we conclude that an increase in Γ'/Γ will surely enhance η if κ'/κ is within area B, and the most rapid increase is in the region near $\kappa'/\kappa \approx 10$.

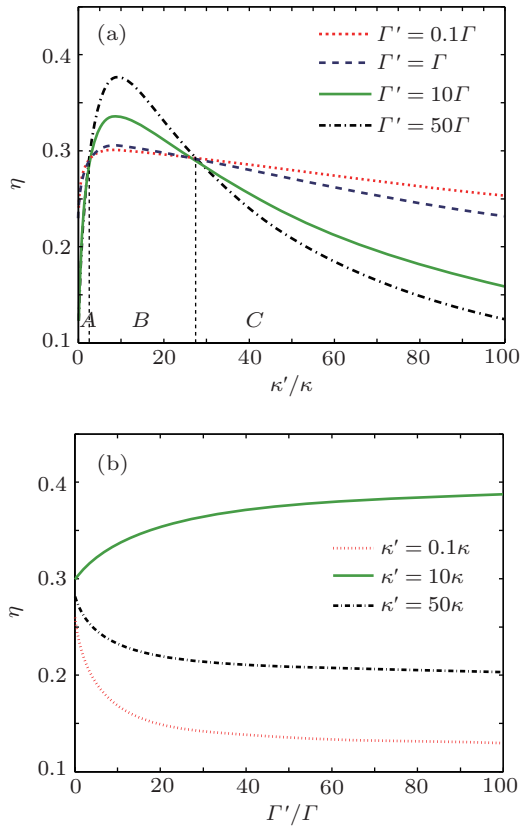


Fig. 4. (color online) (a) The η as a function of ratio κ'/κ; b) η as a function of ratio Γ'/Γ, where $A(0) = 10$.

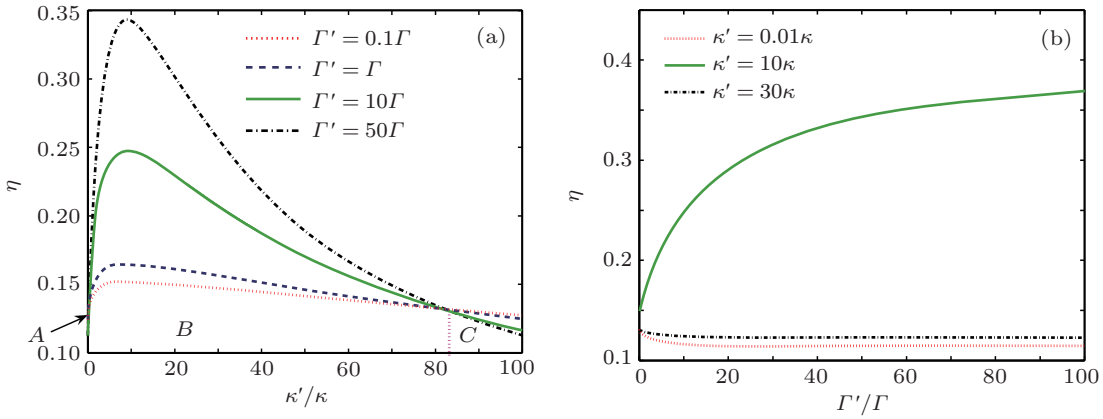


Fig. 5. (color online) (a) The η as a function of ratio κ'/κ; (b) η as a function of ratio Γ'/Γ, where $A(0) = 5$.

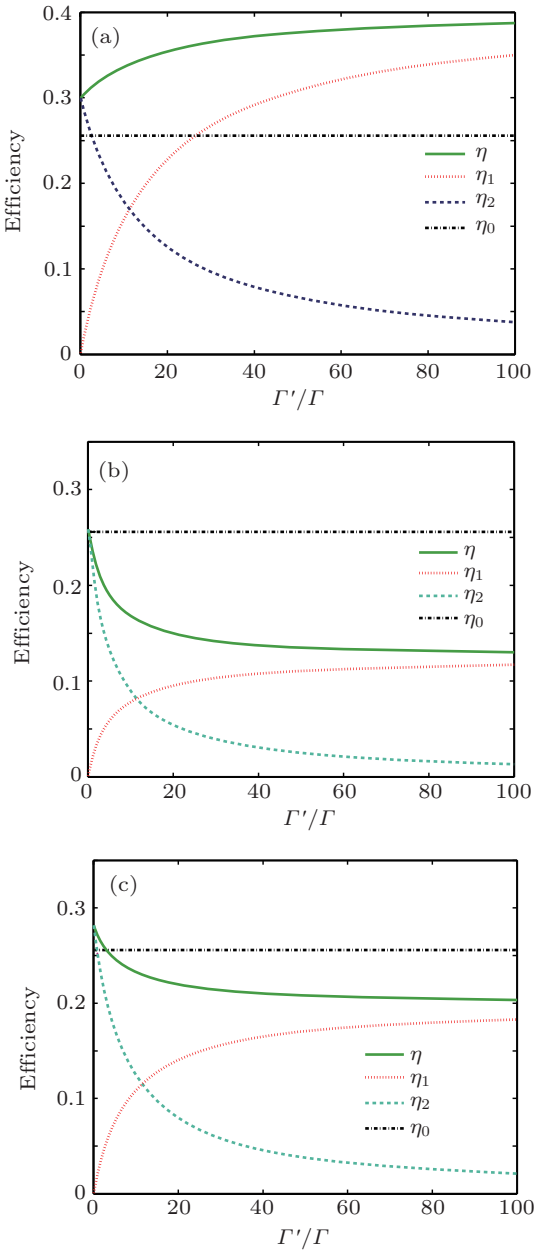


Fig. 6. (color online) The η, η_1, and η_2 each as a function of Γ'/Γ for (a) $\kappa'/\kappa = 10$, (b) $\kappa'/\kappa = 0.1$, and (c) $\kappa'/\kappa = 50$, where $A(0) = 5$.

The above discussion again indicates that the dephasing rate and the initial coherence of the donor aggregate are the key factors for the energy transfer in LHCs.

How do these two factors compete in η? In the previous section, we divided η into two parts, as given in Eqs. (16) and (17), η_1 is proportional to Γ', and η_2 depends on $A(0)$; the comparative relation between the two parts is given by Eq. (20). In Fig. 6, we compare η_1 and η_2 with the total efficiency η to show which part is the main contributor. For reference, we let η_0 correspond to the efficiency without the contribution from the dephasing process, i.e., $\Gamma' = \kappa' = 0$. Figures 6(a)–6(c) show results for $\kappa'/\kappa = 10$ (in area B), $\kappa'/\kappa = 0.1$ (in area A), and $\kappa'/\kappa = 50$ (in area C), respectively. In all three figures, we find that η_1 always increases as Γ'/Γ increases, but η_2 is contrary to Γ'/Γ. The η increases or decreases with Γ'/Γ; before $\Gamma'/\Gamma < A(0) = 10$, $\eta_2 > \eta_1$ is satisfied, however η_1 primarily contributes to the total efficiency when $\Gamma'/\Gamma > A(0) = 10$.

4. Conclusion

In summary, we have theoretically studied the excitation energy transfer between two coupled aggregates in a chromophoric-pigment (protein) system. We employed the theoretical approach of an open quantum system to show that the initial coherence of the donor aggregate, the numbers of pigments in the two aggregates, and the decoherent environmental factors affect the efficiency of energy transfer. We found that there are economical numbers of pigments in the donor and the acceptor aggregates for a high EET, and this finding provides evidence for that the actual numbers of pigments in the chromophoric rings of LHCs under normal environmental conditions should be the natural choices for a high EET. Moreover, we explicitly showed how the environmental factors affect the energy transfer, and found that under certain circumstances, the dephasing processes play a crucial role in the excitation energy transfer. Our results were obtained under a general theoretical model and thus provide a new approach to solve many similar systems.

References

[1] Blankenship R E 2002 *Molecular Mechanisms of Photosynthesis* (Oxford: Blackwell Science)
[2] Fleming G R and Grondelle R V 1994 *Phys. Today* **47** 48
[3] Engel G S, Calhoun T R, Read E L, Ahn T K, Mancal T, Cheng Y C, Blankenship R E and Fleming G R 2007 *Nature* **446** 782
[4] Lee H, Cheng Y C and Fleming G R 2007 *Science* **316** 1462
[5] Collini E and Scholes D 2009 *Science* **323** 369
[6] Collini E, Wong C Y, Wilk K E, Curmi P M G, Brumer P and Scholes G D 2010 *Nature* **463** 644
[7] Mercer I P, El-Taha Y C, Kajumba N, Marangos J P, Tisch J W G, Gabrielsen M, Cogdell R J, Springate E and Turcu E 2009 *Phys. Rev. Lett.* **102** 057402
[8] Panitchayangkoon G, Hayes D, Fransted K A, Caram J R, Harel E, Wen J, Blankenship R E and Engel G S 2010 *Proc. Natl. Acad. Sci. USA* **107** 12766
[9] Cheng Y C and Silbey R J 2006 *Phys. Rev. Lett.* **96** 028103
[10] Castro A O, Lee C F, Olsen F F and Johnson N F 2008 *Phys. Rev. B* **78** 085115
[11] Plenio M B and Huelga S F 2008 *New J. Phys.* **10** 113019
[12] Mohseni M, Rebentrost P, Lloyd S and Aspuru-Guzik A 2008 *J. Chem. Phys.* **129** 174106
[13] Rebentrost P, Mohseni M, Kassal I, Lloyd S and Aspuru-Guzik A 2009 *New J. Phys.* **11** 033003
[14] Caruso F, Chin A W, Datta A, Huelga S F and Plenio M B 2009 *J. Chem. Phys.* **131** 105106
[15] Yang S, Xu D Z, Song Z and Sun C P 2010 *J. Chem. Phys.* **132** 234501
[16] Nazir A 2009 *Phys. Rev. Lett.* **103** 146404
[17] McCutcheon D P S and Nazir A 2011 *Phys. Rev. B* **83** 165101
[18] Sarovar M, Ishizaki A, Fleming G R and Whaley K B 2010 *Nature Phys.* **6** 462
[19] Caruso F, Chin A W, Datta A, Huelga S F and Plenio M B 2010 *Phys. Rev. A* **81** 062346
[20] Fassioli F and Castro A O 2010 *New J. Phys.* **12** 085006
[21] Liao J Q, Huang J F, Kuang L M and Sun C P 2010 *Phys. Rev. A.* **82** 052109
[22] Ghosh P K, Smirnov A Y and Nori F 2011 *Phys. Rev. E* **84** 061138
[23] Briggs J S and Eisfeld A 2011 *Phys. Rev. E* **83** 051911
[24] Eisfeld A and Briggs J S 2012 *Phys. Rev. E* **85** 046118
[25] Caruso F, Saikin S K, Solano E, Huelga S F, Aspuru-Guzik A and Plenio M B 2012 *Phys. Rev. B* **85** 125424
[26] Caruso F, Montangero S, Calarco T, Huelga S F and Plenio M B 2012 *Phys. Rev. A* **85** 042331
[27] Allegra M and Giorda P 2012 *Phys. Rev. E* **85** 051917
[28] Shabani A, Mohseni M, Rabitz H and Lloyd S 2012 *Phys. Rev. E* **86** 011915
[29] Pachn L A and Brumer P 2013 *Phys. Rev. A* **87** 022106
[30] Lloyd S, Mohseni M, Shabani A and Rabitz H 2011 arXiv: 1111.4982v1
[31] Scully M O and Zubairy M S 1997 *Quantum Optics* (Cambridge: Cambridge University Press)
[32] Tan Q S and Kuang L M 2012 *Sci China-Phys. Mech. Astron.* **55** 1541
[33] Hu X and Schulten K 1997 *Phys. Today* **50** 28