Realistic Quantum Control of Energy Transfer in Photosynthetic Processes

Reda M. El-Shishtawy [1], Robert Haddon [2,3], Saleh Al-Heniti [4], Bahaaudin Raffah [4], Sayed Abdel-Khalek [5,*], Kamal Berrada [6] and Yas Al-Hadeethi [4,7]

[1] Department of Chemistry, Faculty of Sciences, King Abdulaziz University, Jeddah 21589, Saudi Arabia; relshishtawy@kau.edu.sa

[2] Center for Nanoscale Science and Engineering, Departments of Chemistry and Chemical and Environmental Engineering, University of California, Riverside, CA 92521-0403, USA; haddon@ucr.edu

[3] Physics Department, King Abdulaziz University, Jeddah 21589, Saudi Arabia

[4] Physics Department, Faculty of Science, King Abdulaziz University, Jeddah 21589, Saudi Arabia; salheniti@kau.edu.sa (S.A.-H.); braffah@kau.edu.sa (B.R.); yalhadeethi@kau.edu.sa (Y.A.-H.)

[5] Mathematics Department, Faculty of Science, Taif University, Taif 009662, Saudi Arabia

[6] Department of Physics, College of Science, Al Imam Mohammad Ibn Saud Islamic University (IMSIU), Riyadh 009661, Saudi Arabia; berradakamal2006@gmail.com

[7] Lithography in Devices Fabrication and Development Research Group, DSR, King Abdulaziz University, Jeddah 21589, Saudi Arabia

*   Correspondence: sayedquantum@yahoo.co.uk

Abstract: The occurrence of coherence phenomenon as a result of the interference of the probability amplitude terms is among the principle features of quantum mechanics concepts. Current experiments display the presence of quantum techniques whose coherence is supplied over large interval times. Specifically, photosynthetic mechanisms in light-harvesting complexes furnish oscillatory behaviors owing to quantum coherence. In this manuscript, we study the coherent quantum energy transfer for a single-excitation and nonlocal correlation in a dimer system (donor+acceptor) displayed by two-level systems (TLSs), interacting with a cavity field with a time-dependent coupling effect considering the realistic situation of coupling between each TLS and the cavity field. We analyze and explore the specific conditions which are viable with real experimental realization for the ultimate transfer of quantum energy and nonlocal quantum correlation. We show that the enhancement of the probability for a single-excitation energy transfer greatly benefits from the energy detuning, photon-number transition, classicality of the field, and the time-dependent coupling effect. We also find that the entanglement between the donor and acceptor is very sensitive to the physical parameters and it can be generated during the coherent energy transfer.

1. Introduction

Aphotosynthetic light-harvesting system transforms the energy from the absorbed photons to the reaction center [1–7]. It employs light-harvesting antennae to absorb and transform solar energy to the reaction center with quantum efficiency. The transfer of the energy of light from an atom to a nearby atom can be performed by electronic energy transfer (see Figure 1), referred to as the resonance energy transfer. Several recent important applications and illustrations of energy transfer are defined, such as the increase of the spectral and spatial cross-section in photosynthetic proteins using resonance energy transfer (light-harvesting proteins), in order to capture the quantum solar

energy by photonic systems. Usually, the quantum transport of energy is described by an incoherent process where a donor atom captures the excitation energy and then transported to an acceptor atom. Here, the electronic coupling elements are used to describe the transferred energy considering the effect of inter-atomic interaction in the framework of dipole-dipole coupling. More recently, coherent energy transfer is provided to be an interesting period inphotosynthesis, in order to produce electronic excitations from photosynthetic pigments and transport this excited energy to a reaction center [8–14]. A simple mechanism is proposed to study the light-harvesting complex in a dimer system consisting of a donor and an acceptor where each one is described by a two-level quantum system. Recently, a number of both theoretical studies have been developed and experimental tools have been conducted using the phenomenon of electronic coherence in the transport of the quantum energy [15,16]. It is shown that exciton delocalization, together with pure dephasing induced by stochastic fluctuations of the external environment, has been considered to improve the efficiency of quantum transfer in multichromophoric quantum systems [17,18].
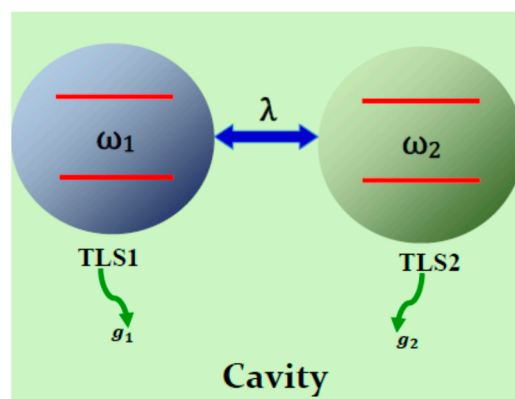

Figure 1. A diagram illustrating our system. TLS1 and TLS2 are both placed inside one cavity. A donor (with resonant frequency ω_1) and an acceptor (with resonant frequency ω_2) are modeled by two-level atoms with strong dipole-dipole interaction. Realistic quantum control almost necessarily implies engagement of continuous variable interaction of TLSs having a finite number of states with a "large" system with a continuous quantum state. The coupling strength that provides the physical mechanism for coherent energy transfer and entanglement generation is considered in the context of the time-dependent coupling effect $G(t)$.

The observation of long-lived coherence in those quantum systems has been a topic of great contemplation. It is shown that the quantum coherence leads to a decrease in the quantum efficiency of the energy transfer and includes dephasing, which combines the tunneling and noise effects leading to a highly efficient quantum energy transfer [19–21]. Recent investigations on photosynthetic complexes show that the coherence resists thepopulation transfer long enough to impact quantum transport dynamics in the time scale [22]. It has been shown that in the photosynthetic antenna quantum systems, the quantum coherences could be removed during the dynamics. However, the long lifeof the quantum coherence, alone, will not be sufficient to result in ahigh quantum efficiency of the transferred energy. The quantum coherences play a role incontrolling the system dynamics and the excitation populations in a given quantum state (quantum transport).

Quantum entanglement is one of the most promising phenomenon in different branches of science, which exhibits the quantum correlations between different physical systems and was introduced by Schrodinger [23,24], as the characteristic trait of quantum mechanics, and the one that enforces its entire departure from classical lines of thought [25]. These quantum correlations quantify from the kind of the quantum states of the composite systems. Nonlocal correlations in the combined system are independent of the spatial separation of the constituents, in order to appear as a lone system. As Schrodinger mentioned, the best possible knowledge of a whole does not necessarily include the

best possible knowledge of its parts [25]. Investigation of this kind of quantum correlation and its consequences for quantum measurements led to solving and understanding many physical problems, including the Bell inequalities [26,27], and verified the experimental part of the spooky action at a distance, as mentioned by Einstein. More recently, the development of quantum information theory has provided a rise in knowledge and also augmented the literature of the entanglement phenomenon, which has played a crucial role in different tasks of quantum information processing and transmission and, more recently, in quantum metrology [28–34]. The significance of this kind of correlation (entanglement) in different applications has led to analysis and investigation of high-dimensional quantum systems and brings a new role and application of these kinds of correlations in many particle quantum systems [35–47]. A measure of entanglement must be invariant under local operations and classical communication. The understanding of derivable measures of quantum entanglement are borne in mind when searching to detect entanglement in large dimensional complex systems, such as those considered in biological systems.

Motivated by the above considerations, this study aims to investigate the influence of the atomic motion effect in the interactions between the pigments on energy transfer considering a model of atom-atom interaction that closely describes a realistic experimental scenario. We will present, in detail, how the time-dependent coupling effect and energy frequencies can influence the efficiency of the coherent energy transfer for a single excitation from a donor to an acceptor where each one is modeled by a two-level atom system. We can obtain physical answers to this very complex problem of quantum energy transfer considering the atomic motion effect. On the other hand, we will study the dynamics of the nonlocal correlation between the pigments during the process of the quantum energy transfer. Furthermore, we will study the dynamic behavior of the quantum variance when performing a measurement on an observable for the density matrix in pigments, which is rather significant in different tasks of quantum information and computational technologies.

This article is organized as follows: in Section 2, we present the model for our system and the details of the formalism that describes the dependence of the physical parameters on the population dynamics of the coherent quantum energy transfer for a single-excitation and nonlocal correlation between the pigments; in Section 3, we discuss the main obtained results. Our aim is to highlight the dependence of the population, entanglement, and classicality of the cavity field on the energy frequencies and the time-dependent coupling effect during the time evolution. Finally, conclusions are drawn in Section 4.

2. Model of the Physical System

The physical system under our investigation is a dimer system considering dipole-dipole interaction with the time-dependent coupling effect. Here, the dimer is defined by a pair of two-level systems (TLSs), which compose TLS1 as the donor system and TLS2 as the acceptor system, with energy separations ω_1 and ω_2, respectively, as depicted in Figure 1. The Hamiltonian for the donor and acceptor systems can be written as:

$$
\begin{aligned}
H_{\text{TLSs}} = &\frac{\omega_1}{2}\sigma_1^z + \frac{\omega_2}{2}\sigma_2^z + \omega a^\dagger a + \lambda\left(\sigma_1^+\sigma_2^- + \sigma_1^-\sigma_2^+\right) \\
&+ G(t)\sum_{j=1}^{2}\left\{\left(a^{\dagger 2}\right)^l\sigma_j^- + \sigma_j^+\left(a^2\right)^l\right\}
\end{aligned}
\tag{1}
$$

where λ is the dipole-dipole interaction strength between TLS1 and TLS2 and the usual Pauli operators $\sigma_j^+ = |e_j\rangle\langle g_j|$ (raising operator for the *j*th two-level atoms), $\sigma_j^- = |g_j\rangle\langle e_j|$ (lowering operator for the *j*th two-level atoms), and $\sigma_j^z = |e_j\rangle\langle e_j| - |g_j\rangle\langle g_j|$, where $|g_j\rangle$ $(|e_j\rangle)$ is the ground (excited) states of the *j*th ($j = 1, 2$) TLS. The coupling provided by the dipole-dipole interaction term exhibits the physical process for excitation energy transfer and the nonlocal correlation between TLS1 and TLS2. $G(t)$ denotes the time-dependent coupling between TLSs and the cavity field and *l* is the photon number transition. The generalization from the constant coupling *g* to a coupling that evolves with

time $G(t)$, will provide new physical phenomena that have not been discussed before. A realization of particular interest, with respect to $G(t)$, may be the time-dependent alignment or orientation of the atomic/molecular dipole moment using a laser pulse [48–53] and the motion of the atom through the cavity. For an atom oscillating back and forth across a narrow cavity within a square trap, the coupling is modelled approximately to be sinusoidal $G(t) = g\sin^2(t)$ [48].

In order to investigate the populations of the transfer of energy in a single excitation from the donor to the acceptor, we consider that the donor is initially defined in an excited state $|e_1\rangle$ and the acceptor is in a ground state $|g_2\rangle$:

$$|\psi_{12}\rangle = |e_1 g_2\rangle \tag{2}$$

and the probability to get the acceptor in an excited state (single excitation energy transfer) at time t will be:

$$P(t) = \mathbf{Tr}\left(\rho_2 \sigma_2^+ \sigma_2^-\right) \tag{3}$$

The entanglement between the donor and acceptor can be quantified via the concurrence defined by [54]:

$$\mathbf{C} = \max\{0, \lambda_1 - \lambda_2 - \lambda_3 - \lambda_4\} \tag{4}$$

where λ_i define the eigenvalues giving in the decreasing order of $\rho_{12}\widetilde{\rho}_{12}$ and $\widetilde{\rho}_{12}$ is defined by:

$$\widetilde{\rho}_{12} = \left(\sigma_y \otimes \sigma_y\right) \rho_{12}^* \left(\sigma_y \otimes \sigma_y\right) \tag{5}$$

where ρ_{12}^* denotes the conjugate of ρ_{12} in the standard basis of two bipartite system and σ_y is the Pauli Y operator. In order to study the nonclassical properties of the cavity field during the energy transfer process, we use Mandel's parameter as a quantifier of the statistical properties and verify the presence of quantumness in the cavity field. This parameter is defined as [55]:

$$M_p = \frac{\langle(\Delta\hat{n})^2\rangle - \langle\hat{n}\rangle}{\langle\hat{n}\rangle}, \tag{6}$$

where $\langle\hat{n}\rangle$ is the average photon number of the cavity field and $\langle(\Delta\hat{n})^2\rangle$ corresponds to the mean-square variance. Mandel's parameter is used to precisely determine whether the photon distribution of the cavity field is sub-Poissonian ($-1 \leq M_p \leq 0$), evidently being the nonclassical state, the Poissonian ($M_p = 0$) corresponds to the case of the classical coherent state, and super-Poissonian ($M_p > 0$). Mandel's parameter can be considered as a tool to investigate the effect of the physical parameters, which contain all information on the statistical properties of the cavity field during the energy transfer process.

3. Results and Discussion

The coherence introduced by the influence of the different terms of the probability amplitude is one of discriminatory characters of quantum mechanics. The coherent phenomena are shown to be responsible for the oscillatory-appearingbehaviors in quantum systems. It is shown experimentally that the light-harvesting provides oscillatory electronic dynamics and explains the nature of such an oscillatory comportment [4,6], showing the importance of quantum mechanics in biological functional systems [4]. Thus, it is important to treat these oscillations realistically in order to disclose and understand the mechanisms adopted by nature, which will lead, in the future, to inspiring new quantum technologies.

Let us investigate the influence of the physical parameters on the dynamic behavior of the probability P for a single-excitation energy transfer and entanglement in the absence and presence of the time-dependent coupling effect when TLSs are interacting with a continuous quantum field following the considered models, as shown in Figure 1. We plot, in Figures 2 and 3, the evolution of the probability and entanglement versus time gt for various values of the energy detuning ω_d without

and with time-dependent effect, respectively. It can be seen that the probability and entanglement experience damped oscillations during the time-evolution and they tend to stabilize at steady behavior in the asymptotic $gt \to \infty$ limit, showing the donor and acceptor are trapped by the cavity field at this limit. We find that the value of the probability and degree of the entanglement during the process of the coherent energy transfer are very sensitive to the energy detuning of the TLSs. Interestingly, the amplitude of the probability (oscillations) decreases (enhances) with the increase of the detuning parameter with and without the time-dependent coupling effect. This means that when the strength of the energy detuning is large, the energy emitted by the donor may be excited more than one acceptor and leads to enhance the probability oscillations. In the presence of the TLSs motion ($G(t) = g\sin^2(t)$), the dynamic behavior of the probability and entanglement are deeply influenced by the coupling between each TLS and the cavity field. In general, we find that the motion effect leads to enhancing the probability of energy transfer and entanglement between the donor and acceptor during the time-evolution. On the other hand, we find that there exists a critical value of the energy detuning for which the amount of correlation is maximal during the process of coherent energy transfer. These results indicate that the improvement of the efficiency of the coherent energy transfer for a single excitation between TLSs greatly benefits from the combination of the energy detuning and time-dependent coupling effect in the TLS-field interaction.
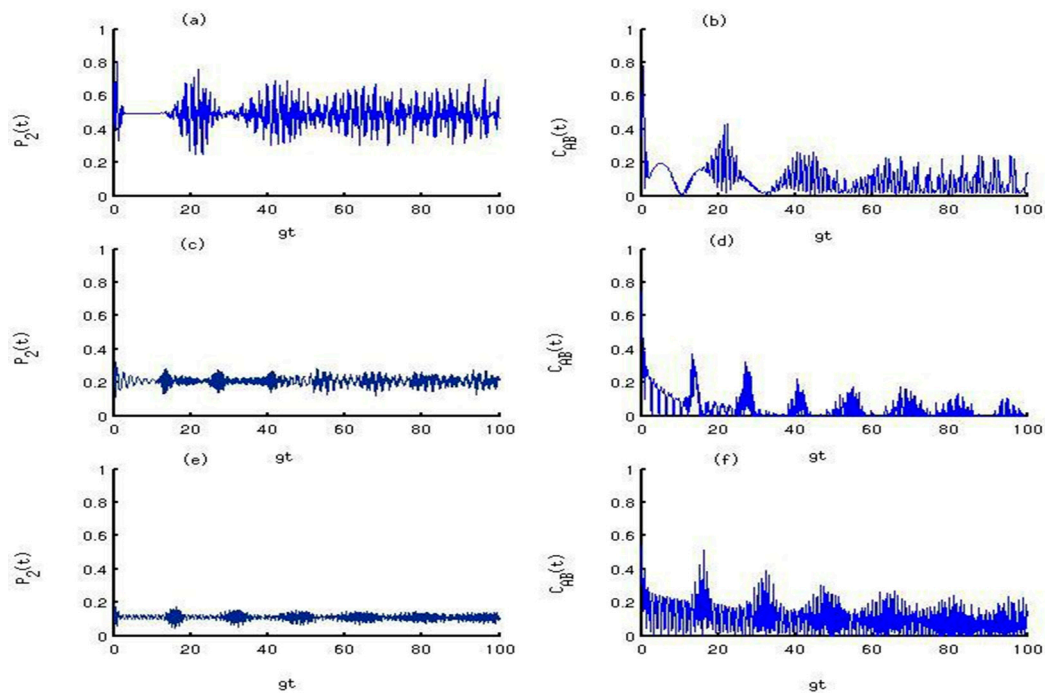

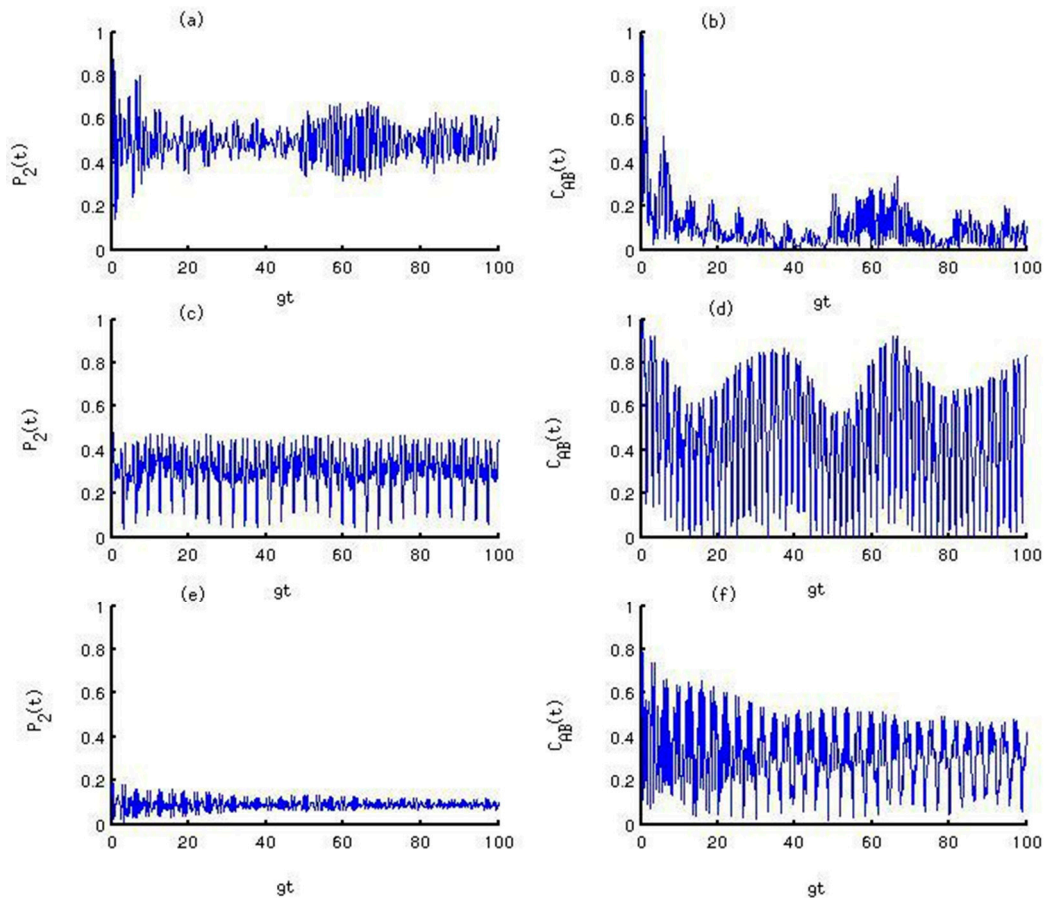**Figure 2.** (Color Online) The probability and entanglement are plotted versus the dimensionless time gt for various values of the energy detuning for one-photon transition ($l = 1$) in the absence of the atomic motion effect $G(t) = g$ with $|\alpha|_2 = 10$. (**a**,**c**,**e**) display the variation of the population for a single-excitation with the time; (**b**,**d**,**f**) present the evolution of the concurrence of the donor-acceptor state during the time evolution. (**a**,**b**) is for $\omega_d = 0$; (**c**,**d**) is for $\omega_d = 2$; and (**e**,**f**) is for $\omega_d = 4$.

Figure 3. The probability and entanglement are plotted versus the dimensionless time *gt* for various values of the energy detuning for one-photon transition ($l = 1$) in presence of atomic motion effect $G(t) = g\sin^2(t)$ with $|\alpha|_2 = 10$. (**a**,**c**,**e**) display the variation of the population for a single-excitation with the time; (**b**,**d**,**f**) present the evolution of the concurrence of the donor-acceptor state during the time evolution. (**a**,**b**) is for $\omega_d = 0$; (**c**,**d**) is for $\omega_d = 2$; and (**e**,**f**) is for $\omega_d = 4$.

In Figures 4 and 5 we present a comparison between the dynamic behavior of the probability and entanglement in the multi-photon process with and without the time-dependent coupling effect, respectively. From the figures, we observe that the number of the photon transition in the interaction between TLSs and cavity field significantly affects the variation of the probability and entanglement between the donor and acceptor system during the energy transfer operation. Interestingly, we find that the photon-transition number leads to a reduction in the oscillations, exhibiting a quasi-periodic behavior of the probability and entanglement when the detuning energy gets close tothe resonance case in the absence of the time-dependent coupling effect.
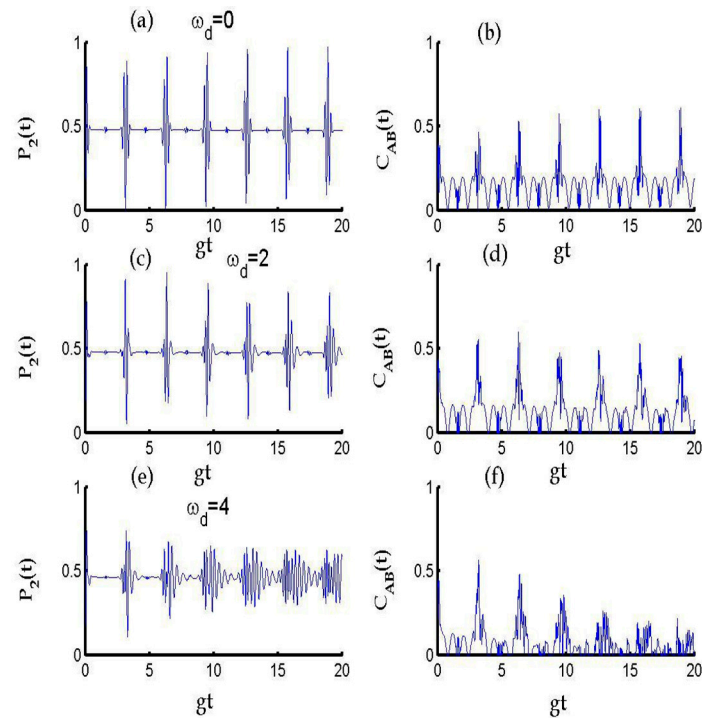
Figure 4. (Color Online) The probability and entanglement are plotted versus the dimensionless time *gt* for various values of the energy detuning for two-photon transition ($l = 2$) in the absence of atomic motion effect $G(t) = g$ with $|\alpha|_2 = 10$. (**a,c,e**) display the variation of the population for a single-excitation with the time; (**b,d,f**)present the evolution of the concurrence of the donor-acceptor state during the time evolution. (**a,b**) is for $\omega_d = 0$; (**c,d**) is for $\omega_d = 2$; and (**e,f**) is for $\omega_d = 4$.
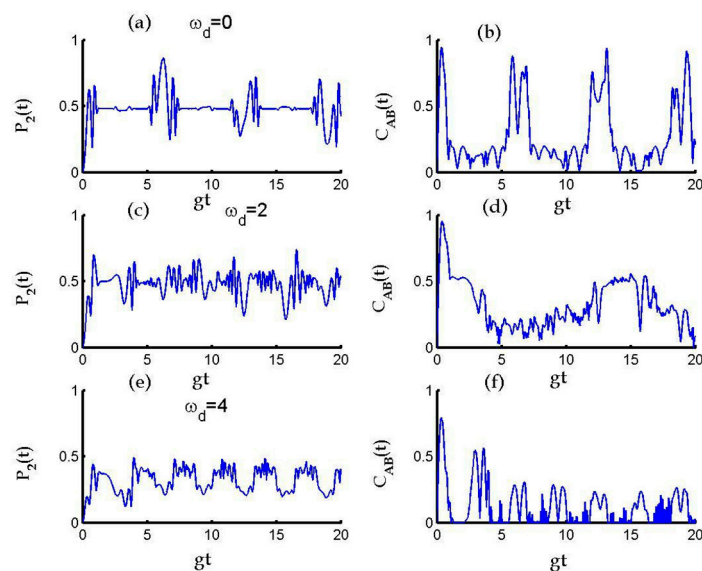

Figure 5. (Color Online) The probability and entanglement are plotted versus the dimensionless time *gt* for various values of the energy detuning for two-photon transition ($l = 2$) in the presence of atomic motion effect $G(t) = g \sin^2(t)$ with $|\alpha|_2 = 10$. (**a,c,e**) display the variation of the population for a single-excitation with the time; (**b,d,f**) present the evolution of the concurrence of the donor-acceptor state during the time evolution. (**a,b**) is for $\omega_d = 0$; (**c,d**) is for $\omega_d = 2$; and (**e,f**) is for $\omega_d = 4$.

Now let us investigate the dynamic behavior of the statistical properties of the cavity field using Mandel's parameter during the quantum energy transfer. In order to explore the influence

of the physical parameters on Mandel's parameter clearly, in Figure 6, we show the variation of Mandel's parameter with respect to different values of the energy detuning ω_d. In order to make results comparable, we have considered both cases without (Figure 6a,c,e) and with (Figure 6b,d,f) time-dependent coupling effects. From the figure, we find that that the parameters ω_d significantly affect the photon distribution of the cavity field, where the increase (decrease) in the parameter ω_d leads to enhancing the non-classicality of the field in the absence (presence) of the time-dependent coupling effect. Interestingly, in the resonance case $\omega_d = 0$ with $G(t) = g$, Mandel's parameter verifyies the inequality $-1 \leq M_p \leq 0$ for small values of time showing the sub-Poissonian distribution of the photons and $M_p > 0$ as the time becomes significantly large, exhibiting super-Poissonian distribution of the photons. Whereas for the case of $G(t) = g \sin^2(t)$, the parameter M_p is always negative during the time evolution. For the off-resonance case ($\omega_d \neq 0$), Mandel's parameter tends to be near to the trivial case $M_p = 0$ as the time becomes large, reflecting that the photon number distribution is Poissonian.
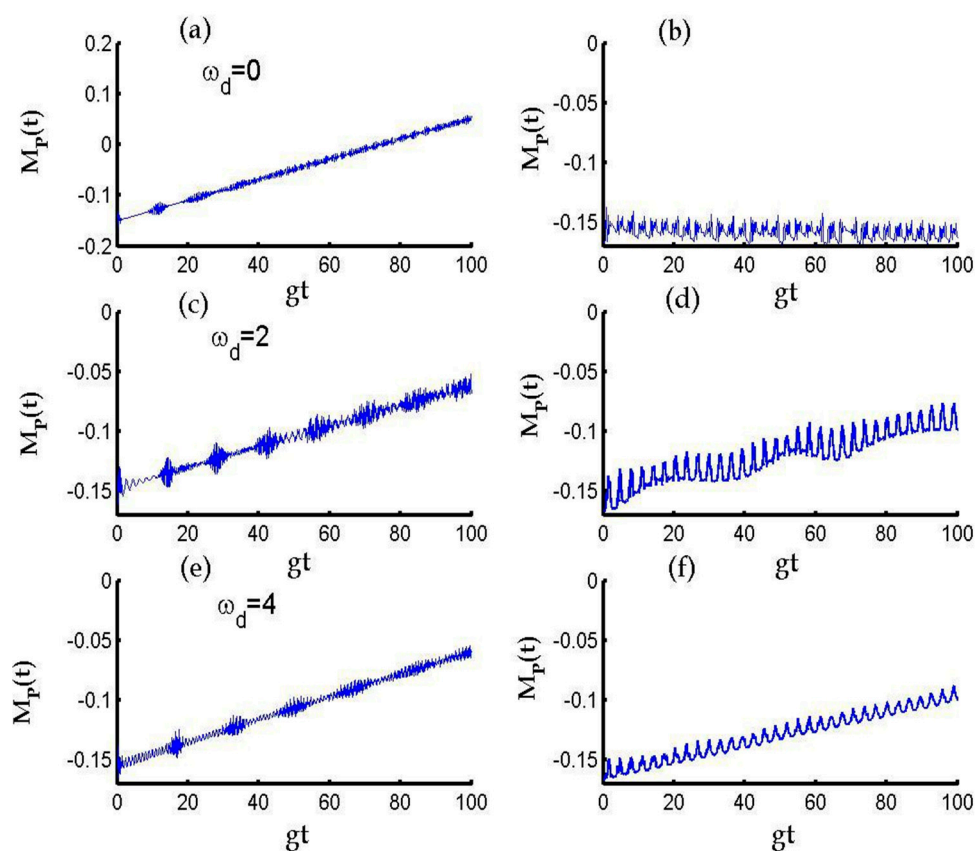

Figure 6. (Color Online) Mandel's parameter given in Equation (6) is plotted vs. the dimensionless time gt for various values of the energy detuning for one-photon transition ($l = 1$) with $|\alpha|_2 = 10$. (**a,c,e**) present the variation of the quantumness in the field in the absence of the time-dependent effect $G(t) = g$; (**b,d,f**) exhibit the variation of the quantumness in the field in the presence of the time-dependent effect $G(t) = g \sin^2(t)$. (**a,b**) is for $\omega_d = 0$; (**c,d**) is for $\omega_d = 2$; and (**e,f**) is for $\omega_d = 4$.

These results indicate that the quantumness in the field can be used as an indicator to enhance and control the quantum energy transfer and entanglement between the donor and acceptor in the photosynthesis process by a proper choice of the detuning parameter and photon-number transition in the presence of the time-dependent effect during the time-evolution.

4. Conclusions

Exploring new fundamental processes responsible for controlling quantum energy transfer in photosynthesis is the crucial aim for both theoretical and experimental investigations. In summary, we have investigated the coherent quantum transfer energy for a single-excitation and nonlocal correlation in a dimer system consisting of a donor and an acceptor, where each one is described by a two-level atom in the absence and presence of the time-dependent coupling effect. We have analyzed and explored the required conditions that are feasible with real experimental realization for optimal transfer of quantum energy and generation of nonlocal quantum correlation. We have shown that the enhancement of the probability for a single-excitation transfer energy is greatly benefits from the combination of energy detuning and time-dependent coupling effect. On the other hand, we have investigated the generation of a degree of quantum nonlocal correlation in the dimer during the process of the energy transfer in terms of the physical parameters using the linear entropy as a quantifier of entanglement. Furthermore, we have studied the dynamic behavior of the quantum variance when performing a measurement on an observable for the density matrix in the pigments. Finally, an interesting relationship between the transfer probability, entanglement, and quantum variance is explored during the time evolution in terms of the physical parameters. We treat here the generation of the entanglement among the pigments during the process of energy transfer in the presence of the atomic motion effect. A study into the initial prepared entanglement among the pigments on the quantum energy transfer in the presence of the time-dependent coupling effect will make for an interesting investigation. Another interesting contribution is to the study of the population of the energy transfer and entanglement in multi-level systems interacting with external fields under the effects of the atomic motions and field parameters.

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