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Structure-dependent wavelike energy transfer on pigment rings of individual light-harvesting-2 complexes from photosynthetic bacteria

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This paper studies the transfer behavior of electronic excitations on the circular and elliptically deformed B850 rings theoretically. It shows that degree of delocalization of the electronic excitation is dependent on the kinds of measured electronically excited states and the kinds of studied structures. It finds that if initial excitation is a coherent exciton, then the elliptical deformation of the B850 ring will work against energy transfer. It depicts wavelike energy transfer persisting for several hundred femtoseconds on the circular and elliptically deformed B850 rings at cryogenic temperature and finds that under the deformation, inducing localized excitation at the site near the elliptical major axis will be in favor of energy transfer, whereas at the minor axis will work against energy transfer both visibly in wavelike region.

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Photosynthetic light-harvesting (LH) antennae have been shown very efficient in solar energy collection and transfer, which is attributed to the spatial arrangement of pigment molecules within protein frame, especially the unique structure of LH complexes of photosynthetic bacteria, in which bacteriochlorophyll a (*Bchla*) molecules are coupled and arranged on a ring. The ring's structures exist in photosynthesis broadly and they have aroused intense interests both in experimental and theoretical studies. Especially wavelike electronic quantum coherence [1–3] and degree of delocalization of electronically excited states are important for efficiency of energy transfer in photosynthesis. The behavior of wavelike energy transfer (WLET) in LH systems is poorly understood. First direct experimental evidence of it had been obtained within Fenna-Matthews-Olson bacteriochlorophyll complex by G. S. Engel *et al.* [1], and they had reported the detection of "quantum beating" signals, coherent electronic oscillations in both donor and acceptor molecules, generated by light-induced energy excitations. Wavelike electronic quantum coherence and degree of delocalization of electronic states are deeply dependent on the structure of LH systems. The crystal structure of LH2 complex from *Rps. acidophila* strain 100050 shows that the active assembly consists of two concentric cylinders of helical protein subunits which enclose the pigment molecules [4,5]. This structure was improved in 2003 to a resolution of 2.0 Å [6,7]. Our calculation was performed on the basis of this structure, and its atomic coordinates were taken from the Brookhaven protein data bank (identification code: 1nkz). Spectroscopy of individual LH2 complexes revealed a deformation of the circular complex into C_2 symmetry, and M. Matsushita *et al.* had shown that the elliptically deformed B850 ring with smaller interpigment distances, where the curvature of ellipse is small, provided the best agreement with the experimental spectra [8]. In this paper, we will investigate transfer behaviors of different electronically excited states and mainly consider the effects of deformation and static disorder on WLET in B850 rings at cryogenic temperature theoretically. We will show that degree of delocalization of the electronic excitation is dependent on the kinds of measured electronically excited states and the kinds of studied structures. We will also show that the elliptical deformation of B850 ring will reduce degree of delocalization of coherent excitons. We will depict WLET on circular and elliptically deformed B850 rings and find that under the deformation, inducing the electronic excitation localized at sites near the elliptical major axis will be in favor of energy transfer, and initially localized excitation at the minor axis will work against energy transfer both visibly in WLET region.

The B850 ring of circular LH2 complex is composed of 18 tightly coupled *Bchla* molecules which are grouped into nine $\alpha\beta$ *Bchla* pairs, and the nine α and nine β *Bchlas* form approximately the α and β circles with radii r_α and r_β, respectively. We only consider the lowest excited state Q_y of individual *Bchla* molecules. The Q_y transition dipole moments (TDMs) are assumed to be orientated along the axes running through the N_I-N_{III} nitrogen atoms of the *Bchlas* [5] and the dipole strength at each pigment is denoted as one. For the elliptical deformation of α or β circle, each *Bchla* is displaced along the line connecting it and the center of the corresponding circle. The orientations of projections of α and β TDMs onto LH2 membrane plane are adjusted to keep fixed angles with the corresponding local tangents of α and β ellipses, respectively [8].

The electronic structures of LH complexes have been investigated intensively by the exciton model [5,8–11]. Assuming that intermolecular interactions are of dipolar origin, we only consider the nearest-neighboring (N-N) and second N-N dipole-dipole interactions on the B850 ring and the system can be described by the Hamiltonian

$$H = \sum_{n=1}^{18} \varepsilon_n |n\rangle\langle n| + \sum_{n=1}^{18}\sum_{m=1}^{2} V_{n,n+m}[|n\rangle\langle n+m| + |n+m\rangle\langle n|], \tag{1}$$

where ε_n represents the excitation energy of individual *Bchla*

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at site n, $V_{n,n+1}$, and $V_{n,n+2}$ are the N-N and second N-N interactions, respectively. In this dipole-dipole approximation, the Coulomb interaction between *Bchlas* n and m can be written as

$$V_{n,m} = V_0 \mathrm{K}_{n,m}, \tag{2}$$

$$\mathrm{K}_{n,m} = [\hat{\mu}_n \cdot \hat{\mu}_m - 3(\hat{\mu}_n \cdot \hat{r}_{n,m})(\hat{\mu}_m \cdot \hat{r}_{n,m})]/r_{n,m}^3, \tag{3}$$

where $\hat{\mu}_n$ and $\hat{\mu}_m$ are the unit vectors of local TDMs at sites n and m, respectively. $r_{n,m}$ is the distance between *Bchlas* n and m with the corresponding unit vector $\hat{r}_{n,m}$ pointing from the *Bchla* n to m or in the reverse direction. The interaction parameter V_0 is determined by taking average value of interactions within $\alpha\beta$ pairs on B850 ring at its different structures always as $V_i = 370\ \mathrm{cm}^{-1}$ [6], and then other interactions can be obtained from Eqs. (2) and (3). The mean values of excitation energies on α and β circles or ellipses, ε_α and ε_β, respectively, are supposed to be independent of the structural deformations of LH2 complex, and their difference had been estimated to be about the same as the average N-N interaction on B850 ring in circular LH2 complex [8,12,13], i.e., $\varepsilon_\alpha - \varepsilon_\beta = (V_i + V_e)/2 \approx 326\ \mathrm{cm}^{-1}$, where $V_e \approx 282\ \mathrm{cm}^{-1}$ is the average value of N-N interactions between $\alpha\beta$ pairs. The physical properties, except the zero point of energy space, of electronically excited states in one LH complex are independent of the intercomplex heterogeneity, hence we only consider the intracomplex disorder of excitation energies with a Gaussian distribution of a full width at half maximum (FWHM) $\Gamma = 125\ \mathrm{cm}^{-1}$ [14]. Monte Carlo simulations are performed by using 10 000 iterations to calculate the ensemble average results.

The coherent one-exciton eigenfunction of the Hamiltonian (1) can be written as

$$|k\rangle = \sum_{n=1}^{18} c_n^k |n\rangle, \tag{4}$$

with the eigenvalue e_k and these coherent excitons from the top to lowest in energy space are denoted as $|k=1\rangle$ to $|k=18\rangle$, respectively. Where c_n^k is the participation of n-th *Bchla* in the state $|k\rangle$. For the elliptically deformed B850 ring, with consideration of the two prominent absorptions being around 856 and 864 nm [10], we let $\varepsilon_\alpha = 12490\ \mathrm{cm}^{-1}$ and assume that deformation amplitudes of the α and β circles both are at $\delta r_\alpha / r_\alpha = \delta r_\beta / r_\beta = 5.7\%$, where δr_α and δr_β are deviations of the major and minor axes of α and β ellipses from the radii r_α and r_β of α and β circles, respectively. With these assumptions, we have following results: (1) the energy splitting between states $|k=16\rangle$ and $|k=17\rangle$ (absorbing averagely at 856.1 and 864.2 nm, respectively) is $109.2\ \mathrm{cm}^{-1}$, which is compatible with the observed value of $110\ \mathrm{cm}^{-1}$; (2) the angle between transition dipoles of these two states is 90.1°, which is in agreement with the experimental observation; (3) the intensity ratio between these two states is 0.95, which is discrepant with the experimentally observed value of 0.7. But the difference between randomly averaged absorption peaks of states $|k=18\rangle$ (at 866.6 nm) and $|k=17\rangle$ is only 2.4 nm, and then the two states may be spectrally overlapped in practical measurement. If we add the absorption of state $|k=18\rangle$ into state $|k=17\rangle$, the intensity ratio between $|k=16\rangle$ and $|k=17\rangle$ becomes 0.78, which is close to the experimental value. In this experimentally matched model, owing to intracomplex disorder and elliptical deformation, the oscillator strength of the lowest state $|k=18\rangle$ now is $D_{k=18} = 1.656$, and the corresponding dipole strength at one *Bchla* molecule is denoted as one. As we know that the excited singlet state lifetime for isolated monomeric *Bchla* is about 1–2 ns [7] and the fluorescence decay time for individual LH2 complexes is about 1 ns [15,16], our result is consistent with these experimental results.

I. DEGREE OF DELOCALIZATION

Degree of delocalization of electronic excitations in molecular aggregates is always debated hotly [5]. We use

$$\zeta_k \equiv \left\langle 1 \bigg/ \sum_{n=1}^{18} (c_n^k)^4 \right\rangle_r, \tag{5}$$

as a measurement of the delocalization length of k-th coherent exciton, where $\langle \ldots \rangle_r$ represents the random ensemble average. The electronic excitation on B850 ring may be not an exciton eigenstate, but may be an exciton wavepacket that is a superposition of exciton eigenstates, and its shape is time dependent. However, in the steady-state limit [17], we can use the function

$$\zeta(T) \equiv \left\langle \left(\sum_{n=1}^{18} a_n^2(T) \right)^2 \bigg/ \sum_{n=1}^{18} a_n^4(T) \right\rangle_r, \tag{6}$$

to estimate the delocalizaton length of the exciton wavepacket at temperature T, where $a_n(T) \equiv \Sigma_{k=1}^{18} p_k^{1/2}(T) c_n^k$, $p_k(T) \equiv \exp(-e_k/k_B T)/\Sigma_{k=1}^{18} \exp(-e_k/k_B T)$ is the Boltzmann distribution of exciton populations and k_B is the Boltzmann constant.

In Fig. 1, we give the relations of randomly averaged delocalization lengths of the lowest exciton and exciton wavepacket at $T = 25\ °\mathrm{C}$ with the FWHM Γ of Gaussian distribution of intracomplex random excitation energies in circular and elliptically deformed LH2 complexes with deformation amplitude at 5.7%. Owing to the random scattering, delocalization length of the lowest exciton, i.e., the band-edge state, will be shortened in both circular and elliptical structures, but even at $\Gamma = 800\ \mathrm{cm}^{-1}$, the lowest exciton is still delocalized on about 3.2 molecules in both two structures. When $0 \le \Gamma < 400\ \mathrm{cm}^{-1}$, the difference of delocalization lengths of the lowest excitons between the two structures is visible. At $\Gamma = 125\ \mathrm{cm}^{-1}$, delocalization lengths of the lowest excitons are $\zeta_{18} \approx 12.8$ and 7.9 molecules in circular and elliptical structures, respectively, and at $\Gamma = 370\ \mathrm{cm}^{-1}$, they are 6.3 and 5.4 molecules, respectively. Hence delocalization lengths of the lowest excitons on different B850 rings are very dissimilar. In Fig. 1, we also find that except close to $\Gamma = 236\ \mathrm{cm}^{-1}$ in circular structure and $\Gamma = 70\ \mathrm{cm}^{-1}$ in elliptical structure, the differences of delocalization lengths between the lowest excitons and exciton wavepackets are also visible. At weak disorder the lowest excitons are more delocalized than the exciton wavepackets

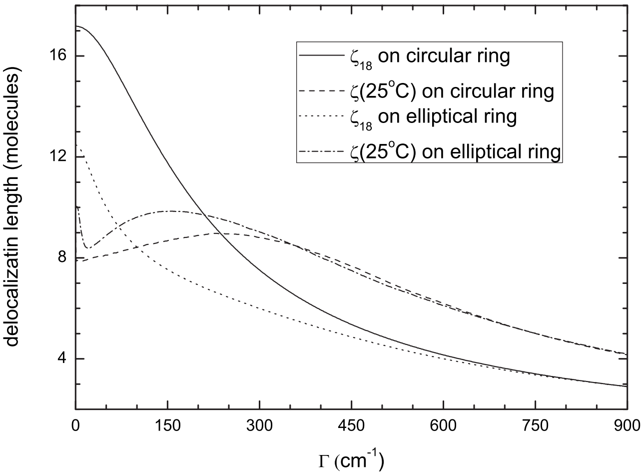

FIG. 1. Randomly averaged delocalization lengths of the lowest exciton and exciton wavepacket at $T=25$ °C, ζ_{18} and $\zeta(25\ °\text{C})$, respectively, as functions of the FWHM Γ, both on circular and elliptically deformed B850 rings with deformation amplitude at 5.7%, and always with $V_i=370\ \text{cm}^{-1}$, $\varepsilon_\alpha=12490\ \text{cm}^{-1}$, and $\varepsilon_\alpha-\varepsilon_\beta=326\ \text{cm}^{-1}$.

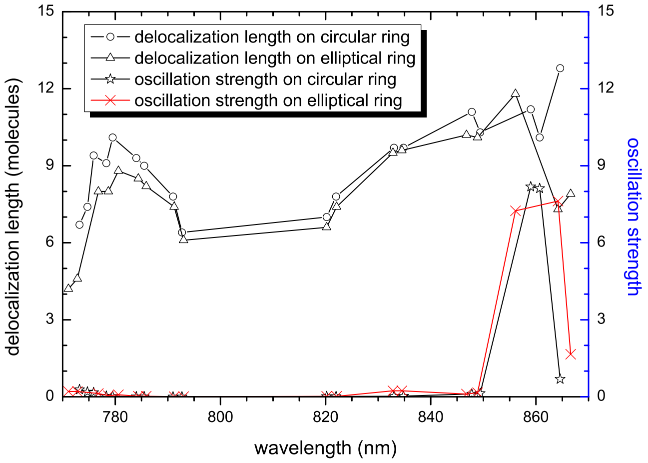

FIG. 2. (Color online) The distributions of randomly averaged delocalization lengths and oscillator strengths of coherent excitons within exciton bands of the circular and elliptically deformed B850 rings with the deformation amplitude of 5.7%, and both with $V_i=370\ \text{cm}^{-1}$, $\varepsilon_\alpha=12490\ \text{cm}^{-1}$, $\varepsilon_\alpha-\varepsilon_\beta=326\ \text{cm}^{-1}$, and $\Gamma=125\ \text{cm}^{-1}$.

and vice versa at strong disorder in both two structures. The reasons for this are thermal mixing of eigenstates in exciton wavepackets and the band-edge states tend to be more localized in general [18]. Hence, delocalization lengths of different electronic excitations in both two structures are very dissimilar. In total, delocalization lengths of electronic excitations on B850 rings are various and are dependent on the kinds of measured electronically excited states and the kinds of studied geometric structures.

II. COHERENT EXCITON EXCITATION

In following, we will study the spatiotemporal evolution of electronic excitations on circular and elliptical B850 rings without taking into account dynamic disorder and exciton-phonon coupling. Owing to the excited singlet state lifetime for isolated monomeric *Bchla* and the fluorescence decay time for individual LH2 complexes are about 1 ns, the effect of the exciton recombination on WLET will be weak and we neglect it here. The time evolution of the density operator $\hat{\rho}(t)$ is

$$\hat{\rho}(t)=\exp(-iHt/\hbar)\hat{\rho}(0)\exp(iHt/\hbar), \tag{7}$$

it is used to calculate the dynamics of the electronic excitation [17,19], where $\hat{\rho}(0)$ is the initial density operator. If an initial excitation is a coherent exciton $|k=k_0\rangle$ on B850 ring, the time evolution of the excitation is represented by the matrix elements $\rho_{k,k'}(t)=\rho_{k,k'}(0)=\delta_{k,k'}\delta_{k,k_0}$ in eigenstate representation and $\rho_{n,n'}(t)=c_n^{k_0}c_{n'}^{k_0}$ in site representation and these matrix elements all are independent of the time t. Different coherent excitons within the exciton bands of different B850 rings will have different delocalization lengths and then will have different contributions to energy transfer.

In Fig. 2, we give the randomly averaged delocalization lengths and oscillator strengths of coherent excitons on the circular and elliptical B850 rings with deformation at 5.7% and both with $V_i=370\ \text{cm}^{-1}$, $\varepsilon_\alpha=12490\ \text{cm}^{-1}$, $\varepsilon_\alpha-\varepsilon_\beta=326\ \text{cm}^{-1}$, and $\Gamma=125\ \text{cm}^{-1}$, where the wavelengths correspond to randomly averaged eigenvalues of excitons at the zero-phonon lines. The oscillator strengths mainly concentrate on states $|k=16\rangle$, $|k=17\rangle$, and $|k=18\rangle$ in both structures. Generally coherent excitons on the circular ring are more delocalized than those on that deformed, and the differences are especially large at states $|k=1\rangle$, $|k=2\rangle$, $|k=17\rangle$, and $|k=18\rangle$, owing to the band-edge states trend to be more localized on the deformed ring. Hence when the initial excitation is a coherent exciton, the elliptical deformation will work against the energy transfer.

III. LOCALIZED EXCITON EXCITATION

If the initial excitation is a localized exciton $|n=n_0\rangle$ at the site n_0, the time evolution of the density matrix in site representation is

$$\rho_{n,n'}(t)\approx\sum_{k_1,k_2=1}^{18}\{\exp[i0.188(\nu_{k_2}-\nu_{k_1})t]\}c_n^{k_1}c_{n_0}^{k_1}c_{n_0}^{k_2}c_{n'}^{k_2}, \tag{8}$$

where ν_{k_1} and ν_{k_2} are the excitation wavenumbers of coherent excitons $|k_1\rangle$ and $|k_2\rangle$ at the zero-phonon lines, respectively, and we use inverse of centimeter (cm^{-1}) as unit. The time t is measured in picosecond (ps).

In Fig. 3, at sites $n=9,10,11$, we give the randomly averaged excitation populations $\langle\rho_{n,n}(t)\rangle_r$ as functions of excited time t for initially localized excitation at the site $n_0=1$ on the elliptical B850 ring with the deformation amplitude of 5.7%, where the site $n_0=1$ is nearby the major axis of the ring and $V_i=370\ \text{cm}^{-1}$, $\varepsilon_\alpha=12490\ \text{cm}^{-1}$, $\varepsilon_\alpha-\varepsilon_\beta=326\ \text{cm}^{-1}$, and $\Gamma=125\ \text{cm}^{-1}$. The energy transfer on the ring from the site $n_0=1$ to its opposite site $n=10$ with a population 0.09 needs 78 femtosecond (fs). Then the popu-

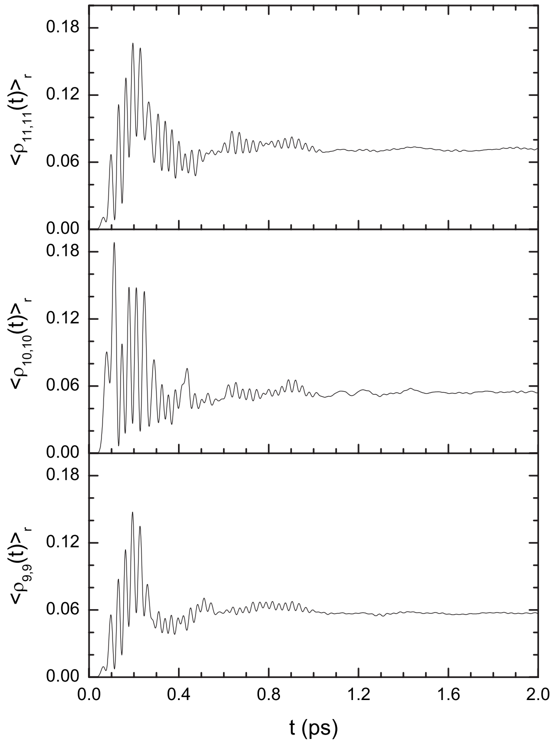

FIG. 3. The randomly averaged excitation populations $\langle\rho_{n,n}(t)\rangle_r$ at sites $n=9,10,11$ as functions of excited time t for initially localized excitation at the site $n_0=1$ on the elliptical B850 ring with the deformation amplitude of 5.7%, and $V_i=370\ \text{cm}^{-1}$, $\varepsilon_\alpha=12490\ \text{cm}^{-1}$, $\varepsilon_\alpha-\varepsilon_\beta=326\ \text{cm}^{-1}$, and $\Gamma=125\ \text{cm}^{-1}$.

lation at site $n=10$ oscillates as excited time t with the maximum peak 0.19 at 112 fs. This wavelike energy transfer on the ring keeps approximately 300 fs. After then the wavelike behavior disappears owing to destroying of the electronic quantum coherence by the random scattering, and after 1 ps of the excitation, the energy distribution on the ring will be almost stable which has a peak at the initially excited site $n=1$ as depicted in Fig. 4, where the excited time is always at $t=10$ ps. During the oscillating region of energy transfer, the populations at sites $n=5,6,7$ are small than those at sites $n=9,10,11$ in our calculation, and what's more, the behavior of energy transfer for initial excitation at the site $n_0=2$ is similar to that for initial excitation at the site $n_0=1$ on the same ring.

In Fig. 5, at sites $n=14,15,16$, we give the populations $\langle\rho_{n,n}(t)\rangle_r$ as functions of excited time t for initially localized excitation at the site $n_0=6$ on the same ring in Fig. 3, where site $n_0=6$ is nearby the elliptical minor axis. Now the energy transfer on the ring behaves as a weakly wavelike character. The transfer from the site $n_0=6$ to its opposite site $n=15$ needs 75 fs only with a population 0.04. Then the population at site $n=15$ weakly oscillates as excited time t with the maximum peak smaller than 0.07 at 249 fs. At the excited time $t=10$ ps, the energy distribution on the ring is depicted also in Fig. 4, and it has a peak at site $n=6$. Comparing the different energy transfers on the elliptical ring, in the wavelike transfer region, we find that if initial excitation is localized at the site near the elliptical major axis, the energy transfer from initial site to its opposite site is more efficient than that if initially excited site is nearby the minor axis. We also find that after 1 ps of the excitation, the energy distribution on the ring will be almost stable and in the case of initially excited site being nearby the major axis, the population at the opposite site of initial excitation is some larger than that in the case of initially excited site being nearby its minor axis as depicted in Fig. 4. Hence we can say that on the ellipti-

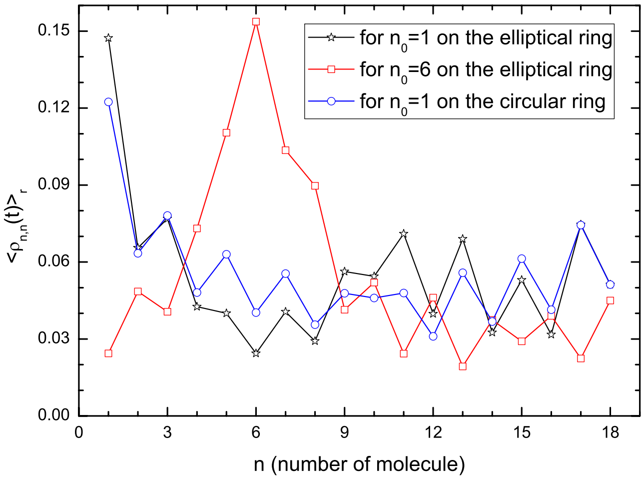

FIG. 4. (Color online) Distributions of populations $\langle\rho_{n,n}(t)\rangle_r$ on the circular B850 ring for initially localized excitation at the site $n_0=1$ and on the elliptical ring with the deformation amplitude of 5.7% for initially localized excitations at the sites $n_0=1$ and 6, where always at the excited time $t=10$ ps and with $V_i=370\ \text{cm}^{-1}$, $\varepsilon_\alpha=12490\ \text{cm}^{-1}$, $\varepsilon_\alpha-\varepsilon_\beta=326\ \text{cm}^{-1}$, and $\Gamma=125\ \text{cm}^{-1}$.

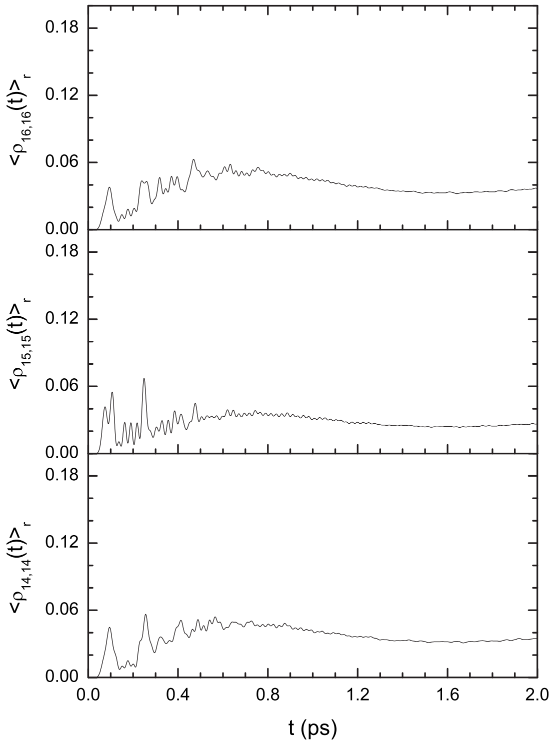

FIG. 5. The populations $\langle\rho_{n,n}(t)\rangle_r$ at sites $n=14,15,16$ as functions of excited time t for initially localized excitation at the site $n_0=6$ on the same ring in Fig. 3.

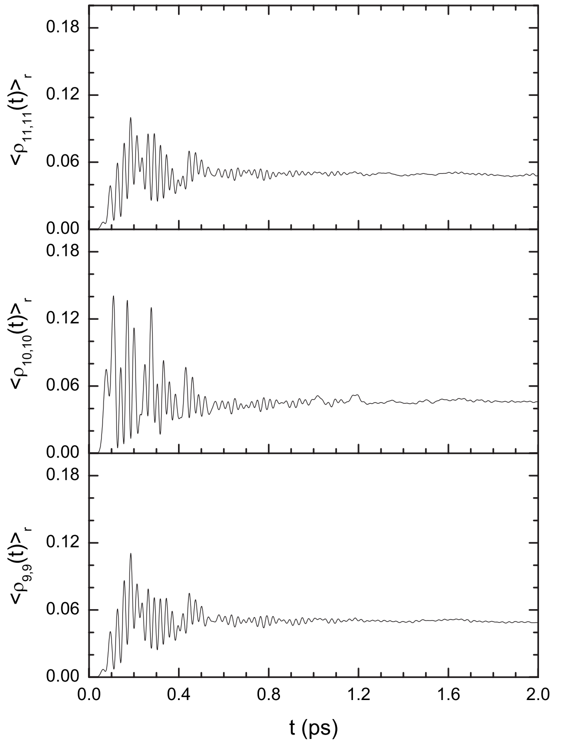

FIG. 6. The populations $\langle\rho_{n,n}(t)\rangle_r$ at sites $n=9,10,11$ as functions of excited time t for initially localized excitation at the site $n_0=1$ on the circular ring with $V_i=370$ cm^{-1}, $\varepsilon_\alpha=12490$ cm^{-1}, $\varepsilon_\alpha-\varepsilon_\beta=326$ cm^{-1}, and $\Gamma=125$ cm^{-1}.

cally deformed B850 ring, if initial excitation is localized at the site near its major axis, the energy transfer will be more efficient.

In Fig. 6, at sites $n=9,10,11$, we give the populations $\langle\rho_{n,n}(t)\rangle_r$ as functions of excited time t for initially localized excitation at the site $n_0=1$ on the circular B850 ring, where $V_i=370$ cm^{-1}, $\varepsilon_\alpha=12490$ cm^{-1}, $\varepsilon_\alpha-\varepsilon_\beta=326$ cm^{-1}, and $\Gamma=125$ cm^{-1}. The energy transfer from site $n_0=1$ to its opposite site $n=10$ with a population 0.07 needs 77 fs. Then the population at site $n=10$ oscillates as excited time t with the maximum peak 0.14 at 109 fs. The temporally averaged oscillation amplitude at the opposite site of initial excitation on the circular ring in WLET region is smaller than that in the case of initial excitation at the site $n_0=1$ of the elliptical ring in Fig. 3, but larger than that of $\langle\rho_{15,15}(t)\rangle_r$ on the same elliptical ring in the case of initial excitation at the site $n_0=6$ in Fig. 5 both visibly. During the stable population distribution, the population at the opposite site of initial excitation on the circular ring is some smaller than that in the case of initial excitation being nearby the elliptical major axis, but is some larger than that in the case of initial excitation being nearby the minor axis as depicted in Fig. 4. In total, if B850 ring is deformed elliptically, the initial excitation localized at the site near the major axis is in favor of energy transfer, but the initial excitation at the minor axis works against the transfer both visibly. The initially localized excitation in LH complexes may be induced via optical pumping through a much small nanoscale tip of an optical fiber [19].

We have investigated the behaviors of different electronic excitations on the circular and elliptically deformed B850 rings theoretically. We have shown that the degree of delocalization of electronic excitation in LH2 complexes is dependent on the kinds of measured electronically excited states and the kinds of studied structures. We also have shown that if initial excitation is a coherent exciton, the elliptical deformation of the B850 ring will work against energy transfer. We have depicted WLET persisting for several hundred femtoseconds on the circular and elliptically deformed B850 rings at cryogenic temperature, and found that if B850 ring is deformed elliptically, inducing the electronic excitation localized at the site near the major axis will be in favor of energy transfer, whereas at the minor axis will work against the transfer both visibly during WLET. In our calculation, we have only considered the Q_y transition, but owing to the fast $Q_x \rightarrow Q_y$ relaxation occurs, for example, in ~100–200 fs in Rs. rubrum [20], the Q_x excitation will indeed change the wavelike dynamics. This and the exciton-phonon interactions in LH2 complexes need further study. The effects of different structures of LH complexes in photosynthesis [21] may also need further investigation, and our study may be useful for designing different artificial systems to harvest light.

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