PHYSICAL REVIEW A **106**, 032218 (2022)

Quantum feedback control in quantum photosynthesis

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(Received 24 May 2022; accepted 12 September 2022; published 22 September 2022)

A model of charge separation in quantum photosynthesis as a model of quantum feedback control in a system of interacting excitons and vibrons is introduced. In this approach quantum feedback induces a nonlinear evolution equation which describes the Landau-Zener transition with decoherence and can be considered a quantum ratchet. It explains both the irreversibility in the process of charge separation for quantum photosynthesis (for this model direct transitions have probabilities close to 1, and reverse transitions have probabilities close to zero) and coincidence of the energy of the vibron paired to the transition and the Bohr frequency of the transition.

DOI: 10.1103/PhysRevA.106.032218

I. INTRODUCTION

Quantum effects in photosynthetic systems attract a lot of attention [1]. In particular, the irreversibility of charge separation in quantum photosynthesis has been discussed [2]. One of the key observations is the presence of vibrons (vibrations of nuclear degrees of freedom of chromophores with a lifetime of the order of several picoseconds) in photosynthetic systems [3]. The following phenomenon has been observed: typically, energy of the vibron is close to the Bohr frequency (i.e., the difference of energies between the levels) for electronic states which interact with the vibron; moreover, one vibron can interact with several transitions with the same Bohr frequency [3,4]. Another important phenomenon in quantum photosynthesis is the coincidence of the lifetime of electronic coherences and the duration of the Landau-Zener transition in the reaction center. In this work, we describe charge separation in quantum photosynthesis as a quantum feedback control in the Landau-Zener system modeling interacting excitons and vibrons, which explains both the irreversibility in the process of charge separation and the coincidence of the energy of the vibron paired to the transition and the Bohr frequency of the transition. The approach of this paper can be considered an attempt to describe the charge-separation process discussed in [4–6] using a classical model for vibrons and a quantum model for excitons; this will help us to understand better the mechanism of exciton-vibron interaction.

Exciton-vibron interaction works as follows. The interaction of electrons at chromophores with light generates excitons. Excitons generate vibrons as vibrations of nuclei in the field of the exciton according to the Franck-Condon principle in its semiclassical form—the landscape of the potential energy for nuclei (which corresponds to Coulomb interaction) changes abruptly when the exciton is excited, and for this new energy landscape the positions of the nuclei are not in equilibrium, and nuclei begin to oscillate around new minima of the potential energy. Transitions between potential-energy surfaces for electronic states happen via the Landau-Zener mechanism in the vicinity of the avoided-crossing point, where the potential-energy surfaces are close. Experimental observations of photosynthetic complexes have revealed many vibrons paired to different transitions between electronic states [4,7].

It is interesting to compare the process of charge separation in the PSII-RC (reaction center of photosystem II) [3–5,8] and in the bacterial reaction center [6,9] [reaction center of the purple bacterium *Rhodobacter* (*Rba.*) *sphaeroides*]. For PSII-RC, charge separation is controlled by the interaction with a vibron with an energy of 340 cm^{-1} which gives a resonance with the energy difference of the energy levels. The process of charge separation in the bacterial reaction center utilizes the interaction of electronic states with two vibrons with energies of 115 and 35 cm^{-1}, which are quite far from the resonance. A vibron with an energy of 115 cm^{-1} is generated when an exciton state is created and oscillates along the reaction coordinate for the transition between the exciton state and the charge-separation state, causing a transition to the charge-separation state. A vibron with an energy of 35 cm^{-1} is generated after this transition. It prevents recombination of the charge-transfer state, providing directionality of the charge-separation process.

For both photosystems it was observed [5,6] that the lifetime of the electronic coherences, which is important for the Landau-Zener mechanism, is close to the transition time between the energy levels for the process of charge separation in photosynthesis. This motivates the necessity to consider a Landau-Zener system with decoherence. The Landau-Zener system, including with decoherence, has been extensively studied to describe various phenomena in many works [10–15], and quantum control in the Landau-Zener system has been studied for different values of coupling strength, temperature, and driving force [16–20].

Various approaches for describing vibrons in quantum photosynthesis have been discussed, such as treating

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excitons and vibrons as collective eigenstates of the Hamiltonian [3,4], master equations in the polaron frame [21–23], and master equations with nonsecular terms [24], where vibrons are considered coherences of the vibrational degrees of freedom generated by interaction with populations of electronic states and are treated by nonsecular terms. Decoherence in the exciton-vibron interaction has been treated by using master equations [25,26]. The formation of collective polariton states in artificial photosynthesis has been studied [27]. Control-based models have been considered for similar problems, including the coherent control protocol for separating energy-transfer pathways for the Fenna-Matthews-Olson (FMO) photosynthetic complex [28], coherent open-loop optimal control of photosynthetic molecules for verification of coherent transport mechanisms in natural and artificial light-harvesting complexes under realistic conditions [29], coherent control of excitonic states in a light-harvesting complex with phase and phase-amplitude control of the electronically excited state populations in the FMO complex [30], controlled excitations induced by entangled photons in photosynthetic complexes to target excitation of specific states [31], and measurement-based incoherent control of retinal isomerization in rhodopsin [32]. Modeling the photosynthetic reaction center as a quantum heat engine has been considered [33]. These works exploit applications of open-loop control, i.e., control without feedback, which is different from the feedback (closed-loop) approach of the present work. Quantum feedback control, which has various applications in quantum technologies [34], has not been applied to this problem. An introduction to quantum feedback control can be found in [35]. An abstract formulation for controlling observable quantum systems was provided in [36]. The development of the formalism for models of quantum optics was given in [37–39]. The use of nonselective quantum measurements for controlling quantum systems and quantum feedback was studied in [40–52]. Quantum feedback control was used to describe the quantum Maxwell's demon in [53] based on the work in [54] (a model of the classical Maxwell's demon was considered by Smoluchowski [55] and discussed by Feynman in his lectures [56]; the demon was based on a ratchet which performs separation of molecules).

In this paper we construct a model of charge separation in quantum photosynthesis based on quantum feedback control and a quantum rachet in the Landau-Zener system. This model is different from other control models considered for photosynthesis in that it takes into account quantum feedback which induces nonlinearity of the evolution equation and leads to decoherence of electronic degrees of freedom during the Landau-Zener transition, which is described by the quantum measurement procedure or the collapse of electronic wave function and forces this transition to be directed. We describe the irreversible transition using a model of the quantum ratchet which is related to the notion of the quantum Maxwell's demon. Charge separation in the photosynthetic reaction center in our model utilizes this quantum ratchet, which performs irreversible transitions between potential-energy surfaces of electronic states using the oscillations of vibrons. The advantage of the considered model is that it allows us to explain both the irreversibility of the charge separation and the coincidence of the energy of the vibron paired to the transition and Bohr frequency of the transition. It is also simpler than common master-equation approaches with tailored parameters. A model of this kind can be considered for the process of avoiding trapping in quantum photosynthesis as described in [7]. In this regard, quantum feedback control can be considered an important mechanism for efficient operation of photosynthetic systems.

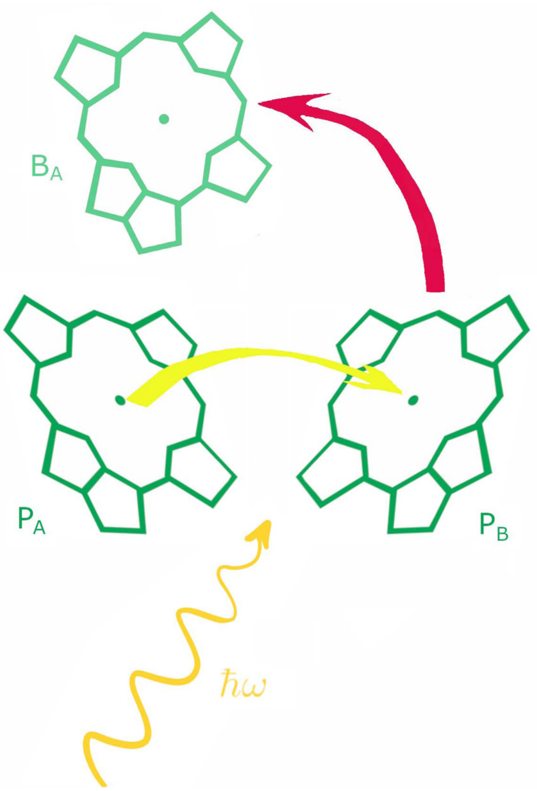

FIG. 1. Schematic picture of charge separation in the photosynthetic reaction center of the purple bacterium *Rhodobacter sphaeroides*. The reaction center contains a pair of strongly excitonically coupled bacteriochlorophylls, P_A and P_B; two monomeric BChl, B_A and B_B; two bacteriopheophytins, H_A and H_B; and two ubiquinone molecules symmetrically arranged in two branches (A and B branches; the A branch is not shown, and only part of the B branch is shown; B_B, H_A, H_B, and two ubiquinone molecules are not shown). Charge separation takes place in P_A and P_B (schematically shown by the yellow arrow), and then the electron is transported through the A branch of the reaction center (shown by the red arrow).

We consider as an example a model of charge separation in the reaction center of the purple bacterium *Rba. sphaeroides*, as schematically shown in Fig. 1; see [6] for a discussion of electron transfer in this system. The reaction center contains a pair P of strongly excitonically coupled bacteriochlorophylls, P_A and P_B; two monomeric BChl, B_A and B_B; two bacteriopheophytins, H_A and H_B; and two ubiquinone molecules symmetrically arranged in two branches (the A and B branches). In Fig. 1, the A branch is not shown, and only part of the B branch is shown. Charge separation takes place in P and is schematically shown by the yellow arrow. Then the electron is transported through the A branch of the reaction center, as schematically shown by the red arrow. The structures of all photosynthetic reaction centers are generally similar, and in particular, PSII-RC has a similar structure, although the energies of the vibrons paired to transitions are different.

The structure of this paper is as follows. In Sec. II, a quantum feedback model for a two-level system modeling the vibron is provided, and simulations are performed (we use parameters for the PSII reaction center since in this case a resonance between the energy of the vibron and the energy difference of the levels is observed). In Sec. III, a simple and intuitively more clear model with the Schrödinger equation and collapse of the wave function is discussed. In Sec. IV, a model of the quantum ratchet is considered. In Secs. III and IV we discuss the bacterial reaction center, where two vibrons are involved in the charge separation. Section V summarizes the results.

II. VIBRONS AND QUANTUM FEEDBACK CONTROL

A. Master equation for a quantum system with feedback control

The mathematical theory of quantum nonlinear filtering was developed by Belavkin [52], and that for physical models was developed by Carmichael [57] and Wiseman [38]. The dynamics of the density matrix ρ of a quantum system evolving under continuous measurement of an observable X (a Hermitian operator) with strength κ and feedback is described [38,58,59] by the stochastic Schrödinger equation,

$$\begin{aligned} d\rho &= -i[H,\rho] - \kappa[X,[X,\rho]]dt \\ &\quad + 4\kappa[X\rho + \rho X - 2\langle X\rangle\rho](dr - \langle X\rangle dt), \end{aligned} \tag{1}$$

where $r(t)$ is the observer's measurement record, which satisfies

$$dr = \langle X\rangle dt + \frac{1}{\sqrt{8\kappa}}dW. \tag{2}$$

Here W is the Wiener process, $dW^2 = dt$. Generalization to non-Hermitian X can be performed, for example, for an indirect quantum measurement which is achieved by causing the measured system to interact with a bath via a system operator L and then making a measurement of the bath [60]. The master equation for the evolution of the qubit density matrix during the measurement process within Bayesian formalism was studied [61,62]. Geometric tools for the quantum filtering equation with examples of one- and two-qubit system were applied [63]. A quantum Fokker-Planck master equation describing joint dynamics of a quantum system and a detector with finite bandwidth for feedback control was recently considered [64–66].

The control Hamiltonian H is a function of the measurement record $r(t')$ at $t' < t$, $H = H(r)$. The function $H(r)$ can be nonlinear in general. In the case of a strong measurement ($\kappa \gg 1$) one has $dr \approx \langle X\rangle dt$ and, approximately,

$$\frac{d\rho}{dt} = -i[H,\rho] - \kappa[X,[X,\rho]], \tag{3}$$

where $H = H(\langle X_{t'<t}\rangle)$. In the case of a negligible time delay in the feedback loop, one can consider in particular $H = H(\langle X_t\rangle)$, where $\langle X_t\rangle = \mathrm{Tr}\rho_t X$. The term $-[X,[X,\rho]] = 2X\rho X - X^2\rho - \rho X^2$ for the Hermitian observable $X = X^\dagger$ has a standard Lindblad form and describes the collapse of the wave function of the system due to measurement back-action or interaction with the environment. Hence, Eq. (3) is a (in general, nonlinear) Markovian master equation. Below we explicitly write terms with observable X, which is a combination of $|1\rangle\langle 2|$ and conjugated and Hamiltonian H for the Landau-Zener transition with dissipation and feedback for the description of the quantum photosynthetic system.

B. Master equation with vibrons

We describe the process of charge separation in quantum photosynthesis using the master equation for the 2×2 density matrix $\rho(t)$ with quantum feedback, dissipation, and sink,

$$\frac{d}{dt}\rho(t) = -i[H(\rho,t),\rho(t)] + \mathcal{L}(\rho(t)) + \mathcal{S}(\rho(t)), \tag{4}$$

$$H(\rho,t) = \begin{pmatrix} E_1 + \mathbf{u}\cdot\mathbf{q}(\rho,t) & J \\ J & E_2 + \mathbf{v}\cdot\mathbf{q}(\rho,t) \end{pmatrix}, \tag{5}$$

where $\mathbf{u}\cdot\mathbf{q}$ and $\mathbf{v}\cdot\mathbf{q}$ describe the interaction with the vibrons with feedback in $\mathbf{q}$,

$$\mathbf{q}(\rho,t) = \rho_{11}(t)\mathbf{q}_1(t) + \rho_{22}(t)[\mathbf{q}_1(t) + \mathbf{q}_2(t)]. \tag{6}$$

The vibrons $\mathbf{q}_1(t)$ and $\mathbf{q}_2(t)$ can be compared with the vibron observed in PSII-RC with an energy of 340 cm^{-1} [5] and with vibrons observed in the bacterial reaction center of *Rba. sphaeroides* with energies of 115 and 35 cm^{-1} [6].

Decoherence and dissipation during the transition are described by the Lindblad term

$$\begin{aligned} \mathcal{L}(\rho) &= \gamma^+\left(\langle 2|\rho|2\rangle|1\rangle\langle 1| - \frac{1}{2}\{\rho,|2\rangle\langle 2|\}\right) \\ &\quad + \gamma^-\left(\langle 1|\rho|1\rangle|2\rangle\langle 2| - \frac{1}{2}\{\rho,|1\rangle\langle 1|\}\right), \\ |1\rangle &= \begin{pmatrix}1\\0\end{pmatrix}, \quad |2\rangle = \begin{pmatrix}0\\1\end{pmatrix}. \end{aligned} \tag{7}$$

Here $\gamma^+/\gamma^- = e^{-\beta(E_1-E_2)}$, E_1 and E_2 ($E_1 > E_2$) are the energies of levels $|1\rangle$ and $|2\rangle$, respectively, β is the inverse temperature for this transition ($\beta^{-1} \approx 300$ K at room temperature), and $\{A,B\} := AB + BA$ denotes the anticommutator of operators A and B.

The physical meaning of this model is as follows: excitations are created by absorption of light; excitation is level $|1\rangle$ of the system. Charge separation corresponds to the transition from level $|1\rangle$ to level $|2\rangle$ and is described by the first two terms in (4). The term

$$\mathcal{S}(\rho) = -s_1\langle 1|\rho|1\rangle|1\rangle\langle 1| - s_2\langle 2|\rho|2\rangle|2\rangle\langle 2|, \quad s_1, s_2 > 0, \tag{8}$$

describes the sink of the electronic states. The physical meaning is that the second term, $-s_2\langle 2|\rho|2\rangle|2\rangle\langle 2|$, describes the transport of the electron from the special pair of chromophores in the reaction center along the charge-transfer chain and the first term, $-s_1\langle 1|\rho|1\rangle|1\rangle\langle 1|$, describes the recombination of excitons.

Master equations with nonsecular terms (terms which describe interaction between populations and coherences, i.e., diagonal and off-diagonal elements of the density matrix) were discussed for quantum photosynthesis, in particular in [24], where the nonsecular term was called the "laser-like

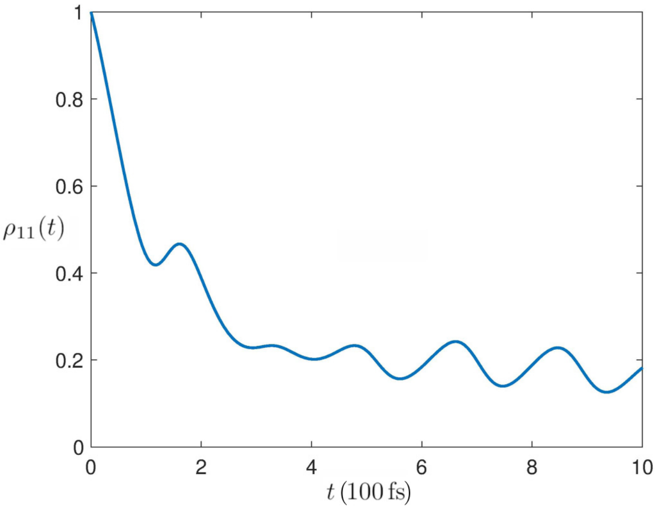

FIG. 2. The time dependence of the matrix element $\rho_{11}(t)$ of the density matrix $\rho(t)$ which is the solution of Eq. (4) with the parameters described in Sec. II C. Time is in units of 100 fs. To decrease recombination, one has to minimize the area under the curve, which is the effective exciton recombination time $\bar{T}$.

driving" term which maintains electronic coherences. In the approach of the present paper we make this nonsecular term dependent on the electronic state through interaction with the vibron (6); this makes the above master equation a model of the quantum feedback control.

In order to make the operation of quantum photosynthesis more effective one has to minimize recombination through the first term of the sink operator (8). Exciton recombination is proportional to the effective time the system spends in state $|1\rangle$, i.e., to

$$\bar{T} = \int_0^\infty \rho_{11}(t)dt, \tag{9}$$

where $\rho(t)$ is a solution of (4) with the initial condition $|1\rangle\langle 1|$.

The goal is to find the regime which minimizes $\bar{T}$. This minimization can be achieved if the charge-separation transition $|1\rangle \to |2\rangle$ is as fast and irreversible as possible; see the discussion in [5]. We have performed a simulation of the evolution governed by the dynamical equation (4) for the parameters (10), as shown in Fig. 2. This simulation shows that the recombination time (9) is small if the energy of the vibron (as discussed below) matches the difference $E_1 - E_2$ of the energy levels of the system and if the time of transition $|1\rangle \to |2\rangle$ is close to the decoherence time for the corresponding system state defined by the dissipative operator (7). Both these conditions are satisfied in nature and have been extensively discussed in the literature (see, for example, [3–6]).

C. Simulation

For the simulation, we consider Eqs. (4), (5), (7), and (8) with the vibron (6) chosen as follows: we ignore the second vibron $\mathbf{q}_2(t) = 0$ and choose the first vibron $\mathbf{q}_1(t)$ in the form $2\sin(3.35t)$; that is, the parameters are [in the notation of (5) and (14)]

$$\mathbf{u} = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad \mathbf{v} = \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \quad \mathbf{q}_0 = 0, \quad \mathbf{v}_1 = \begin{pmatrix} 0 \\ 1 \end{pmatrix},$$
$$E_1 = 3, \quad E_2 = 0, \quad \omega_1 = 3.35, \quad J = 0.75. \tag{10}$$

With these parameters the Hamiltonian (5) takes the explicit form

$$H(\rho, t) = \begin{pmatrix} 3 & 0.75 \\ 0.75 & 2\sin(3.35t)[\rho_{11}(t) + \rho_{22}(t)] \end{pmatrix}.$$

Due to the sink the trace of the density matrix is not conserved. We also choose

$$s_2 = 0.1, \quad \gamma^- = 0.5, \quad \gamma^+ = 0.22, \quad \gamma^- = 0.11.$$

These parameters describe the PSII reaction center; values are given in hundreds of cm^{-1} (i.e., $E_1 - E_2 = 300$ cm^{-1}, $J = 75$ cm^{-1}) [4,5]. The energy of the vibron ω_1 is chosen to be 335 cm^{-1} [the exact resonance is $2\sqrt{(E_1 - E_2)^2/4 + J^2} = 335$, and the experimental value is 340 cm^{-1}]. The ratio $\gamma^+/\gamma^- = \exp[-(E_1 - E_2)/kT] = 0.22$ corresponds to $E_1 - E_2 = 300$ cm^{-1} and $T = 300$ K. The decoherence is strong enough to make the direct transition $|1\rangle \to |2\rangle$ coherent and the reverse transition $|2\rangle \to |1\rangle$ incoherent, as discussed in [5] (the decoherence time is close to the half period of the transition). We choose the parameters of the PSII reaction center for the simulation because for this case the exact resonance with the vibron is observed [4,5].

The result of the simulation for the matrix element $\rho_{11}(t)$ of the density matrix which is a solution of the master equation (4) with the initial condition $\rho(0) = |1\rangle\langle 1|$ is shown in Fig. 2 (for simplicity we choose $s_1 = 0$ in the simulation for this solution; the time axis unit is 100 fs). With the parameters chosen, the system performs the fast transition $|1\rangle \to |2\rangle$ (with the help of the oscillation of the vibron, which reduces the difference in energies of the energy levels), and the reverse transitions will be suppressed [when the system arrives at level $|2\rangle$, the vibron oscillates in the reverse direction, and the energy difference increases; moreover, decoherence reduces transitions by the action of the first term in (4)].

III. MODEL WITH SCHRÖDINGER'S EQUATION

In this section we analyze an intuitively simpler model with quantum feedback which utilizes pure states, where instead of dissipation we consider the collapse of the wave function. This model allows us to understand on the physical level of rigor how the above quantum feedback equation generates irreversible transitions. The idea is as follows: instead of an investigation of the quantum open system with a sink considered above one can discuss only the transition between levels $|1\rangle$ and $|2\rangle$, using pure states and the collapse of wave functions to describe the decoherence. This should allow us, on the physical level of rigor, to explain the nonsymmetry of the transition between $|1\rangle$ and $|2\rangle$ (although it should be symmetric using the Landau-Zener formula).

A. Landau-Zener formula

The model by Landau and Zener investigates the dynamics of a two-level quantum system with Schrödinger's equation,

$$\frac{d}{dt}\psi(t) = -iH(t)\psi(t), \quad \psi(t) = \begin{pmatrix}\psi_1(t)\\ \psi_2(t)\end{pmatrix},$$

where the time-dependent Hamiltonian acts in the basis of states $|1\rangle$ and $|2\rangle$ as

$$H(t) = \begin{pmatrix} ut & J \\ J & vt\end{pmatrix}, \quad |1\rangle = \begin{pmatrix}1\\0\end{pmatrix}, \quad |2\rangle = \begin{pmatrix}0\\1\end{pmatrix}.$$

States $|1\rangle$ and $|2\rangle$ are called diabatic energy levels, whereas the eigenlevels of the time-dependent Hamiltonian $H(t)$ are called adiabatic levels. For this system, the probability of a transition between diabatic states at large times was computed analytically [67–70] to be equal to $1 - P$, where

$$P = e^{-2\pi\gamma}, \quad \gamma = \frac{J^2}{|u-v|}. \tag{11}$$

This transition occurs in the vicinity of the avoided-crossing point, where matrix elements at the diagonal of the generator are equal, i.e., in the vicinity of the point $t = 0$. The value of $1 - P$ is large either for a large interaction J or for a low difference $|u - v|$ between the velocities of the passage through the avoided-crossing point at different slopes of the transition.

B. Landau-Zener transition with dissipation

The Landau-Zener theory with dissipation was considered, e.g., in [10–15]. In all these papers the assumption $u = -v$ for the velocities of the transition (sweep velocity) was made. This assumption describes the general situation for the case without dissipation since in this case it reduces to subtraction of a term wt times the unity matrix from the Hamiltonian (where $2w = u + v$), but for the case with dissipation it is not general—coherences in the system decay in the process of the transition, which makes different sides of the avoided crossing point (with velocities u and v) unequal. This is especially important in the adiabatic case, where the system spends a considerable time in the vicinity of the avoided crossing and decoherence in the process of the transition cannot be ignored. We will consider a model in which decoherence of the wave function is described by a projection operator applied at the avoided-crossing point.

Without decoherence the electronic wave function feels both sides of the transition due to quantum nonlocality, which gives the contribution $|u - v|$ in the Landau-Zener formula $P = e^{-\frac{2\pi J^2}{|u-v|}}$, and the probabilities of direct and reverse transitions are equal, but if this wave function is collapsed in the avoided-crossing point between slopes u and v, then the probabilities of the direct and reverse transitions become different (with $|u|$ and $|v|$ instead of $|u - v|$). It is natural to choose the avoided crossing as the point of collapse of the wave function for the regime when the vibron is slow in the avoided crossing—the vicinity of the avoided crossing is small, but the system spends a lot of time there, as discussed below.

The analysis of Landau [67] involved quasiclassical arguments, in which the transition probability $P_{1\to 2}$ is given by integration over complex time, where t_1 is real and t_2 is the branch point where the two eigenvalues of the Hamiltonian are equal,

$$P_{1\to 2} = \exp\left(-\frac{2}{\hbar}\mathrm{Im}\int_{t_1}^{t_2}[E_2(t) - E_1(t)]dt\right).$$

The difference of eigenvalues $E_2(t) - E_1(t)$ arises from the integral over a loop in a contour in the complex plane where the integral on the left side of the loop contains $E_1(t)$ and the integral on the right side of the loop contains $E_2(t)$.

If the wave function collapses at the branch point t_2, then only half of the integral remains, and we obtain, for the transition probability $|1\rangle \to |2\rangle$, the expression

$$P_{1\to 2} = \exp\left(\frac{2}{\hbar}\mathrm{Im}\int_{t_1}^{t_2} E_1(t)dt\right).$$

An analogous expression with the integral of $E_2(t)$ exists for the probability of transition $|2\rangle \to |1\rangle$,

$$P_{2\to 1} = \exp\left(-\frac{2}{\hbar}\mathrm{Im}\int_{t_1}^{t_2} E_2(t)dt\right).$$

That is, transition probabilities of the direct and reverse transitions for the case with the collapse of the wave function are different; generally, $P_{1\to 2} \neq P_{2\to 1}$, and the transition becomes directed.

Landau-Zener transitions with decoherence induced by measurement-assisted transitions were considered in [46]. This approach is not valid if projection occurs at the avoided crossing. It might seem that the probability of the collapse of a wave function in the vicinity of an avoided crossing is rather low, but important regimes exist where this probability cannot be neglected.

For the case of quantum photosynthesis the transition time $|1\rangle \to |2\rangle$ of charge separation in [6] is approximately 100 fs, which is close to the lifetime of electronic coherences. Therefore, we obtain exactly the regime discussed above.

C. Exciton-phonon Hamiltonian

The Hamiltonian for interaction between excitons and phonons has the form [71]

$$H = H_{\mathrm{el}} + H_{\mathrm{Coul}} + H_{\mathrm{ph}} + H_{\mathrm{el-ph}} + H_{\mathrm{reorg}},$$

where

$$H_{\mathrm{el}} = \sum_n \varepsilon_n |n\rangle\langle n|, \quad H_{\mathrm{reorg}} = \sum_{n,k} |n\rangle\langle n| \int \frac{h_{nk}^2(\xi)}{\omega_k(\xi)} d\xi,$$

$$H_{\mathrm{ph}} = \sum_k \int \omega_k(\xi) c_k^+(\xi) c_k^-(\xi) d\xi,$$

$$H_{\mathrm{el-ph}} = \sum_{n,k} |n\rangle\langle n| \int h_{nk}(\xi)[c_k^+(\xi) + c_k^-(\xi)]d\xi,$$

$$H_{\mathrm{Coul}} = \frac{1}{2}\sum_{m,n,\, m\neq n} J_{mn}|m\rangle\langle n|, \quad J_{nm} = J_{mn}^*.$$

Here $|n\rangle$ are electronic states (excitons), $c_k^{\pm}$ are creation and annihilation operators for phonons, and the values of J_{mn} describe Coulomb interaction between excitons.

D. Vibrons and quantum feedback control

An important observation in quantum photosynthesis is that excitation of electronic states generates vibrons that describes self-interaction, which we will describe via a quantum feedback control approach. To describe the process of charge separation in quantum photosynthesis, we need three electronic states in the bacteriochlorophyll pair P: $|0\rangle$. the ground state; $|1\rangle$. the exciton; and $|2\rangle$, the charge-separation state, with energies such that $E_1 > E_2 > E_0$. Charge separation, which we discuss below, is the transition between $|1\rangle$ and $|2\rangle$.

The above Hamiltonian contains the transition term between states $|1\rangle$ and $|2\rangle$ (where J is real):

$$J(|1\rangle\langle 2| + |2\rangle\langle 1|).$$

We will consider vibrons classically as vector-valued functions of time, so that to describe the interaction between electrons and vibrons we will substitute the quantum operator $c_k^+ + c_k^-$ in the above Hamiltonian $H_{\text{el-ph}}$ by the classical value q_k; it corresponds to the semiclassical Franck-Condon principle. Then the interaction of the vibron with the pair of electronic states will be described by the interaction Hamiltonian,

$$\sum_{n=1,2;k} |n\rangle\langle n|h_{nk}q_k(t) = |1\rangle\langle 1|\mathbf{u}\cdot\mathbf{q}(t) + |2\rangle\langle 2|\mathbf{v}\cdot\mathbf{q}(t).$$

Here the vibron is described by the vector $\mathbf{q}(t) = [q_k(t)]$ in the scalar products with vectors $\mathbf{u}$ and $\mathbf{v}$ of the interactions with electronic states.

We also have to take into account that vibrons are switched on by transitions between electronic states: excitation of the exciton $|1\rangle$ generates the vibron $\mathbf{q}_1(t)$, and the transition to the charge-transfer state $|2\rangle$ generates the second vibron $\mathbf{q}_2(t)$. Following the above discussion, we assume that at the moment of excitation of a vibron a collapse of electronic wave functions occurs. To describe this behavior we will multiply projectors $P_n = |n\rangle\langle n|$ for $n = 1, 2$ onto electronic states in the above formula by populations $\rho_{nn}(t)$ of these states. This procedure describes phenomenologically the effect of measurement, i.e., decoherence of the electronic wave function due to interaction with the environment; moreover, the measurement occurs at the avoided-crossing point. Therefore, we obtain feedback (the quantum dynamics controls itself): the generation of vibrons is controlled by populations of corresponding electronic states, and vibrons initiate transitions between electronic states.

The quantum ratchet contains two vibrons as in [6]. Vibron $\mathbf{q}_1(t)$, with an energy of 115 cm^{-1}, moves the electronic state $|1\rangle$ to the avoided-crossing point, and vibron $\mathbf{q}_2(t)$, with an energy of 35 cm^{-1}, repels state $|2\rangle$ from the avoided-crossing point after the transition. These two vibrons together control the irreversible charge-separation transition. They can be treated as the right and left hands of the quantum Maxwell's demon. State $|\psi(t)\rangle$ describes the electronic degrees of freedom which correspond to the two potential-energy surfaces for exciton $|1\rangle$ and charge-separation state $|2\rangle$. In this section we construct a model of the quantum feedback control for the quantum photosynthetic system discussed in [6].

For the dynamics of exciton-vibron interaction we get the Landau-Zener Hamiltonian for quantum feedback control master equations (1) and (3), which describe the collapse of wave function:

$$H(\rho,t) = \begin{pmatrix} E_1 + \mathbf{u}\cdot\mathbf{q}_1(t)\rho_{11}(t) & J \\ J & E_2 + \mathbf{v}\cdot[\mathbf{q}_1(t)+\mathbf{q}_2(t)]\rho_{22}(t) \end{pmatrix},$$

where all vibrons as a function of time should be continuous and should have continuous first derivatives since a vibron after excitation starts moving from some initial position and zero velocity.

For the considered case of pure quantum states we have

$$\rho_{11}(t) = |\langle 1|\psi(t)\rangle|^2, \quad \rho_{22}(t) = |\langle 2|\psi(t)\rangle|^2,$$

where ψ is the wave function of the electron. Experimental observations of photosynthetic complexes reveal many vibrons paired to different transitions between electronic states [4,7]. Charge separation as a principal process is paired with two vibrons, which allows us to make this transition directed. Other transitions are paired to a single vibron $\mathbf{q}(t)$ with a corresponding quantum feedback control equation. Thus, finally, we get the (phenomenological) quantum feedback control Hamiltonian,

$$H(\psi,t) = \begin{pmatrix} E_1 + \mathbf{u}\cdot\mathbf{q}_1(t)|\langle 1|\psi(t)\rangle|^2 & J \\ J & E_2 + \mathbf{v}\cdot[\mathbf{q}_1(t)+\mathbf{q}_2(t)]|\langle 2|\psi(t)\rangle|^2 \end{pmatrix}, \tag{12}$$

and the corresponding Schrödinger equation with quantum feedback.

$$\frac{d}{dt}\psi(t) = -iH(\psi,t)\psi(t). \tag{13}$$

Squared moduli of quantum amplitudes at the diagonal of the generator $H(\psi,t)$ describe the collapse of the electronic wave function at the avoided-crossing point for the Landau-Zener transition with friction. The quantum feedback Hamiltonian (12) has cubic nonlinearity like for the nonlinear Schrödinger equation. We claim that this model allows us to obtain different probabilities for direct $|1\rangle \to |2\rangle$ and reverse $|2\rangle \to |1\rangle$ transitions, and hence, it describes the quantum ratchet, although the standard Landau-Zener formula predicts equal probabilities for direct and reverse transitions. Below we perform a rough estimate of probabilities of transitions for the quantum ratchet with Hamiltonian (12).

First, we consider the direct move of the quantum ratchet, that is, the transition $|1\rangle \to |2\rangle$, where $\langle 1|\psi(t)\rangle = 1$ and $\langle 2|\psi(t)\rangle = 0$. Then the generator of the transition takes the form

$$\begin{pmatrix} E_1 + \mathbf{u}\cdot\mathbf{q}_1(t) & J \\ J & 0 \end{pmatrix}.$$

The avoided-crossing point for the direct move of the quantum ratchet is defined by the condition

$$E_1 + \mathbf{u}\cdot\mathbf{q}_1(t) = 0.$$

Since vibronic oscillations are at least an order of magnitude slower than electronic oscillations, these oscillations can be linearized in the vicinity of the avoided-crossing point. Then the probability of the transition $|1\rangle \to |2\rangle$ between diabatic states for one passage of the vibron through the avoided-crossing point can be approximated by the Landau-Zener formula with quantum feedback; that is, the electronic wave function is collapsed in the avoided-crossing point: this probability equals $1 - P$, where

$$P = e^{-2\pi\gamma}, \quad \gamma = \frac{J^2}{|\mathbf{u}\cdot\mathbf{w}|},$$

and $\mathbf{w}$ is the velocity of the vibron in the moment of passage of the avoided-crossing point. By decelerating the vibron at the moment of the passage, it is possible to make the transition probability close to 1, $1 - P \approx 1$.

Now we consider the reverse move of the quantum ratchet, that is, the transition $|2\rangle \to |1\rangle$, where $\langle 1|\psi(t)\rangle = 0$ and $\langle 2|\psi(t)\rangle = 1$. The generator of the transition (we again take into account the quantum feedback) is

$$\begin{pmatrix} 0 & J \\ J & E_2 + \mathbf{v}\cdot[\mathbf{q}_1(t) + \mathbf{q}_2(t)] \end{pmatrix}.$$

For the reverse movement of the quantum ratchet the avoided-crossing point satisfies

$$E_2 + \mathbf{v}\cdot[\mathbf{q}_1(t) + \mathbf{q}_2(t)] = 0.$$

Instead of a single vibron $\mathbf{q}_1(t)$ (which was the case for the direct move) this transition is driven by the sum of vibrons $\mathbf{q}_1(t)$ and $\mathbf{q}_2(t)$. Hence, it might be that the above equation has no solutions and the transition probability is close to zero, $1 - P \approx 0$.

In general, the problem of quantum control for the quantum ratchet with the Hamiltonian (12) can be formulated as follows: find parameters for vibrons $\mathbf{q}_1(t)$ and $\mathbf{q}_2(t)$ to make the transition probability $P_{1\to 2}$ for the direct move $|1\rangle \to |2\rangle$ of the ratchet close to 1 and the transition probability $P_{2\to 1}$ for the reverse move $|2\rangle \to |1\rangle$ of the ratchet close to zero.

IV. A MODEL OF THE QUANTUM RATCHET

Here we consider a solution for a model of vibrons and show that the operation of the quantum ratchet described in the previous section indeed takes place.

We take the following ansatz for the first vibron:

$$\mathbf{q}_1(t) = \mathbf{q}_0 + \mathbf{v}_1 \sin(\omega_1 t). \tag{14}$$

That is, the vibron is switched on when the exciton is excited and oscillates with frequency ω_1 along the vector $\mathbf{v}_1$ starting from the initial position $\mathbf{q}_0$. For the second vibron we take the following ansatz:

$$\mathbf{q}_2(t) = \mathbf{v}_2[\sin(\omega_2 t) - 1]. \tag{15}$$

That is, the vibron oscillates with frequency ω_2 along vector $\mathbf{v}_2$.

Then the equation for the avoided-crossing point for the direct move of the quantum ratchet is

$$E_1 + \mathbf{u}\cdot[\mathbf{q}_0 + \mathbf{v}_1 \sin(\omega_1 t)] = 0. \tag{16}$$

This equation can be satisfied if the amplitude of oscillations of the vibron is large enough. Let us assume, as in Sec. II, that $E_1 = 1$, $E_2 = 0$, and $\mathbf{u}\cdot\mathbf{q}_0 = 0$.

To elevate the transition probability one has to make the velocity of the vibron (14) in the moment of passage through the avoided-crossing point (16) close to zero, i.e., to make $\mathbf{u}\cdot\mathbf{v}_1 = -1$. Thus, it is the adiabatic regime. To amplify transitions to make the transition probability for the direct move of the ratchet close to 1, one has to decrease the amplitude of the vibron almost to the minimal value which still allows the existence of a solution of Eq. (16) for the crossing point.

The energy difference between adiabatic levels (the eigenlevels of the time-dependent Hamiltonian) at the initial time point $t = 0$ is close to $E_1 - E_2 = 1$, and in the avoided-crossing point it almost equals zero (since J is small); that is, in the adiabatic regime the energy of the vibron matches the energy difference between the levels. The approximate coincidence of the energy of the vibron and the energy difference of levels coupled to this vibron has widely been discussed in the literature, in particular in [3,4,6].

The equation for the avoided-crossing point for the reverse move of the quantum ratchet has the form

$$\begin{aligned}\Delta E(t) &:= E_2 + \mathbf{v}\cdot\{\mathbf{q}_0 + \mathbf{v}_1 \sin(\omega_1 t) + \mathbf{v}_2[\sin(\omega_2 t) - 1]\} \\ &= 0.\end{aligned} \tag{17}$$

The solvability of this equation can be easily broken; this is obvious if we choose, as in (10), $E_2 = 0$, $\mathbf{v}\cdot\mathbf{q}_0 = -1$, and $\mathbf{v}\cdot\mathbf{v}_1 = 0$ and choose $\mathbf{v}\cdot\mathbf{v}_2 > 0$. Then this equation will not have a solution. Let us consider a more complex case to imitate the discussion in [4,6], where the first vibron was used to put the system in the vicinity of the avoided crossing and the second vibron was used to repel the system from the avoided crossing and prevent reverse transitions.

Let us choose $E_2 = 0$, $\mathbf{v}\cdot\mathbf{q}_0 = -1$, $\mathbf{v}\cdot\mathbf{v}_1 = 1$, and $\mathbf{v}\cdot\mathbf{v}_2 = 1$ (the exact value of this scalar product is not very important, but it should be positive). Then to satisfy (17) we need simultaneously $\sin(\omega_1 t) = 1$ and $\sin(\omega_2 t) = 1$, which is a rare event (even in the approximate sense). Hence, reverse transitions will be effectively reduced. The simulation with the above parameters in Fig. 3 (we also choose $\omega_1 = 2$ and $\omega_2 = 3.7$) shows that due to the interference of the two vibrons the energy difference in (17) is close to zero in comparably infrequent cases.

Remark. The model of charge separation discussed in this paper has the following feature: in order to make charge transfer faster, the vibron (14) should operate in the adiabatic regime; that is, the amplitude of the vibron has the minimal value which allows us to satisfy Eq. (16) for the avoided-crossing point. As mentioned above, in this regime the energy of the vibron matches the energy difference between the levels. Also in this regime the vibron is "fragile"—perturbations of the parameters of the vibron related to mutations can break the solvability of (16) and block the charge transfer. In [6],

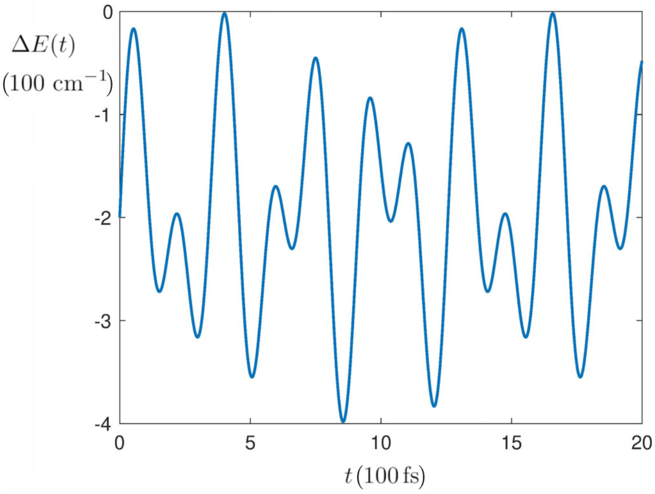

FIG. 3. Plot of $\Delta E(t)$, which is defined by Eq. (17). Time is in units of 100 fs. The energy difference $\Delta E(t)$ is in units of 100 cm^{-1}.

mutations in the photosynthetic reaction center were investigated with the following observations: the YM210W mutation which causes small changes in geometry and potential in the reaction center modifies the operation of the vibron with an energy of 115 cm^{-1} and slows the excitation transfer by two orders of magnitude. One can consider this observation to be proof that a vibron with energy of 115 cm^{-1} works in the adiabatic regime (the velocity of the vibron in the avoided-crossing point is slow). It is interesting that the GM203L mutation which removes the second vibron (with energy of 35 cm^{-1}) slows the excitations transfer by only one order of magnitude [6]. The discussion in this and the previous section uses the bacterial reaction center as in [6], while for the simulation in Sec. II we use the PSII reaction center as in [4,5].

V. CONCLUSIONS

We considered a model of charge-separation transition in quantum photosynthesis based on the quantum feedback control equation. This quantum feedback control model is explained as follows: for a Landau-Zener transition with dissipation that is close to adiabatic transitions, when the vibron is slow at the avoided crossing, the collapse of the electronic wave function should occur at the avoided-crossing point. This process can be described by a quantum feedback control model which allows to make the probabilities of the direct and reverse transitions different so that the transition becomes directed. Taking into account excitation of two vibrons, it is possible to make this transition maximally irreversible, i.e., to make the probability of the direct transition maximal and the probability of the reverse transition minimal. The nonlinearity in this approach can be considered to be an effect of the approximation of self-interaction as in the method of self-consistent field. To make the probability of the direct transition maximal, the energy of the vibron should be close to the Bohr frequency of the transition. Another indication of the regime of this quantum control model is the coincidence of the lifetime of the electronic coherences and the duration of the Landau-Zener transitions. Both of these properties—resonance between energies of the vibron and the Bohr frequency of the transition and the similarity of the lifetime of the electronic coherence and the duration of the transition—are observed in experiments. Numerical simulations were performed for the parameters describing the PSII and bacterial reaction centers.

ACKNOWLEDGMENTS

The authors are grateful to I. V. Volovich and A. S. Trushechkin for discussions. This work was funded by the Ministry of Science and Higher Education of the Russian Federation (Grant No. 075-15-2020-788).

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