Model of Vibrons in Quantum Photosynthesis as an Analog of a Model of Laser

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Received August 27, 2018; revised October 7, 2018; accepted May 1, 2019

Abstract—The mechanism of vibronic amplification of transport of excitons has been discussed in connection with quantum photosynthesis. Vibrons (some modes of vibrations of molecules) have been observed experimentally in photosynthetic systems. In the present paper we consider models of vibronic amplification of quantum transfer in which the generation of vibrons as a coherent vibrational mode is described by an analog of the semiclassical laser theory. We study two models: a model of nonequilibrium three-level system with vibronic mode, and a variant of a model of lasing without inversion. We conjecture that the dark states discussed in connection with quantum photosynthesis might be related to the mechanism of vibronic “laser” without inversion, which amplifies the transfer of excitons. We prove that in the presence of a vibronic mode the transfer rate of excitons increases, and compute the dependence of the transfer rate on the parameters of the model.

DOI: 10.1134/S0081543819050146

1. INTRODUCTION

Quantum effects in photosynthesis attract much attention [4, 21]. In quantum photosynthesis the effect of generation of vibrons is observed. Vibrons are some modes of vibrations of molecules related to transitions between electronic states (the energies of vibrons are equal to the Bohr frequencies for the corresponding transitions). A relation of vibrons to the amplification of transfer of excitons to the photosynthetic reaction center has also been discussed [6, 7, 11, 16, 20].

In the present paper we propose a model of vibronic amplification of quantum transport of excitons in quantum photosynthesis. The model is based on the idea that vibrons can be regarded as a phononic analog of the laser mode. Then, to describe vibrons, we apply the semiclassical laser theory.

We discuss a variant of the semiclassical laser theory in which the equations of state for a medium are described by quantum dissipative dynamics in the Lindblad form. We use the dynamics given by the quantum stochastic limit method [1, 2]. To complete the model, we supplement the Lindblad equations for a medium with an equation for the laser mode from the semiclassical laser theory. This approach is a particular case of the standard semiclassical laser theory [8, 22].

We show that the generation of a coherent vibronic mode indeed increases the transfer rate. For the laser mechanism to operate, population inversion is necessary; moreover, the inversion should exceed some threshold. However, there exist models of lasing without inversion that involve so-called quantum dark states of the system, which interact with the laser mode (see [9, 10, 15, 22]).

Dark states attract attention in quantum optics and in applications to quantum informatics [5]. Quantum dark states have been investigated experimentally in photosynthetic systems [6, 7, 16]. Here we consider a modification of the laser model of vibrons in which quantum dark states are generated and vibrons are described by a model of lasing without inversion.

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In [3, 12–14, 24] the nonequilibrium quantum dissipative dynamics of the stochastic limit method for quantum theory was used to model quantum photosynthesis. Manipulations of quantum states and their applications to quantum computations and quantum control were considered in [14, 18, 19, 24]. In [17] the application of quantum methods to computations and biology was discussed. The relation between “global” and “local” approaches in the theory of open quantum systems was studied in [23].

The paper is organized as follows. In Section 2 we discuss a variant of the semiclassical laser theory for a three-level system interacting with a nonequilibrium environment. We compute the transfer rate in the stationary state, find a condition for the generation of a coherent “laser” mode, and show that the presence of the coherent vibronic mode amplifies the transfer rate. In Section 3 we examine a similar model for a four-level system, which is an analog of “laser without inversion” involving quantum dark states. We show that in this model it is easier to satisfy the condition for the generation of a coherent mode (vibrons). Section 4 contains concluding remarks.

2. LASER MECHANISM FOR VIBRONS

We consider quantum dissipative dynamics of a system with three energy levels \( \varepsilon_0 < \varepsilon_1 < \varepsilon_2 \) and the Hamiltonian

\[
H_S = \varepsilon_0 |0\rangle\langle 0| + \varepsilon_1 |1\rangle\langle 1| + \varepsilon_2 |2\rangle\langle 2|.
\]

The physical meaning is as follows: the system describes excitons in quantum photosynthesis in one-exciton approximation; the lower state \( |0\rangle \) is the state without excitons; the state \( |1\rangle \) describes an exciton in the reaction center; and the state \( |2\rangle \) describes an exciton on chromophore. Here \( \{|0\rangle, |1\rangle, |2\rangle\} \) is an orthonormal basis in the Hilbert space \( \mathcal{H}_S \) of the system.

The system interacts with a nonequilibrium environment given by three reservoirs (Bose fields in temperature states with different temperatures). The interaction of the system with light leads to creation of excitons; the interaction of excitons with phonons (vibrations of molecules) is related to the transfer of excitons to the reaction center; and the absorption of excitons in the reaction center is described by an additional sink field. The total Hamiltonian of the system and environment has the form

\[
H = H_S + H_{em} + H_{ph} + H_{sink} + \lambda (H_{1,em} + H_{1,ph} + H_{1,sink}).
\]

(2.2)

Here \( H_R, R = \text{em, ph, sink} \), are the Hamiltonians of the reservoirs (Bose fields) with dispersions \( \omega_R(k) \),

\[
H_R = \int_{\mathbb{R}^3} \omega_R(k)a_R^\dagger(k)a_R(k) \, dk,
\]

(2.3)

which act in Bose Hilbert spaces of the reservoirs \( \mathcal{H}_R \). The total Hamiltonian of the system and environment acts in the tensor product \( \mathcal{H}_S \otimes \mathcal{H}_{em} \otimes \mathcal{H}_{ph} \otimes \mathcal{H}_{sink} \) of the spaces of the system and reservoirs.

Each of the reservoirs is in a temperature state, a Gaussian mean-zero state with quadratic correlation function

\[
\langle a_R^\dagger(k)a_R(k') \rangle = N_R(k)\delta(k - k'),
\]

where \( N_R(k) \) has the form

\[
N_R(k) = \frac{1}{e^{\beta_R \omega_R(k)} - 1};
\]

(2.4)

here \( \beta_R \) is the inverse temperature of the reservoir.
The form of the interaction Hamiltonians \( H_{\text{em}}, H_{\text{ph}}, \) and \( H_{\text{sink}} \) is described below in formulas (2.5), (2.6), and (2.7), respectively, where \( g_R(k) \) are form factors of the interaction of the system with the reservoirs (complex-valued functions).

The interaction of the system with light is described by the interaction Hamiltonian

\[
H_{\text{em}} = A_{\text{em}}|2\rangle \langle 0| + A_{\text{em}}^*|0\rangle \langle 2|,
\]

\[
A_{\text{em}}^* = \int_{\mathbb{R}^3} g_{\text{em}}(k)a_{\text{em}}^*(k) \, dk.
\]  

(2.5)

The transport of excitons to the reaction center is related to the interaction with phonons:

\[
H_{\text{ph}} = A_{\text{ph}}|2\rangle \langle 1| + A_{\text{ph}}^*|1\rangle \langle 2|,
\]

\[
A_{\text{ph}}^* = \int_{\mathbb{R}^3} g_{\text{ph}}(k)a_{\text{ph}}^*(k) \, dk.
\]

(2.6)

The absorption of excitons in the reaction center is described by the interaction with the sink reservoir:

\[
H_{\text{sink}} = A_{\text{sink}}|1\rangle \langle 0| + A_{\text{sink}}^*|0\rangle \langle 1|,
\]

\[
A_{\text{sink}}^* = \int_{\mathbb{R}^3} g_{\text{sink}}(k)a_{\text{sink}}^*(k) \, dk.
\]

(2.7)

For the model under investigation, the dynamics of the reduced density matrix of the system interacting with three reservoirs is generated by the sum of four generators:

\[
\frac{d}{dt} \rho(t) = (n_{\text{em}}\theta_{\text{em}} + n_{\text{ph}}\theta_{\text{ph}} + i[\cdot, H_{\text{eff}}] + n_{\text{sink}}\theta_{\text{sink}})(\rho(t)),
\]

\[
\quad \text{where } \theta_R, R = \text{em}, \text{ph}, \text{sink}, \text{are quantum dissipative generators in the Lindblad form and } n_R \text{ is the total number of quanta for reservoir } R.
\]

The contribution \( i[\cdot, H_{\text{eff}}] \) with effective Hamiltonian

\[
H_{\text{eff}} = s(|2\rangle \langle 1| + |1\rangle \langle 2|), \quad s \in \mathbb{R},
\]

describes a vibronic mechanism (an analog of the interaction with the laser mode). Here \( s \) is the amplitude of the vibronic mode. The generators \( \theta_R \) are obtained using the procedure of the stochastic limit of quantum theory [2] (the Lamb shift is ignored) and have the following form.

The generation of excitons is described by the photonic generator, \( R = \text{em} \), the Bohr frequency equals \( \omega_{\text{em}} = \varepsilon_2 - \varepsilon_0 \), and the temperature is \( \beta_{\text{ph}}^{-1} = 6000 \text{ K} \):

\[
\theta_{\text{em}}(\rho) = 2\gamma_{\text{em}}^+(|2\rangle \langle 2|) - 2\gamma_{\text{em}}^-(|0\rangle \langle 2|) + 2\gamma_{\text{em}}^+(0\rangle |0\rangle \langle 2| - \frac{1}{2} |\rho\rangle \langle 2|)
\]

(2.9)

The transport of excitons is described by the phononic generator, \( R = \text{ph} \), the Bohr frequency equals \( \omega_{\text{ph}} = \varepsilon_2 - \varepsilon_1 \), and the temperature is \( \beta_{\text{ph}}^{-1} = 300 \text{ K} \):

\[
\theta_{\text{ph}}(\rho) = 2\gamma_{\text{ph}}^-|2\rangle \langle 1| - \frac{1}{2} \rho - \frac{1}{2} |\rho\rangle \langle 1|) + 2\gamma_{\text{ph}}^+|1\rangle \langle 2| - \frac{1}{2} |\rho\rangle \langle 2|)
\]

(2.10)

The absorption of excitons is described by the sink generator, \( R = \text{sink} \), the Bohr frequency equals \( \omega_{\text{sink}} = \varepsilon_1 - \varepsilon_0 \), and the temperature is \( \beta_{\text{sink}}^{-1} = 300 \text{ K} \):

\[
\theta_{\text{sink}}(\rho) = 2\gamma_{\text{sink}}^-|1\rangle \langle 0| - \frac{1}{2} \rho - \frac{1}{2} |\rho\rangle \langle 1|) + 2\gamma_{\text{sink}}^+|0\rangle \langle 1| - \frac{1}{2} |\rho\rangle \langle 0|)
\]

(2.11)
The coefficients $\gamma_R^\pm$ have the form
\[
\gamma_R^+ = \pi \int |g_R(k)|^2 \delta(\omega_R(k) - \omega_R)N_R(k) \, dk, \tag{2.12}
\]
\[
\gamma_R^- = \pi \int |g_R(k)|^2 \delta(\omega_R(k) - \omega_R)(N_R(k) + 1) \, dk. \tag{2.13}
\]

The value of $\gamma_R^+$ is the rate of induced transitions between the corresponding energy levels (pair of levels with energy difference $\omega_R$) due to interaction with the reservoir $R$, and the value of $\gamma_R^-$ is the sum of rates of spontaneous and induced transitions.

The rates of transitions (for the temperature state of the reservoir) satisfy the relation
\[
\frac{\gamma_R^+}{\gamma_R^-} = e^{-\beta_R \omega_R},
\]
where $\omega_R$ is the Bohr frequency of the transition and $\beta_R$ is the inverse temperature of the reservoir; i.e.,
\[
\frac{\gamma_{\text{em}}^+}{\gamma_{\text{em}}^-} = e^{-\beta_{\text{em}}(\varepsilon_2 - \varepsilon_0)}, \quad \frac{\gamma_{\text{ph}}^+}{\gamma_{\text{ph}}^-} = e^{-\beta_{\text{ph}}(\varepsilon_2 - \varepsilon_1)}, \quad \frac{\gamma_{\text{sink}}^+}{\gamma_{\text{sink}}^-} = e^{-\beta_{\text{sink}}(\varepsilon_1 - \varepsilon_0)}.
\]

**Remark 1.** In the above formulas, for the regime under study, it holds that $\beta_{\text{em}}$ is approximately 20 times less than $\beta_{\text{ph}}$, $\beta_{\text{ph}} \leq \beta_{\text{sink}}$, $\omega_{\text{em}}$ is approximately 5 or 10 times greater than $\omega_{\text{ph}}$, and $\omega_{\text{em}} = \omega_{\text{ph}} + \omega_{\text{sink}}$.

**Remark 2.** The coefficients $n_R$ in (2.8) describe the intensity of interaction of the system with the reservoirs $R$. The reservoirs are in temperature states. Light in a temperature state can be characterized not only by temperature but also by brightness: one can vary the brightness for the same temperature. The coefficient $n_R$ allows one to take this effect into account; its value describes the intensity of interaction with the reservoir. The coefficients $\gamma_R^\pm$ describe the spectral properties of the reservoir.

We will consider the dynamics of the system described by equation (2.8) in the space of density matrices (containing diagonal entries and one pair of off-diagonal entries) of the form
\[
\rho = \rho_{22}|2\rangle\langle 2| + \rho_{11}|1\rangle\langle 1| + \rho_{00}|0\rangle\langle 0| + \rho_{21}|2\rangle\langle 1| + \rho_{12}|1\rangle\langle 2|. \tag{2.14}
\]

The generator of the dynamics has the form
\[
L = n_{\text{em}}\theta_{\text{em}} + n_{\text{ph}}\theta_{\text{ph}} + i[\cdot, H_{\text{eff}}] + n_{\text{sink}}\theta_{\text{sink}}. \tag{2.15}
\]

The other off-diagonal entries of the density matrix will decay exponentially with this dynamics (i.e., we will have decoherence). The elements $\rho_{21}$ and $\rho_{12}$ can be generated due to interaction with the vibronic mode (contribution $i[\cdot, H_{\text{eff}}]$ to the generator) with nonzero $s$.

To describe the generation of the vibronic mode by transitions between the energy levels $|2\rangle$ and $|1\rangle$, we use the following equation of the semiclassical laser theory [8]:
\[
\frac{d}{dt}s = -\kappa s - i\rho_{21}. \tag{2.16}
\]

Here $\kappa$ is the rate of dissipation of the vibronic mode.

We will investigate the stationary solution for the dynamics ($L(\rho) = 0$) and the amplitude of the vibronic mode $s$ in the stationary state (using (2.16)). We will compute the transition rate of excitons to the sink and prove that in the presence of a nonzero vibronic mode the rate of transport of excitons increases. We propose to consider this effect as a model of a vibronic mechanism of amplification of exciton transport.
Claim. The stationary nonequilibrium state \((L(\rho) = 0)\) without vibrons \((s = 0)\) has the form

\[
\rho_{21} = \rho_{12} = 0,
\]

\[
\rho_{22} = \frac{n_{\text{em}} n_{\text{ph}} \gamma_{\text{em}}^+ \gamma_{\text{ph}}^+ + n_{\text{em}} n_{\text{sink}} \gamma_{\text{em}}^- \gamma_{\text{sink}}^- + n_{\text{ph}} n_{\text{sink}} \gamma_{\text{ph}}^- \gamma_{\text{sink}}^-}{\Delta}, \tag{2.17}
\]

\[
\rho_{11} = \frac{n_{\text{em}} n_{\text{ph}} \gamma_{\text{em}}^- \gamma_{\text{ph}}^- + n_{\text{em}} n_{\text{sink}} \gamma_{\text{em}}^+ \gamma_{\text{sink}}^+ + n_{\text{ph}} n_{\text{sink}} \gamma_{\text{ph}}^+ \gamma_{\text{sink}}^+}{\Delta}, \tag{2.18}
\]

\[
\rho_{00} = \frac{n_{\text{em}} n_{\text{ph}} \gamma_{\text{em}}^+ \gamma_{\text{ph}}^- + n_{\text{em}} n_{\text{sink}} \gamma_{\text{em}}^- \gamma_{\text{sink}}^+ + n_{\text{ph}} n_{\text{sink}} \gamma_{\text{ph}}^+ \gamma_{\text{sink}}^-}{\Delta}, \tag{2.19}
\]

where the normalization \(\Delta\) ensures that the trace of density matrix is equal to one:

\[
\Delta = n_{\text{em}} n_{\text{ph}} (\gamma_{\text{em}}^+ \gamma_{\text{ph}}^+ + \gamma_{\text{em}}^- \gamma_{\text{ph}}^- + \gamma_{\text{em}}^+ \gamma_{\text{ph}}^- + \gamma_{\text{em}}^- \gamma_{\text{ph}}^+) + n_{\text{ph}} n_{\text{sink}} (\gamma_{\text{ph}}^+ \gamma_{\text{sink}}^- + \gamma_{\text{ph}}^- \gamma_{\text{sink}}^+) + n_{\text{em}} n_{\text{sink}} (\gamma_{\text{em}}^+ \gamma_{\text{sink}}^- + \gamma_{\text{em}}^- \gamma_{\text{sink}}^+) + n_{\text{em}} n_{\text{sink}} (\gamma_{\text{em}}^- \gamma_{\text{sink}}^+ + \gamma_{\text{em}}^+ \gamma_{\text{sink}}^-). \tag{2.20}
\]

Proof. One can compute the stationary state \((L(\rho) = 0)\) in the absence of vibrons \((s = 0)\) as follows. Note that in this case the off-diagonal entries \(\rho_{21}\) and \(\rho_{12}\) in the stationary state are equal to zero (since for \(s = 0\) these entries are eigenvectors of the matrix \(L\) with negative eigenvalues).

In the diagonal subspace (of dimension 3) the matrix \(L\) is degenerate (this is a degenerate square matrix with the sum of lines equal to zero). Therefore, the solution of the system of linear equations \(L\rho = 0\) can be found if we take the entries \(\rho_{22}, \rho_{11},\) and \(\rho_{00}\) to be equal to the minors with alternating signs of the first line of \(L\) (in the basis \(\rho_{22}, \rho_{11}, \rho_{00}\)). The equation corresponding to the first line of the matrix then coincides with the expansion of the determinant of the matrix (which is zero) along the first line and is satisfied identically.

Let us consider the second equation of the system \(L\rho = 0\) and substitute into it the same set of variables. The resulting expression can be regarded as the expansion along the first line of the determinant of the matrix obtained from the matrix of the system by substituting the second line of the matrix for the first line. Since the obtained matrix has two identical lines, its determinant is equal to zero. Therefore, the second equation is satisfied for the described set of variables, and a similar statement holds for the other equations. Therefore, the set of minors with alternating signs for the first line of the matrix \(L\) gives a solution of the system of equations.

If the obtained solution is not equal to zero identically and the matrix \(L\) has corank 1 (which holds in the generic case), then we obtain a general form of the stationary state in the absence of vibrons \((s = 0)\). The normalization condition \(\rho_{22} + \rho_{11} + \rho_{00} = 1\) gives the solution (2.17)–(2.20) for the stationary state.

The transfer rate of excitons to the sink (in view of the expression for the sink generator) is given by

\[
F = n_{\text{sink}} \langle 0 | \theta_{\text{sink}}(\rho) | 0 \rangle = 2n_{\text{sink}} (\gamma_{\text{sink}}^- \rho_{11} - \gamma_{\text{sink}}^- \rho_{00}).
\]

For the flow of excitons to the sink in the above stationary state (2.17)–(2.20), we obtain (taking into account that \(\beta_{\text{ph}} = \beta_{\text{sink}}\))

\[
F = \frac{2n_{\text{em}} n_{\text{ph}} n_{\text{sink}} (\gamma_{\text{em}}^- \gamma_{\text{ph}}^- \gamma_{\text{sink}}^- - \gamma_{\text{em}}^+ \gamma_{\text{ph}}^+ \gamma_{\text{sink}}^+)}{\Delta}
\]

\[
= \frac{2n_{\text{em}} n_{\text{ph}} n_{\text{sink}} \gamma_{\text{em}}^- \gamma_{\text{ph}}^- \gamma_{\text{sink}}^+}{\Delta} (e^{(\beta_{\text{ph}} - \beta_{\text{em}})(\varepsilon_2 - \varepsilon_0)} - 1). \tag{2.21}
\]

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The dependence of the transfer rate $F$ on $n_R$ exhibits saturating behavior: for any reservoir $R$, when the number $n_R$ of quanta of the reservoir is small and the parameters $n_{R'}$ of the other reservoirs are fixed, the flow is proportional to $n_R$; and as $n_R \to \infty$ the flow saturates, i.e., tends to a constant.

The case of presence of vibrons. For the stationary state in the presence of vibrons, the equation $L(\rho) = 0$ implies

$$
\rho_{21} = -\frac{i s}{\mu} (\rho_{22} - \rho_{11}), \quad \rho_{12} = \frac{i s}{\mu} (\rho_{22} - \rho_{11}), \quad \mu = -n_{ph} (\gamma_{ph}^- + \gamma_{ph}^+) \;;
$$

i.e., the off-diagonal entries are proportional to the inversion $\rho_{22} - \rho_{11}$ (for some fixed $s$). Substituting these expressions into the system of equations for the stationary state yields

$$
\begin{pmatrix}
-2n_{em} \gamma_{em}^- - 2n_{ph} \gamma_{ph}^- + 2s^2 \mu & 2n_{ph} \gamma_{ph}^+ - 2s^2 \mu & 2n_{em} \gamma_{em}^+ \\
2n_{ph} \gamma_{ph}^- - 2s^2 \mu & -2n_{ph} \gamma_{ph}^+ - 2n_{sink} \gamma_{sink}^- + 2s^2 \mu & 2n_{sink} \gamma_{sink}^+ \\
2n_{em} \gamma_{em}^- & 2n_{sink} \gamma_{sink}^- & -2n_{em} \gamma_{em}^+ - 2n_{sink} \gamma_{sink}^+
\end{pmatrix}
\begin{pmatrix}
\rho_{22} \\
\rho_{11} \\
\rho_{00}
\end{pmatrix}
= 0.
$$

We obtain the following recipe for describing the stationary state: the diagonal entries of the stationary density matrix have the form (2.17)-(2.20), where the following transformations should be made:

$$
n_{ph} \gamma_{ph}^- \mapsto n_{ph} \gamma_{ph}^- - \frac{s^2}{\mu}, \quad n_{ph} \gamma_{ph}^+ \mapsto n_{ph} \gamma_{ph}^+ - \frac{s^2}{\mu} \;.
$$

The value of $\mu$ is negative; i.e., for nonzero $s$ this transformation increases the parameters $n_{ph} \gamma_{ph}^\pm$. For the inversion, equations (2.17) and (2.18) yield

$$
\rho_{22} - \rho_{11} = \frac{(n_{em} \gamma_{em}^+ + n_{sink} \gamma_{sink}^+) n_{ph} (\gamma_{ph}^+ - \gamma_{ph}^-) + n_{em} n_{sink} (\gamma_{em}^+ \gamma_{sink}^- - \gamma_{em}^+ \gamma_{sink}^+)}{\Delta(s)},
$$

where $\Delta(s)$ is obtained from (2.20) by the substitutions (2.22), i.e.,

$$
\Delta(s) = \Delta - \frac{s^2}{\mu} (n_{em} (2 \gamma_{em}^+ + \gamma_{em}^-) + n_{sink} (2 \gamma_{sink}^+ + \gamma_{sink}^-)),
$$

and the inversion decreases with $s$ (note that $\mu$ is negative, so $\Delta$ increases under this transformation).

Expression (2.23) for the inversion in the regime of irreversible absorption of excitons in the reaction center ($\gamma_{sink}^+ = 0$) takes the form

$$
\rho_{22} - \rho_{11} = \frac{n_{em} \gamma_{em}^+ n_{sink} \gamma_{sink}^- - n_{ph} (\gamma_{ph}^- - \gamma_{ph}^+))}{\Delta(s)};
$$

i.e., the inversion is positive if the rate $n_{sink} \gamma_{sink}^-$ of absorption of excitons in the reaction center is greater than the rate of transfer of excitons to the reaction center due to spontaneous transitions $n_{ph} (\gamma_{ph}^- - \gamma_{ph}^+)$.

The flow of excitons to the sink in the presence of vibrons takes the form

$$
F = \frac{2n_{em} n_{ph} n_{sink} (\gamma_{em}^+ \gamma_{sink}^- - \gamma_{em}^- \gamma_{sink}^+)}{\Delta(s)} - \frac{s^2 2n_{em} n_{sink} (\gamma_{em}^+ \gamma_{sink}^- - \gamma_{em}^- \gamma_{sink}^+)}{\Delta(s)}; \quad (2.26)
$$

i.e., the flow of excitons increases in the presence of vibrons. We propose to consider this effect as a description of the vibronic mechanism of amplification of the exciton transfer rate.
Remark 3. The flow $F$ increases with $s^2$ if the parameters of the system take realistic values (see Remark 1). In particular, for (2.26) we get the following necessary condition for the positivity of the second term: $\beta_{em}\omega_{em} < \beta_{sink}\omega_{sink}$. This condition is satisfied with approximately ten-fold redundancy. The derivative of $F$ with respect to $s^2$ is positive if

$$\left(\gamma^+_\text{em} - \gamma^-_{\text{em}}\right)\Delta > n_{\text{ph}}\left(\gamma^+_\text{em} \gamma^-_{\text{em}} - \gamma^-_{\text{em}} \gamma^+_\text{em}\right)\left(n_{\text{em}}\left(2\gamma^+_\text{em} + \gamma^-_{\text{em}}\right) + n_{\text{sink}}\left(2\gamma^+_\text{em} + \gamma^-_{\text{em}}\right)\right),$$

where $\Delta$ is given by (2.20).

The expression in parentheses on the left-hand side of the above inequality is positive for $\beta_{em}\omega_{em} < \beta_{sink}\omega_{sink}$. In the first parentheses on the right-hand side, we take the approximation $\gamma^+_{\text{ph}} = \gamma^-_{\text{ph}}$ (this approximation holds for the regime of small Bohr frequency $\omega_{ph}$, which is lower by an order of magnitude than $\omega_{sink}$). In this case one can factor out $\gamma^+_{\text{ph}}$ from the parentheses on the right-hand side, and the resulting inequality will be satisfied due to the form of $\Delta$.

The laser generation is known to exhibit threshold behavior; i.e., in the model under consideration, the vibronic mechanism of transport amplification operates only for sufficiently large inversion. Let us consider the equation of generation of vibrons (2.16) in the stationary state:

$$\gamma s = -i\rho_{21} = -\frac{s}{\mu}(\rho_{22} - \rho_{11}).$$

For all values of the parameters we have the solution $s = 0$ (without vibronic mode). A necessary condition for the existence of a solution with nonzero $s$ is the positivity of the inversion $\rho_{22} - \rho_{11}$ (since $\mu$ is negative). For a solution with nonzero vibronic mode, we find the squared amplitude of the vibronic mode to be

$$s^2 = \frac{n_{\text{em}}\gamma^+_\text{em} + n_{\text{sink}}\gamma^-_{\text{sink}}}{\Delta(n_{\text{em}}(2\gamma^+_{\text{em}} + \gamma^-_{\text{em}}) + n_{\text{sink}}(2\gamma^-_{\text{em}} + \gamma^-_{\text{sink}}))}(\gamma^+_{\text{em}} - \gamma^+_{\text{em}} - \gamma^-_{\text{em}} - \gamma^-_{\text{em}}) - \Delta n_{\text{ph}}(\gamma^-_{\text{ph}} + \gamma^+_{\text{ph}})\gamma\frac{s^2}{\mu}.$$

Here $\Delta$ is given by (2.20) (i.e., it corresponds to the case without vibrons). The positivity of the numerator of the above expression is sufficient (in our model) for the existence of a vibronic mode.

The positivity of (2.27) can be achieved only for sufficiently small $\gamma$ (when the numerator is positive); moreover, the smaller the $\gamma$ (dissipation of vibrons), the greater the absolute value of $s$ (vibronic mode) that satisfies the equation for the stationary state. As discussed above, when $s$ increases, the flow of excitons $F$ also increases and tends to a constant value determined by the level of light and the rate of absorption of excitons in the reaction center.

In the approximation of irreversible absorption of excitons in the reaction center ($\gamma^-_{\text{sink}} = 0$), for the squared amplitude of the vibronic mode (2.27) we obtain

$$s^2 = \frac{n_{\text{em}}\gamma^+_\text{em} - n_{\text{ph}}(\gamma^-_{\text{ph}} + \gamma^+_{\text{ph}}) - \Delta n_{\text{ph}}(\gamma^-_{\text{ph}} + \gamma^+_{\text{ph}})\gamma}{\Delta(n_{\text{em}}(2\gamma^+_{\text{em}} + \gamma^-_{\text{em}}) + n_{\text{sink}}\gamma^-_{\text{sink}})}.$$

and for the flow of excitons to the sink (2.26) we get

$$F = \frac{2n_{\text{em}}n_{\text{sink}}\gamma^+_\text{em} \gamma^-_{\text{sink}}}{\Delta(s)}\left(n_{\text{ph}}\gamma^-_{\text{ph}} - \frac{s^2}{\mu}\right),$$

where $s^2 \geq 0$ and $\mu < 0$.

Therefore, in the presence of vibrons the transfer rate of excitons to the sink increases (due to the presence of the positive contribution $-s^2/\mu$ in $n_{\text{ph}}\gamma^-_{\text{ph}} - s^2/\mu$).
3. VIBRONIC LASER WITHOUT INVERSION

For the model described in the previous section, it is difficult to generate sufficiently large inversion to overcome the laser (vibronic) generation threshold (2.27), which is possible only for small dissipation $\varkappa$. For small inversion, the vibronic mechanism of amplification of the exciton transfer rate ceases to work. To solve this problem, we can use the well-known mechanism of inversion-free laser (see [9, 10, 15, 22]). Lasers of this kind involve the so-called quantum dark states. For an inversion-free laser, it is sufficient to create inversion in a subspace where the quantum transport operates (rather than in the total space of density matrices, which includes dark states).

Let us discuss the following modification of the model introduced in the previous section. In this model the second (middle) energy level is almost degenerate. In particular, this level may correspond to two interacting molecules (note that the photosynthetic reaction center contains a special pair of chlorophylls). In this case the energy level splits: instead of the state $|1\rangle$ the middle energy level now contains two states $|1\rangle$ and $|1'\rangle$, and the following symmetric and antisymmetric states are eigenvectors of the Hamiltonian of the system:

$$|S\rangle = \frac{1}{\sqrt{2}}(|1\rangle + |1'\rangle), \quad |A\rangle = \frac{1}{\sqrt{2}}(|1\rangle - |1'\rangle).$$

The Hamiltonian of the system takes the form

$$H_S = \varepsilon_0 |0\rangle\langle 0| + \varepsilon_S |S\rangle\langle S| + \varepsilon_A |A\rangle\langle A| + \varepsilon_2 |2\rangle\langle 2|, \quad \varepsilon_0 < \varepsilon_S < \varepsilon_A < \varepsilon_2. \quad (3.1)$$

We assume for simplicity that the energy difference between $\varepsilon_S$ and $\varepsilon_A$ is small compared with the other Bohr frequencies in the system.

We also assume that (due to some additional interactions) there is an exchange of population between the symmetric and antisymmetric states. This process can be described using the following additional generator of the dynamics of the density matrix of the system:

$$\theta_{\text{ex}}(\rho) = 2\gamma_{\text{ex}} \left( |A\rangle\langle A| |S\rangle\langle S| - \frac{1}{2} |A\rangle\langle A| \right) + 2\gamma_{\text{ex}} \left( |S\rangle\langle S| |A\rangle\langle A| - \frac{1}{2} |S\rangle\langle S| \right). \quad (3.2)$$

Here we suppose for simplicity that the coefficients $\gamma_{\text{ex}}$ of the two terms in the generator can be assumed to be equal.

Thus, the total generator of the dynamics of the density matrix of the system has the form

$$L = n_{\text{em}} \theta_{\text{em}} + n_{\text{ph}} \theta_{\text{ph}} + i[\cdot, H_{\text{eff}}] + n_{\text{ex}} \theta_{\text{ex}} + n_{\text{sink}} \theta_{\text{sink}}. \quad (3.3)$$

Here the generator $\theta_{\text{em}}$ has the same form (2.9) as in the previous section, while the generators $\theta_{\text{ph}}$, $i[\cdot, H_{\text{eff}}]$, and $\theta_{\text{sink}}$ are modified as follows.

The sink generator (2.11) (which describes the absorption of excitons in the reaction center) is replaced by a sum of two terms that contain $|S\rangle$ and $|A\rangle$, respectively, instead of $|1\rangle$ (i.e., excitons are absorbed in both states $|S\rangle$ and $|A\rangle$ and the coefficients $\gamma_{\text{sink}}^\pm$ for the symmetric and antisymmetric states are taken to be equal):

$$\theta_{\text{sink}}(\rho) = 2\gamma_{\text{sink}}^- \left( |S\rangle\langle S| |0\rangle\langle 0| - \frac{1}{2} |A\rangle\langle A| \right) + 2\gamma_{\text{sink}}^+ \left( |0\rangle\langle 0| |S\rangle\langle S| - \frac{1}{2} |A\rangle\langle A| \right)$$

$$+ 2\gamma_{\text{sink}}^- \left( |A\rangle\langle A| |0\rangle\langle 0| - \frac{1}{2} |A\rangle\langle A| \right) + 2\gamma_{\text{sink}}^+ \left( |0\rangle\langle 0| |A\rangle\langle A| - \frac{1}{2} |A\rangle\langle A| \right). \quad (3.4)$$

The generator (2.10) is replaced by a generator of the form

$$\theta_{\text{ph}}(\rho) = 2\gamma_{\text{ph}}^- \left( |2\rangle\langle 2| |S\rangle\langle S| - \frac{1}{2} |A\rangle\langle A| \right) + 2\gamma_{\text{ph}}^+ \left( |S\rangle\langle S| |2\rangle\langle 2| - \frac{1}{2} |A\rangle\langle A| \right). \quad (3.5)$$
i.e., the antisymmetric state \( |A \rangle \) is not connected by a transition to the upper level (since the amplitudes of transitions between the upper level and the states \( |1 \rangle \) and \( |1' \rangle \) are equal, the amplitudes cancel each other out for the antisymmetric state). Hence the state \( |A \rangle \) will be a dark state.

The vibronic “laser” is generated by the contribution

\[
H_{\text{eff}} = s(2|S\rangle\langle S| + |S\rangle\langle 2|), \quad s \in \mathbb{R},
\]

with the following equation of generation of the coherent mode:

\[
\frac{d}{dt}s = -\kappa s - i\rho_{2S}.
\]

We will study the dynamics of the system in the space of density matrices that contain diagonal entries and one pair of off-diagonal entries (the other entries will decay exponentially due to decoherence):

\[
\rho = \rho_{22}|2\rangle\langle 2| + \rho_{SS}|S\rangle\langle S| + \rho_{AA}|A\rangle\langle A| + \rho_{00}|0\rangle\langle 0| + \rho_{2S}|2\rangle\langle S| + \rho_{S2}|S\rangle\langle 2|.
\]

(3.6)

**Stationary nonequilibrium state.** In the stationary state \((L(\rho) = 0 \text{ with } L \text{ given by (3.3)), the diagonal entries of the density matrix in the case without vibrons are computed in the same way as in the previous section, i.e., as the minors with alternating signs for the first line of the matrix \(L\) in the subspace of diagonal density matrices of the form (3.6)).

In the presence of vibrons, the off-diagonal entries \(\rho_{2S}\) and \(\rho_{S2}\) of the density matrix in the stationary state are proportional to the inversion \(\rho_{22} - \rho_{SS}\) (difference of populations of the upper level and the symmetric state):

\[
\rho_{2S} = -\frac{i\kappa}{\mu}(\rho_{22} - \rho_{SS}), \quad \rho_{S2} = \frac{i\kappa}{\mu}(\rho_{22} - \rho_{SS}), \quad \mu = -n_{\text{ph}}(\gamma_{\text{ph}}^{-} + \gamma_{\text{ph}}^{+}).
\]

Therefore, as in the previous section, the introduction of vibrons reduces to the following transformation of transition rates for the diagonal entries of the stationary density matrix:

\[
n_{\text{ph}}\gamma_{\text{ph}}^{\pm} \mapsto n_{\text{ph}}\gamma_{\text{ph}}^{\pm} - \frac{s^2}{\mu},
\]

and the off-diagonal entries are given by the formulas presented above.

The flow of excitons to the sink equals (in view of (3.4))

\[
F = 2n_{\text{sink}}(\gamma_{\text{sink}}^{-}(\rho_{SS} + \rho_{AA}) - 2\gamma_{\text{sink}}^{+}n_{\text{sink}})
= \frac{8}{\Delta(s)}(4n_{\text{ex}}\gamma_{\text{ex}} + 2n_{\text{sink}}\gamma_{\text{sink}}^{-})n_{\text{em}}n_{\text{sink}}\left[-\left(n_{\text{ph}}\gamma_{\text{ph}}^{-} - \frac{s^2}{\mu}\right)\gamma_{\text{em}}\gamma_{\text{sink}}^{-} + \left(n_{\text{ph}}\gamma_{\text{ph}}^{+} - \frac{s^2}{\mu}\right)\gamma_{\text{em}}\gamma_{\text{sink}}^{+}\right],
\]

where \(\Delta(s)\) is the normalization that makes the trace of the density matrix equal to one.

This normalization has the form

\[
\Delta(s) = \left(2n_{\text{ph}}\gamma_{\text{ph}}^{+} - \frac{s^2}{\mu}\right)\left(2n_{\text{sink}}\gamma_{\text{sink}}^{+}(-2n_{\text{em}}\gamma_{\text{em}}^{-} + 2n_{\text{sink}}\gamma_{\text{sink}}^{-})
- (-2n_{\text{ex}}\gamma_{\text{ex}} - 2n_{\text{sink}}\gamma_{\text{sink}}^{-})(-2n_{\text{em}}\gamma_{\text{em}}^{-} - 2n_{\text{em}}\gamma_{\text{em}}^{+} - 4n_{\text{sink}}\gamma_{\text{sink}}^{+})
+ (4n_{\text{ex}}\gamma_{\text{ex}} + 2n_{\text{sink}}\gamma_{\text{sink}}^{-})\left((-2n_{\text{em}}\gamma_{\text{em}}^{-} - 2n_{\text{ph}}\gamma_{\text{ph}}^{-} + \frac{s^2}{\mu}\right)
\times (2n_{\text{sink}}\gamma_{\text{sink}}^{-} + 2n_{\text{em}}\gamma_{\text{em}}^{+} + 4n_{\text{sink}}\gamma_{\text{sink}}^{+}) - 2n_{\text{em}}\gamma_{\text{em}}^{+}(-2n_{\text{em}}\gamma_{\text{em}}^{-} + 2n_{\text{sink}}\gamma_{\text{sink}}^{-})\right).
\]

(note that \(\Delta(s)\) in the present section is negative; see formula (3.8) below for a particular case).
As in the previous section, the values of the off-diagonal entries and the amplitude of the vibronic mode for the model under consideration are related to the inversion \( \rho_{22} - \rho_{SS} \). In particular, vibronic generation occurs only when there exists a nonzero solution of the equation

\[
x s = -\frac{8}{\mu}(\rho_{22} - \rho_{SS}).
\]

In the stationary state the inversion has the form

\[
\rho_{22} - \rho_{SS} = \frac{1}{\Delta(s)} [(2n_{em}\gamma_{em}^+ + 2n_{ph}(\gamma_{ph}^- - \gamma_{ph}^+)) \times (2n_{ex}\gamma_{ex}^- + 2n_{sink}(\gamma_{sink}^+ - 4n_{sink}\gamma_{sink}^- - 4n_{sink}\gamma_{sink}^- - 2n_{em}\gamma_{em}^-) (2n_{ex}\gamma_{ex}^- + 2n_{sink}\gamma_{sink}^-) (2n_{ex}\gamma_{ex}^- + 2n_{sink}\gamma_{sink}^-)].
\]

To simplify this expression, let us study the regime where the absorption of excitons in the reaction center is irreversible (\( \gamma_{sink}^+ = 0 \)) and assume additionally that the rate of exchange between the symmetric and antisymmetric states \( n_{ex}\gamma_{ex} \) is much greater than the rate of absorption of excitons in the reaction center \( n_{sink}\gamma_{sink}^- \). We obtain (recall that in this section \( \Delta(s) < 0 \))

\[
\rho_{22} - \rho_{SS} = -\frac{8n_{em}n_{em}n_{ex}^\prime}{\Delta(s)} (n_{ph}(\gamma_{ph}^+ - \gamma_{ph}^-) + 2n_{sink}\gamma_{sink}^-). \tag{3.7}
\]

It is worth comparing this formula with a similar formula (2.25) for the inversion in the model of the previous section (without the degeneracy of the middle level) in the considered approximation:

\[
\rho_{22} - \rho_{11} = \frac{n_{em}\gamma_{em}^+(n_{ph}(\gamma_{ph}^+ - \gamma_{ph}^-) + n_{sink}\gamma_{sink}^-)}{\Delta(s)}
\]

(one should keep in mind that the normalizations \( \Delta(s) \) in this formula and formula (3.7) above are different).

It is easy to see that in (3.7) (where the system contains dark states \( |A| \)) we get an additional positive contribution for the inversion (given by the coefficient 2 of \( n_{sink}\gamma_{sink}^- \); recall that \( \gamma_{ph}^+ - \gamma_{ph}^- < 0 \)). This makes it easier to achieve positive inversion and the occurrence of vibronic generation using the scheme analogous to the one described in the previous section. Therefore, in the regime of small \( n_{sink}\gamma_{sink}^- \) and large \( n_{ex}\gamma_{ex} \), the effect of the presence of dark states reduces to doubling the positive contribution to the inversion (cf. (2.25) and (3.7)).

In the regime of \( \gamma_{sink}^+ = 0 \) and \( n_{ex}\gamma_{ex} \gg n_{sink}\gamma_{sink}^- \), for the transfer rate of excitons to the sink and for the normalization we obtain the expressions

\[
F = -\frac{32}{\Delta(s)} n_{ex}n_{em}n_{sink}\gamma_{ex}\gamma_{em}^+\gamma_{sink}^-(n_{ph}\gamma_{ph}^- - \frac{s^2}{\mu}),
\]

\[
\Delta(s) = -8n_{ex} \left[ (n_{ph}\gamma_{ph}^- - \frac{s^2}{\mu}) n_{em}(\gamma_{em}^- + \gamma_{em}^+) + (n_{ph}\gamma_{ph}^- - \frac{s^2}{\mu})(2n_{sink}\gamma_{sink}^- + 2n_{em}\gamma_{em}^+) \right]
\]

\[
+ 2n_{sink}\gamma_{sink}^- n_{em}(\gamma_{em}^- + \gamma_{em}^+). \tag{3.8}
\]

Thus, we arrive at the following conclusion, which is similar to the results of the previous section: In the presence of vibrons, the transfer rate of excitons to the sink increases, but in the model with dark states the positive contribution to the inversion is twice greater than in the model without dark states. Therefore, it is easier to satisfy the condition for the existence of a nonzero vibronic mode.
4. CONCLUSIONS

The vibronic mechanism of amplification of quantum transport of excitons has been widely discussed in the studies of quantum photosynthesis [6, 7, 11, 16, 20]. Vibrons are collective electronic–vibrational excitations which are observed experimentally in photosynthetic systems. Unlike phonons, vibrons are some modes that are in resonance with some transitions for electronic states.

In the present paper we propose a model describing the vibronic mechanism of amplification of quantum transport by analogy with the semiclassical model of lasing. In this approach vibrons are described by the coherent “laser” mode (i.e., by phononic laser). In the presence of the coherent mode, the transition rate in the system (flow of excitons) increases.

Our approach is based on the semiclassical laser model in which the dissipative dynamics of quantum states of molecules is described by equations in the Lindblad form obtained by the quantum stochastic limit method. We have examined two models (two variants of the model): a model with a three-level system and a model of “laser without inversion” with a four-level system (the laser without inversion involves quantum dark states).

In Section 2 we showed that in the presence of the coherent vibronic mode the rate of transport of excitons increases. We computed the dependence of the transfer rate of excitons on the parameters of the model and found a condition for the generation of the vibronic mode. In particular, the population inversion is necessary for the generation of the vibronic mode (the upper level should be more populated than the lower one).

In Section 3 we considered a vibronic analog of the “laser without inversion.” The effect of the inversion-free laser weakens the conditions necessary for the generation of the coherent mode and for the vibronic amplification of quantum transport. For inversion-free lasers, the system should contain quantum “dark” states. Dark states in quantum photosynthesis have been discussed in the literature [6, 7, 16], which gives an argument in favor of the approach proposed in the present paper.

FUNDING

This work is supported by the Russian Science Foundation under grant 17-71-20154.

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*This article was submitted by the author simultaneously in Russian and English*