Supplementary Information: On thermodynamic inconsistencies in several photosynthetic and solar cell models and how to fix them

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I. Energy conversion models

We derive the evolution equations for some examples of two types of energy conversion models. The results of this section are used to generate Figure 2 in the main text, as well as Eq. 3.

IA. Donor-acceptor models

As examples of these models, we analyze below two particular donor-acceptor models that use a decay transfer scheme. This kind of analysis may be expanded to models that include coherent vibronic evolution such as proposed in reference [1].

1) We consider the biological quantum heat engine model proposed in [2] (see in particular Eqs. S34-S37 in reference [2]). It consists of a four level system coupled to a hot bath, a cold bath, and to the reaction center/circuit (also termed "the load"). $T_{h(c)}$ is the hot (cold) bath temperature. The different decay rates are shown in Figure S1a. The equations of motion are

$$\begin{gathered}\dot{\rho}_{aa} = -\gamma_c\left[\left(1+\bar{n}_c\right)\rho_{aa} - \bar{n}_c\rho_{\alpha\alpha}\right] - \gamma_h\left[\left(1+\bar{n}_h\right)\rho_{aa} - \bar{n}_h\rho_{bb}\right],\\ \dot{\rho}_{\alpha\alpha} = \gamma_c\left[\left(1+\bar{n}_c\right)\rho_{aa} - \bar{n}_c\rho_{\alpha\alpha}\right] - \Gamma\rho_{\alpha\alpha},\\ \dot{\rho}_{bb} = \gamma_h\left[\left(1+\bar{n}_h\right)\rho_{aa} - \bar{n}_h\rho_{bb}\right] + \Gamma_c\left[\left(1+\bar{N}_c\right)\rho_{\beta\beta} - \bar{N}_c\rho_{bb}\right],\\ \rho_{aa}+\rho_{bb}+\rho_{\alpha\alpha}+\rho_{\beta\beta}=1,\end{gathered} \tag{S1}$$

where we have kept the original paper notation. ρ_{ii} is the level population of state i and $\bar{n}_i$ or $\bar{N}_i$ are the relevant i- bath mode population. For details on the derivation of Eq. S1, we refer the reader to the original paper [2]. The free Hamiltonian of the four level system is

$$H = \sum_{i\in\{a,b,\alpha,\beta\}} \omega_i |i\rangle\langle i|. \tag{S2}$$

The steady state populations are

$$\frac{\rho_{aa}^{ss}}{\rho_{\alpha\alpha}^{ss}} = \frac{\gamma_c\bar{n}_c+\Gamma}{\gamma_c(\bar{n}_c+1)}, \tag{S3}$$

$$\frac{\rho_{bb}^{ss}}{\rho_{\alpha\alpha}^{ss}} = \frac{\Gamma\left[\gamma_c\left(\bar{n}_c+1\right)+\gamma_h\left(\bar{n}_h+1\right)\right]+\gamma_h\gamma_c\bar{n}_c\left(1+\bar{n}_h\right)}{\gamma_h\bar{n}_h\gamma_c\left(\bar{n}_c+1\right)}, \tag{S4}$$

$$\frac{\rho_{aa}^{ss}}{\rho_{bb}^{ss}} = \frac{\left(\gamma_c\bar{n}_c+\Gamma\right)\gamma_h\bar{n}_h}{\Gamma\left\{\gamma_c\left(\bar{n}_c+1\right)+\gamma_h\left(\bar{n}_h+1\right)\right\}+\gamma_h\gamma_c\bar{n}_c\left(1+\bar{n}_h\right)}, \tag{S5}$$

$$\frac{\rho_{\beta\beta}^{ss}}{\rho_{bb}^{ss}} = e^{-\hbar(\omega_\beta-\omega_b)/k_BT_c} + \frac{\gamma_h\bar{n}_h\Gamma\gamma_c\left(\bar{n}_c+1\right)}{\left(1+\bar{N}_c\right)\Gamma_c\left[\Gamma\left\{\gamma_c\left(\bar{n}_c+1\right)+\gamma_h\left(\bar{n}_h+1\right)\right\}+\gamma_h\gamma_c\bar{n}_c\left(1+\bar{n}_h\right)\right]}. \tag{S6}$$

The evolution induced by the hot bath can be obtained from Eq (S1) by setting $\gamma_c=\Gamma=\Gamma_c=0$. The evolution induced by the cold bath is obtained by setting in the same equations $\gamma_h=\Gamma=0$. The heat currents at steady state are (see Eq.2 in the main text) obtained using the induced evolution of each bath and the Hamiltonian (S2),

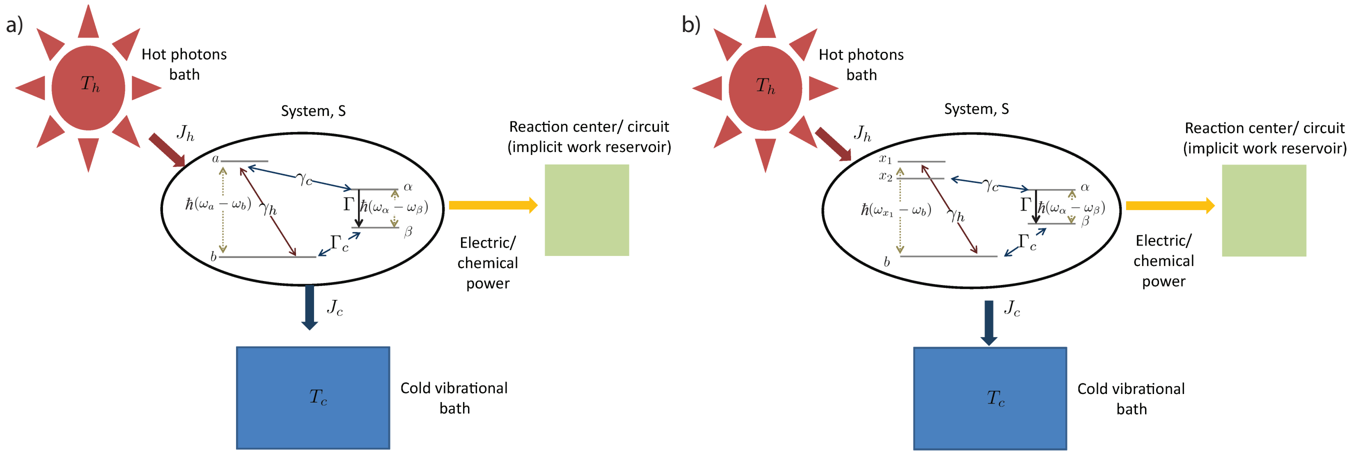

Figure S1: Donor-acceptor models: a) Biological quantum heat engine model from [2]; b) Photocell model proposed in reference [4].

$$J_h = \hbar(\omega_a-\omega_b)\left(1+\bar{n}_h\right)\gamma_h\rho^{ss}_{bb}\left(e^{-\hbar(\omega_a-\omega_b)/k_BT_h}-\frac{\rho^{ss}_{aa}}{\rho^{ss}_{bb}}\right) =$$

$$\frac{\hbar\left(\omega_a-\omega_b\right)\left(1+\bar{n}_h\right)\gamma_h\rho^{ss}_{bb}}{\Gamma\left[\gamma_c\left(\bar{n}_c+1\right)+\gamma_h\left(\bar{n}_h+1\right)\right]+\gamma_h\gamma_c\bar{n}_c\left(1+\bar{n}_h\right)}\left(e^{-\hbar(\omega_a-\omega_b)/k_BT_h}\Gamma\gamma_c(\bar{n}_c+1)\right), \tag{S7}$$

$$J_c = \hbar\left(\omega_a-\omega_\alpha\right)\left(1+\bar{n}_c\right)\gamma_c\rho^{ss}_{\alpha\alpha}\left(e^{-\hbar(\omega_a-\omega_\alpha)/k_BT_c}-\frac{\rho^{ss}_{aa}}{\rho^{ss}_{\alpha\alpha}}\right)+\hbar\left(\omega_\beta-\omega_b\right)\left(1+\bar{N}_c\right)\Gamma_c\rho^{ss}_{bb}\left(e^{-\hbar(\omega_\beta-\omega_b)/k_BT_c}-\frac{\rho^{ss}_{\beta\beta}}{\rho^{ss}_{bb}}\right) =$$

$$-\hbar\left(\omega_a-\omega_\alpha\right)\rho^{ss}_{\alpha\alpha}\Gamma-\hbar\left(\omega_\beta-\omega_b\right)\Gamma_c\left(1+\bar{N}_c\right)\rho^{ss}_{bb}\left(\frac{\gamma_h\bar{n}_h\Gamma\gamma_c\left(\bar{n}_c+1\right)}{\left(1+\bar{N}_c\right)\Gamma_c\left[\Gamma\left\{\gamma_c\left(\bar{n}_c+1\right)+\gamma_h\left(\bar{n}_h+1\right)\right\}+\gamma_h\gamma_c\bar{n}_c\left(1+\bar{n}_h\right)\right]}\right) =$$

$$-\frac{\rho^{ss}_{bb}\gamma_h\bar{n}_h\gamma_c\left(\bar{n}_c+1\right)\Gamma}{\Gamma\left[\gamma_c\left(\bar{n}_c+1\right)+\gamma_h\left(\bar{n}_h+1\right)\right]+\gamma_h\gamma_c\bar{n}_c\left(1+\bar{n}_h\right)}\hbar\left[\omega_a-\omega_\alpha+\omega_\beta-\omega_b\right], \tag{S8}$$

$$-\frac{J_c}{J_h} = \frac{\omega_a-\omega_\alpha+\omega_\beta-\omega_b}{\omega_a-\omega_b} = 1+\frac{\omega_\beta-\omega_\alpha}{\omega_a-\omega_b}, \tag{S9}$$

where $\hbar\omega_a-\hbar\omega_b$ $(\hbar\omega_\alpha-\hbar\omega_\beta)$ is the energy of the absorbed (emitted) quanta from the hot bath (to the RC/circuit). Therefore they are equivalent to $\hbar\omega_{abs}(\hbar\omega_{rc})$. Using this paper notation,

$$\begin{aligned} J_h \to J_{abs}, &\quad J_c \to J_{loss}, \\ \hbar\omega_\beta-\hbar\omega_\alpha \to -\hbar\omega_{rc}, &\quad \hbar\omega_a-\hbar\omega_b \to \hbar\omega_{abs}, \\ T_h \to T_{abs}, &\quad T_c \to T_{loss}, \end{aligned}$$

we obtain Eq. 3 in the main text. A similar analysis can be done for the coherence-assisted biological quantum heat engine model proposed also in the same paper and to the model proposed in reference [3].

2) We consider the photocell model proposed in reference [4]. It consists of a five level system coupled to a hot bath, a cold bath and to the reaction center/circuit (also termed "the load"). $T_{h(c)}$ is the hot (cold) bath temperature. The decay rates are shown in Figure S1b. For the sake of simplicity we assume there is no acceptor-to-donor recombination ($\chi = 0$, in the original paper notation). The equations of motion are

$$\begin{aligned}
\dot{\rho}_{\alpha\alpha} &= \gamma_c \left[(1+n_{2c})\, \rho_{x2x2} - n_{2c}\rho_{\alpha\alpha} \right] - \Gamma \rho_{\alpha\alpha}, \\
\dot{\rho}_{x2x2} &= \gamma_x \left[(1+n_x)\rho_{x1x1} - n_x \rho_{x2x2} \right] - \gamma_c \left[(1+n_{2c})\rho_{x2x2} - n_{2c}\rho_{\alpha\alpha} \right], \\
\dot{\rho}_{bb} &= - \left[\gamma_h n_h + \Gamma_c N_c \right] \rho_{bb} + \gamma_h \left(n_h + 1 \right) \rho_{x1x1} + \Gamma_c (N_c+1) \rho_{\beta\beta}, \\
\dot{\rho}_{x1x1} &= -\gamma_x \left[(1+n_x)\rho_{x1x1} - n_x \rho_{x2x2} \right] - \gamma_h \left[(1+n_h)\rho_{x1x1} - n_h \rho_{bb} \right], \\
& \rho_{x1x1} + \rho_{x2x2} + \rho_{bb} + \rho_{\alpha\alpha} + \rho_{\beta\beta} = 1.
\end{aligned} \tag{S10}$$

where we have kept the original paper notation. ρ_{ii} is the level population of state i and n_i or N_i are the relevant i- bath mode population. For details on the derivation of Eq. S10, we refer the reader to the original paper [4].

The free Hamiltonian of the five level system is

$$H = \sum_{i \in \{x_1, x_2, b, \alpha, \beta\}} \omega_i |i\rangle\langle i|. \tag{S11}$$

The steady state populations are

$$\frac{\rho^{ss}_{x2x2}}{\rho^{ss}_{\alpha\alpha}} = \frac{\Gamma + \gamma_c n_{2c}}{\gamma_c \left(1+n_{2c}\right)}, \tag{S12}$$

$$\frac{\rho^{ss}_{x1x1}}{\rho^{ss}_{x2x2}} = \frac{\gamma_x n_x + \gamma c \left(1+n_{2c}\right) - \gamma_c n_{2c} \frac{\gamma_c (1+n_{2c})}{\Gamma + \gamma_c n_{2c}}}{\gamma_x \left(1+n_x\right)} = \frac{\gamma_x n_x \left(\Gamma + \gamma_c n_{2c}\right) + \gamma c \left(1+n_{2c}\right) \Gamma}{\gamma_x \left(1+n_x\right) \left(\Gamma + \gamma_c n_{2c}\right)}, \tag{S13}$$

$$\frac{\rho^{ss}_{x1x1}}{\rho^{ss}_{bb}} = \frac{\gamma_h n_h \left[\gamma_x n_x \left(\Gamma + \gamma_c n_{2c}\right) + \gamma_c (1+n_{2c}) \Gamma \right]}{\gamma_x \gamma_c \Gamma \left(1+n_x\right) \left(1+n_{2c}\right) + \gamma_h \left(1+n_h\right) \left[\gamma_x n_x \left(\Gamma + \gamma_c n_{2c}\right) + \gamma_c (1+n_{2c}) \Gamma \right]}, \tag{S14}$$

$$\frac{\rho^{ss}_{\beta\beta}}{\rho^{ss}_{bb}} = e^{-\hbar(\omega_\beta - \omega_b)/k_B T_c} + \frac{\gamma_h n_h \gamma_x \gamma_c \Gamma \left(1+n_x\right) \left(1+n_{2c}\right)}{\Gamma_c \left(1+N_c\right) \left\{ \gamma_x \gamma_c \Gamma \left(1+n_x\right) \left(1+n_{2c}\right) + \gamma_h \left(1+n_h\right) \left[\gamma_x n_x \left(\Gamma + \gamma_c n_{2c}\right) + \gamma_c \left(1+n_{2c}\right) \Gamma \right] \right\}}. \tag{S15}$$

The evolution induced by the hot bath can be obtained from Eq (S10) by setting $\gamma_c = \Gamma = \Gamma_c = \gamma_x = 0$. The evolution induced by the cold bath is obtained by setting in the same equations $\gamma_h = \Gamma = 0$. The heat currents at steady state are (see Eq.2 in the main text) obtained using the induced evolution of each bath and the Hamiltonian (S11),

$$\begin{aligned}
J_h = \hbar \left(\omega_{x1} - \omega_b\right) \left(1 + \bar{n}_h\right) \gamma_h \rho^{ss}_{bb} \left(e^{-\hbar(\omega_{x1} - \omega_b)/k_B T_h} - \frac{\rho^{ss}_{x1x1}}{\rho^{ss}_{bb}} \right) = \\
\hbar \left(\omega_{x1} - \omega_b\right) \frac{\rho^{ss}_{bb} \gamma_h n_h \gamma_x \gamma_c \Gamma \left(1+n_x\right) \left(1+n_{2c}\right)}{\gamma_x \gamma_c \Gamma \left(1+n_x\right) \left(1+n_{2c}\right) + \gamma_h \left(1+n_h\right) \left[\gamma_x n_x \left(\Gamma + \gamma_c n_{2c}\right) + \gamma_c \left(1+n_{2c}\right) \Gamma \right]},
\end{aligned} \tag{S16}$$

$$\begin{aligned}
J_c = \hbar \left(\omega_{x1} - \omega_{x2}\right) \left(1 + \bar{n}_x\right) \gamma_x \rho^{ss}_{x2x2} \left(e^{-\hbar(\omega_{x1} - \omega_{x2})/k_B T_c} - \frac{\rho^{ss}_{x1x1}}{\rho^{ss}_{x2x2}} \right) + \\
\hbar \left(\omega_{x2} - \omega_\alpha\right) \left(1 + \bar{n}_{2c}\right) \gamma_c \rho^{ss}_{\alpha\alpha} \left(e^{-\hbar(\omega_{x2} - \omega_\alpha)/k_B T_c} - \frac{\rho^{ss}_{x2x2}}{\rho^{ss}_{\alpha\alpha}} \right) + \hbar \left(\omega_\beta - \omega_b\right) \left(1 + \bar{N}_c\right) \Gamma_c \rho^{ss}_{bb} \left(e^{-\hbar(\omega_\beta - \omega_b)/k_B T_c} - \frac{\rho^{ss}_{\beta\beta}}{\rho^{ss}_{bb}} \right) = \\
- \frac{\rho^{ss}_{bb} \gamma_h n_h \gamma_x \gamma_c \Gamma \left(1+n_x\right) \left(1+n_{2c}\right)}{\gamma_x \gamma_c \Gamma \left(1+n_x\right) \left(1+n_{2c}\right) + \gamma_h \left(1+n_h\right) \left[\gamma_x n_x \left(\Gamma + \gamma_c n_{2c}\right) + \gamma_c \left(1+n_{2c}\right) \Gamma \right]} \hbar \left(\omega_{x1} + \omega_\beta - \omega_\alpha - \omega_b\right),
\end{aligned} \tag{S17}$$

$$\frac{-J_c}{J_h} = \frac{\omega_{x1} + \omega_\beta - \omega_\alpha - \omega_b}{\omega_{x1} - \omega_b} = 1 + \frac{\omega_\beta - \omega_\alpha}{\omega_{x1} - \omega_b}, \tag{S18}$$

where $\hbar\omega_{x1} - \hbar\omega_b$ ($\hbar\omega_\alpha - \hbar\omega_\beta$) is the energy of the absorbed (emitted) quanta from the hot bath (to the RC/circuit), therefore equivalent to $\hbar\omega_{abs}(\hbar\omega_{rc})$. Using this paper notation,

$$J_h \to J_{abs}, \quad J_c \to J_{loss},$$
$$\hbar\omega_\beta - \hbar\omega_\alpha \to -\hbar\omega_{rc}, \quad \hbar\omega_{x1} - \hbar\omega_b \to \hbar\omega_{abs},$$
$$T_h \to T_{abs}, \quad T_c \to T_{loss},$$

we obtain Eq. 3 in the main text.

IB. Standard FMO models

We use the standard model proposed in reference [5] for the Fenna-Mathews-Olson (FMO) complex of the green sulfur bacterium *Prosthecochloris aestuarii*. Its dynamics is governed by the following Hamiltonian,

$$H_{FMO} = H_{sites} + H_{FMO-vib} + H_{vib} + H_{Dec}, \tag{S19}$$

where H_{vib} is the free Hamiltonian for the vibrational degrees of freedom of the pigments and proteins, which we assume to be at equilibrium at a temperature $T_{loss} = 300K$. H_{sites} is the exciton Hamiltonian,

$$H_{sites} = \sum_{m\in FMO} E_m|m\rangle\langle m| + \sum_{m\neq n\in FMO} V_{mn}|m\rangle\langle n|, \tag{S20}$$

where $|m\rangle$ is the excited state of the m site, and the sum is over all the FMO sites. $H_{FMO-vib}$ represents the interaction between the excitons and the vibrational bath,

$$H_{FMO-vib} = \sum_{m\in FMO,\xi} k_\xi^m |m\rangle\langle m| \otimes Q_\xi, \tag{S21}$$

where Q_ξ operates on the vibrational degrees of freedom. All the parameters for this Hamiltonian can be found in reference [5].

Transmission of energy to the reaction center

The Hamiltonian H_{Dec} governs the transmission of energy to the reaction center and is typically modeled [1, 6–12] as an irreversible decay term from the FMO site 3 to 8,

$$H_{Dec} = \sqrt{\Gamma_{3,8}}|8\rangle\langle 3|. \tag{S22}$$

We use a typical value for this rate, $\Gamma_{3,8} = 62.8/1.88\, cm^{-1}$[6–8, 12].

Thermal radiation

Even though at the surface of the Sun the emitted thermal radiation is at thermal equilibrium with the same temperature as the Sun, T_S, due to geometric considerations, only a small fraction of those photons reaches the Earth. This is quantified by a geometric factor $\lambda = 2 * 10^{-5}$ equal to the angle subtended by the Sun seen from the Earth. If $n_{T_S}[\omega]$ photons of frequency ω are emitted from the Sun, only λn_{T_s} reach the Earth. This radiation is no longer at thermal equilibrium, but rather is a non-equilibrium bath at an effective temperature [13–15],

$$e^{-\hbar\omega_{ant}/k_B T_{abs}} = \frac{\lambda n_{T_S}[\omega_{ant}]}{\lambda n_{T_S}[\omega_{ant}] + 1} \to T_{abs} \sim 1356K. \tag{S23}$$

where $n_{T_S}[\omega] = (e^{\hbar\omega/k_B T_S} - 1)^{-1}$. The dilution of the photon numbers turns the effective temperature, T_{abs}, frequency dependent. Nevertheless, the frequency variation between the antenna and the FMO site is small, therefore we assume the same T_{abs} for the antenna and the FMO sites.

II. Dynamic equations for simple models for the RC/circuit

In this section, we explain the preliminary steps taken for deriving the dynamics of the simple models for the RC/circuit (Eqs. 8 and 12 in the main text).

We consider a three level system (3LS), S, coupled to the reaction center (RC) or electric circuit. The later is a reservoir of independent quinones/sites, each of them represented by a single two level system (TLS). Its ground state represents an empty quinone/site and the excited state corresponds to a full quinone/site. Besides, the 3LS is coupled to a photon (hot) bath and a vibrational (cold) bath (see Figure 3 in the main text). The total Hamiltonian is

$$H_S + H_B + H_{SB}, \tag{S24}$$

where $H_B = H_{photons} + H_{phonons}$ is the baths free Hamiltonian. The S-baths coupling Hamiltonian is given by

$$H_{SB} = S \otimes (B_h + B_c) = \sum_\lambda g_{h,\lambda} \left(|2\rangle\langle 0|a_\lambda + |0\rangle\langle 2|a_\lambda^\dagger\right) + \sum_\lambda g_{c,\lambda} \left(|2\rangle\langle 1|b_\lambda + |1\rangle\langle 2|b_\lambda^\dagger\right), \tag{S25}$$

where a_λ,$a_\lambda^\dagger$ (b_λ, $b_\lambda^\dagger$) are the annihilation and creation operators of photons (phonons) modes. The S + RC/circuit Hamiltonian is

$$H_S = H_0 + H_{trans}, \qquad H_0 = \hbar\omega_{abs}|2\rangle\langle 2| + \frac{\hbar\omega_{rc}}{2}\left(|1\rangle\langle 1| - |0\rangle\langle 0|\right), \tag{S26}$$

where H_0 is the 3LS free Hamiltonian and H_{trans} describes the energy transfer to the RC/circuit. We compare between two possible schemes: i) a decay transfer described by a non-Hermitian H_{trans}; ii) a Hamiltonian transfer, represented by a Hermitian H_{trans}.

IIA. Decay transfer

The decay transfer is described by the following non-Hermitian term,

$$H_{tranf}^{Dec} = \sqrt{\Gamma}|0\rangle\langle 1|. \tag{S27}$$

As a first step we transform the S-bath interaction and the transfer Hamiltonian to the interaction picture

$$H_{SB} \to e^{iH_0 t} H_{SB} e^{-iH_0 t}, \qquad H_{tranf}^{Dec} \to e^{iH_0 t} H_{tranf}^{Dec} e^{-iH_0 t}. \tag{S28}$$

H_{tranf}^{Dec} is a fictitious Hamiltonian due to its lack of Hermiticity, therefore cannot form part of the rotation, $e^{iH_0 t}$, which has to be unitary. Besides, we derive the reduced dynamics only for S. The operators in the interaction picture are:

$$|2\rangle\langle 0|\,[t] = e^{it\hbar\left(\omega_{abs} + \frac{\omega_{rc}}{2}\right)}|2\rangle\langle 0|, \qquad |2\rangle\langle 1|\,[t] = e^{it\hbar\left(\omega_{abs} - \frac{\omega_{rc}}{2}\right)}|2\rangle\langle 1|, \qquad |0\rangle\langle 1|\,[t] = e^{-it\hbar\omega_{rec}}|0\rangle\langle 1|. \tag{S29}$$

Using the standard Born-Markov approximation, the Lindblad equation [16] for S is obtained (Eq.8 in the main text). The steady state populations are

$$\frac{\rho_{22}^{ss}}{\rho_{11}^{ss}} = 1, \qquad \frac{\rho_{22}^{ss}}{\rho_{00}^{ss}} = e^{-\hbar\omega_+/k_B T_{abs}} - \frac{\rho_{11}^{ss}}{\widetilde{n}_{abs}\rho_{00}^{ss}}, \qquad \frac{\rho_{11}^{ss}}{\rho_{00}^{ss}} = \frac{n_{abs}\widetilde{n}_{loss}}{n_{loss}\widetilde{n}_{abs} + \widetilde{n}_{abs} + \widetilde{n}_{loss}},$$
$$\rho_{11}^{ss} = \frac{1}{1 + \frac{\rho_{00}^{ss}}{\rho_{11}^{ss}} + \frac{\rho_{22}^{ss}}{\rho_{11}^{ss}}} = \frac{1}{1 + 2e^{\hbar\omega_+/k_B T_{abs}}}. \tag{S30}$$

where $n_{abs(loss)}$ is the photon (vibrational) bath population of mode $\omega_\pm = \omega_{abs} \pm \frac{\omega_{rc}}{2}$ and $\widetilde{n}_{abs(loss)} = n_{abs(loss)} + 1$.

IIB. Hamiltonian transfer

Here we explicitly consider the RC/circuit and its coupling to S, by considering H_{trans} as an Hermitian Hamiltonian. The RC/circuit is composed of identical and independent TLSs. The S + RC/circuit Hamiltonian is:

$$H_S = \hbar\omega_{abs}|2\rangle\langle 2| + \frac{\hbar\omega_{rc}}{2}\left(|1\rangle\langle 1| - |0\rangle\langle 0|\right) + \sum_k^j \sqrt{\frac{\Gamma}{2j}}\left(\sigma_-^k|1\rangle\langle 0| + \sigma_+^k|0\rangle\langle 1|\right) + \hbar\omega_{rc}\sum_k \sigma_z^k, \tag{S31}$$

where j is the number of TLSs.

In order to find the energy that is being transferred to the RC/circuit, we start by diagonalizing the S + RC circuit. This is achieved by first applying the Holstein-Primakoff transformation [17], that consists on the introduction of the following collective operators:

$$\sum_k \sigma_-^k = \left(\sqrt{2j - c^\dagger c}\right)c, \qquad \sum_k \sigma_+^k = c^\dagger\left(\sqrt{2j - c^\dagger c}\right), \qquad \sum_k \sigma_z^k = c^\dagger c - j. \tag{S32}$$

The new Hamiltonian is

$$H_S = \hbar\omega_{abs}|2\rangle\langle 2| + \frac{\hbar\omega_{rc}}{2}\left(|1\rangle\langle 1| - |0\rangle\langle 0|\right) + \sqrt{\frac{\Gamma}{2j}}\left[\left(\sqrt{2j - c^\dagger c}\right)c|1\rangle\langle 0| + c^\dagger\left(\sqrt{2j - c^\dagger c}\right)|0\rangle\langle 1|\right] + \hbar\omega_{rc}\left(c^\dagger c - j\right). \tag{S33}$$

At this point, the modes are displaced, $c \to c - \sqrt{\epsilon}$,

$$H_S = \hbar\omega_{abs}|2\rangle\langle 2| + \frac{\hbar\omega_{rc}}{2}\left(|1\rangle\langle 1| - |0\rangle\langle 0|\right) + \sqrt{\frac{\Gamma k\eta}{2j}}\left(c|1\rangle\langle 0| + c^\dagger|0\rangle\langle 1|\right) - \sqrt{\frac{\Gamma k\eta\epsilon}{2j}}\left(|1\rangle\langle 0| + |0\rangle\langle 1|\right) + \hbar\omega_{rc}\left(c^\dagger c - \sqrt{\epsilon}(c + c^\dagger) + \epsilon - j\right), \tag{S34}$$

where $k = 2j - \epsilon$ and $\eta = 1 - \frac{c^\dagger c - \sqrt{\epsilon}(c^\dagger + c)}{k}$. We assume that the number of TLSs is large, $\frac{c^\dagger c - \sqrt{\epsilon}(c^\dagger + c)}{k} \ll 1$. The physical interpretation of this approximation is clarified below. Under this assumptions, we expand $\sqrt{\eta} \approx 1 - \frac{c^\dagger c - \sqrt{\epsilon}(c^\dagger + c)}{2k} - \frac{\epsilon(c^\dagger + c)^2}{8k^2}$ and keep terms up to order $\frac{1}{\sqrt{j}}$,

$$\begin{aligned} H_S = {} & \hbar\omega_{abs}|2\rangle\langle 2| + \frac{\omega_{rc}}{2}\left(|1\rangle\langle 1| - |0\rangle\langle 0|\right) - \sqrt{\frac{\Gamma k\epsilon}{2j}}\left(|1\rangle\langle 0| + |0\rangle\langle 1|\right) + \sqrt{\frac{\Gamma k}{2j}}\left(c|1\rangle\langle 0| + c^\dagger|0\rangle\langle 1|\right) - \\ & \frac{\epsilon}{2}\sqrt{\frac{\Gamma}{2jk}}\left(c^\dagger + c\right)\left(|1\rangle\langle 0| + |0\rangle\langle 1|\right) + \hbar\omega_{rc}\left(c^\dagger c - \sqrt{\epsilon}\left(c + c^\dagger\right) + \epsilon - j\right). \end{aligned} \tag{S35}$$

Setting $\epsilon = 0$, the Hamiltonian is simplified to

$$H_S = \hbar\omega_{abs}|2\rangle\langle 2| + \frac{\hbar\omega_{rc}}{2}\left(|1\rangle\langle 1| - |0\rangle\langle 0|\right) + \sqrt{\Gamma}\left(c|1\rangle\langle 0| + c^\dagger|0\rangle\langle 1|\right) + \hbar\omega_{rc}\left(c^\dagger c - j\right) \tag{S36}$$

and the approximation to $\frac{c^\dagger c}{2j} \ll 1$. Therefore, we are just assuming that the total number of excitations in the RC/circuit is very small compared to the number of quinones/sites, so energy may always be transferred to the RC/circuit. From Eq. S36, we derive Eq. 8 in the main text,

$$H_{trasns}^{Ham} = \sqrt{\Gamma}\left(c|1\rangle\langle 0| + c^\dagger|0\rangle\langle 1|\right) + \hbar\omega_{rc}\left(c^\dagger c - j\right). \tag{S37}$$

which is an effective equation for a harmonic oscillator (HO) coupled to S. Therefore, from now on we model the RC/circuit as a HO. Next we diagonalize Eq. S36. The Hamiltonian eigenvectors are

$$|+,n\rangle = \frac{1}{\sqrt{2}}\left(|1,n\rangle + |0,n+1\rangle\right), \qquad |-,n\rangle = \frac{1}{\sqrt{2}}\left(|0,n+1\rangle - |1,n\rangle\right), \tag{S38}$$

$$E_{\pm} = \hbar\omega_{rc}\left(n+\frac{1}{2}\right) \pm \frac{\hbar\Omega_n}{2} - j\hbar\omega_{rc}, \tag{S39}$$

$$H_S + H_{trans}^{Ham} = \hbar\omega_{abs}|2,n\rangle\langle 2,n| + \hbar\omega_{rc}\left(\tilde{c}^\dagger\tilde{c} - j\right) + \sum_n \frac{\hbar\omega_{rc}}{2}\left\{\left(1+\frac{\Omega_n}{\omega_{rc}}\right)|+,n\rangle\langle +,n| + \left(1-\frac{\Omega_n}{\omega_{rc}}\right)|-,n\rangle\langle -,n|\right\}, \tag{S40}$$

where $\tilde{c}^\dagger(\tilde{c})$ is the creation (annihilation) operator in the new basis and $\Omega_n = 2\sqrt{\Gamma(n+1)}$. The inverse transformations are

$$|1,n\rangle = \frac{1}{\sqrt{2}}\left(|+,n\rangle - |-,n\rangle\right), \qquad |0,n+1\rangle = \frac{1}{\sqrt{2}}\left(|+,n\rangle + |-,n\rangle\right). \tag{S41}$$

Rewriting the S-bath Hamiltonian, Eq. S25 , in the new basis,

$$|2\rangle\langle 0| = \sum_n \frac{1}{\sqrt{2}}\left(|2,n+1\rangle\langle +,n| + |2,n+1\rangle\langle -,n|\right), \qquad |2\rangle\langle 1| = \sum_n \frac{1}{\sqrt{2}}\left(|2,n\rangle\langle +,n| - |2,n\rangle\langle -,n|\right), \tag{S42}$$

and transforming to the interaction picture,

$$H_{SB} \to e^{iH_S t} H_{SB} e^{-iH_S t},$$

$$|2\rangle\langle 0|\,[t] = \sum_n \frac{1}{\sqrt{2}}\left(e^{it\hbar\left(\omega_+ - \frac{\Omega_n}{2}\right)}|2,n+1\rangle\langle +,n| + e^{it\hbar\left(\omega_+ + \frac{\Omega_n}{2}\right)}|2,n+1\rangle\langle -,n|\right),$$

$$|2\rangle\langle 1|\,[t] = \sum_n \frac{1}{\sqrt{2}}\left(e^{it\hbar\left(\omega_- - \frac{\Omega_n}{2}\right)}|2,n\rangle\langle +,n| - e^{it\hbar\left(\omega_- + \frac{\Omega_n}{2}\right)}|2,n\rangle\langle -,n|\right). \tag{S43}$$

In contrast to the decay transfer scheme (Eq. S28), here H_{trans}^{Ham} is Hermitian and we derive the reduced dynamics for the S + RC/circuit. Therefore H_{trans}^{Ham} is included in the rotation, $e^{iH_S t}$. Using the standard Born-Markov approximation, the Lindblad equation [16] for S + RC/circuit is obtained, and from it the evolution equations are derived,

$$\dot{\rho}_{+,n} = \frac{\Gamma}{2}\left\{-\left(n_{abs}\left[\omega_+ - \frac{\Omega_n}{2}\right] + n_{loss}\left[\omega_- - \frac{\Omega_n}{2}\right]\right)\rho_{+,n} + \widetilde{n}_{loss}\left[\omega_- - \frac{\Omega_n}{2}\right]\rho_{2,n} + \widetilde{n}_{abs}\left[\omega_+ - \frac{\Omega_n}{2}\right]\rho_{2,n+1}\right\},$$

$$\dot{\rho}_{-,n} = \frac{\Gamma}{2}\left\{-\left(n_{abs}\left[\omega_+ + \frac{\Omega_n}{2}\right] + n_{loss}\left[\omega_- + \frac{\Omega_n}{2}\right]\right)\rho_{-,n} + \widetilde{n}_{loss}\left[\omega_- + \frac{\Omega_n}{2}\right]\rho_{2,n} + \widetilde{n}_{abs}\left[\omega_+ + \frac{\Omega_n}{2}\right]\rho_{2,n+1}\right\},$$

$$\dot{\rho}_{2,n} = \frac{\Gamma}{2}\left\{-\left(\widetilde{n}_{abs}\left[\omega_+ - \frac{\Omega_{n-1}}{2}\right] + \widetilde{n}_{abs}\left[\omega_+ + \frac{\Omega_{n-1}}{2}\right] + \widetilde{n}_{loss}\left[\omega_- + \frac{\Omega_n}{2}\right] + \widetilde{n}_{loss}\left[\omega_- - \frac{\Omega_n}{2}\right]\right)\rho_{2,n}\right.$$
$$\left. n_{loss}\left[\omega_- - \frac{\Omega_n}{2}\right]\rho_{+,n} + n_{loss}\left[\omega_- + \frac{\Omega_n}{2}\right]\rho_{-,n} + n_{abs}\left[\omega_+ - \frac{\Omega_{n-1}}{2}\right]\rho_{+,n-1} + n_{abs}\left[\omega_+ + \frac{\Omega_{n-1}}{2}\right]\rho_{-,n-1}\right\}, \tag{S44}$$

where ρ_i is the population of the combined state i (S + RC/circuit), Γ_i and $n_i\,[\omega]$ are the decay rate and the ω-mode population of the i-bath, respectively, and $\widetilde{n}_i\,[\omega] = n_i\,[\omega] + 1$. The equations for the off-diagonal terms are decoupled from the populations and for simplicity we assume that the off-diagonal terms are zero. In order to simplify the evolution equations we assume that the mode population does not change on shift of the order of Ω_n, therefore $n_i\left[\omega_\pm \pm \frac{\Omega_n}{2}\right] \approx n_i\,[\omega_\pm]$. The resulting equations are shown in the main text, Eqs. 12.

The 3LS steady state is obtained by summing over n Eqs. 12 in the main text. It is

$$\frac{\rho_2^{ss}}{\rho_+^{ss}} = \frac{n_{abs} + n_{loss}}{\widetilde{n}_{abs} + \widetilde{n}_{loss}}, \qquad \rho_+^{ss} = \frac{\widetilde{n}_{abs} + \widetilde{n}_{loss}}{3\widetilde{n}_{abs} + 3\widetilde{n}_{loss} - 2}, \tag{S45}$$

where $n_{abs(loss)}$ is the photon (vibrational) bath population of mode $\omega_\pm = \omega_{abs} \pm \frac{\omega_{rc}}{2}$. Using these expressions, we can find the evolution for the HO excitation energy,

$$\hbar\omega_{rc}\langle\dot{n}\rangle = \hbar\omega_{rc}\left(s-r\right), \tag{S46}$$

which is equal to $-P^{Ham}$ (the used sign convention can be found below Eq. 1 in the main text). Thus, $s > r$ is required in order to increase the RC/circuit energy. At the 3LS steady state, this implies,

$$s-r = \frac{\Gamma\widetilde{n}_{loss}\widetilde{n}_{abs}}{3\widetilde{n}_{abs}+3\widetilde{n}_{loss}-2}\left(e^{-\hbar\omega_+/k_B T_{abs}} - e^{-\hbar\omega_-/k_B T_{loss}}\right) = K_1\left(e^{-\hbar\omega_+/k_B T_{abs}} - e^{-\hbar\omega_-/k_B T_{loss}}\right) > 0, \tag{S47}$$

where $K_1 = \frac{\Gamma\widetilde{n}_{loss}\widetilde{n}}{3\widetilde{n}_h+3\widetilde{n}_c-2} > 0$ and the energy gain condition is

$$\frac{T_{loss}}{T_{abs}} < \frac{\omega_-}{\omega_+}. \tag{S48}$$

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