Supplementary Material—Quantum Correlation and Synchronisation-Enhanced Energy Transfer in Driven-Dissipative Light-Harvesting Dimers

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DERIVATION OF RATE EQUATIONS

The set of rate equations (7) in the main text is derived by taking the expectation values of the following operators $|1\rangle\langle 1| \equiv \hat{\sigma}_1^+\hat{\sigma}_1^-(\hat{1}-\hat{\sigma}_2^+\hat{\sigma}_2^-)$, $|2\rangle\langle 2| \equiv (\hat{1}-\hat{\sigma}_1^+\hat{\sigma}_1^-)\hat{\sigma}_2^+\hat{\sigma}_2^-$, $\hat{\sigma}_1^+\hat{\sigma}_2^-$, $\hat{\sigma}_2^+\hat{\sigma}_1^-$, $\hat{b}_1^\dagger+\hat{b}_1$, $\hat{b}_2^\dagger+\hat{b}_2$, $\mathrm{i}(\hat{b}_1^\dagger-\hat{b}_1)$, and $\mathrm{i}(\hat{b}_2^\dagger-\hat{b}_2)$ [1]. For example,

$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}\operatorname{tr}\{|1\rangle\langle 1|\,\hat{\rho}\} \equiv & \frac{\mathrm{d}}{\mathrm{d}t}\rho_{11} = -\mathrm{i}\,\langle[|1\rangle\langle 1|\,, \frac{\Delta\varepsilon}{2}\big(\hat{\sigma}_1^+\hat{\sigma}_1^- - \hat{\sigma}_2^+\hat{\sigma}_2^-\big) + J\big(\hat{\sigma}_1^+\hat{\sigma}_2^- + \hat{\sigma}_2^+\hat{\sigma}_1^-\big) + g_1\,|1\rangle\langle 1|\,(\hat{b}_1^\dagger+\hat{b}_1) + g_2\,|2\rangle\langle 2|\,(\hat{b}_2^\dagger+\hat{b}_2)]\rangle \\
& + \Gamma_{\mathrm{deph}}\sum_m \langle\hat{\sigma}_m^+\,|1\rangle\langle 1|\,\hat{\sigma}_m^- - \frac{1}{2}\,|1\rangle\langle 1|\,\hat{\sigma}_m^+\hat{\sigma}_m^- - \frac{1}{2}\hat{\sigma}_m^+\hat{\sigma}_m^-\,|1\rangle\langle 1| + \hat{\sigma}_m^-\,|1\rangle\langle 1|\,\hat{\sigma}_m^+ - \frac{1}{2}\,|1\rangle\langle 1|\,\hat{\sigma}_m^-\hat{\sigma}_m^+ - \frac{1}{2}\hat{\sigma}_m^-\hat{\sigma}_m^+\,|1\rangle\langle 1|\rangle \\
= & -\mathrm{i}J\,\langle\hat{\sigma}_1^+\hat{\sigma}_2^- - \hat{\sigma}_2^+\hat{\sigma}_1^-\rangle - \Gamma_{\mathrm{deph}}(\rho_{11}-\rho_{22}),
\end{aligned}
\tag{1}
$$

$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle = & -\mathrm{i}\,\langle[\hat{\sigma}_1^+\hat{\sigma}_2^-, \frac{\Delta\varepsilon}{2}\big(\hat{\sigma}_1^+\hat{\sigma}_1^- - \hat{\sigma}_2^+\hat{\sigma}_2^-\big) + J\big(\hat{\sigma}_1^+\hat{\sigma}_2^- + \hat{\sigma}_2^+\hat{\sigma}_1^-\big) + g_1\,|1\rangle\langle 1|\,(\hat{b}_1^\dagger+\hat{b}_1) + g_2\,|2\rangle\langle 2|\,(\hat{b}_2^\dagger+\hat{b}_2)]\rangle \\
& + \Gamma_{\mathrm{deph}}\sum_m \langle\hat{\sigma}_m^+\hat{\sigma}_1^+\hat{\sigma}_2^-\hat{\sigma}_m^- - \frac{1}{2}\hat{\sigma}_1^+\hat{\sigma}_2^-\hat{\sigma}_m^+\hat{\sigma}_m^- - \frac{1}{2}\hat{\sigma}_m^+\hat{\sigma}_m^-\hat{\sigma}_1^+\hat{\sigma}_2^- + \hat{\sigma}_m^-\hat{\sigma}_1^+\hat{\sigma}_2^-\hat{\sigma}_m^+ - \frac{1}{2}\hat{\sigma}_1^+\hat{\sigma}_2^-\hat{\sigma}_m^-\hat{\sigma}_m^+ - \frac{1}{2}\hat{\sigma}_m^-\hat{\sigma}_m^+\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle \\
= & \mathrm{i}\Delta\varepsilon\,\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle + \mathrm{i}J(\rho_{11}-\rho_{22}) + \mathrm{i}g_1\,\langle\hat{\sigma}_1^+\hat{\sigma}_2^-(\hat{b}_1^\dagger+\hat{b}_1)\rangle - \mathrm{i}g_2\,\langle\hat{\sigma}_1^+\hat{\sigma}_2^-(\hat{b}_2^\dagger+\hat{b}_2)\rangle - \Gamma_{\mathrm{deph}}\,\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle\,,
\end{aligned}
\tag{2}
$$

and

$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}\langle\hat{b}_1^\dagger \pm \hat{b}_1\rangle = & -\mathrm{i}\,\langle[\hat{b}_1^\dagger \pm \hat{b}_1, \omega_1\hat{b}_1^\dagger\hat{b}_1 + g_1\,|1\rangle\langle 1|\,(\hat{b}_1^\dagger+\hat{b}_1) + g_2\,|2\rangle\langle 2|\,(\hat{b}_2^\dagger+\hat{b}_2)]\rangle \\
& + \Gamma_{\mathrm{ph}}(\bar{n}_{\mathrm{ph}}+1)\,\langle\hat{b}_1^\dagger(\hat{b}_1^\dagger \pm \hat{b}_1)\hat{b}_1 - \frac{1}{2}(\hat{b}_1^\dagger \pm \hat{b}_1)\hat{b}_1^\dagger\hat{b}_1 - \frac{1}{2}\hat{b}_1^\dagger\hat{b}_1(\hat{b}_1^\dagger \pm \hat{b}_1)\rangle \\
& + \Gamma_{\mathrm{ph}}\bar{n}_{\mathrm{ph}}\,\langle\hat{b}_1(\hat{b}_1^\dagger \pm \hat{b}_1)\hat{b}_1^\dagger - \frac{1}{2}(\hat{b}_1^\dagger \pm \hat{b}_1)\hat{b}_1\hat{b}_1^\dagger - \frac{1}{2}\hat{b}_1\hat{b}_1^\dagger(\hat{b}_1^\dagger \pm \hat{b}_1)\rangle \\
= & -\mathrm{i}\,\langle -\hat{b}_1^\dagger \pm \hat{b}_1\rangle - \frac{\Gamma_{\mathrm{th}}}{2}\,\langle\hat{b}_1^\dagger \pm \hat{b}_1\rangle\,,
\end{aligned}
\tag{3}
$$

where we have used the cyclic property of the trace, and $\bar{n}_{\mathrm{ph}}(x) = 1/(e^{\beta x}-1)$ denotes the Planck distribution at inverse temperature β. At transition frequencies ω_ν, we notice that when $T = 298$ K,

$$
\bar{n}_{\mathrm{ph}}(\Delta\varepsilon/\hbar) \approx 0. \tag{4}
$$

ENVIRONMENT-ASSISTED TRANSFER

We consider the dipole-dipole interaction can be assisted by environmental phonon [2] or photons. The Lindblad master equation of interest reads

$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}\hat{\rho}(t) = -\mathrm{i}[\hat{H}_{\mathrm{el}} + \hat{H}_{\mathrm{vib}} + \hat{V}_{\mathrm{dis}}, \hat{\rho}(t)] & + \Gamma_{\mathrm{dip}}(\bar{n}_{\mathrm{ph}}+1)\bigg(\hat{\sigma}_2^+\hat{\sigma}_1^-\,\hat{\rho}(t)\hat{\sigma}_1^+\hat{\sigma}_2^- - \frac{1}{2}\hat{\sigma}_1^+\hat{\sigma}_2^-\hat{\sigma}_2^+\hat{\sigma}_1^-\,\hat{\rho}(t) - \frac{1}{2}\hat{\rho}(t)\hat{\sigma}_1^+\hat{\sigma}_2^-\hat{\sigma}_2^+\hat{\sigma}_1^-\bigg) \\
& + \Gamma_{\mathrm{dip}}\bar{n}_{\mathrm{ph}}\bigg(\hat{\sigma}_1^+\hat{\sigma}_2^-\,\hat{\rho}(t)\hat{\sigma}_2^+\hat{\sigma}_1^- - \frac{1}{2}\hat{\sigma}_2^+\hat{\sigma}_1^-\hat{\sigma}_1^+\hat{\sigma}_2^-\,\hat{\rho}(t) - \frac{1}{2}\hat{\rho}(t)\hat{\sigma}_2^+\hat{\sigma}_1^-\hat{\sigma}_1^+\hat{\sigma}_2^-\bigg),
\end{aligned}
\tag{5}
$$

where the transition frequency $\Gamma_{\mathrm{dip}} = 100$ THz is set in simulation. The physical picture of $\hat{\sigma}_2^+\hat{\sigma}_1^-$ here implies that a phonon is created due to a electronic transition from higher to lower excitation states, while the reverse process hardly appears.

CLASSICAL NONLINEAR DYNAMICS OF RATE EQUATIONS

Fig. (1) compares the open quantum and classical dynamics when $\hat{\sigma}_m$ electronic dephasing is absent. We can observe that no energy transfer takes place in the ODE simulation, while the OPS simulation shows. The quantum synchronisation appears to facilitate the transfer and lead to the crossover between ρ_{11} and ρ_{22}. However, the ODE simulation shows incorrect dynamics where ρ_{22} oscillates near 0 and has negative value. This error is same with ρ_{11}, where it oscillates around 1 and has values greater than 1. These are nonphysical due to obvious quantum correlations predicted in the last row of Fig. (1). The above observations manifest the non-classicality of the system and the energy transfer process.

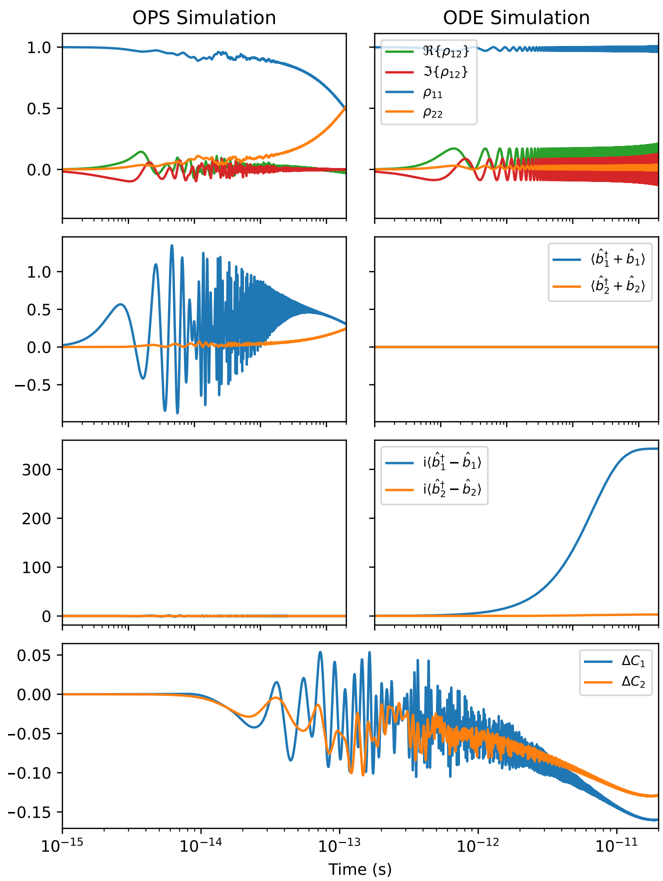

FIG. 1. Numerical simulation of the quantum dynamics without $\hat{\sigma}_m$ electronic dephasing compared to classical ODE simulation. The first row shows electronic density matrix elements. The second and third rows depict $\langle \hat{b}_m^\dagger + \hat{b}_m \rangle$ and $\mathrm{i}\,\langle \hat{b}_m^\dagger - \hat{b}_m \rangle$, respectively. The last row plots the quantum correlations due to the factorised-state approximation. Parameters used are from in Table. (I) in the main text.

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[1] H.-P. Breuer and F. Petruccione, *The theory of open quantum systems* (Oxford University Press, USA, 2002).
[2] H. Chen, X. Wang, A.-P. Fang, and H.-R. Li, Phonon-assisted excitation energy transfer in photosynthetic systems, Chinese Physics B **25**, 098201 (2016).