Enhancing light-harvesting power with coherent vibrational interactions: a quantum heat engine picture

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Recent evidence suggests that quantum effects may have functional importance in biological light-harvesting systems. Along with delocalized electronic excitations, it is now suspected that quantum coherent interactions with certain near-resonant vibrations contribute to light-harvesting performance. However, the actual quantum advantage offered by such coherent vibrational interactions has not yet been established. We investigate a quantum design principle, whereby coherent exchange of single energy quanta between electronic and vibrational degrees of freedom can enhance a light-harvesting system’s power above what is possible by thermal mechanisms alone. We present a prototype quantum heat engine which cleanly illustrates this quantum design principle, and quantify its quantum advantage using thermodynamic measures of performance. We also demonstrate the principle’s applicability for realistic biological structures.

The primary insight of quantum information science is that, by exploiting quantum effects, we can enhance performance over comparable classical devices. So it is humbling to think that nature may have taken advantage of quantum effects long before we had the idea. Indeed, there are several scenarios where quantum phenomena could play a role in biological function. One instance is biological light-harvesting; growing experimental evidence \[8\] and theoretical models \[9,15\] now suggest that quantum processes contribute beneficially to the high efficiency of biological light-harvesting complexes (LHCs) (see \[10\] for a recent review). Identifying quantum-mechanical design principles in such natural systems can, in turn, inspire new artificial light-harvesting technologies \[17,21\].

Attention has focused of late on the role of latent vibrational structures in LHCs \[16,22,28\]. In a similar vein, vibronic coherence was recently discovered during charge separation in Photosystem II \[7,8\]. While most vibrations can be coarse-grained into a background environment, it is now recognized that a discrete subset of strongly coupled modes should be considered on the same footing as the electronic light-harvesting system itself. Models where electrons interact strongly with near-resonant underdamped vibrations \[24,27,29,30\] can explain the unusually long-lived coherence observed in photosynthetic complexes \[2,6\] and can exhibit enhanced transport properties \[22,28,31,33\]. Most interestingly, with such a model, the vibrations can develop unambiguous signatures of non-classicality \[35\], which themselves correlate suggestively with energy transport. Despite these intriguing observations, the ‘quantum advantage’ to light-harvesting provided by non-classical vibrations has not yet been fully elucidated.

In this work, we present a quantum design principle in action, whereby coherent interactions between electrons and surrounding near-resonant vibrations lead to enhanced light-harvesting capabilities for the collective system. Taking a thermodynamic perspective, we focus on quantumness in the light-harvesting dynamics rather than in the states. Combining typical network transport models with a quantum heat engine picture \[36,38\], we present a prototype light-harvester which clearly reveals the vibrational quantum advantage. Specifically, near-resonant quantized vibrations have a catalytic role, opening up new energy transfer pathways and allowing the light-harvesting process to occur at a higher rate. These additional pathways are intrinsically quantum mechanical, involving coherent energy exchange between the electronic and vibrational subsystems. Even with limited coherence, these processes can outpace thermal mechanisms at natural conditions, leading to larger energy currents and an overall enhancement in light-harvesting power. In addition to our idealized light-harvester, we will also demonstrate these principles in action for several biologically relevant scenarios.

*Light-harvesting framework.* — Both natural and artificial light-harvesting can be understood within a common framework (Fig. 1). Two subsets of electronic states (ground/excited subspaces in LHCs; valence/conduction bands in semiconductors) are separated by a large energy gap $E_g$. Incoming solar energy pumps an electron out of a low-energy state, across the energy gap, into a high-energy excited state. The electron is then transported through a network of excited states to the edge of the gap. Here, we picture an abstract load bridging the separated electronic ‘terminals,’ as in an electric circuit. This load might represent a LHC’s reaction center, an electronic battery, or a further subnetwork of electronic states. By giving its excess energy to the load, the electron can return to the low-energy subspace. The electron can then recombine with the remaining low-energy hole, completing the cycle.

We consider the single excitation subspace, with cor-
responding electronic Hamiltonian

\[ H_e = E_0 |\varepsilon_0\rangle \langle \varepsilon_0| + \sum_{i=1}^{N} E_i |s_i\rangle \langle s_i| + \sum_{i \neq j}^{N} J_{ij} |s_i\rangle \langle s_j| . \]

The orthogonal states \(|s_i\rangle\) correspond to excitations spatially localized at specific sites within the network. For our considerations, the low-energy subspace will contain only the ground state \(|\varepsilon_0\rangle\). In many situations of interest, the couplings \(J_{ij}\) between different sites are non-zero, leading to delocalized exciton eigenstates \(|\varepsilon_i\rangle\) with energies \(\varepsilon_i\). Such delocalization can be remarkably robust, even in noisy conditions [11, 39, 40].

Our light-harvester’s evolution will be described by a Lindblad-type master equation [41-43, 44].

\[ \dot{\rho}_S(t) = i[H_S, \rho_S(t)] + \sum_{\alpha} \mathcal{L}_{\alpha}[\rho_S(t)]. \]

The desired ‘system’ degrees of freedom are included in the density matrix \(\rho_S\) and the (time-independent) Hamiltonian \(H_S\). We consider the electronic level scheme in the exciton basis \(|\varepsilon_i\rangle\) of the electronic Hamiltonian \(H_e\) alone does not cause excitonic transitions. Important exchanges with external systems (incoming solar energy, the dissipative background, and the load to which work is extracted) are mediated by thermal reservoirs at fixed temperatures \(T_H > T_C\) and \(T_L = 0\), respectively (see Fig. 1). This lets us consider the detailed inner workings of our light-harvester, while simplifying the sources, sinks, and load to which it connects.

Explicitly, we have the following electronic transitions:

- \(|\varepsilon_0\rangle\) to \(|\varepsilon_1\rangle\)
- \(|\varepsilon_1\rangle\) to \(|\varepsilon_0\rangle\)

where \(\gamma_H, \gamma_C\) are coupling parameters and the \(\pi_a\) are the mean populations of the corresponding reservoirs at the given transition energies and temperatures. While the source \(\mathcal{L}_H\) and load \(\mathcal{L}_L\) link only two levels each, the background \(\mathcal{L}_C\) can facilitate all possible transitions \(|\varepsilon_i\rangle \leftrightarrow |\varepsilon_j\rangle, i \neq j \in \{0, \ldots, N\}\). The operators with \(i, j = 0\) model an excitation decaying back to the ground state by giving its energy to the background instead of the load (or the reverse).

We analyze our light-harvester as a quantum heat engine (QHE) [18, 29, 37]. This perspective has revealed functional roles for quantum coherence in both natural and artificial systems. QHE performance is determined by the rate at which useful energy is transferred to the load, i.e., by the power \(P = I_L V_L\), where \(I_L\) and \(V_L\) are the current and voltage across the load. The current is simply \(I_L = e \rho_{NN} \Gamma_L\), with \(\rho_{NN}\) the population of the lowest excited state (\(e\) is the electron charge). The voltage is given by \(V_L = E_0 + k_B T_C \ln(\rho_{NN}/\rho_0)\), where \(E_0 = \varepsilon_N - \varepsilon_0\) and \(k_B\) is Boltzmann’s constant. This quantifies the energy that could be released by equilibrating a given population ratio at temperature \(T_C\). Using detailed balance arguments, Shockley and Queisser [37] showed that the maximum achievable voltage is \(e V_L \leq \eta_C E_g\), where \(\eta_C := 1 - \frac{T_L}{T_C}\) is the Carnot efficiency. Since \(V_L\) is fundamentally limited, the best strategy to increase power may therefore be to increase \(I_L\), which is directly related to the population \(\rho_{NN}\). Thus, the more population in this lowest excited state during operation, the more power the heat engine can deliver. As we will now show, this is the key advantage offered by coherently coupling electron and vibration systems.

Vibrionic light-harvester.— Our basic prototype will have \(N = 3\) sites, and we consider additional quantized modes at sites 1 and 2, with ladder operators \(\hat{a}_1, \hat{a}_2^\dagger\) and frequencies \(\omega_1/2\). The mode Hamiltonian and electron-mode interaction are given by

\[ H_m = \hbar \omega_i \sum_{i=1}^{2} \hat{a}_i^\dagger \hat{a}_i, \quad H_I = \sum_{i=1}^{2} \hbar g_i |s_i\rangle \langle s_i| (\hat{a}_i + \hat{a}_i^\dagger). \]

We take \(\omega_i = \omega_m\) and \(g_i = g\) for each mode. The system Hamiltonian \(H_S := H_e + H_m + H_I\) describes the coherent part of the evolution in Eq. 2. We note that the quantum design principle also works in larger networks, as well as less idealized scenarios (see Supplementary Material (SM)).
Motivated by values found in biological systems, we take site energies $E_1 = E_2 = 300 \text{ cm}^{-1}$, $E_3 = 0 \text{ cm}^{-1}$, and ground state energy $E_0 = -E_g = -10000 \text{ cm}^{-1}$. The two highest excited states are coupled, $J_{12} = J_{21} = 100 \text{ cm}^{-1}$, while the remaining couplings $J_{13} = J_{31}$ and $J_{32} = J_{23}$ are considered negligible in comparison and set to zero. We do this to isolate a single dimer; in a detailed microscopic model, the couplings $J_{13}$ and $J_{23}$ would be small but non-zero, or else no energy transfer could take place (we recover this phenomenologically via the Lindblad transitions). These values lead to electronic eigenstates $\{|\varepsilon_i; n\rangle\}$, where $\varepsilon_{1/2} = \pm \sqrt{3}|\varepsilon_3| \pm |\varepsilon_2|$ are delocalized, while $\varepsilon_3 = |s_3|$ and $\varepsilon_0$ is unchanged. The corresponding energies are $\varepsilon_{1/2} = E_{1/2} \pm J_{12} = 300 \pm 100 \text{ cm}^{-1}$, with $\varepsilon_0 = E_0$ and $\varepsilon_3 = E_3$ as before. Finally, we set the mode frequency resonant with delocalized exciton spacing $\hbar \omega_m = \varepsilon_1 - \varepsilon_2 = 200 \text{ cm}^{-1}$.

In this regime, the interaction becomes $H_I = H_{CM} + H_{RD}$, with
\begin{align*}
H_{CM} &= \frac{\hbar g}{\sqrt{2}} \left(|\varepsilon_1\rangle\langle\varepsilon_1| + |\varepsilon_2\rangle\langle\varepsilon_2|\right) \otimes (\hat{a}_{CM} + \hat{a}_{CM}^\dagger), \\
H_{RD} &= \frac{\hbar g}{\sqrt{2}} \left(|\varepsilon_1\rangle\langle\varepsilon_2| + |\varepsilon_2\rangle\langle\varepsilon_1|\right) \otimes (\hat{a}_{RD} + \hat{a}_{RD}^\dagger).
\end{align*}
In these mutually commuting terms, we have introduced centre of mass/relative displacement modes $\hat{a}_{CM/BD} = \frac{1}{\sqrt{2}}(\hat{a}_{CM} \pm \hat{a}_{BD})$. Both electron-vibration coupling and delocalized excitons were important for arriving at this form. The term $H_{RD}$ is a Jaynes-Cummings (JC) interaction, whose eigenstates, within the rotating wave approximation (RWA), are $\{|\varepsilon_i; n\rangle\}_{n=0}^\infty$, where the states $|\pm; n\rangle := \frac{1}{\sqrt{2}}(|\varepsilon_1; n\rangle \pm |\varepsilon_2; n+1\rangle)$ are entangled between the electronic and vibrational subsystems (we use the shorthand $|\varepsilon_i; n\rangle := |\varepsilon_i\rangle \otimes |n\rangle_{RD}$). The JC interaction induces coherent Rabi oscillations between the states $|\varepsilon_1; n\rangle$ and $|\varepsilon_2; n+1\rangle$ at frequencies $\omega_{\text{coh}}(n) \sim 2\hbar g \sqrt{n+1}$.

The Rabi oscillations provide a new mechanism for electronic population to transfer between $|\varepsilon_1\rangle$ and $|\varepsilon_2\rangle$. Consider the electron/mode state $|\varepsilon_1; n\rangle$. Without coherent interactions, the electron would dissipate some of its energy to the background reservoir, relaxing to the nearby state $|\varepsilon_3\rangle$ at the rate $\Gamma_{C12}$ (the transition $|\varepsilon_1\rangle \rightarrow |\varepsilon_3\rangle$ is suppressed because of the larger energy difference). On the other hand, the vibronic transfer $|\varepsilon_1; n\rangle \rightarrow |\varepsilon_2; n+1\rangle$ provides a coherent shortcut bypassing the thermal electron-only transition $|\varepsilon_1; n\rangle \rightarrow |\varepsilon_2; n\rangle$ (Fig. 2), allowing $|\varepsilon_2\rangle$ to be populated at earlier times. In turn, the load-connected state $|\varepsilon_3\rangle$ will also be populated earlier.

Coherent exchange can also transfer population back from $|\varepsilon_2; n+1\rangle \rightarrow |\varepsilon_1; n\rangle$. However, the thermal transition $|\varepsilon_2; n+1\rangle \rightarrow |\varepsilon_3; n+1\rangle$ will transfer some population out of the two-level subspace, suppressing its revival in $|\varepsilon_1; n\rangle$ and enforcing directionality. Since the load is connected only to the electronic subsystem, it is insensitive to the mode. By coherently absorbing one quanta of energy, the mode thus catalyzes faster electron transfer through the network. Additionally, re-excitation of the electron can occur even before the mode has dissipated the extra energy (this typically happens at a slower rate), allowing multiple mode levels to contribute in parallel. Over time, dissipative processes on the mode will regulate the available oscillation frequencies $\omega_{\text{coh}}(n)$, suppressing excessive back-transfer. At steady-state, the interplay of all these coherent and incoherent mechanisms contributes to a higher overall load current $I_L$. In Fig. 3, we plot the...
numerical I-V characteristic for our prototype, with and without coherent coupling, demonstrating the enhanced power made possible by coherent electron-vibration evolution. Note that for the numerics, we use the full system Hamiltonian, without the RWA.

To make the quantum design principle most clear, the prototype was designed to give an exactly resonant JC-like model where neither incoherent nor coherent processes dominate. In more realistic systems, things will not be so ideal, yet the principle still holds. Even resonance-detuned modes and partially delocalized excitons can combine to produce coherent oscillations. Whenever these oscillations have non-negligible overlap with $|\epsilon_1\rangle$ and $|\epsilon_2\rangle$, they will provide beneficial alternate transfer pathways (for full details on all models, see SM).

**Linking coherence to performance.**— To quantify the influence of electron-vibration coherence, we define currents

$$I_H := \gamma_H (\rho_{\epsilon_0}^e(t)\rho_H - \rho_{\epsilon_1}^e(t)(\rho_H + 1)), \quad I_C^{\epsilon_{ij}} := \gamma_C (\rho_{\epsilon_i}^e(t)(\rho_{\epsilon_j}^e(t) + 1) - \rho_{\epsilon_j}^e(t)(\rho_{\epsilon_i}^e(t))),$$

$$I_L := \Gamma_L \rho_{\epsilon_3}^e(t). \quad (7)$$

We track the flow of energy in our heat engine using the electronic Hamiltonian $H_e$. This flow contains two contributions, due to the coherent (Hamiltonian) and incoherent (Lindbladian) parts of the master equation:

$$\frac{d}{dt} \langle H_e \rangle = i\text{Tr}(\rho(t)|H|H_e) + \text{Tr}(\mathcal{L}(\rho(t))H_e). \quad (9)$$

The incoherent part takes the form

$$\text{Tr}(\mathcal{L}(\rho(t))H_e) = \frac{I_{coh}}{Q_H} - \sum_{\epsilon_{ij}} \frac{\epsilon_{ij} I_C^{\epsilon_{ij}}} {Q_H} - \sum_{\epsilon_{ij}} \frac{\epsilon_{ij} I_L^{\epsilon_{ij}}} {Q_L} \quad (10)$$

where $Q_\alpha$ are net energy flows and $\epsilon_{ij} := \epsilon_i - \epsilon_j$ are energy differences between the various states. Without coherent interactions, the electronic populations $\rho_{\epsilon k} := \text{Tr}(\rho|\epsilon_k\rangle\langle\epsilon_k| \otimes 1_m)$ satisfy classical balance equations which dictate the value of the load current $I_L$ in steady state.

If the interaction $H_I$ is present, then $[H_S, H_e] \neq 0$, and there is an additional coherent energy exchange between the electron and the mode, $Q_{e-m} := i\text{Tr}(\rho(t)|H_S|H_e)$. Simplifying, we find

$$Q_{e-m} = \frac{1}{\varepsilon_{30}} \left[ \varepsilon_{10} I_H - \varepsilon_{12} I_{coh} - \sum_{\epsilon_i > \epsilon_j} \varepsilon_{ij} I_C^{\epsilon_{ij}} \right]. \quad (12)$$

Thus, the coherent current pushes the steady state away from the rate equation solution (involving only the incoherent terms $I_H$ and $I_C^{\epsilon_{ij}}$) expected by detailed balance arguments, thereby allowing the load current to be increased overall.

**Biological example.**— Delocalized excitons and strongly coupled near-resonant modes appear in several biological LHCs, so the design principle has wide applicability. For illustration, we consider the light-harvesting complex Phycocyanin-645 (PC645), found in the cryptophyte algae *Chroomeonas* CCMP270 (see SM for another example). Cryptophytes are noteworthy for their ability to absorb solar energy even under low-light conditions. The PC645 system contains a pair of energetically similar and strongly coupled sites (called DBV C and DBV D) $[4, 45, 46]$, leading to a delocalized exciton dimer. As well, vibrational structures are believed to be important for understanding experimentally observed coherences $[47, 48]$, giving us the second ingredient of the design principle.

We model this system using 4 electronic levels. A ground state, a coupled dimer pair, and an extra uncoupled lowest excited state based on the chromophore site PCB 158 D, which represents one of the major transfer pathways $[49]$ out of the DBV C/D dimer (the other pathways have similar energies). The energy and cou-
pling values are taken from [45, 46]. We also include quantized vibrational modes at sites 1 and 2, each with a frequency, based on the analysis of [48], which is near-resonant with the excitonic splitting. A full list of parameter values can be found in Table 1. The numerical I-V and power curves are presented in Fig. 4, showing a maximum power enhancement of 10.6%.

Conclusion.— We have outlined the quantum advantage offered by coherent electron-vibration evolution in light-harvesting systems. Strong vibrational interactions open up alternate, coherence-mediated, pathways for excitation transfer, allowing the total system to deliver energy faster than possible by incoherent thermal processes alone. This mechanism was illustrated in both an idealized prototype and for more realistic model systems of biological LHCs. Our results build a quantitative link between coherent evolution and functional advantage and inspire potential coherence-based designs for artificial light harvesting systems.

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Supplementary Material

Model details

Quantum heat engines, interacting incoherently with external sources and sinks, are naturally described as open quantum systems. Accordingly, we use a Lindblad master equation,

\[ \dot{\rho}_S(t) = i[\rho_S(t), H_S] + \sum_{\alpha} \mathcal{L}_\alpha[\rho_S(t)], \]

(13)
to describe the open system dynamics. Each Lindblad superoperator \( \mathcal{L}_\alpha \) is associated with a transition rate \( \Gamma_\alpha \geq 0 \) and a transition operator \( A_\alpha \):

\[ \mathcal{L}_\alpha(\rho_S) = \Gamma_\alpha [A_\alpha \rho_S A_\alpha^\dagger - \frac{1}{2} \{A_\alpha^\dagger A_\alpha, \rho_S\}] . \]

(14)

While other types of equations exist for describing open system evolution (e.g., Redfield equations), Lindblad-type master equations have the desirable feature that they guarantee completely positive (i.e., physically consistent) evolution of the quantum system [11,13]. For one, this means that no population can ever become negative, an issue which has been a catch in previous heat engine models based on perturbative equations (as pointed out in [20]).

For our prototype model, the system Hamiltonian \( H_S \) consists of three parts: the electronic Hamiltonian within the single excitation subspace,

\[ H_e = E_0 |\varepsilon_0\rangle\langle \varepsilon_0 | + \sum_{i=1}^3 E_i |s_i\rangle\langle s_i | + \sum_{i\neq j} J_{ij} |s_i\rangle\langle s_j | , \]

(15)

where \( |s_i\rangle \) denotes an excitation localized on site \( i \); the Hamiltonian for the quantized modes (one each at sites 1 and 2),

\[ H_m = \hbar \omega_1 \sum_{i=1}^2 \hat{a}_i^\dagger \hat{a}_i ; \]

(16)

and a linear electron-mode interaction

\[ H_I = \sum_{i=1}^2 \hbar g_i |s_i\rangle\langle s_i | \otimes (\hat{a}_i + \hat{a}_i^\dagger) . \]

(17)

The interaction of the electron with the dissipative background modes can also be thought to originate from many localized microscopic terms of this form.

Assuming similar structure at each site, we fix \( \omega_1 = \omega_2 = \omega_m \) and \( g_1 = g_2 = g \) to have the same values for each mode. We emphasize that electron-vibration coupling alone does not lead to any population/energy transfer since the interaction Hamiltonian does not connect different sites. Excitonic energy transfer only becomes possible with this interaction if the electronic eigenstates have some amount of delocalization. Thus, delocalization can be thought of as a tool for “unlocking” transfer capabilities from the available interaction. In the prototype and cryptophyte example, only excitons 1 and 2 are delocalized, and a quantized mode at site 3 would not contribute to the evolution. On the other hand, for the FMO example of the next section, the coupling between sites 2 and 3 is non-negligible. Having a quantized mode at site 3 can thus open up a second coherent pathway, which further enhances transport.

Our QHE connects to three important external systems: a “hot” source of incoming energy, a “cold” dissipative background, and an abstract load. We model each of these external systems as thermal reservoirs at temperatures \( T_H, T_C, \) and \( T_L \), respectively, with \( T_H > T_C \). To model an irreversible transition for the load, we use \( T_L = 0 \). We use the excitonic basis to study the dynamics; thus, the electronic Hamiltonian \( H_e \) does not contribute to the dynamics. Instead, the three reservoirs facilitate all possible transitions between the exciton states \( |\varepsilon_i \rangle \). Aside from the ground state \( |\varepsilon_0 \rangle \), the electronic eigenstates are ordered by their energy (\( |\varepsilon_1 \rangle \) highest, \( |\varepsilon_N \rangle \) lowest).

The hot reservoir drives transitions only between the ground state \( |\varepsilon_0 \rangle \) and the highest energy exciton state \( |\varepsilon_N \rangle \). Conversely, the load transfers the electron only from the lowest excited state \( |\varepsilon_N \rangle \) back to the ground state \( |\varepsilon_0 \rangle \). On the other hand, the dissipative cold background reservoir causes transitions between all pairs of excitons. This includes not only beneficial transitions \( |\varepsilon_i \rangle \to |\varepsilon_j \rangle \) (\( i, j \neq 0 \)) which assist transport through the excited state network, but also deleterious transitions \( |\varepsilon_i \rangle \to |\varepsilon_0 \rangle \) (\( i \neq 0 \)) which represent unwanted but physically important decays of excitons back to the ground state without giving their energy to the load.

The interaction of the QHE with each of the thermal reservoirs is parameterized by two quantities, namely a coupling \( \gamma_\alpha, \alpha \in \{ H, C, L \} \), and the reservoir’s mean occupation number \( n_{\alpha ij}, i, j \in \{ 0, 1, \ldots, N \} \), at the given transition energy \( \varepsilon_{ij} = \varepsilon_i - \varepsilon_j \). Because the hot reservoir and the load only link two levels, we can omit the redundant \( ij \) indices for these. The full set of employed Lindblad terms is as in the main text:

| \( \mathcal{L}_H \) | \{transition \( A_\alpha \), rate \( \Gamma_\alpha \)\} |
|---|---|
| \( \mathcal{L}_{H_{\varepsilon_0}} \) | \{\( |\varepsilon_0\rangle |\varepsilon_0\rangle, \gamma_H |\pi_H\rangle \} |
| \( \mathcal{L}_{c_{\varepsilon_{ij}}} \) | \{\( |\varepsilon_i\rangle |\varepsilon_i\rangle, \gamma_C (|\pi_{c_{ij}}\rangle + 1) \} |
| \( \mathcal{L}_{L_{\varepsilon_0}} \) | \{\( |\varepsilon_0\rangle |\varepsilon_0\rangle, |\varepsilon_3\rangle, \gamma_L \} |

Except for \( \mathcal{L}_H \), all forward transitions proceed to lower energies, indicating the desired directionality for the QHE circuit. For the hot reservoir, we fix the occupa-
tion $\pi_H = 60000$ to represent concentrated solar energy, in line with previous work [20]. The thermal occupations of the cold reservoir are determined from the Planck distribution

$$\pi_{Cij} = \frac{1}{\exp(\epsilon_{ij}/(k_BT_C)) - 1}. \quad (18)$$

To make the load transition irreversible, we have fixed $T_L = 0$, so the load has $n_L = 0$. Thus, the Lindblad rate $\Gamma_L$ and the coupling $\gamma_L$ are equal. Different impedances of the load are modelled by varying the parameter $\Gamma_L$.

The remaining ingredients of the model are the quantized modes. Numerically, we truncate the Hilbert space of each mode at some fixed dimension $D = N_{\text{max}} + 1$. For all examples except the FMO, $D = 6$; for the FMO example, we used $D = 4$ because the added numerical expense of including the mode at site 3. Physically, the modes should also undergo relaxation, though at a somewhat slower rate than the electronic subsystem. Accordingly, we include additional Lindblad terms $E_{M}^{\downarrow \uparrow}$ for each quantized mode. As with the electrons, these terms represent local interaction with a thermal reservoir, and are parameterized by couplings $\gamma_M$ and mean bath occupations $\pi_M$ (assumed to be the same at each site). The Lindblad rates are thus $\Gamma_M = \gamma_M(\pi_M + 1)$ (damping) and $\Gamma_M = \gamma_M\pi_M$ (excitation), while the transition operators are simply the ladder operators $A_M^\dagger = \hat{a}_i$ (damping) and $A_M^\dagger = \hat{a}_i^\dagger$ (excitation). We have set $\gamma_M$ to give a rate $1/(1\text{ ps})$. The occupation numbers $\pi_M$ are calculated using a Planck distribution (Eq. (18)) at temperature $T_M$ for a level spacing $\epsilon_{i+1,i} = \hbar\omega_m$. The specific numerical parameter values for all examples are listed in Table I.

Finally, we remind that although the RWA is helpful for elucidating the underlying mechanisms of coherent energy transfer, all numerics are done without making the RWA. Even when the RWA is valid for a closed electron/vibration system, the evolution also contains a number of Lindblad transitions which will interact with the Hamiltonian evolution. The energy scales and interplay from the open system evolution may make the omitted RWA terms non-negligible, and we therefore use the unsimplified Hamiltonian in the simulations.

### Another LHC example

The design principle we have presented relies primarily on two ingredients: delocalization of excitons and near-resonant modes. These conditions can be found in several biological LHCs. We have already demonstrated the benefit for the cryptophyte PC645 in the main text; we now provide a further example: the FMO light-harvesting complex [51], found in green sulfur bacteria. A single FMO unit consists of 7 sites, with two main transfer pathways [12,13,52]. We focus on the pathway containing sites 1-3, which qualitatively resembles our prototype system, with sites 1/2 forming a delocalized dimer (energies/couplings taken from [53]). An interesting distinction from the prototype is that sites 2 and 3 also have non-negligible coupling, allowing for a second coherent transfer pathway $|\varepsilon_2\rangle \rightarrow |\varepsilon_3\rangle$. We add quantized modes at sites 1-3 with frequency $\omega_m = 180 \text{ cm}^{-1}$ [52], which is close to resonance with both excitonic transitions $\varepsilon_{12}$ and $\varepsilon_{23}$. The performance of the three-site FMO model is presented in Fig. 5. We see a modest enhancement of power when coherent coupling is included, despite the relatively high employed incoherent transfer rate, $\gamma_C = 37 \text{ cm}^{-1}$. Around the maximum power point, coherent transfer pathways for both $|\varepsilon_1\rangle \rightarrow |\varepsilon_2\rangle$ and $|\varepsilon_2\rangle \rightarrow |\varepsilon_3\rangle$ contribute to an overall power enhancement of 2.2%.

| Prototype | PC645 | FMO |
|-----------|-------|-----|
| $E_1$     | 300   | 1226| 310 |
| $E_2$     | 300   | 1145| 230 |
| $E_3$     | 0     | 0   | 0   |
| $E_0$     | -10000| -1588| -12195|
| $E_{12} = E_{21}$ | 100 | 319.4 | 94.8 |
| $E_{13} = E_{31}$ | 0 | 0 | 5.5 |
| $E_{23} = E_{32}$ | 0 | 0 | 29.8 |
| $\hbar\omega_m$ | 200 | 807 | 180 |
| $\hbar\gamma$ | 55 | 200 | 55 |
| $\pi_H$ | 60000 | 60000 | 60000 |
| $T_C$     | 293   | 293 | 293 |
| $T_M$     | 293   | 293 | 293 |

TABLE I. Numerical parameters used in the examples, both from the main text (prototype and FMO) and Supplementary Material (PC645). Energies, couplings, and rates are in units of cm$^{-1}$, mean phonon numbers are unitless, and temperatures are in K.

Non-ideal situations

The idealized prototype in the main text is meant to reveal the design principle and its advantages most clearly. Yet it is important to show that the design principle can work even in less idealized situations. To this end, we will examine separately three sources of imperfection. These are quantized modes which are detuned from the excitonic transition frequencies, excitonic states which are only partially delocalized, and additional decoherence effects beyond those induced by the thermal transitions. For all, we take the same basic Hamiltoni-
and eigenenergies

\[ \varepsilon_{\pm n} := \hbar \omega_m (n + \frac{1}{2}) \pm \frac{1}{2} \Omega_n, \]

where \( \Omega_n := \sqrt{4\hbar^2 g^2(n + 1) + \delta^2} \) is the Rabi frequency and \( \tan(\phi_n) = 2h g \sqrt{n + 1}/\delta \). Using the same intuition as the prototype model (Eqs. (15)-(17)). As before, the system evolution is described by a Lindblad master equation, with hot, cold, and load transitions occurring incoherently between the relevant excitonic eigenstates. Possible modifications of the system-bath interactions in these non-ideal cases, which should be based on a more detailed microscopic model, are beyond the scope of the present work. Finally, any parameter values which are not explicitly mentioned in the following examples are the same as in Table I.

**Off-resonant vibrations:** Another deviation from the ideal situation is when there are large mismatches between the exciton energy splitting and the frequency of the quantized vibrations. Similar to the partial delocalization example, such mismatches can suppress the coherent current, but will not destroy it altogether. To see this, we consider our prototype model, but with a mode frequency which is detuned by \( \delta \) from the transition energy, i.e., \( \hbar \omega_m = \varepsilon_{12} - \delta \). The interaction of Eq. (17) and the remaining parameters from Table I are kept the same as the prototype.

When the rotating wave approximation is valid, the total Hamiltonian \( H_S = H_c + H_m + H_I \) (a detuned Jaynes-Cummings model) can be diagonalized to obtain entangled electron-vibration eigenstates

\[ |\pm; n\rangle = \cos(\frac{\phi_n}{2}) |\varepsilon_1; n\rangle \pm \sin(\frac{\phi_n}{2}) |\varepsilon_2; n + 1\rangle \]  

\[ |-; n\rangle = \sin(\frac{\phi_n}{2}) |\varepsilon_1; n\rangle - \cos(\frac{\phi_n}{2}) |\varepsilon_2; n + 1\rangle \]

and eigenenergies

\[ \varepsilon_{\pm n} := \hbar \omega_m (n + \frac{1}{2}) \pm \frac{1}{2} \Omega_n, \]

where \( \Omega_n := \sqrt{4\hbar^2 g^2(n + 1) + \delta^2} \) is the Rabi frequency and \( \tan(\phi_n) = 2h g \sqrt{n + 1}/\delta \). Using the same intuition as the main text, for every level \( n \), the Hamiltonian will cause coherent rotations on the Bloch sphere defined for the two states \( \{|\varepsilon_1; n\rangle, |\varepsilon_2; n + 1\rangle\} \). In the ideal prototype, population was rotated directly along the equator; with detuning, the axis of rotation will point somewhere else on the sphere. As long as \( \phi_n \) is not an integer multiple of \( \pi \) (which happens when the coupling \( g \) is zero or the detuning \( \delta \) is infinitely far away), this rotation will still lead to partial population oscillations between the levels \( |\varepsilon_1; n\rangle \) and \( |\varepsilon_2; n + 1\rangle \). We also note that detuning modulates the rotation speed, which can in some situations lead to faster overall transfer even with a tilted axis of rotation. We plot the I-V and power curves of this detuned system in Fig. 6 with \( \delta = 50 \text{ cm}^{-1} \), demonstrating that the design principle continues to yield a power enhancement in this situation. As in the main text, we do not make the RWA in the actual numerics.

**Partial delocalization:** Electronic delocalization was important in our prototype because it enabled the quantized vibrations to accept energy from the electronic subsystem under the given interaction. Delocalization effects can be examined by varying the dipole energies \( E_{1/2} \) and coupling \( J_{12} \) from the prototype model. In general, the eigenstates in the 1-2 subspace are \( |\varepsilon_1\rangle = \cos(\frac{\theta_J}{2}) |s_1\rangle + \sin(\frac{\theta_J}{2}) |s_2\rangle \) and \( |\varepsilon_2\rangle = \sin(\frac{\theta_J}{2}) |s_1\rangle - \cos(\frac{\theta_J}{2}) |s_2\rangle \), where \( \tan(\theta_J) := \frac{2J_{12}}{E_{12} - E_2} \) captures the degree of delocalization. The associated eigenenergies are \( \varepsilon_{1/2} = \frac{1}{2}(E_1 + E_2 \pm \sqrt{(E_1 - E_2)^2 + 4J_{12}^2}) \). With respect to these eigenstates, the interaction Hamiltonian becomes \( H_I = H_{12} + H_{RD} \), where
the states

Since the design principle depends on coherence between the states $|\varepsilon_1; n\rangle$ and $|\varepsilon_2; n + 1\rangle$, the Lindblad operator

$$H_{12} = g(|\varepsilon_1\rangle\langle\varepsilon_1| \otimes \cos(\frac{\theta_J}{2})(\hat{a}_1 + \hat{a}_1^\dagger) + \sin(\frac{\theta_J}{2})(\hat{a}_2 + \hat{a}_2^\dagger)) + |\varepsilon_2\rangle\langle\varepsilon_2| \otimes \sin(\frac{\theta_J}{2})(\hat{a}_1 + \hat{a}_1^\dagger) + \cos(\frac{\theta_J}{2})(\hat{a}_2 + \hat{a}_2^\dagger))\]}

$$H_{RD} = \sqrt{2}g\sin(\frac{\theta_J}{2})\cos(\frac{\theta_J}{2})(|\varepsilon_1\rangle\langle\varepsilon_2| + |\varepsilon_2\rangle\langle\varepsilon_1|) \otimes (\hat{a}_RD + \hat{a}_RD^\dagger).$$

So long as the delocalization angle $\theta_J$ is not an integer multiple of $\pi$ (which represents completely localized excitons), this interaction will still lead to coherent population oscillations between $|\varepsilon_1\rangle$ and $|\varepsilon_2\rangle$ via Eq. (13), with the amplitude of the oscillations depending on the degree of delocalization via the prefactor $\sqrt{2}g\sin(\frac{\theta_J}{2})\cos(\frac{\theta_J}{2})$. But we can see that a coherent current will be active, and the quantized modes contributing to the dynamics, for any non-zero amount of delocalization. In Fig. 7 we plot the I-V and power curves for our prototype with $E_1 = 310$ cm$^{-1}$, $E_2 = 290$ cm$^{-1}$, and $J_{12} = 50$ cm$^{-1}$. These values lead to partially delocalized excitons with $\cos(\frac{\theta_J}{2}) \sim 0.83$ and $\sin(\frac{\theta_J}{2}) \sim 0.56$, yet we still gain a power enhancement with the quantized modes.

*Additional decoherence:* Finally, we consider the influence of decoherence on our prototype. As we saw in the FMO example, strong interactions with the outside baths can suppress the flow of coherent current via quantum Zeno mechanisms, i.e., by applying the incoherent transition operators too frequently. Yet there may be other sources of decoherence present in the system, for instance due to microscopic details not accounted for in our master equation. To study this potential, we introduce an extra decoherence mechanism phenomenologically via a Lindblad term $\mathcal{L}_{\text{decoh}}$ in the master equation.

Since the design principle depends on coherence between the states $|\varepsilon_1; n\rangle$ and $|\varepsilon_2; n + 1\rangle$, the Lindblad operator

$$\mathcal{L}_{\text{decoh}} = \gamma_{\text{decoh}} \sum_{n=0}^{N_{\text{max}}} \mathcal{L}_{\text{decoh}}^{(n)},$$

where each $\mathcal{L}_{\text{decoh}}^{(n)}$ has a transition operator

$$A^{(n)} = |\varepsilon_1; n\rangle\langle\varepsilon_2; n + 1| - |\varepsilon_2; n + 1\rangle\langle\varepsilon_1; n|.$$  

We show the I-V and power curves for the decoherence rate $\gamma_{\text{decoh}} = 1.0$ eV in Fig. 8 where we can see that the power enhancement is only partially suppressed by decoherence.

*Quantum advantage for a model with optimum incoherent transport*

The design principle outlined in this paper relies on the interplay of three primary components. These are incoherent baths, delocalized electronic states, and coherently coupled vibrational modes. The incoherent baths provide directionality through driving and damping; the delocalized electrons allow for transfer to even take place at all given the local form of the baths; and the coherently coupled modes increase the overall rate of transfer by offering alternate pathways. Clearly, the design principle will work best when neither the incoherent nor the coherent processes dominate. However, in our idealized model, the possibility exists to turn up the coupling $\gamma_C$ to the cold bath until incoherent transport speeds dominate coherent oscillation frequencies. In such a regime, the coherent vibrational coupling would operate on too slow a timescale to provide any quantum advantage. Of course, one could also increase the coupling $g$ to the expotional vibrations in the model until the quantum nature of evolution again provided an advantage. While our idealized
FIG. 9. a) Maximum power point versus $\gamma_C$ in a model with bath-induced relocalization, both with (solid lines) and without (dashed lines) coherent coupling to the quantized modes. In an appreciable region around the optimum, the coherently coupled modes provide higher maximum power than is possible for any choice of bath strength parameter $\gamma_C$.

The model allows for this kind of parameter manipulation, doing so is not really in the spirit of what is happening physically.

We would expect that our prototype model only describes the physical system well for some appropriate range of parameters. In particular, we should not expect the physics to be the same for all bath strengths. When the bath strength dominates over the electronic degrees of freedom, the preferred electronic states which undergo transitions will be effectively relocalized [40], since the underlying environmental interaction is typically thought to be itself local,

$$H_{SE} = \sum_{i=1}^{N} \sum_k \hbar g_{ik} |s_i\rangle\langle s_i| \otimes (\hat{a}_{ik} + \hat{a}_{ik}^\dagger),$$

where the $\hat{a}_{ik}$ represent modes only at site $i$ (cf. [17]). But the more localized the preferred electronic states, the less an interaction of the form of Eq. 23 can actually facilitate transitions between them. Thus, bath-induced relocalization can suppress the actual transition rates when the bath strength goes beyond some optimal value.

We model this situation by introducing a background-induced localization angle $\theta_B := \arctan(\frac{\alpha}{\gamma_C})$, where $\alpha = 1$ meV is a constant (this should be distinguished from the delocalization angle caused purely by electronic coupling). The relocalized excitonic states are

$$|\varepsilon_1(\theta_B)\rangle := \cos(\frac{\theta_B}{2}) |s_1\rangle + \sin(\frac{\theta_B}{2}) |s_2\rangle,$$

$$|\varepsilon_2(\theta_B)\rangle := \sin(\frac{\theta_B}{2}) |s_1\rangle - \cos(\frac{\theta_B}{2}) |s_2\rangle,$$

with $|\varepsilon_3(\theta_B)\rangle = |s_3\rangle$ and $|\varepsilon_0(\theta_B)\rangle = |\varepsilon_0\rangle$ as before. When $\gamma_C \ll \alpha$, the excitons are completely delocalized, while if $\gamma_C \gg \alpha$, they become localized. Finally the Lindblad transition operators are modified to take place between the $\theta_B$-exciton states with new rates $\gamma_C(\theta_B) = \gamma_C \sin^2(\theta_B)$ which are weighted by the degree of localization. As well, the bath occupations $n_{Cij}(\theta_B)$ will also depend on $\theta_B$ since the energy differences for Eq. (18) are modified by the delocalization angle. In summary, the Lindblad components are exactly as in the earlier table, except with the replacements $|\varepsilon_i\rangle \rightarrow |\varepsilon_i(\theta)\rangle$, $\gamma_C \rightarrow \gamma_C(\theta_B)$, and $n_{Cij} \rightarrow n_{Cij}(\theta_B)$.

Numerically, we determine the steady state of this incoherent transport model for various values of the coupling parameter $\gamma_C$, first with no quantized vibrations. We show the maximum power point for each value of $\gamma_C$ in Fig. 9 (dashed curve). As expected, the incoherent model has an optimal choice of $\gamma_C$, with relocalization effects suppressing the power after this optimum. We then add in quantized modes at sites 1 and 2, at frequency $\hbar \omega_m = 200$ cm$^{-1}$. The max power points for this situation are given by the solid curve in Fig. 9. It is clear that even if we are at the regime of optimal incoherent transport, the inclusion of coherently coupled vibrations provides an additional quantum advantage to the power of the light-harvesting system.