

Delocalized Excitation Transfer in Open Quantum Systems with Long-Range Interactions

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The interplay between coherence and system-environment interactions is at the basis of a wide range of phenomena, from quantum information processing to charge and energy transfer in molecular systems, biomolecules, and photochemical materials. In this work, we use a Frenkel exciton model with long-range interacting qubits coupled to a damped collective bosonic mode to investigate vibrationally assisted transfer processes in donor-acceptor systems featuring internal substructures analogous to light-harvesting complexes. We find that certain delocalized excitonic states maximize the transfer rate and that the entanglement is preserved during the dissipative transfer over a wide range of parameters. We investigate the reduction in transfer caused by static disorder, white noise, and finite temperature and study how transfer efficiency scales as a function of the number of dimerized monomers and the component number of each monomer, finding which excitonic states lead to optimal transfer. Finally, we provide a realistic experimental setting to realize this model in analog trapped-ion quantum simulators. Analog quantum simulation of systems comprising many and increasingly complex monomers could offer valuable insights into the design of light-harvesting materials, particularly in the nonperturbative intermediate parameter regime examined in this study, where classical simulation methods are resource intensive.

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I. INTRODUCTION

Understanding how quantum systems interact with their surrounding environment and how coherence and quantum correlations affect the nonequilibrium dynamics in the presence of decoherence and dissipation remains one of the outstanding challenges in quantum information science, condensed matter physics, and physical chemistry. The delicate interplay of dissipative and coherent processes in driven-dissipative quantum systems can lead to non-trivial out-of-equilibrium dynamics [1], dissipative phase transitions that do not always have an equilibrium counterpart [2–5], and dissipative engineering of correlated many-body steady states [6–16].

Open quantum systems are also used to model the transfer of excitations coupled to nuclear vibrations in

the presence of a complex environment, which occurs in charge transfer (CT) and energy transfer (ET) phenomena in various systems in molecular electronics [17], biomolecules [18,19], perovskite photochemical cells [20], and bilayer materials [21]. In these systems, environmental fluctuations and vibrations play a crucial role in assisting the transfer of excitations to the lowest-energy state by dissipating energy and destroying coherent superpositions, ensuring irreversible transfer [22,23]. For example, the role of vibronic coherence and delocalization has been suggested to be connected to the fast transfer of excitons in light-harvesting complexes [24–27] and organic semiconductors [28,29].

CT and ET in the presence of a complex environment can be generally described by Frenkel exciton models [30–33] that employ discrete Hilbert spaces to encode the electronic states along with harmonic oscillators to describe molecular and solvent vibrations. Recently, it has been shown that both electronic and vibrational degrees of freedom can be directly mapped onto state-of-the-art analog quantum simulators based on trapped atomic ions [34] and superconducting qubits [35]. These platforms allow for precise control of the coherent part of the evolution [36–42] and, in some cases, also provide turning knobs

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to tailor the system-bath coupling implementing reservoir engineering [43–46].

In this work, we theoretically and numerically investigate a minimal Frenkel exciton model, where the donor and acceptor have an internal substructure [47] similar to light-harvesting complexes in which different monomers feature strongly coupled molecular sites. Here, the molecular sites are encoded in collections of two-level systems, among which the excitation can be shared. The two-level systems are coupled to a damped collective bosonic mode, where the damping arises from the presence of an Ohmic bath.

Previous studies focused on the incoherent transfer among weakly coupled monomers [47] or coherent exciton transfer with nearest-neighbor couplings [48]. Conversely, in this work, we study a regime in which intermonomer coherences play a key role in the excitation transfer, and the molecular sites possess long-range interactions. The model studied here singles out the role of delocalization in excitation transfer and its robustness against static disorder in the energy landscape and the vibronic couplings, as well as the effect of the temperature. We also generalize this setting for a larger number of monomers and for monomers with increasingly complex internal substructures.

In addition to its fundamental theoretical interest, this model is also motivated by being experimentally realizable on an analog trapped-ion simulator, where the electronic degrees of freedom can be encoded in the internal atomic states, and the molecular vibrations in the collective vibrations of the ions around their equilibrium positions (see Sec. VI). Finally, in this work, we provide experimentally accessible parameters to realize this model in a trapped-ion simulator using a chain of qubit and coolant ions [44].

This paper is organized as follows. In Sec. II, we introduce the model for excitation transfer. Section III focuses on the transfer rate of an initially delocalized excitation in a dimer, where each monomer is realized by two ions, both in the perturbative and nonperturbative regimes. Additionally, we investigate the effect of static disorder and noise on the transfer rate. In Sec. IV, we investigate the effect of temperature on transfer rates. In Sec. V, we discuss how this model can be extended to longer chains with more monomers and with monomers composed of more than two qubits. In Sec. VI, we propose an experimental implementation of the model in a trapped-ion simulator. Finally, in Sec. VII, we summarize our findings and provide an outlook for future studies.

II. MODEL

We consider a one-dimensional system of N spin-1/2 qubit ions interleaved with M coolant ions, which are used to perform reservoir engineering on the shared bosonic mode [44], as shown in Fig. 1(a). The qubit ions interact via long-range spin exchange interactions J_{ij} [49] and are

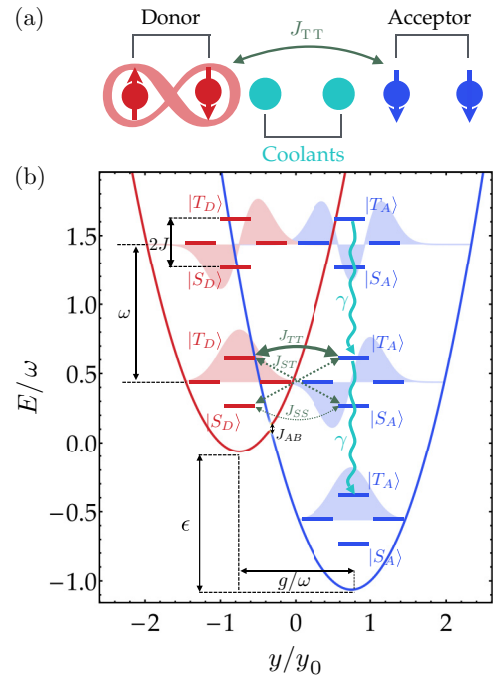


FIG. 1. Model and trapped-ion implementation. (a) A chain of four qubit ions and two coolants can realize the Frenkel exciton model (see the main text). The red and blue ions on the sides represent donor and acceptor monomers, respectively, and the information in each ion is encoded in a qubit. The two cyan ions in the middle represent the cooling ions used for the sympathetic cooling of the ion chain motion. (b) Pictorial representation of the spectrum of the total Hamiltonian in the symmetric case, described by Eqs. (10) and (12). Donor (red) and acceptor (blue) surfaces are shown as a function of the normalized reaction coordinate $y/y_0 = 1/2(a^\dagger + a)$ with their respective noninteracting harmonic wave functions. The effect of the bath is illustrated by the cyan wavy arrows that represent the relaxation with rate γ . The qubits in the donor and acceptor sites are strongly coupled and can be described in terms of local triplet and singlet states $|T_{D,A}\rangle, |S_{D,A}\rangle$ separated by an energy $2J$. These states are, in turn, coupled by intermonomer couplings J_{AB} , where $AB = TT, ST, SS$ (see the main text). The intermonomer couplings J_{AB} induce anticrossings between the two surfaces, allowing the donor-to-acceptor transfer.

subject to local energy shifts ϵ_i . At the same time, the two-level systems are coupled to the collective bosonic mode defined by creation (annihilation) operators $a^\dagger$ (a) with frequency ω , resulting in the total Hamiltonian

$$\begin{aligned}
 H_s &= H_{\text{el}} + H_v + H_{\text{el-v}} \\
 &= \left(\sum_i^N \frac{\epsilon_i}{2} \sigma_i^z + \sum_{i<j}^N J_{ij} (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+) \right) \\
 &\quad + \omega a^\dagger a + \sum_i^N \frac{g_i}{2} \sigma_i^z (a^\dagger + a). \tag{1}
 \end{aligned}$$

Hamiltonian H_{el} encodes the electronic degrees of freedom with the energy landscape that determines the roles of the molecular sites and their mutual electronic couplings. We assume a power-law decay for the electronic couplings $J_{ij} = J/d_{ij}^p$ that can be natively realized in trapped-ion systems with $J > 0$ [49]. Here, d_{ij} is the distance between the qubit ions and p is the power-law exponent. In the study of energy transfer in molecular systems, the interaction between donor and acceptor electronic states is often found to be mediated by dipole-dipole interactions with $p = 3$. However, it has been broadly discussed that the power-law exponent p depends on the separation of the molecular sites and its relative scale with respect to the dimensions of the molecular sites [50–53], with p in the range of $1 \leq p \leq 6$ for the distinct cases. Here, we choose $p = 1$ and small intermonomer distances (similar in magnitude to the intramonomer distances) because this configuration is more readily realizable in experiments with trapped ions ($0.5 \lesssim p \lesssim 2$; see Sec. VI below), and it leads to a faster numerical convergence than $p > 1$. However, in Appendix B, we comment that very similar behavior is expected for $p = 3$ and larger intermonomer distances, up to an overall slowdown of the dynamics.

Moreover, systems in $\mathcal{D}$ dimensions with $p \leq 2\mathcal{D} + 1$ have generated a lot of interest recently because deriving tight bounds on the ultimate speed of quantum information transfer is challenging in this regime [54–58]. Hamiltonian H_v describes a collective vibrational mode of motion that can be encoded in a normal mode of the ion chain, defining a reaction coordinate $y/y_0 = 1/2(a^\dagger + a)$, with $y_0 = \sqrt{\hbar/2m\omega}$ and m the ion mass [59]. The reaction coordinate determines the progress of a given process or reaction; in this case, the value of the reaction coordinate helps us differentiate between the different sites, where the excitation can be located, namely, the donor or acceptor sites [see Fig. 1(b)]. Finally, $H_{\text{el-v}}$ represents the interaction between the electron and vibration with site-dependent couplings g_i .

The total Hamiltonian in Eq. (1) conserves the total number of spin excitations $\Sigma_z = \sum_i \sigma_i^z$. In this work, we focus on the *single excitation manifold*, where the allowed number of spin states is reduced to N . For example, in the two-monomer case with two sites per monomer (see Fig. 1), the two donor states are given by $|\uparrow\downarrow\downarrow\downarrow\rangle$ and $|\downarrow\uparrow\downarrow\downarrow\rangle$, while the two acceptor states are given by $|\downarrow\downarrow\uparrow\downarrow\rangle$ and $|\downarrow\downarrow\downarrow\uparrow\rangle$.

Here, we consider a single, damped bosonic mode to model the relaxation driven by the interaction with the environment. This setting can be realized experimentally in trapped-ion systems through the process of sympathetic cooling [44,59] (see Sec. VI below for details). The system dynamics is described by a master equation with Lindbladian superoperators $\mathcal{L}_c[\rho]$, where c is a generic jump operator:

$$\frac{\partial \rho}{\partial t} = -i[H_s, \rho] + \gamma(\bar{n} + 1)\mathcal{L}_a[\rho] + \gamma\bar{n}\mathcal{L}_{a^\dagger}[\rho], \quad (2)$$

$$\mathcal{L}_c[\rho] = c\rho c^\dagger - \frac{1}{2}\{c^\dagger c, \rho\}. \quad (3)$$

Here, ρ is the total density matrix of the system, γ is the motional relaxation rate, and $\bar{n}$ is the phonon population determined by the temperature of the bath $k_B T = \omega/\log(1 + 1/\bar{n})$. This description is motivated by the fact that, under the assumption of weak damping ($\gamma \ll \omega$), where the relaxation rate is weaker than both the system's vibrational ($\gamma \ll \omega$) and bath thermal energies ($\gamma\beta \ll 1$, with $\beta = 1/k_B T$), the dynamics of the spin and the bosonic observables predicted by Eq. (2) are a very good approximation of those of a system in contact with an Ohmic bath described by the spectral density $J(\omega) \sim \omega$, a common choice in electron transfer literature [60–62].

III. TWO-MONOMER MODEL

Let us consider a model with only two monomers, which has been used as a minimal model of charge transfer in photosynthetic complexes [22,23,63–65]. In this case, each monomer is composed of two sites and separated by $M = 2$ coolant ions (see Sec. VIB below), as shown in Fig. 1. In this case, we can use the following notation:

$$\begin{aligned} |1\rangle &= |\uparrow\downarrow\downarrow\downarrow\rangle, & |2\rangle &= |\downarrow\uparrow\downarrow\downarrow\rangle, \\ |3\rangle &= |\downarrow\downarrow\uparrow\downarrow\rangle, & |4\rangle &= |\downarrow\downarrow\downarrow\uparrow\rangle. \end{aligned} \quad (4)$$

Expressed in this basis, J_{ij} is given by

$$J_{ij} = \begin{pmatrix} 0 & J & J/4^p & J/5^p \\ J & 0 & J/3^p & J/4^p \\ J/4^p & J/3^p & 0 & J \\ J/5^p & J/4^p & J & 0 \end{pmatrix}. \quad (5)$$

We note that apart from their dissipative role, the number of coolants also determines the distance between neighboring monomers, in this case, $d_{23} = 3$. We refer to the nearest-neighbor terms $J_{12}=J_{34}=J$ as the intramonomer couplings since they connect two donor (acceptor) states. All other J_{ij} terms correspond to intermonomer couplings. In the configuration shown in Fig. 1, the dominant intermonomer coupling is $J_{23} = J/3^p$. The choice of two coolant ions separating monomers is motivated by the following reasons: (i) it imposes a hierarchy in the electronic configuration where the intermonomer couplings are smaller than the intramonomer ones [47] without making the former too small; (ii) it is more convenient to have even chains in the experimental implementation proposed here (see Sec. VI below). We keep the coolant configuration when considering longer chains in Sec. V below. In the following, we study the transfer of a single excitation from donor sites to acceptor sites as a function of different Hamiltonian parameters.

For simplicity, we consider the symmetric case $\epsilon_1 = \epsilon_2 = -\epsilon_3 = -\epsilon_4 = \epsilon$. With this choice, both donor and acceptor states are shifted symmetrically with respect to zero energy, and the energy gap between donor and acceptor sites is ϵ . This configuration facilitates the irreversible transfer into the acceptor sites in the dissipative dynamics. Unless specified otherwise, we consider $p = 1$ in the following. The main goal of this analysis is to understand whether and, if so, which entangled delocalized initial donor states enable faster transfer compared to the initial product states.

A. Perturbative regime

We first consider the perturbative regime in which the electronic coupling is weak compared to the other energy scales in the model ($J \ll g_i, \epsilon, \omega$). In this case, the total Hamiltonian in Eq. (1) can be divided as $H_{\text{tot}} = H_0 + H_J$, where the electronic coupling defined by Eq. (5) is the perturbation, and the unperturbed Hamiltonian is given by

$$H_0 = \frac{\epsilon}{2}(|1\rangle\langle 1| + |2\rangle\langle 2| - |3\rangle\langle 3| - |4\rangle\langle 4|) + \omega a^\dagger a + \sum_{j=1}^4 \frac{g_j}{2} (a + a^\dagger) |j\rangle\langle j|. \quad (6)$$

The unperturbed Hamiltonian (6) is diagonal in the basis $\{|j\rangle\}$, and thus the unperturbed eigenstates are determined by the diagonalization of the bosonic part of the Hamiltonian:

$$H_{Bj} = \omega a^\dagger a + \frac{g_j}{2} (a + a^\dagger). \quad (7)$$

As shown in Appendix A, this Hamiltonian corresponds to a displaced harmonic oscillator with displacement $\alpha_j = -g_j/2\omega$ [see Fig. 1(b)]. The eigenstates of the bare system are $|\psi_{j,n}\rangle = |j\rangle \otimes |n, \alpha_j\rangle$, where $|n, \alpha_j\rangle$ denote the Fock states of the displaced operator at site j , and states $|j\rangle$ are defined in Eq. (4). The eigenenergies of H_0 are therefore given by

$$E_{j,n} = n\omega - \frac{g_j^2}{4\omega} + \frac{\epsilon_j}{2}, \quad (8)$$

where n is the number of bosonic excitations. It is convenient to define the reorganization energy for the j th site $\lambda_j = g_j^2/\omega$, which quantifies the amount of energy needed to displace the motional wavepacket by g_j/ω without any excitation transfer.

Since H_J is a perturbation, we can express the transfer rate between the donor and acceptor states using the Fermi golden rule (FGR) [59,60]. Considering an initial state $|\psi_{i,0}\rangle$ with zero bosonic excitations, the transfer rate

to a final state $|\psi_{f,n}\rangle$ (see Appendix A) can be expressed in the zero temperature case as

$$\frac{k_T^{if}}{\gamma} = \sum_{n=0}^{\infty} \frac{|J_{if}|^2}{(\Delta E_{if})^2 + \gamma^2/4} \left(\frac{\exp(-\tilde{n}) \tilde{n}^n}{n!} \right), \quad (9)$$

where the Franck-Condon factor $|\langle n, \alpha_f | 0, \alpha_i \rangle|^2$ follows a Poissonian distribution determined by $\tilde{n} = (\alpha_f - \alpha_i)^2 = (g_f/2\omega - g_i/2\omega)^2$. Here, the dissipation is taken into account by considering a Lorentzian broadening of the delta functions in the original FGR [59,60]. The total transfer rate to the acceptor site is defined by the sum over all possible acceptor states $|\psi_{f,n}\rangle$ as $k_T^{\text{FGR}} = \sum_{f \in A} k_T^{if}$. In this regime, the transfer is maximized when the energy is conserved, namely, when $\Delta E_{if} = E_{f,n} - E_{i,0} = 0$, which leads to the resonance condition $\epsilon = n\omega + (g_f^2/4\omega - g_i^2/4\omega)$.

In order to gain an intuitive understanding of the role of delocalized states in the transfer process, we choose a specific configuration for the values of g_j . Since we want to identify states $|1\rangle$ and $|2\rangle$ to be in the donor site and states $|3\rangle$ and $|4\rangle$ to be in the acceptor site, they need to be associated with different values (displacements) of the reaction coordinate. Because the coupling of the electronic states to the vibrational mode is proportional to g_j , we thus set $g_1 = g_2 = g \neq g_3 = g_4 = \tilde{g}$, to differentiate between donor and acceptor states. This means that donor and acceptor sites have their energy surfaces displaced by different amounts [see Fig. 1(b)]. For this parameter choice, we can include the intramonomer nearest-neighbor coupling terms in the unperturbed Hamiltonian, that is,

$$H_0 = \omega a^\dagger a + \frac{\epsilon}{2}(|1\rangle\langle 1| + |2\rangle\langle 2| - |3\rangle\langle 3| - |4\rangle\langle 4|) + \frac{1}{2}(a + a^\dagger)(g(|1\rangle\langle 1| + |2\rangle\langle 2|) + \tilde{g}(|3\rangle\langle 3| + |4\rangle\langle 4|)) + J(|1\rangle\langle 2| + |2\rangle\langle 1| + |3\rangle\langle 4| + |4\rangle\langle 3|). \quad (10)$$

For any choice of g and $\tilde{g}$, the unperturbed vibrational eigenstates are unchanged, and the spin eigenstates are now defined by the triplet and singlet excitonic states of the donor and acceptor sites, respectively:

$$\begin{aligned} |T_D\rangle &= \frac{1}{\sqrt{2}}(|1\rangle + |2\rangle), & |S_D\rangle &= \frac{1}{\sqrt{2}}(|1\rangle - |2\rangle), \\ |T_A\rangle &= \frac{1}{\sqrt{2}}(|3\rangle + |4\rangle), & |S_A\rangle &= \frac{1}{\sqrt{2}}(|3\rangle - |4\rangle). \end{aligned} \quad (11)$$

For simplicity, we set $g = -\tilde{g}$ to reduce the number of parameters and comment on the effect of different values of these parameters in Appendix B. The new eigenstates can still be defined as $|\psi_{j,n}\rangle$ with $j = T_D, S_D, T_A, S_A$. In this excitonic basis, the FGR in Eq. (9) considers

indices $i = T_D, S_D$ and $f = T_A, S_A$. These eigenstates have energies identical to those in Eq. (8) with the difference that the triplet (singlet) states are shifted by $+J$ ($-J$). A pictorial representation of the energetics in this new basis is provided in Fig. 1(b). In this new notation, the perturbation Hamiltonian H_J now corresponds to all the intermonomer coupling terms [see the green arrows in Fig. 1(b)]:

$$H_J = J_{TT}(|T_D\rangle\langle T_A| + \text{H.c.}) + J_{ST}(|S_D\rangle\langle T_A| + \text{H.c.}) \\ + J_{TS}(|T_D\rangle\langle S_A| + \text{H.c.}) + J_{SS}(|S_D\rangle\langle S_A| + \text{H.c.}). \quad (12)$$

We note that there are no intramonomer couplings in this basis, thanks to the values chosen for g_j . The coupling strengths are explicitly given by

$$J_{TT} = \left(\frac{1}{2} \frac{J}{3^p} + \frac{J}{4^p} + \frac{1}{2} \frac{J}{5^p} \right), \quad (13a)$$

$$J_{TS} = -J_{ST} = \left(\frac{1}{2} \frac{J}{3^p} - \frac{1}{2} \frac{J}{5^p} \right), \quad (13b)$$

$$J_{SS} = \left(-\frac{1}{2} \frac{J}{3^p} + \frac{J}{4^p} - \frac{1}{2} \frac{J}{5^p} \right). \quad (13c)$$

A plot of the ratio of these intermonomer couplings as a function of p is shown in Fig. 7 in Appendix B. For values of $p \leq 3$, the coefficient J_{TT} is the dominant term, with its value being significantly larger than both J_{ST} and J_{SS} . This means that if the system is initialized in a triplet donor state $|\psi_{T_D,0}\rangle$, the triplet-to-triplet transfer will be faster than that associated with any initial donor product state since J_{TT} is greater than any individual intermonomer coupling. Additionally, because both triplet states, $|T_D\rangle$ and $|T_A\rangle$, have their energies shifted by $+J$, the resonance condition remains $\epsilon = n\omega$, which is independent of the spin-phonon coupling strengths g_j for the current parameter choice [see Fig. 1(b)].

We check this prediction numerically by integrating Eq. (2) for different initial states up until $\omega t_{\text{sim}}/2\pi = 100$ (see Appendix C for details on the dynamics of the bosonic degrees of freedom and truncation of the bosonic Hilbert space). In Figs. 2(a)–2(b), we show the transfer rate k_T from donor to acceptor as a function of ϵ for $J = 0.03\omega$, which is well within the perturbative regime. The transfer rate obtained numerically is defined in terms of the population in the donor state $P_D = \langle (|1\rangle\langle 1| + |2\rangle\langle 2|) \rangle$ as [44,59,66]

$$k_T = \frac{\int_{t=0}^{\infty} P_D(t) dt}{\int_{t=0}^{\infty} t P_D(t) dt}. \quad (14)$$

This definition can characterize the transfer rate for arbitrary donor population dynamics and depends on both the speed at which the system reaches the steady state and

the value of the steady-state population itself. In the special case when the donor population dynamics follows an exponential decay, we can also define an equilibration rate Γ as

$$P_D = (1 - P_D^{\text{SS}}) \exp(-\Gamma t) + P_D^{\text{SS}}, \quad (15)$$

where P_D^{SS} is the donor population in the steady state. Differently from the transfer rate k_T , the equilibration rate Γ characterizes only how fast the steady state is reached, independent of the specific value of P_D^{SS} . For a complete population transfer ($P_D^{\text{SS}} = 0$) and exponential decay, the two rates coincide.

Figure 2(a) shows the expected peaks in the transfer rate at the resonances $\epsilon = n\omega$ for triplet, singlet, and product states. The relative height of the peaks is determined by the Franck-Condon factor, which, following the Poissonian distribution dependent on g and ω [see Eq. (9)], peaks at $\tilde{n}=1$ since $g = \omega$ for all the simulations. We note that we choose this value of g to explore the regime in which the donor and acceptor sites are described by displaced potential energy surfaces with quantized vibrational levels [see Fig. 1(b)]. The weakly coupled regime with $g/\omega \lesssim 0.1$ has been explored before; see, for example, Refs. [36,67]. The numerically obtained transfer rates qualitatively agree with those predicted by the FGR, as shown by the black solid line (note that we display $\tilde{k}_T^{\text{FGR}} = k_T^{\text{FGR}}/5$ in the plot). However, the FGR predicts a higher transfer rate, which can be attributed to the fact that this expression is derived under the assumption that $\gamma \gg J$ [66]. This condition differs from the parameters used in panel (a), where $J = 0.03\omega$ and $\gamma = 0.015\omega$.

Remarkably, in the perturbative regime, the transfer rate of a symmetric spin triplet superposition ($|T_D\rangle$) is significantly higher than the transfer rate starting in either an antisymmetric singlet superposition ($|S_D\rangle$) or a product state $|2\rangle$. This observation can be understood as follows: since the transfer rate is proportional to J_{ij}^2 in the perturbative FGR regime, the $|T_D\rangle \rightarrow |T_A\rangle$ transfer is more efficient than the $|S_D\rangle \rightarrow |S_A\rangle$ case by a factor proportional to $(J_{TT}/J_{SS})^2 \gg 1$. Similarly, the triplet state will transfer faster than the most favorable product state $|2\rangle$ by at least $(J_{TT}/J_{23})^2 > 1$ (see Appendix B).

We note that the roles of triplet and singlet states are determined by the sign of the nearest-neighbor electronic coupling $J > 0$ and that they can be exchanged by programming the electronic couplings to have alternating signs ($J_{ij} \rightarrow (-1)^{|i-j|} |J_{ij}|$) [68].

In Fig. 2(b), we show the transfer dynamics from the initial state $|\psi_{T_D,0}\rangle$ for the three main resonances $\epsilon = \omega, 2\omega, 3\omega$. The donor population P_D shows an approximately exponential decay, as expected from Eq. (9), into the acceptor state. Importantly, Fig. 2(b) also shows $1 - P_{T_A}$ (signaled by x markers), where $P_{T_A} = \langle |T_A\rangle\langle T_A| \rangle$, confirming that the transfer into the acceptor is a coherent

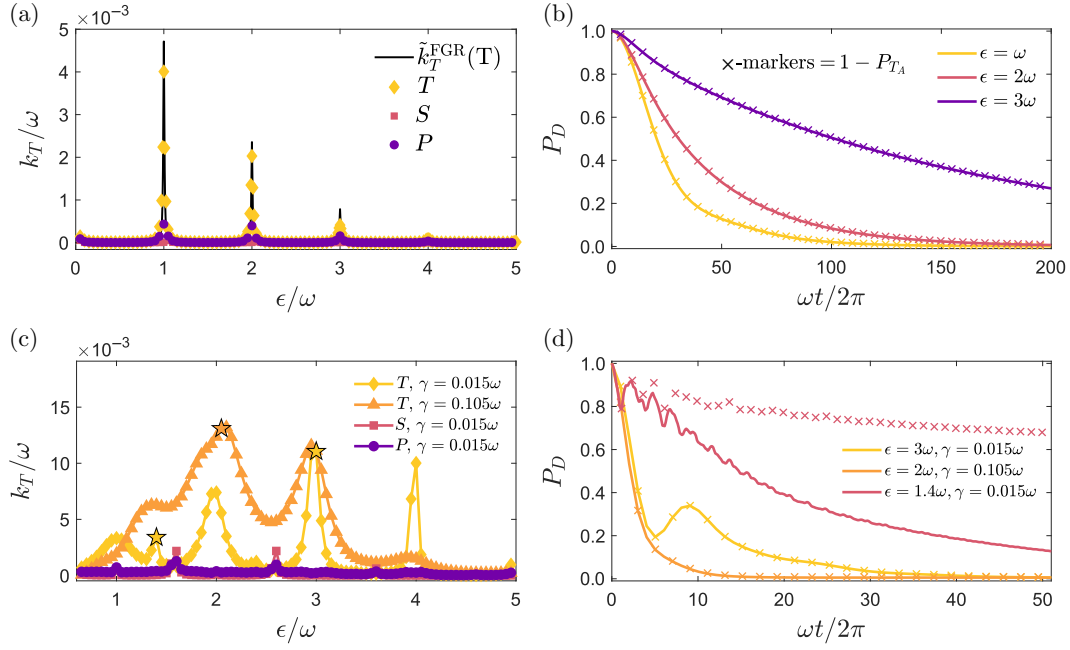


FIG. 2. Excitation transfer: transfer rate k_T in the perturbative (a),(b) and nonperturbative regimes (c),(d). (a) Transfer rate k_T as a function of ϵ in the perturbative regime ($J, \gamma \ll \omega$) for the different initial states $|T_D\rangle$ (T), $|S_A\rangle$ (S), and $|I\rangle$ (P). The black solid line represents the transfer predicted for initial state $|T_D\rangle$ using the Fermi golden rule $\tilde{k}_T^{\text{FGR}} = \frac{1}{5}k_T^{\text{FGR}}$, where we rescale by a factor of $\frac{1}{5}$ to display it clearly in the same plot. (b) Dynamics at the highest resonances of (a). Solid lines represent the population in the donor sites P_D , while the x markers illustrate the behavior of $1 - P_{T_A}$ with P_{T_A} being the population of $|T_A\rangle$. All curves correspond to the initial state $|T_D\rangle$. The parameters for (a) and (b) not specified in the plots are $g = \omega$, $J = 0.03\omega$, $\gamma = 0.015\omega$, $p = 1$, and $\bar{n} = 0.01$. (c) Transfer rate k_T as a function of ϵ in the nonperturbative regime ($J \sim \lambda$) for different initial conditions and parameter values. The parameter sets of data points marked by stars are analyzed further in panel (d). (d) Dynamics of the donor population for the parameters signaled by stars in (c). Solid lines and x markers are defined as in (b). The parameters for (c) and (d) not specified in the plots are $g = \omega$, $J = 0.3\omega$, $p = 1$, and $\bar{n} = 0.01$.

process in which not only the population but the full quantum state is transferred to the acceptor site. Given that $|T_D\rangle$ and $|T_A\rangle$ are maximally entangled states of the qubits in the donor and acceptor, respectively, the dynamics can also be understood in terms of entanglement transfer; see Appendix D for more details. It is worth noting that, when starting in the triplet state, the system behaves approximately as a two-level system defined by the $\{|T_D\rangle, |T_A\rangle\}$ subspace. In this electronic two-level description, an initial state $|\psi_{T_D,0}\rangle$ is coupled to state $|\psi_{T_A,n}\rangle$ through the electronic coupling J_{TT} when $\epsilon = n\omega$. Decay through the relaxation rate γ couples the latter to lower-energy vibrational states $|\psi_{T_A,m}\rangle$ with $m < n$. Eventually, the system decays to state $|\psi_{T_A,0}\rangle$ where, in the case of near-zero temperature, it gets trapped as this state is off resonant with respect to any donor state (see Sec. IV below for the effects of finite temperature). For the population to remain trapped in $|\psi_{T_A,0}\rangle$, it is necessary that $\epsilon > 2J$; otherwise, state $|\psi_{S_D,0}\rangle$ could get populated even if this process is slower (rate proportional to J_{ST}^2). We adopt the condition $\epsilon > 2J$ throughout this work.

For all these reasons, the results in the nonperturbative regime presented in Fig. 2(a) show good agreement with

previous studies on electron-transfer models for a single donor and acceptor site [44,59]. The more complex structure of the monomer, however, plays an important role when the system is outside the perturbative regime, which will be addressed in the next section.

B. Nonperturbative regime

When the intermonomer coupling becomes comparable to the reorganization energy $J \sim \lambda$, the intermonomer coupling cannot be treated as a perturbation. This nonperturbative regime can be accessed by increasing J or decreasing the power-law exponent p . In Fig. 2(c), we consider the first case as we take $J = 0.3\omega$ while keeping p fixed. The role of p will be discussed in more detail in the following sections and in Appendix B.

In this regime, we note that the resonances at $\epsilon = n\omega$ are still present despite the relative height not following the pattern expected from the effective vibronic couplings determined by the Franck-Condon factors. Additionally, new resonances appear at noninteger values of ω . These resonances can be understood as a transfer from an initial state $|\psi_{T_D,0}\rangle$ to a final state $|\psi_{S_A,n}\rangle$. The energy difference

between such states is given by $\Delta E_{if} = n\omega - 2J$, which, for the value of $J = 0.3\omega$ considered here, yields resonances at $\epsilon/\omega = 0.4, 1.4, 2.4$, and so on. This is confirmed numerically in Fig. 2(c) (see the yellow diamonds). Similarly, if the system is initialized in state $|\psi_{SD,0}\rangle$, the transfer to state $|\psi_{TA,n}\rangle$ becomes resonant at values $\epsilon/\omega = 0.6, 1.6, 2.6, \dots$ (pink squares). Although these processes are still slower than the triplet-to-triplet transfer, the height difference between the triplet-triplet and the singlet-triplet peaks is smaller than the ratio J_{TT}^2/J_{TS}^2 , illustrating the departure from the perturbative description of the dynamics. To understand this behavior in depth, we show the time dynamics of the transfer for different values of ϵ in Fig. 2(d). In the case of $\epsilon = 1.4\omega$ (pink lines and markers), the total donor population is not predicted only by the population in the triplet acceptor ($P_D \neq 1 - P_{TA}$), which means that the population is transferred to both the triplet and singlet acceptor states. This is the result of the competition between two processes: first, the transfer from $|\psi_{TD,0}\rangle$ to $|\psi_{TA,n}\rangle$ features a larger coupling ($\propto J_{TT}$), but the energy difference between them is finite since ϵ is not an integer of ω and acts as an effective detuning; second, the transfer from $|\psi_{TD,0}\rangle$ to $|\psi_{SA,n}\rangle$ has a lower coupling ($\propto J_{TS}$), but the states are resonant. The combination of these two processes results in an overall higher transfer rate k_T compared to that expected in the perturbative regime.

Similarly, the triplet-to-triplet transfer is favored for values of ϵ being integers of ω both from a large transfer rate and a resonant energy, making the transfer at these parameter values considerably faster than any other configuration with initial singlet or product states. As shown in Fig. 2(d), at these resonances, the triplet state gets fully transferred to the acceptor site, as signaled by the x markers ($P_D \simeq 1 - P_{TA}$).

In Eq. (9), we incorporated the motional relaxation rate γ as an effective broadening of the transfer resonances. In the nonperturbative regime, the effect of γ is similar, as shown numerically in Fig. 2(c) with orange triangles. We note that all resonances broaden as γ increases, with the peaks slightly shifting from the expected perturbative values. Comparing the results for low and high γ (triangles and diamonds), we note that, for certain resonances, an increase of γ enhances the transfer rate ($\epsilon = 2\omega$), while, for other cases, it has the opposite effect in the $\epsilon = 4\omega$ case [see also Fig. 2(d), yellow line and markers]. This behavior poses the question of how we can optimize the transfer rate k_T by modifying the different parameter values, which will be discussed next.

C. Optimal transfer, static disorder, and noise

As shown in Figs. 2(c) and 2(d), the ratio between the electronic coupling J_{TT} and the relaxation rate γ is a crucial parameter in determining the transfer rate in the nonperturbative regime. Simply increasing the electronic

coupling J_{TT} might induce oscillations between the donor and acceptor before the steady state is reached, therefore reducing the transfer rate. On the other hand, setting a larger relaxation rate γ broadens the vibrational levels, making the resonant process less prominent. Consequently, all else being equal, we expect an optimal transfer when J_{TT} and γ are of the same order, similar to a critically damped classical oscillator. This is reminiscent of what is believed to happen in light-harvesting materials, where the relaxation of the environment occurs on similar timescales as the coherent electronic couplings [26].

In Fig. 3(a), we report numerical results of the transfer rate k_T as a function of J_{TT}/γ . Each transfer rate curve is obtained by varying γ while keeping all other parameters fixed. We note that, given g and ϵ , the transfer peaks at the same value of J_{TT}/γ regardless of the specific values of J , γ , and p . The position of the peak, however, can be shifted if a different value of ϵ or g is chosen (see the purple dashed line). We note that a similar optimization behavior of the transfer was reported when considering a single donor and acceptor states [44,59].

So far, we considered a symmetric configuration with $|\epsilon_j| = \epsilon$ and $|g_j| = g$, finding that it favors the transfer from triplet to triplet states. However, it is important to consider the effect of static disorder that can impact the transfer rate by localizing the excitation either due to heterogeneity of site energies ϵ_j or inducing “dynamical localization” via the spin-phonon couplings g_j [26]. In both cases, we expect the excitons to localize and the transfer to be suppressed.

To study the effect of static disorder on the transfer rate, we consider four parameter configurations p_1, p_2, p_3 , and p_4 , signaled with markers in Fig. 3(a), and we average over 100 disorder realizations. Here p_1 represents the optimal J_{TT}/γ ratio, while p_2, p_3 , and p_4 are chosen such that they all yield almost identical transfer rates despite representing different parameter values [see the horizontal dashed line in Fig. 3(a)].

To model the static disorder, we consider $g_j = \bar{g}_j(1 + \delta_j^g)$, with $\bar{g}_1 = \bar{g}_2 = -\bar{g}_3 = -\bar{g}_4 = g$ and δ_j^g being a Gaussian random variable with zero average and standard deviation σ_g .

In Fig. 3(b), results for different values of σ_g are shown. The vertical axis is normalized with respect to the optimal value without disorder k_T^* for each parameter set p_j . As expected, when the disorder increases, the transfer rate is lowered. A site dependent g_j changes the energy of the vibronic states [see Eq. (8)], affecting the resonance condition. Furthermore, the disorder in the displacements g_j/ω associated with the donor (acceptor) sites also reduces the transfer rate by decreasing the Franck-Condon factor. The decrease in transfer rate can be understood by noting that the reorganization energies of each qubit $\lambda_j = g_j^2/\omega$ are, on average, always larger than the electronic coupling J for our parameter choice, resulting in dynamical

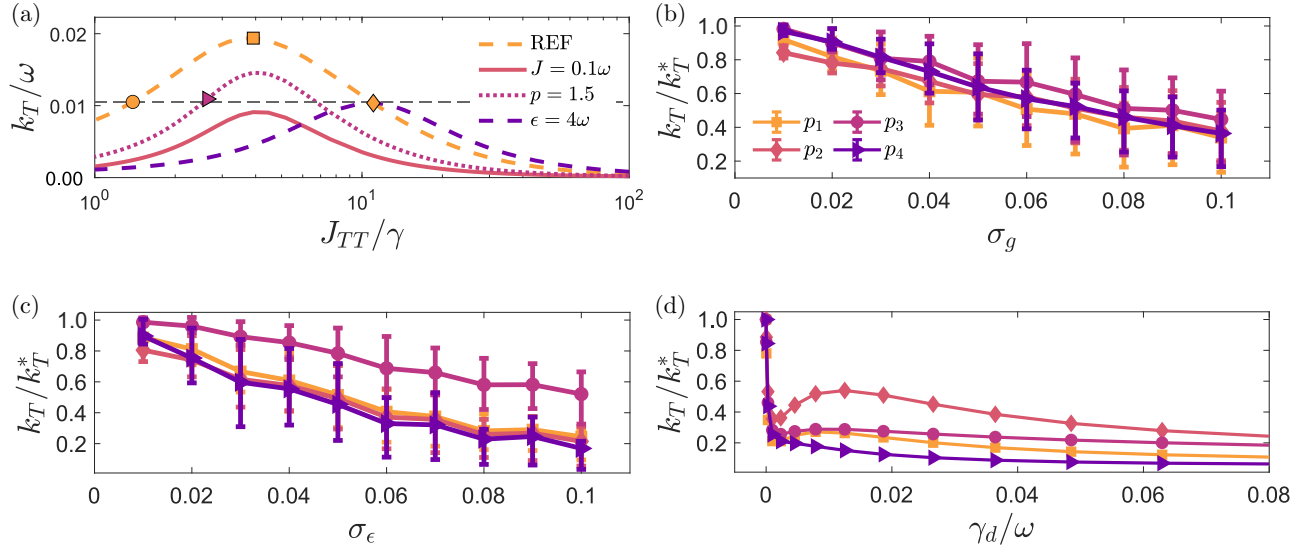


FIG. 3. Optimal transfer and static disorder. (a) The transfer rate as a function of J_{TT}/γ is presented for different parameter conditions. For each plot, J is kept fixed and γ is varied. The reference parameter set (REF in the legend) corresponds to $J = 0.3\omega$, $p = 1$, $g = \omega$, $\epsilon = 3\omega$, and $\bar{n} = 0.01$. For all other curves, one of these parameters is modified, as indicated in the legend, while all others remain the same. Four γ values are chosen for further analysis (see Table II in Appendix E for the exact values of γ). These parameter sets are labeled as p_1 , p_2 , p_3 , and p_4 , corresponding to the square, circle, diamond, and triangle markers, respectively. The horizontal dashed line is used to illustrate that p_2 , p_3 , and p_4 yield almost identical transfer rates. (b) Transfer rate as a function of the standard deviation of the static disorder in the g_j couplings. Here k_T^* represents the transfer value without disorder. Each data point is the average over 100 disorder realizations, and error bars represent the 25th and 75th percentiles. (c) Same as in (b), but for disordered on-site energies ϵ_j . (d) Transfer rate as a function of the dephasing rate γ_d . Here k_T^* represents the transfer rate value without noise. All markers and colors are defined as in (b) and (c).

localization of the excitonic states. As σ_g increases, we note a higher variance of the possible transfer rates. Indeed, in the presence of static disorder, the different g_j can favor transfer processes that are not resonant when all couplings have the same magnitude $|g_j| = g$. This may result in the prominence of slower transfer triplet-to-singlet or singlet-to-singlet processes. Moreover, certain disorder realizations can lead to a larger donor population in the steady state, decreasing the transfer rate [see the discussion after Eq. (14) and Appendix E]. Additionally, the transfer rates in the four configurations in Fig. 3(b) are all affected in a similar way by disorder in the spin-phonon coupling g_j . However, the optimal parameter set p_1 , on average, always yields a higher transfer rate in absolute terms since the vertical axis in Fig. 3(b) is normalized with respect to k_T^* .

In Fig. 3(c), we consider the case of static disorder in the on-site energies ϵ_j , and we assume an identical disorder with standard deviation σ_ϵ for each qubit. A difference in ϵ_j only modifies the energy shifts of the different states, differently from the site-dependent g_j couplings that also modify the Franck-Condon factors. In general, we expect to still have delocalized states contributing to the dynamics if the energy variations are small compared to the electronic coupling ($\sigma_\epsilon \ll J/\omega$), while a more disordered energy landscape ($\sigma_\epsilon \gg J/\omega$) localizes the excitations,

decreasing the transfer rate. Additionally, as shown in Fig. 2(c), a faster relaxation rate γ broadens the width of the resonances in the spectrum, therefore making the resonance conditions more robust against variations in ϵ_j . The results in Fig. 3(c) confirm this behavior as the parameter set p_3 (circle markers) with $\gamma \approx 0.1\omega$ exhibits a considerably slower decrease in the transfer rate than all other parameter choices with $0.01 \lesssim \gamma/\omega \lesssim 0.04$. Interestingly, at high disorder $\sigma_\epsilon = 0.1$, the parameter set p_3 with larger γ also exhibits a larger transfer k_T in absolute terms compared to the other configurations, differently from the case of small disorder, when the p_1 configuration is optimal. Consequently, for fixed electronic coupling J_{TT} , the presence of static disorder changes the relaxation rate γ that maximizes the transfer. The interplay of dissipation and disorder has also been explored in the case of Anderson localization [69] and light-harvesting complexes [70].

Apart from static disorder, time-dependent noise in the system parameters has shown to be important in different transfer setups [71–73], especially in the context of environment-assisted quantum transfer, where moderate noise strengths might enhance excitation transfer rates [70,74,75]. In our case, we incorporate noise by considering the parameters $\epsilon_j(t)$ to change in time. As shown in Ref. [76], if the fluctuations on $\epsilon_j(t)$ can be described by white-noise stochastic processes, the dynamics can be

equivalently modeled by a time-independent Hamiltonian and a set of Lindbladian jump operators of the form $\mathcal{L}_{\sigma_j^\pm}[\rho]$ [see Eq. (3)]. Here we consider the same approach and add the terms $\gamma_d \sum_{j=1}^N \mathcal{L}_{\sigma_j^\pm}[\rho]$ to the master equation (2), where the dephasing rate γ_d is proportional to the width of the white noise. Here, we consider the same dephasing rate for all qubits.

In Fig. 3(d) we show the normalized transfer rate as a function of γ_d for the four sets of parameters p_j in Table II in Appendix E. In general, dephasing leads to a slower transfer rate, as compared to the noise-free case. However, the dependence on γ_d is not monotonic for all cases. For parameter sets p_1 , p_2 , and p_3 , the transfer rapidly reaches a minimum, and then it increases again up to a local maximum. Similar cases in which the transfer rate has a nonmonotonic behavior as a function of the noise strength have been reported in similar setups [71,73]. We should note, however, that we do not observe a case for the chosen parameter sets, where the presence of noise leads to a faster transfer with respect to the noise-free case. This is expected as the transfer speed in the model studied here is enhanced due to the coherences between the qubits of each monomer, and dephasing naturally destroys these coherences.

IV. EFFECT OF TEMPERATURE

So far, we have considered the bath to be at very low temperatures ($\bar{n} = 0.01$) and assumed the system to start from the ground state of the displaced harmonic oscillator, namely, $|\psi_{j,0}\rangle$, where $j = T_D$ or $j = S_D$, which is a good approximation for a low-temperature thermal state. In this case, the picture of the population getting fully transferred in the acceptor sites in the steady state is accurate, except for the disordered cases discussed in the previous section. Nonetheless, temperature can play a key role in the transfer of excitons when thermal energies become comparable to the other energy scales of the system [77].

Here, we consider the case of higher temperatures, which affect both the steady-state population and the transfer timescales. We consider thermal energies of the order of the vibrational energy, that is, $0 < k_B T \lesssim \omega$, which translates to $0 < \bar{n} \lesssim 0.7$. We assume the initial state to be a thermal state of the form

$$\rho_j = \sum_n \frac{\bar{n}^n}{(1 + \bar{n})^{n+1}} |\psi_{j,n}\rangle \langle \psi_{j,n}|, \quad (16)$$

where $\bar{n}$ corresponds to the temperature set by the bath, and all the calculations are performed for the triplet donor state with $|\psi_{j,n}\rangle = |\psi_{T_D,n}\rangle$. We report in Fig. 4(a) the transfer rate as a function of ϵ for different temperatures. First, when the temperature increases, the transfer rate at the resonant peaks (integer values of ϵ/ω) decreases. However, it is important to point out that the transfer rates at higher

values of ϵ are more resilient to the temperature increase than those for lower ϵ .

We can understand this behavior through the schematics presented in Fig. 4(b). Initially, a thermal state [Eq. (16)] is coherently coupled to a family of states $\{|\psi_{T_A,n}\rangle\}$ with coupling strength J_{TT} . In the acceptor sites, the dissipative dynamics couples different vibrational states until a thermal distribution is reached in the steady state. Indeed, the heating term in Eq. (2) is relevant when $\bar{n}$ is close to 1.

As shown in Fig. 4(b), depending on the energy offset between the donor and acceptor, the population in the low-lying acceptor states is not coupled with the states in the donor sites because of the energy penalty. On the other hand, higher thermally populated vibronic acceptor states are resonant with the donor states, resulting in a finite donor population P_D in the steady state. What portion of the population remains in the donor sites in the steady state depends on the bath thermal distribution, determined by $\bar{n}$, and by the value of ϵ . Higher values of donor-acceptor imbalance ϵ always lead to transfers that are more robust with respect to the temperature because, if all other parameters are fixed, more energy is needed to overcome the energy barrier.

As briefly discussed in Sec. III B, the transfer rate k_T defined in Eq. (14) may differ from the equilibration rate Γ defined in Eq. (15). The transfer rate k_T can yield a lower value if (i) the speed at which the steady state is reached decreases or (ii) if the steady-state population of the donor sites P_D^{SS} increases. We show in Fig. 4(c) the time evolution of the donor population $P_D(t)$ for different temperatures to distinguish between these two causes of the decreased k_T as a function of $\bar{n}$ observed in Fig. 4(a). We note that, as expected from the schematics in Fig. 4(b), for a fixed value of $\epsilon = 3\omega$, P_D^{SS} increases as $\bar{n}$ increases. However, the time it takes to effectively reach the steady state, quantified by Γ , is mildly affected by the increase in temperature.

To highlight this point, we fit the curves in Fig. 4(c) to Eq. (15). The results of this fit are reported in Fig. 4(d), where they are contrasted with the behavior of k_T . The response of Γ to variations in $\bar{n}$ shows that the donor population equilibration rate is quite robust to temperature in the nonperturbative case.

This is especially significant in processes whose rates are more relevant than the specific values of P_D^{SS} , such as those involving a single elementary step of a more complicated charge transfer reaction. When the donor and/or acceptor are intermediate products of a multistep process, the population transfer is generally not complete, but the equilibration rate is a crucial factor in determining the total rate of the reaction [78]. Alternatively, when the donor and acceptor are coupled to large source and drain reservoirs, the equilibration rate is the main factor that defines the dynamics of the entire process [79].

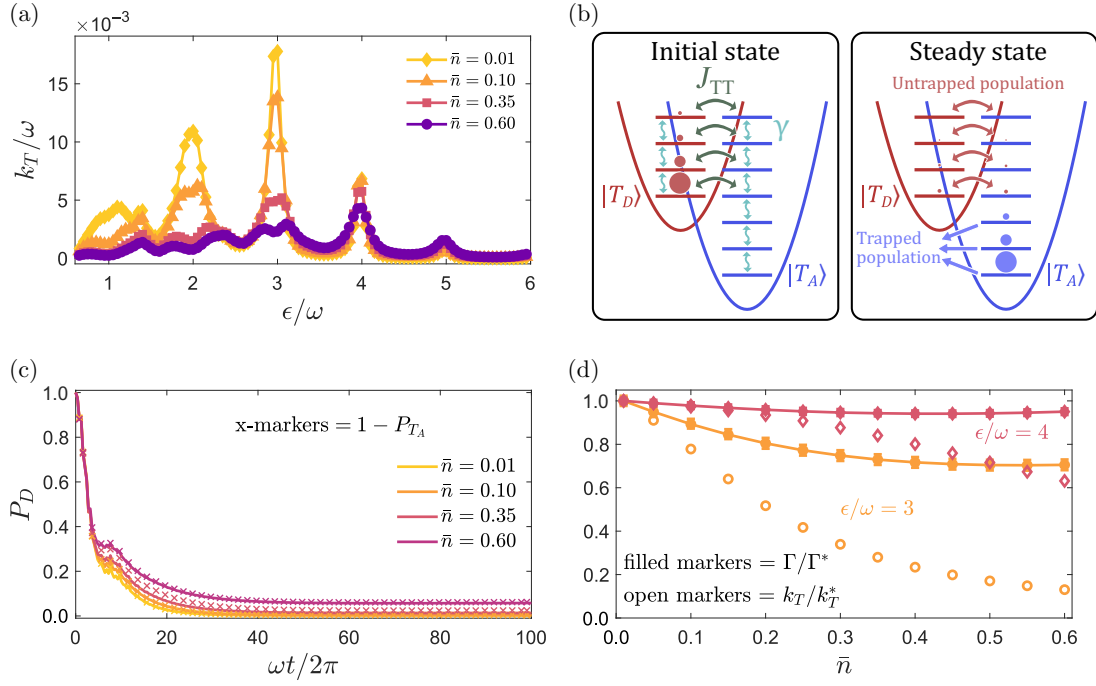


FIG. 4. Effects of temperature. (a) Transfer rate k_T as a function of ϵ for different values of $\bar{n}$. All other parameters are defined as in parameter set p_1 (see Table II in Appendix E) for this and subsequent panels. (b) Schematics of the triplet-triplet transfer for a thermal state. An initial thermal state with electronic state $|T_D\rangle$ gets coupled to the acceptor states through the coherent coupling J_{TT} . Coherent evolution and dissipative dynamics (γ) produce a thermal distribution in the acceptor site in the steady state. A portion of the population remains trapped in the acceptor, while the rest remains coherently coupled to some donor states. The ratio of these two populations depends on the value of $\bar{n}$. (c) Population of the donor as a function of time for different temperatures. We denote $1 - P_{T_A}$ with x markers. Here $\epsilon = 3\omega$. (d) Transfer rate k_T and equilibration rate Γ as a function of temperature. Open markers denote k_T/k_T^* , while filled markers denote Γ/Γ^* , with k_T^* and Γ^* being the rates at zero temperature. The error bars for filled markers denote the 95% confidence intervals of the exponential fit. Different colors signal different values of ϵ/ω .

V. HIGHER-COMPLEXITY MONOMER STRUCTURES

The model studied in the previous sections represents a simple design compared to models describing excitation transfer in biological and chemical systems, where each monomer is composed of several molecular sites, and multiple monomers participate in the excitation transfer. For example, the Fenna-Matthews-Olson complex and the light-harvesting complex II are trimers, where each monomer has 8 [80] and 12 molecules [27,81], respectively. In this section, we consider two different ways to add complexity to the model: first, we consider monomers with a larger number of two-level systems per monomer [see the inset of Fig. 5(a)]; secondly, we consider a larger number of monomers, each one composed of two qubits [inset of Fig. 5(c)].

A. Larger monomers

We start with the case where each monomer is composed of three qubits. Here, we denote the number of qubits per monomer by L (in this case, $L = 3$). The dimension of the electronic Hilbert space is $N = 2L = 6$, and it can

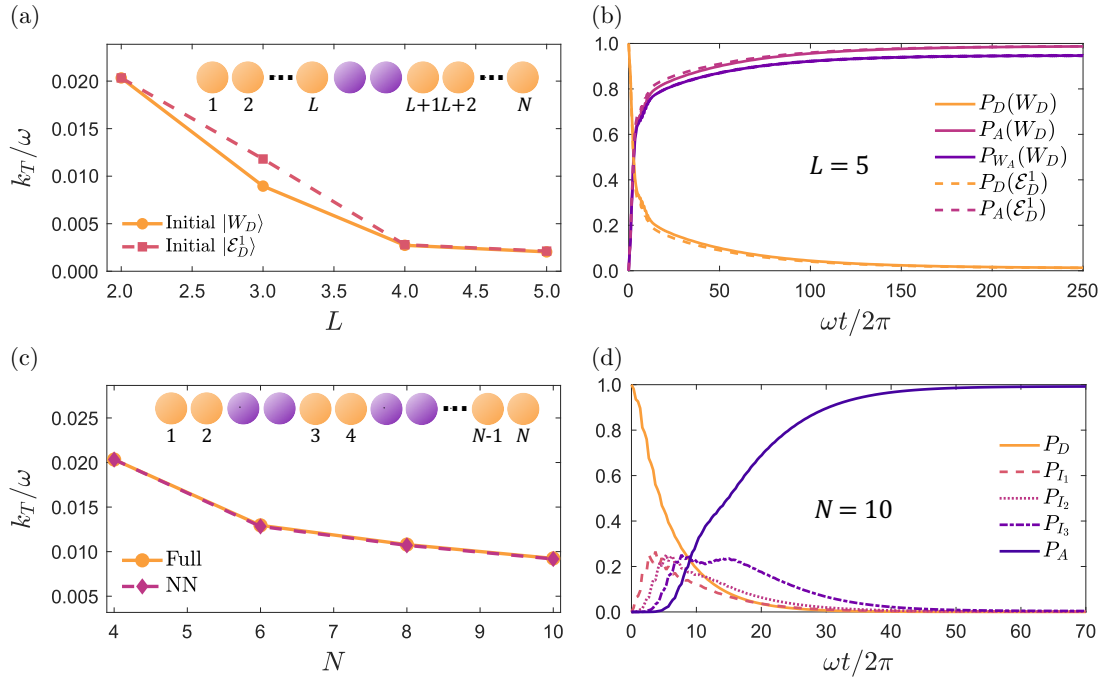
be spanned in terms of the product states with five spins down and one spin up $|j\rangle$, where $j = 1, 2, 3 \equiv j_D$ for the donor site and $j = 4, 5, 6 \equiv j_A$ for the acceptor site. We adopt a similar parameter choice as in the previous sections with $\epsilon_{jD} = -\epsilon_{jA} = \epsilon$ and $g_{jD} = -g_{jA} = g$, and we consider the same power-law decay form for J_{ij} . Note that the latter implies that now the intramonomer couplings can have different strengths, for example, $J_{12} > J_{13}$. Similar to the $L = 2$ case studied above, we can diagonalize the unperturbed Hamiltonian in the donor site while ignoring the intermonomer couplings and obtain the following eigenstates for the $p = 1$ case:

$$|\mathcal{E}_D^1\rangle = \frac{1}{\mathcal{N}_1} \left(|1\rangle + \frac{2(\sqrt{33} + 3)}{\sqrt{33} + 9} |2\rangle + |3\rangle \right), \quad (17a)$$

$$|\mathcal{E}_D^2\rangle = \frac{1}{\mathcal{N}_2} \left(|1\rangle + \frac{2(\sqrt{33} - 3)}{\sqrt{33} - 9} |2\rangle + |3\rangle \right), \quad (17b)$$

$$|\mathcal{E}_D^3\rangle = \frac{1}{\sqrt{2}} (|1\rangle - |3\rangle). \quad (17c)$$

Here $\mathcal{N}_1$ and $\mathcal{N}_2$ are normalization constants (see Appendix F). Similarly, acceptor states $|\mathcal{E}_A^1\rangle$, $|\mathcal{E}_A^2\rangle$, and $|\mathcal{E}_A^3\rangle$



can be equivalently expressed in terms of the states $|j_A\rangle$. We can write Hamiltonian (1) in this basis (see Appendix F for details). The resulting couplings can be written in the notation J_{AB} , where $A, B = \mathcal{E}^1, \mathcal{E}^2, \mathcal{E}^3$.

As shown explicitly in Appendix F, the larger of the intermonomer couplings for $p = 1$ is $J_{\mathcal{E}^1\mathcal{E}^1}$. Importantly, state $|\mathcal{E}_D^1\rangle$ has a large overlap with the W state $|W_D\rangle = (|1\rangle + |2\rangle + |3\rangle)/\sqrt{3}$. Similar to the $L = 2$ case studied previously, the transfer is also favored by an initial symmetric superposition of product states if $J_{ij} > 0$. Since the W state is a paradigmatic state in quantum information science [82] that can be efficiently prepared with digital protocols [83], we study how transfer from an initial $|W_D\rangle$ state behaves in comparison to the initial state $|\mathcal{E}_D^1\rangle$. Specifically, in Figs. 5(a)–5(b), we study how the transfer rate changes as the number of qubits per monomer L is increased. The number of eigenstates $|\mathcal{E}_D^{jD}\rangle$ increases linearly with the number of qubits in each monomer L . Here, for consistency, $|\mathcal{E}_D^1\rangle$ is defined as the state with the largest

overlap to the corresponding W state for each L . As shown in Appendix F, this state has the highest eigenenergy and always yields the largest intermonomer coupling strength for any monomer size L considered here.

In Fig. 5(a), we show the dependence of the transfer rate defined in Eq. (14) on the number of qubits per monomer L . As expected, for larger system sizes, the transfer rate decreases. For each L , the transfer from the initial state $|\mathcal{E}_D^1\rangle$ is always comparable to the W state because of the large overlap of these two states. However, since the fastest processes favor the transfer to the $|\mathcal{E}_A^1\rangle$ state (see Appendix F), the final state of the system will not be the $|W_A\rangle$ state, but it will be closer to $|\mathcal{E}_A^1\rangle$, even when the system is initialized in $|W_D\rangle$, as is illustrated in the dynamics depicted in Fig. 5(b). Given that the system is initialized in the entangled state $|W_A\rangle$ and evolves almost completely into state $|\mathcal{E}_A^1\rangle$, a large portion of the entanglement is transferred from the donor to acceptor, similar to the case of simpler monomers (see Appendix D for more details).

B. Beyond two monomers

Finally, we consider the case where the number of monomers is increased, while the number of qubits per monomer is kept fixed to $L = 2$, as shown in the inset of Fig. 5(c). In the case of $N = 6$ (three monomers), the product basis is the same as for the $L = 3$ case, namely, $|j\rangle$ with $j = 1, \dots, 6$. As each monomer is composed of only two qubits, the triplet-singlet basis is still the eigenbasis of the unperturbed Hamiltonian with

$$\begin{aligned} |T_D\rangle &= \frac{1}{\sqrt{2}}(|1\rangle + |2\rangle), & |S_D\rangle &= \frac{1}{\sqrt{2}}(|1\rangle - |2\rangle), \\ |T_I\rangle &= \frac{1}{\sqrt{2}}(|3\rangle + |4\rangle), & |S_I\rangle &= \frac{1}{\sqrt{2}}(|3\rangle - |4\rangle), \\ |T_A\rangle &= \frac{1}{\sqrt{2}}(|5\rangle + |6\rangle), & |S_A\rangle &= \frac{1}{\sqrt{2}}(|5\rangle - |6\rangle). \end{aligned}$$

Here, label I is used for the intermediate site. As we are interested in irreversible dynamics to the acceptor, we require the energy shift on the acceptor sites to be the lowest. Consequently, we choose a tilted energy configuration $\epsilon_1 = \epsilon_2 = -\epsilon_3 = -\epsilon_4 = -\frac{1}{3}\epsilon_5 = -\frac{1}{3}\epsilon_6 = \epsilon$. For the spin-phonon coupling strengths, we choose the alternating sign configuration $g_1 = g_2 = -g_3 = -g_4 = g_5 = g_6 = g$, which ensures that neighboring sites have nonzero Franck-Condon factor [see Eq. (9) and the text below]. We keep this same prescription for larger values of N , where qubits 1 and 2 always correspond to the donor site, while qubits $N - 1$ and N correspond to the acceptor site, and all other qubits correspond to different intermediate sites. An alternative configuration is considered in Ref. [48], where the spin-phonon couplings are decreasing, and the energies are staggered.

The excitation dynamics in Fig. 5(d) shows that negligible population is trapped in the intermediate states $|T_I\rangle$. This happens because a chain of irreversible transfers occurs between $|T_I, n\rangle$ and $|T_{I+1}, n'\rangle$, with $n' < n$ due to the interplay between coherent transfer and dissipation. The process is iterated until the population is trapped in state $|T_A, 0\rangle$ for zero or very low temperature. We note that, although any triplet state in one site is resonant with multiple triplet states with different numbers of phononic excitations in subsequent sites, these processes are suppressed by the power-law decay coupling strengths and the Franck-Condon factors. This setting makes the interaction between neighboring sites the dominant process. We show in Fig. 5(c) that the change of the transfer rate is negligible when setting $J_{ij} = J/d_{ij}^p = 0$ for $d_{ij} > 5$ (see the inset), which eliminates the next-nearest-neighbor couplings between monomers. However, even in this case, long-range interactions between different qubits in neighboring monomers still play a crucial role in favoring the transfer of delocalized states. It is important to note that,

for $N > 4$, it is necessary to substitute the donor population P_D by $1 - P_A$ in the definition of the transfer rate in Eq. (14).

As described above, in the absence of disorder, the dynamics for multiple monomers involves predominantly the triplet states in each site, given that the system is initialized in $|\psi_{TD,0}\rangle$. In this case, the dynamics reduces to the quantum state transfer from donor site 1 to acceptor site $N/2$ in a chain of $N/2$ sites made of N qubit ions. Recently, a bound for the optimal quantum state transfer was found in the case of a spin chain with power-law decaying interactions and conservation of the number of excitations [58]. While the model presented here fulfills those two conditions, it also considers the spatially inhomogeneous spin-phonon interaction and the dissipation induced by the bath.

The time to reach the steady state [see Fig. 5(d) for example] is always considerably larger than the optimal time $t_{\text{opt}} = \pi/J\sqrt{N}$ reported in Ref. [58]. An interesting open question is whether a state transfer tight bound can be found for power-law systems featuring spin-phonon coupling and dissipation.

VI. EXPERIMENTAL DESIGN

In this section, we show how the master equation (2) can be realized natively in a linear trapped-ion chain consisting of N site ions and M coolant ions with state-of-the-art experimental techniques. We also provide realistic experimental parameters that would allow the analog quantum simulation of the transfer dynamics studied in the previous sections with precise control over the relevant system parameters.

A. Trapped-ion model of excitation transfer

Two long-lived internal states with a frequency difference ω_0 can be used to encode the electronic state of the site ions, and, thus, the N -qubit state represents the electronic configuration of the system that can be manipulated with coherent Raman drives. At the same time, the M coolant ions can be employed to engineer the reservoir for the collective bosonic mode via sympathetic cooling by using ground-state cooling with either Raman [84] or resolved sideband one-photon transitions [44,85]. For example, the setup can be operated with either a chain of $N + M$ $^{171}\text{Yb}^+$ ions, where the laser beam arrangement assigns the N qubit and M coolant ions, or a $^{171}\text{Yb}^+ - ^{172}\text{Yb}^+$ dual-species crystal, where the $^{171}\text{Yb}^+$ ions encode the electronic state of the system, and the $^{172}\text{Yb}^+$ ions undergo cooling [44]. We note that the cooling efficiency relies on the degrees of participation between the coolant ions and the motional mode that encodes the vibrational degree of freedom in Eq. (1). For this reason, an even-numbered $N + M$ ion chain is preferred because all the ions would have nonvanishing participation in all the

collective modes of the crystal due to its mirror symmetry, which maximizes the number of modes available for vibrational mode encoding and reservoir engineering [86].

Individual Raman beams with multiple frequency beatnotes of different amplitudes and phases on the qubit ions can be used to generate the desired Hamiltonian in Eq. (1). A frequency beatnote on resonance with the qubit frequency ω_0 (carrier drive) allows for individual energy shifts to modulate the energy landscape of the system, $\sum_i \sigma_i^z \epsilon_i/2$, in the appropriate spin basis. The site-to-site electronic coupling term, $\sum_{i<j} J_{ij}(\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+)$, can be realized natively by simultaneously driving two symmetric beatnotes $\omega_0 \pm (\omega_N + \delta_J)$, where ω_N is the secular frequency of the center-of-mass collective mode. This results in a long-range Ising XX interaction with the coupling strength $J_{ij} = J/d_{ij}^p$, where p can be adjusted by changing the axial frequency of the ion chain and the detuning δ_J . The tunable range of p is between 0 and 3 [49], but the experimentally accessible range is approximately between 0.5 and 2. A strong transverse field $B \sum_i \sigma_i^z$ ($J \ll B \ll \delta_J$) can be applied to suppress the double excitation processes present in the native Ising interaction, therefore enforcing the conservation of the total number of spin excitations along the transverse field direction. This can be realized either by shifting the two symmetric sideband frequency tones used for generating the Ising interaction with a common detuning or by implementing an additional qubit frequency resonant tone, depending

on the spin basis chosen for the Hamiltonian engineering [49]. In the following section, which describes an experimental setup for simulating the two-monomer case, we demonstrate the use of the latter approach. The residual B field only adds a global offset to the energy landscape of the system and does not affect its dynamics. Similarly, both the vibrational and vibronic coupling terms of the model, $\omega a^\dagger a + \sum_i (g_i/2) \sigma_i^z (a^\dagger + a)$, can be engineered with another pair of symmetric beatnotes $\omega_0 \pm (\omega_k - \omega)$, where ω_k is the secular frequency of a chosen reservoir collective motional mode described by $a^\dagger$ (a) operators, and ω is the detuning from the reservoir motional mode with $\omega \sim g_i$ [44,87].

We note that if a single set of motional modes were used for both bichromatic drives, the approximate effective Hamiltonian stemming from the Magnus expansion of the time-ordering evolution operator would give rise to higher-order, noncommuting electronic coupling terms [88]. To mitigate this problem, we propose to use orthogonal sets of motional modes to generate the two noncommuting parts of the Hamiltonian separately [89,90]: the electronic coupling interactions can be generated by driving the higher-frequency radial mode set along the x principal axis of the trap, and the vibronic coupling interactions can be realized by using one mode from the lower-frequency radial mode set along the y principal axis of the trap (see Fig. 6) since it is standard practice in trapped-ion systems to break the degeneracy of the radial modes into the higher- and

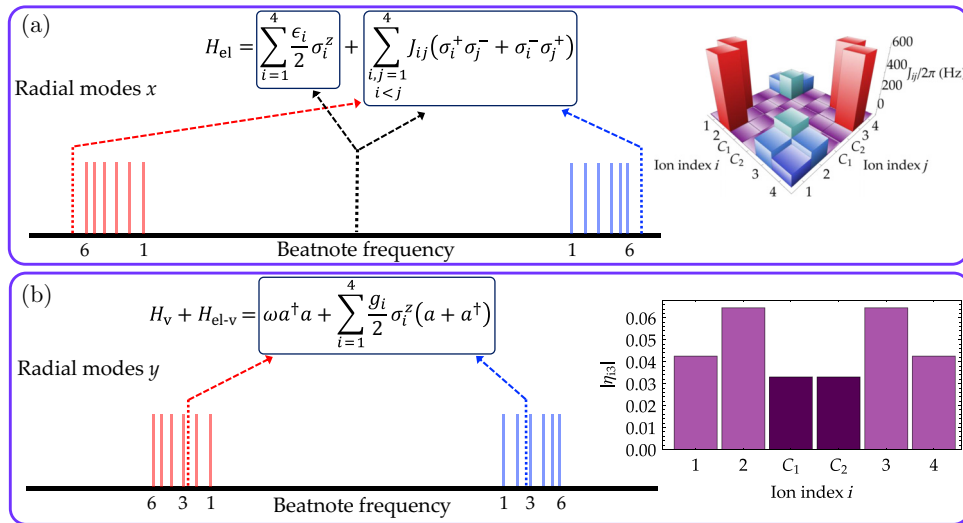


FIG. 6. Experimental configuration for simulating the two-monomer model with a chain of six $^{171}\text{Yb}^+$ ions. (a) Multifrequency drives on the qubit ions (represented by labels 1, 2, 3, and 4) to generate the electronic terms H_{el} of the system Hamiltonian and the corresponding J_{ij} matrix with $J/2\pi = 0.62$ kHz and $p \approx 1$. (b) Bichromatic drive on the qubit ions to generate the vibrational H_v and vibronic coupling $H_{\text{el-v}}$ terms using the third motional mode. The solid (dashed) vertical lines represent the motional sidebands (355-nm laser beatnote frequency tones). The J_{ij} matrix and Lamb-Dicke parameters η_{13} are calculated with the trapping condition, where the center-of-mass mode frequencies along the three principal axes are $\{\omega_6^x, \omega_6^y, \omega_1^z\} = 2\pi \times \{3.2, 2.8, 0.5\}$ MHz. Simultaneously, the two central coolant ions (denoted by C_1 and C_2) undergo continuous resolved sideband cooling on the third motional mode of the radial modes y to emulate the dissipation process of the vibrational mode of the two-monomer system.

lower-frequency sets by applying a radial static quadrupole potential upon the radial confinement potential induced by the radio-frequency drive. This choice has the advantage of ensuring spectral separation between the two drives, reducing crosstalks, and resulting in two drives having very different detunings with respect to their respective normal modes.

We also note that the proposed scheme to generate the vibronic coupling and the harmonic terms can off-resonantly couple to other collective motional modes ($m \neq k$) and induce additional two-body interactions. However, these terms result only in state-dependent shifts to the energy landscape of the system and do not affect its single electronic excitation manifold description. These energy shifts can be corrected experimentally by adjusting the single-site energy offsets (see Appendix G).

Finally, the simultaneous application of the four sideband tones in both bichromatic drives can induce ac Stark shifts, which introduce spurious terms into the engineered trapped-ion Hamiltonian, deviating away from the desired model. Therefore, in practice, one also must compensate for these shifts with a proper choice of the amplitudes and frequencies of the laser tones, which have been successfully achieved by several analog trapped-ion experiments with a similar setup using two simultaneous bichromatic tones [39,67,91].

Combining these drives leads to a trapped-ion Hamiltonian that directly maps to the model in Eq. (1) with individual control knobs on the system parameters, ϵ_i , g_i , J_{ij} , and ω , through the optical power, frequency, and phase of the individual-addressing Raman beams. Likewise, the local implementation of continuous resolved sideband cooling of the collective motional mode associated with $a^\dagger$ (a) operators on the M coolant ions generates the dissipator in Eq. (2) with tunable dissipation rate γ and bath temperature $\bar{n}$ via the optical powers and frequency detunings of the cooling laser beams [44,61,92]. Table I summarizes the experimental ranges for the relevant system parameters accessible by a standard analog trapped-ion quantum simulator, which allows vast combinations of parameters for extensive studies of the model discussed above.

TABLE I. Typical ranges for the relevant system parameters accessible by an analog trapped-ion quantum simulator. The typical average phonon number is $\bar{n} \gtrsim 0.01$.

System parameter ($\hbar = 1$)	Experimental range ($2\pi \times \text{kHz}$)
Local energy shift ϵ_i	$ \epsilon_i \leq 100$
Site-to-site electronic coupling $J_{ij} = J/d_{ij}^p$	$\lesssim 1$ with $0.5 \lesssim p \lesssim 2$
Vibrational energy ω	2–40
Vibronic coupling g_i	$\lesssim 40$
Dissipation rate γ	$\lesssim 5$

B. Two-monomer model

The experimental demonstration of our results on the two-monomer case in Sec. III can be realized with the setup in Fig. 6. The simulator consists of six $^{171}\text{Yb}^+$ ions trapped at the center-of-mass secular frequencies of $\{\omega_6^x, \omega_6^y, \omega_1^z\} = 2\pi \times \{3.2, 2.8, 0.5\}$ MHz along the three principal axes, where z is the axial direction of the trap. We note that $\alpha = x, y, z$ in ω_m^α is different from β in operator σ_i^β , which describes the spin basis in the Bloch sphere. This trapping configuration would provide an average spacing between the neighboring ions of approximately $3.50 \mu\text{m}$. In the driven rotating frame, where two $\pi/2$ pulses are employed to map the z spin basis of Eq. (1) onto the y basis of the trapped-ion Hamiltonian, the combination of a bichromatic 355-nm Raman drive with frequency beatnotes of $\omega_0 \pm (\omega_6^x + \delta_J)$ and a carrier drive at the beatnote frequency of ω_0 on the qubit ions induces a power-law decay Ising Hamiltonian with a transverse field, $\sum_{i<j} J_{ij} \sigma_i^x \sigma_j^x + \sum_i B \sigma_i^z$, where ω_0 is the frequency difference between the clock hyperfine states of $^{171}\text{Yb}^+$ that represent the qubit, and $J_{ij} = J/d_{ij}^p$. For the experimentally feasible values of $\{J, B, \delta_J\} = 2\pi \times \{0.62, 6.4, 66\}$ kHz, the condition $J \ll B \ll \delta_J$ is sufficiently satisfied, resulting in the spin-hopping interactions $\sum_{i<j} J_{ij} (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+)$ with $p \approx 1$. Moreover, another carrier drive is used to generate the energy landscape of the system, and the combined interactions become the electronic terms of the two-monomer Hamiltonian.

As mentioned above, using a suitable mode from the other radial mode set to generate the vibrational and vibronic coupling terms eliminates higher-order, noncommuting electronic coupling terms from the Magnus expansion of the time-evolution operator. To realize our results, it is important to balance the vibronic coupling strengths ($|g_i| = g$) among the qubit ions. Since $|g_i| = |\eta_{ik}| \Omega_i$, where Ω_i is the Rabi coupling strength to ion i and depends on the laser beam power, the choice of mode k must consider the overall absolute differences among the Lamb-Dicke parameters $|\eta_{ik}|$ of the qubit ions, which determine the degree of participation of ion i to the motional mode k . Hence, the required laser powers to compensate for the different couplings are reduced by choosing a mode k that has the least differences among the participation coefficients $|\eta_{ik}|$. A more balanced participation of the ions to the mode of choice also leads to a more efficient sideband cooling on the coolant ions. Although the center-of-mass mode seems like an obvious choice for this criteria due to its intrinsic balanced participation coefficients $|\eta_{i6}| = \eta/\sqrt{6}$, it is the most sensitive mode to electric field noise that typically limits the motional coherence time of trapped-ion experiments. Therefore, in the case of six ions, the best option for this scheme is mode 3 [see Fig. 6(b)] that can be driven with a bichromatic 355-nm Raman drive with frequency beatnotes of $\omega_0 \pm (\omega_3^y - \omega)$,

appropriate phases, and offset-corrected laser powers to generate the vibrational and vibronic coupling terms of the two-monomer Hamiltonian with realistically achievable $\omega = g = |\eta_{i3}|\Omega_i = 2\pi \times 2$ kHz.

Throughout the simulation, it is possible to use individual laser cooling beams on the two central ions to continuously remove the system's vibrational excitations. This can be achieved, for example, via continuous resolved sideband cooling on the third motional mode in the y direction of the trap axes using the narrow-linewidth optical transition of $^2S_{1/2} \leftrightarrow ^2D_{5/2}$ at 411 nm along with a repumper at 1650 nm [93].

In conclusion, owing to the tunability of the trapped-ion system, it will also be possible to investigate the Frenkel exciton model studied in this paper experimentally by singling out the individual roles of the system parameters at the site level (ϵ_i , g_i , J_{ij} , and ω), bath properties (γ and $\bar{n}$), and different electronic configurations in governing the transfer dynamics over wide ranges of parameters. The extension to longer chains with multiple intermediate sites or more complex monomer structures in our proposed setup can be achieved by adding more ions and more coolants. For long chains, a combination of quadratic and quartic axial potentials [94] can be used to achieve quasiuniform ion spacing, decreasing crosstalk in addressing and detection. For example, a quantum simulator of 28 $^{171}\text{Yb}^+$ ions can be used to simulate six monomers composed of three qubit ions each and interleaved with two coolant ions. The center-of-mass secular frequencies can be set to $\{\omega_{28}^x, \omega_{28}^y, \omega_1^z\} = 2\pi \times \{3.2, 2.8, 0.238\}$ MHz, leading to an average spacing between the neighboring ions of approximately $3.50 \mu\text{m}$, similar to the proposed setup for simulating the two-monomer model. With appropriate laser powers, the system parameter values of $\{J, B, \delta_J, p, \omega, g\} = 2\pi \times \{0.58, 5.38, 50, 1.28, 2, 2\}$ are experimentally accessible for realizing the dynamics of vibrationally assisted exciton transfer in a six-monomer system.

As an alternative strategy to scale up to larger systems, it is possible to use qudits instead of qubits, where we encode the monomer's electronic configuration with more than two internal states of the ions [92]. However, this comes at the cost of engineering complex electronic interactions with additional lasers to create the desired Hamiltonian.

Lastly, a key component of the simulation of more complex monomers proposed here is the preparation of W states (or states with a large overlap with W states), which is achievable through digital [83] or analog methods [95]. This hybrid digital-analog approach is also ideal for implementing the recently proposed simulation of two-dimensional spectroscopy using a probe qubit [96], which significantly reduces measurement overhead compared to standard digital protocols [97].

VII. DISCUSSION AND OUTLOOK

In this work, we characterized the role of entanglement and delocalization in long-range interacting spin chains interacting with an engineered reservoir. We found that the spin-spin interaction defines a specific class of entangled states that optimize the transport of excitations in the presence of an engineered environment. This conclusion still holds when accounting for static disorder, noise, and thermal excitations.

Our work is motivated by very recent experimental advances [36,37,44,46,91] and aims at providing a blueprint for the analog quantum simulation of Frenkel exciton models based on trapped-ion hardware. Our work is a step towards the direct trapped-ion simulation of vibrationally assisted exciton transfer in chemical and biological complexes, where the role of quantum coherence and delocalization in electron transfer is still debated [22,24–29,32]. A trapped-ion quantum simulator offers, on the one hand, full tunability of both the coherent and dissipative parts of the evolution while, on the other hand, granting access to site-resolved observables that are crucial to understanding the energy flow dynamics [22].

Trapped-ion quantum simulators can naturally access the intermediate coupling regime [26], where the reorganization energy λ is of the same order or larger than the electronic coupling between different molecular sites. This regime poses significant computational challenges for classical methods, such as tensor networks, especially when considering long-range interactions [34,98].

In this work, we focused on a spin chain coupled to a single, damped phonon mode. However, electron transfer in realistic systems often involves multiple vibrational modes. Therefore, the inclusion of multiple damped modes of motion will be paramount for the simulation of quantum dynamics of complex excitonic models. This can be achieved natively on trapped-ion quantum hardware, where multiple coolant ions can be used to control the individual cooling rates and the temperature of multiple bosonic modes. This capability supports the engineering of structured spectral densities [61], paving the way for studying non-Markovian effects in quantum dynamics [4,5,45] where strong, ultrastrong, and time-dependent system-environment couplings can be studied. Furthermore, the realization of Frenkel exciton models with multiple modes with engineered spectral density will allow simulation across diverse vibrational frequency ranges, capturing both inter- and intramolecular vibrations and low-frequency solvent environments [32,65].

Including more than one bosonic mode in this model will also allow the simulation of multiply bridged donor-acceptor molecules [99], where intermediate sites experience distinct environments or couplings to different vibrational modes [100,101]. Additionally, this setup provides an opportunity to introduce tunable anharmonic couplings

among bosonic modes in the trapped-ion chain [102], facilitating the study of anharmonic effects, which are believed to influence electron transfer [103].

In the work presented here, we focused on the role that coherent superpositions of states within each monomer play in determining the transfer efficiency. Considering more than two monomers, as we did in Sec. V, opens the possibility of studying the transfer of an excitation initially delocalized over multiple monomers. The presence of different types of quantum coherences and their survival over time has been found to be related to the high efficiency of excitation transfer in photosynthetic complexes [24,104]. Previous studies [31,105–107] have pointed out that the consideration of non-Markovian baths is essential to model correctly the survival of these coherences, which, as pointed out earlier, represents an interesting future research direction both experimentally and theoretically.

Here, we explore the effect of static disorder and white-noise fluctuations of the energy shifts through electronic dephasing, with the latter as well as motional dephasing common noise sources in realistic setups that play a key role in transfer dynamics [70,73,108]. In trapped-ion systems, electronic dephasing can be mimicked by introducing noisy light shifts [71,92], and motional dephasing can be realized by controlling the trap frequencies [109]. As shown in Ref. [71], more complicated noise profiles can be realized in these platforms, opening the possibility to study richer coherence dynamics. Although our study focuses on excitation-conserving $\sim \sigma_z(a^\dagger + a)$ spin-phonon coupling, real systems feature relaxation of excitations, which limits the transfer efficiency. In trapped-ion systems, it is possible to include spin relaxation experimentally by using a red-sideband (Jaynes-Cummings-type) drive with dissipation.

A further extension of our model involves generalizing qubits to qudits, allowing the simulation of other transfer phenomena such as singlet fission processes [110,111]. Trapped-ion systems allow for coherent coupling of multiple atomic states to bosonic modes. Spin-1 Hamiltonians, for example, have been implemented with trapped ions [112], and qudit-based gates have been realized [113].

From a quantum information perspective, our model raises intriguing questions about generalizing recently established bounds on quantum state transfer in long-range interacting systems [56–58,114] to open quantum systems with vibronic couplings. Such bounds could help optimize state transfer across the large parameter space $(J_{ij}, \epsilon_i, g_i, \gamma)$ inherent in these models. Exploring larger collections of monomers with intermediate coupling, long-range interactions, and dissipation could also illuminate parameter regimes where classical methods are computationally prohibitive, offering insights into the design of light-harvesting materials.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [115].

APPENDIX A: MODEL

To find the eigenstates of Hamiltonian (7) for a given j , we displace the bosonic mode as $a = \alpha_j + \tilde{a}_j$, where α_j is a complex number and $\tilde{a}_j$ is another bosonic annihilation operator. In terms of the new variables,

$$H_{Bj} = \omega \tilde{a}_j^\dagger \tilde{a}_j + \omega(\alpha_j)^2 + \omega \alpha_j (\tilde{a}_j + \tilde{a}_j^\dagger) + g_j \alpha_j + \frac{g_j}{2} (\tilde{a}_j + \tilde{a}_j^\dagger). \quad (\text{A1})$$

To eliminate the linear terms on the annihilation and creation operators, we set $\alpha_j = -g_j/4\omega$. The Hamiltonian becomes

$$H_{Bj} = \omega \tilde{a}_j^\dagger \tilde{a}_j - \frac{g_j^2}{4\omega}, \quad (\text{A2})$$

corresponding to a harmonic oscillator with a constant energy shift caused by the displacement of the oscillator by $\alpha_j = -g_j/2\omega$. The eigenstates of Eq. (7) are known as displaced Fock states [116] and can be represented by $D(\alpha_j)|n\rangle = |n, \alpha_j\rangle$, where n is the number of excitations in the displaced oscillator and $D(\alpha)$ is the bosonic displace operator. The corresponding eigenvalues are given by $n\omega - g_j^2/(4\omega)$. Consequently, the eigenstates of the full unperturbed Hamiltonian (6) are given by $|\psi_{j,n}\rangle = |j\rangle \otimes |n, \alpha_j\rangle$ with eigenenergies given by Eq. (8).

To derive the expression for the FGR in Eq. (9), we start by considering an initial state $|\psi_{i,0}\rangle = |i\rangle \otimes |0, \alpha_i\rangle$ characterized by zero bosonic excitations $n = 0$ and an initial electronic state $|i\rangle$ (for most of our two-monomer results, $|i\rangle = |T_D\rangle$), and final states of the form

$|\psi_{f,n}\rangle = |f\rangle \otimes |n, \alpha_f\rangle$. The FGR is defined as

$$k_T^{if} = \sum_{n=0}^{\infty} |\langle \psi_{i,0} | \psi_{f,n} \rangle|^2 \frac{\gamma}{(E_{i,0} - E_{f,n})^2 + \gamma^2/4}, \quad (\text{A3})$$

where γ has been incorporated through a Lorentzian broadening of the delta functions that appear in the conventional definition of the FGR. Here, $|\langle \psi_{i,0} | \psi_{f,n} \rangle|^2 = |J_{if}|^2 |\langle 0, \alpha_i | n \alpha_f \rangle|^2$, where J_{if} is the corresponding intermonomer coupling. In this case, the Franck-Condon factors can be simplified further as

$$\begin{aligned} \langle 0, \alpha_i | n \alpha_f \rangle &= \langle 0 | D(-\alpha_i) D(\alpha_f) | n \rangle \\ &= \langle 0, \alpha_i - \alpha_f | n \rangle \\ &= e^{-(\alpha_i - \alpha_f)^2/2} \frac{(\alpha_i - \alpha_f)^n}{\sqrt{n!}}, \end{aligned} \quad (\text{A4})$$

where in the last line we have used the expansion of a displaced state in terms of the Fock basis and the orthonormality condition $\langle m | n \rangle = \delta_{nm}$. Using this simplified form of the Franck-Condon factor, we obtain Eq. (9).

To compare the analytical expression $k_T = \sum_{f \in A} k_T^{if}$ with numerical results, we define the transfer rate in Eq. (14). To extract the transfer rate, we simulate the dynamics for a total time $\omega t_{\text{sim}}/2\pi = 100$, which means that, in practice, we use the expression

$$k_T = \frac{\int_{t=0}^{t_{\text{sim}}} P_D(t) dt}{\int_{t=0}^{t_{\text{sim}}} t P_D(t) dt} - \frac{2}{t_{\text{sim}}}, \quad (\text{A5})$$

where the second term is a correction for considering a finite time t_{sim} .

APPENDIX B: PARAMETER CHOICES

In Sec. III, we commented on the choice $g = g_1 = g_2 \neq g_3 = g_4 = \tilde{g}$ to differentiate between the donor and acceptor sites. There, we chose $g = -\tilde{g}$. We note that, for general g and $\tilde{g}$, we can still write the unperturbed Hamiltonian in terms of the triplet and singlet states [see Eq. (10)] and obtain the same form of the intermonomer couplings in Eqs. (13). This means that we observe a very similar behavior for general g and $\tilde{g}$, where the transfer from initial triplet states has a considerably faster transfer than from other initial states under specific resonant conditions, such as those shown in Figs. 2(a) and 2(c). Now, both the peak position and height of the resonances will be modified by the choice of g and $\tilde{g}$; for instance, the positions will be located at $\epsilon = n\omega + g^2/(4\omega) - \tilde{g}^2/(4\omega)$ with n being an integer. The height is modulated by the Franck-Condon factor in Eq. (9), which depends only on the displacement $|g - \tilde{g}|$. For example, if we chose $\tilde{g} = 2g$, we would have the same modulation as for the parameter choice in the

main text, namely, $g = -\tilde{g}$. However, we would have a different position for the resonances since $g^2 - \tilde{g}^2$ is different for both cases. Again, this would modify the position and height of the peaks, but the conclusion that a delocalized initial state has a faster transfer with respect to the product (localized) state counterpart will hold for all choices of g and $\tilde{g}$. Moreover, in Sec. III C, we relaxed the condition of $g_1 = g_2$ and $g_3 = g_4$ by considering disorder in these parameters.

In Sec. III, we introduced the expression for the intermonomer couplings in the triplet-singlet basis in Eq. (13b). In that expression, we used the convention of considering two coolants [see the discussion below Eq. (5)] that sets $d_{23} = 3$. More generally, the couplings are given by

$$J_{\text{TT}} = \left(\frac{1}{2} \frac{J}{(1+d)^p} + \frac{J}{(2+d)^p} + \frac{1}{2} \frac{J}{(3+d)^p} \right), \quad (\text{B1a})$$

$$J_{\text{SS}} = \left(-\frac{1}{2} \frac{J}{(1+d)^p} + \frac{J}{(2+d)^p} - \frac{1}{2} \frac{J}{(3+d)^p} \right), \quad (\text{B1b})$$

$$J_{\text{TS}} = -J_{\text{ST}} = \left(\frac{1}{2} \frac{J}{(1+d)^p} - \frac{1}{2} \frac{J}{(3+d)^p} \right), \quad (\text{B1c})$$

where the distance between neighboring ions is set equal to 1, and d is the number of coolants. The ratio of the different couplings in Eqs. (B1) for different values of p and d is shown in Fig. 7.

Having a preferred initial delocalized state with an enhanced transfer rate relies on the dominance of one of the couplings (here J_{TT}) over all others. The results reported in Fig. 7(a) show that J_{TT} is the dominant process for all $p \lesssim 3$ and $d = 2$. Around $p = 3$, J_{TT} becomes comparable to $J/(3^p)$, which is the largest intermonomer coupling in the product basis. For all $p < 3$, we would expect that the transfer from the triplet state is faster than from any other product or entangled state.

For $p = 1$, the results in Fig. 7(b) show that the ratio between J_{TT} and any other coupling becomes larger as d increases. This means that increasing d results in a more dominant triplet-triplet transfer, of course, at the cost of the transfer becoming slower. Our choice of $p = 1$ and small intermonomer distance ($d_{23} = 3$) for the results in Sec. III speeds up the numerical calculations as inter- and intramonomer interactions have similar scales with overall faster transfer dynamics. However, we note that the case of $p = 3$ representing the dipole interaction, which is native in many systems, shows a similar trend as a function of d , as shown in Fig. 7(c). Then, for a large intermonomer distance d and $p = 3$, we expect a clear dominance of the triplet-triplet process (J_{TT} much larger than any other coupling), leading to very similar dynamics, but on an overall slower time scale.

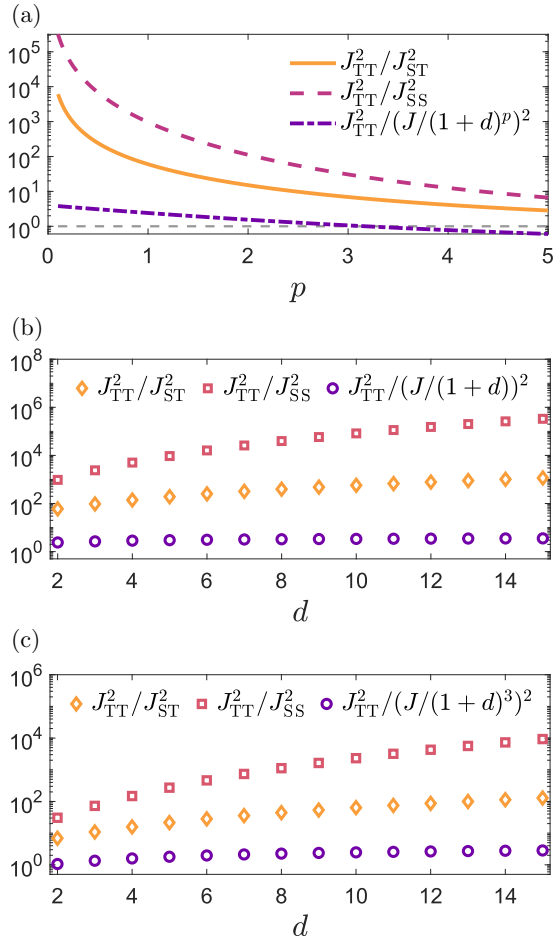


FIG. 7. Intermonomer couplings in the triplet-singlet basis. (a) Ratio of the different interdimer couplings as a function of p for $d = 2$. The horizontal gray dashed line is used as a guide to highlight the point where $J_{TT}^2/(J/3^p)^2 = 1$ at $p \approx 3$. (b) Ratio of the different interdimer couplings as a function of d for $p = 1$. (c) Ratio of the different interdimer couplings as a function of d for $p = 3$.

APPENDIX C: NUMERICAL SIMULATION DETAILS

Since the dynamics described in the excitation transfer involve a nonzero number of excitations in the phonon mode, especially in the case of higher temperatures, the numerical integration of Eq. (2) requires choosing an appropriate cutoff number of phonon excitations N_{ph} . In Fig. 8, we show how the evolution of different observables is affected by the choice of this cutoff.

As depicted in the figure, for these parameters, a cutoff of $N_{\text{ph}} > 10$ excitations already provides an appropriate size of the phonon mode Hilbert space, as no significant changes can be noticed in the observables when the value is increased to $N_{\text{ph}} = 15$ (the two curves lie on top of each other). We adjust the phonon cutoff accordingly for the different parameter sets used in this work.

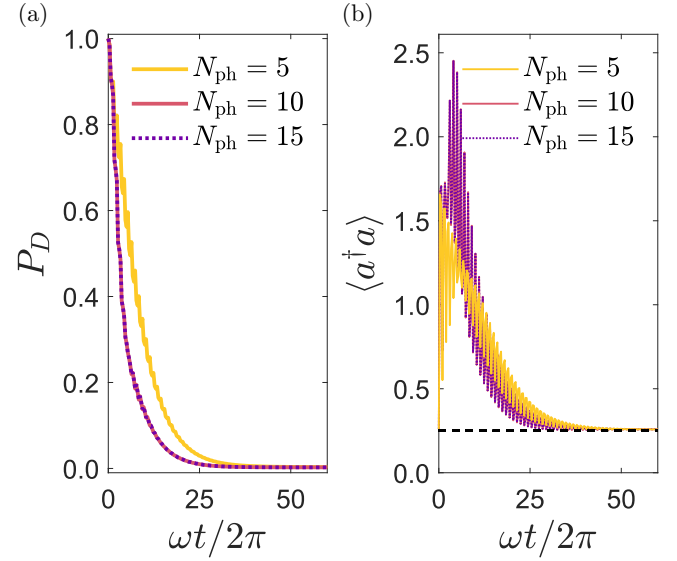


FIG. 8. Phonon cutoff dependence: time evolution of observables for different values of the phonon cutoff N_{ph} , represented by distinct curves. The chosen parameters are the same as the parameter set p_1 in Table II in Appendix E. (a) Population in the donor as a function of time. (b) Population in the phonon mode as a function of time. The black dashed line represents $\alpha^2 = (g/2\omega)^2 = 0.25$, which is the expected value of the population in the steady state.

APPENDIX D: ENTANGLEMENT TRANSFER

One of the main results in this work is that the initialization of the system in an entangled (delocalized) state of the qubits in the donor site speeds up the transfer into the acceptor site. Under ideal conditions (i.e., at low temperature and on resonance), the steady state in the acceptor is also an entangled state, as illustrated in Fig. 2. For example, this is evidenced by the fact that $P_D \approx 1 - P_{T_A}$ when the system is initialized in the electronic entangled state $|T_D\rangle$. However, we make this argument more concrete here, where we explicitly show the dynamics of the qubit's entanglement in terms of the concurrence.

For a two-qubit state, pure or mixed, the concurrence can be used to determine the entanglement. Mathematically, the concurrence is defined as [117,118]

$$\mathcal{C}_{AB}(\rho) = \max(0, \mu_1 - \mu_2 - \mu_3 - \mu_4), \quad (\text{D1})$$

where ρ is the quantum state of the two qubits A and B , and μ_j are the eigenvalues of the matrix $\tilde{\rho} = \rho S \rho^* S$, with $S = \sigma_y \otimes \sigma_y$. Here, σ_y is the conventional Pauli matrix in the y direction. The eigenvalues are defined in descending order $\mu_j > \mu_{j+1}$. If $\mathcal{C}_{AB}(\rho) = 0$, the state has no entanglement (separable state), and if $\mathcal{C}_{AB}(\rho) = 1$, the system is maximally entangled. In Fig. 9, we show the evolution of the concurrence between the donor qubits $\mathcal{C}_{12}$ as well as the concurrence between the acceptor qubits $\mathcal{C}_{34}$ for the two monomer case, described in Sec. III.

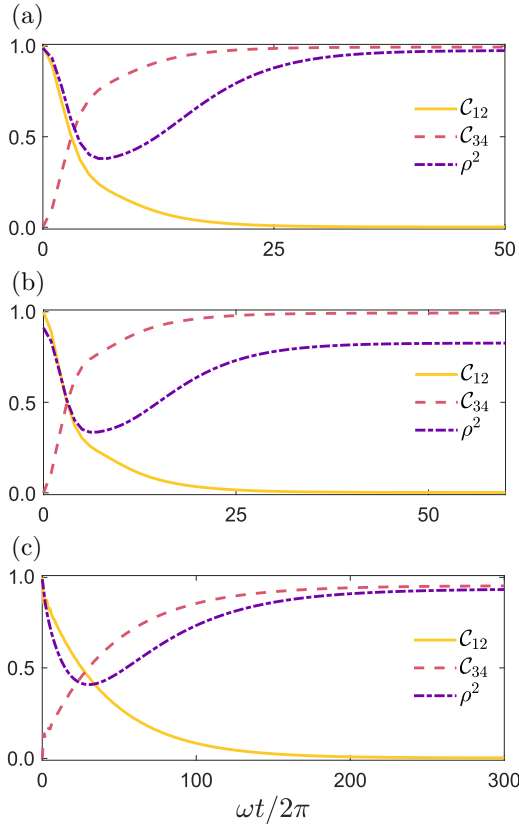


FIG. 9. Entanglement transfer: time evolution of the concurrence between different qubits and the purity of the state. (a) The chosen parameters are those in parameter set p_1 in Table II in Appendix E. (b) Same as in (a), but with $\bar{n} = 0.1$. (c) Same as in (a), but with $\epsilon = 0.26\omega$.

Initially, since the system is prepared in state $|T_D\rangle$, which is maximally entangled, the concurrence is maximal, i.e., $C_{12} = 1$. As time elapses, the two qubits in the donor site lose all entanglement as they reach the separable state $|\downarrow\downarrow\rangle$ when the excitation has been transferred to the acceptor. In Fig. 9(a), where the system is set at a resonance ($\epsilon = 3\omega$), and the temperature is low ($\bar{n} = 0.01$), the steady state of the system is the electronic state $|T_A\rangle$, which is also a maximally entangled state of the two qubits; this is visible in the figure: $C_{34} \rightarrow 1$ at long times.

In the ideal conditions of Fig. 9(a), the state of the phonons is the displaced vacuum state, so the steady state of the total system is the pure state $|\psi_{T_D,0}\rangle$. Consequently, $\rho^2 \rightarrow 1$ at long times, as reported in the plot. If we increase $\bar{n}$, as in panel (b), the steady state can be a mixed state of multiple phonon states, as shown by $\rho^2 < 1$. Nonetheless, the electronic steady state can still be purely $|T_D\rangle$, and in that case, all the initial entanglement between qubits in the donor site is still transferred to the acceptor site.

In Fig. 9(c), we move away from resonance by setting $\epsilon = 0.26\omega$, such that the initial state $|T_D\rangle$ can weakly couple to $|S_A\rangle$. In that case, the electronic steady state is a

mixture of $|T_A\rangle$ and $|S_A\rangle$; consequently, not all the initial entanglement is transferred to the acceptor, namely, $C_{34} < 1$ in the steady state.

When the donor and acceptor sites contain more than two qubits (see Sec. V), other entanglement measures different from concurrence would be needed to characterize the entanglement dynamics of the mixed multiqubit states. However, we expect similar entanglement dynamics. For instance, for the parameters chosen here, if the system is initialized in $|\mathcal{E}_D^1\rangle$, ideal conditions would lead to transfer into state $|\mathcal{E}_A^1\rangle$ [see Fig. 5(b)]. This state has a high overlap with state $|W_A\rangle$, as shown in Appendix F, which implies that the steady state of the system has a large entanglement. For instance, if we consider the donor or acceptor to have N qubits, with N being even, and consider the bipartition of $N/2$ qubits in each subsystem, the bipartite entanglement that can be transferred from the donor to the acceptor grows approximately as $S \propto \log_2(N) + C$ [82] when the system is initialized in $|\mathcal{E}_D^1\rangle$.

APPENDIX E: CONFIGURATIONS WITH STATIC DISORDER

When considering the effects of static disorder, we defined the parameter sets p_1, p_2, p_3 , and p_4 signaled with markers in Fig. 3(a). In Table II, we report the values of all parameters for each of these parameter sets.

As discussed in the main text, static disorder generally reduces the transfer rate; however, the effect of disorder in the dynamics can vary depending on the specific disorder realization. In Fig. 10, we consider two particular disordered configurations to illustrate this point.

The dynamics reported in Fig. 10(a) show that the disorder can cause other processes to become resonant. In this case, we see a coupling between the two acceptor states, increasing the population of the singlet state $|S_A\rangle$. At the perturbative level and in the absence of disorder, there is no coupling between these two states; however, the presence of disorder, even for small J , can modify this picture as $|T_A\rangle$ and $|S_A\rangle$ are not eigenstates of the unperturbed Hamiltonian anymore. However, we should note that this oscillation in the acceptor states might not significantly affect the transfer rate as the population of the donor decreases rapidly and remains close to zero.

For the disorder realization in Fig. 10(b), on the other hand, we see that the population transfer to the acceptor

TABLE II. Parameter sets used for the disorder analysis. All sets consider $J = 0.3\omega$, $\epsilon = 3\omega$, $\bar{n} = 0.01$, and $g = \omega$.

Label	γ/ω	p
p_1	0.039 552	1.0
p_2	0.010 506	1.0
p_3	0.111 707	1.0
p_4	0.030 263	1.5

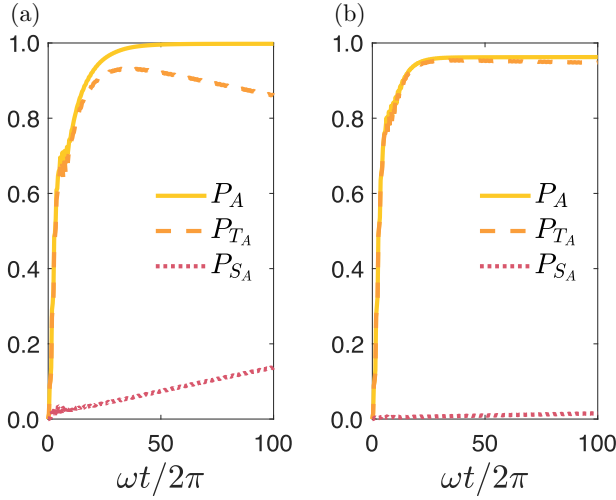


FIG. 10. Dynamics of specific disorder realizations: time evolution of the electronic state populations for configurations with disordered parameters g_j . The yellow solid line, orange dashed line, and pink dotted line represent the populations in the acceptor site, the triplet acceptor state, and the singlet acceptor state, respectively. (a) The disordered parameters are given by $g_1/\omega = 0.9883$, $g_2/\omega = 0.9906$, $g_3/\omega = -0.9638$, and $g_4/\omega = -1.1056$. (b) The disordered parameters are given by $g_1/\omega = 1.0923$, $g_2/\omega = 0.9405$, $g_3/\omega = -1.0220$, and $g_4/\omega = -0.9806$.

is incomplete, namely, some population always remains in the donor. As we discussed in Sec. IV, a higher donor population in the steady state can decrease the value of the transfer rate in Eq. (14).

APPENDIX F: HIGHER-COMPLEXITY MONOMERS AND MORE MONOMERS

We start by considering the case of two monomers with three qubits per monomer ($L = 3$) that we analyzed in Sec. V. The unperturbed Hamiltonian in the donor site is given by

$$H_{0,D} = \frac{\epsilon}{2}(|1\rangle\langle 1| + |2\rangle\langle 2| + |3\rangle\langle 3|) + \omega a^\dagger a + J \left[\left(|1\rangle\langle 2| + |2\rangle\langle 3| + \frac{1}{2}|1\rangle\langle 2| \right) + \text{H.c.} \right] + \frac{g}{2}(|1\rangle\langle 1| + |2\rangle\langle 2| + |3\rangle\langle 3|), \quad (\text{F1})$$

where we have kept $p = 1$. Diagonalization of Eq. (F1) yields the three eigenstates in Eqs. (17), where $\mathcal{N}_1 = \frac{1}{2}\sqrt{\frac{1}{2}(33 - \sqrt{33})}$ and $\mathcal{N}_2 = \frac{1}{2}\sqrt{\frac{1}{2}(\sqrt{33} + 33)}$. The corresponding eigenenergies are given by

$$E_{\mathcal{E}_D^1} = \frac{1}{4}(\sqrt{33}J + J + 4\epsilon), \quad (\text{F2a})$$

$$E_{\mathcal{E}_D^2} = \frac{1}{4}(-\sqrt{33}J + J + 4\epsilon), \quad (\text{F2b})$$

$$E_{\mathcal{E}_D^3} = \epsilon - \frac{J}{2}. \quad (\text{F2c})$$

State $|\mathcal{E}_D^1\rangle$ is the state with the largest overlap with the W state $|W_D\rangle$ and also the state with the highest eigenenergy. Similarly, equivalent results for states $|\mathcal{E}_A^{1,2,3}\rangle$ can be found for the acceptor sites in terms of states $|4\rangle$, $|5\rangle$, and $|6\rangle$. The eigenenergies of these states are identical to those in Eqs. (F2), but with negative on-site energy $\epsilon \rightarrow -\epsilon$. State $|\mathcal{E}_A^1\rangle$ also corresponds to the acceptor state with the highest energy and largest overlap with $|W_A\rangle$.

The perturbation Hamiltonian containing all the intermonomer couplings can be written as

$$H_J = \sum_{A,B} J_{AB}|A\rangle\langle B| + \text{H.c.}, \quad (\text{F3})$$

where $A = \mathcal{E}_A^1, \mathcal{E}_A^2, \mathcal{E}_A^3$ and $B = \mathcal{E}_D^1, \mathcal{E}_D^2, \mathcal{E}_D^3$. In Table III, we report all the values of J_{AB} , where it can be noted that the process where the population is transferred between $|\mathcal{E}_D^1\rangle$ and $|\mathcal{E}_A^1\rangle$ is the fastest. Moreover, if the system is initially prepared in another state in the donor ($|\mathcal{E}_D^2\rangle$ or $|\mathcal{E}_D^3\rangle$), transfer to the $|\mathcal{E}_A^1\rangle$ state is always favored at least from the perspective of having a stronger coupling J_{AB} (see the fourth and seventh rows of Table III).

In the case of transfer from the initial $|W_D\rangle$ state [see Figs. 5(a)–5(b)], the transfer rate is very similar to the case of initial $|\mathcal{E}_D^1\rangle$ for two reasons: (i) the hierarchy of the J_{AB} favors the transfer from any donor state to the $|\mathcal{E}_A^1\rangle$ state, as discussed above, and (ii) these two initial states have a very large overlap $|\langle \mathcal{E}_D^1 | W_D \rangle|^2 = 0.993$. This overlap decreases as L increases, but remains very large, for instance $|\langle \mathcal{E}_D^1 | W_D \rangle|^2 = 0.984$ for $L = 5$.

The transfer from $|\mathcal{E}_D^1\rangle$ to $|\mathcal{E}_A^1\rangle$ also remains the fastest process for larger L . As an example, we consider the case

TABLE III. Intermonomer couplings J_{AB} in the basis of the eigenstates of Eq. (F1).

Donor state	Acceptor state	$ J_{AB}/J $
$\mathcal{E}_D^1$	$\mathcal{E}_A^1$	0.6299
$\mathcal{E}_D^1$	$\mathcal{E}_A^2$	0.0553
$\mathcal{E}_D^1$	$\mathcal{E}_A^3$	0.1446
$\mathcal{E}_D^2$	$\mathcal{E}_A^1$	0.0786
$\mathcal{E}_D^2$	$\mathcal{E}_A^2$	0.0082
$\mathcal{E}_D^2$	$\mathcal{E}_A^3$	0.0250
$\mathcal{E}_D^3$	$\mathcal{E}_A^1$	0.0849
$\mathcal{E}_D^3$	$\mathcal{E}_A^2$	0.0103
$\mathcal{E}_D^3$	$\mathcal{E}_A^3$	0.0381

TABLE IV. Intermonomer couplings J_{AB} in the basis of the eigenstates of the unperturbed Hamiltonian [see Eq. (F3)] for $L = 4$.

Donor state	Acceptor state	$ J_{AB}/J $
$\mathcal{E}^1$	$\mathcal{E}^1$	0.7083
$\mathcal{E}^1$	$\mathcal{E}^2$	0.1280
$\mathcal{E}^1$	$\mathcal{E}^3$	0.2036
$\mathcal{E}^1$	$\mathcal{E}^4$	0.0211
$\mathcal{E}^2$	$\mathcal{E}^1$	0.0823
$\mathcal{E}^2$	$\mathcal{E}^2$	0.0186
$\mathcal{E}^2$	$\mathcal{E}^3$	0.0342
$\mathcal{E}^2$	$\mathcal{E}^4$	0.0039
$\mathcal{E}^3$	$\mathcal{E}^1$	0.1025
$\mathcal{E}^3$	$\mathcal{E}^2$	0.0267
$\mathcal{E}^3$	$\mathcal{E}^3$	0.0576
$\mathcal{E}^3$	$\mathcal{E}^4$	0.0065
$\mathcal{E}^4$	$\mathcal{E}^1$	0.0371
$\mathcal{E}^4$	$\mathcal{E}^2$	0.0106
$\mathcal{E}^4$	$\mathcal{E}^3$	0.0228
$\mathcal{E}^4$	$\mathcal{E}^4$	0.0027

$L = 4$ with donor eigenstates:

$$|\mathcal{E}_D^m\rangle = \frac{1}{\mathcal{N}_m}(|1\rangle + v_{m,2}|2\rangle + v_{m,3}|3\rangle + |4\rangle), \quad (\text{F4a})$$

$$v_{1,2} = v_{1,3} = \frac{1}{9}(2 + \sqrt{85}), \quad (\text{F4b})$$

$$v_{2,2} = v_{2,3} = \frac{1}{9}(2 - \sqrt{85}), \quad (\text{F4c})$$

$$v_{3,2} = -v_{3,3} = \frac{1}{3}(2 - \sqrt{13}), \quad (\text{F4d})$$

$$v_{4,2} = -v_{4,3} = \frac{1}{9}(2 + \sqrt{13}). \quad (\text{F4e})$$

Here the normalization constants are defined by $\mathcal{N}_m = \sqrt{2 + v_{m,2}^2 + v_{m,3}^2}$. Consistent with our notation, $|\mathcal{E}_D^1\rangle$ is the state with largest overlap with the W state ($|\langle \mathcal{E}_D^1 | W_D \rangle|^2 = 0.988$) and with the largest eigenenergy $E_{\mathcal{E}_D^1} = \frac{1}{6}(\sqrt{85} + 4)J + \epsilon$. In Table IV, we report the magnitude of all intermonomer couplings when the perturbation Hamiltonian is written in the form of Eq. (F3). Apart from the transfer from $|\mathcal{E}_D^1\rangle$ to $|\mathcal{E}_A^1\rangle$ being the fastest, we also note that the transfer to state $|\mathcal{E}_A^1\rangle$ is always favored over other processes regardless of the choice of initial state on this basis.

APPENDIX G: EXPERIMENTAL REALIZATION OF THE VIBRATIONAL AND VIBRONIC COUPLING TERMS

Here we outline in detail the derivation of the trapped-ion Hamiltonian corresponding to the vibrational and vibronic coupling terms in Eq. (1) via the proposed experimental scheme in Sec. VIA and the discussion on the effect of the unwanted interactions arising from this scheme.

Consider the application of individual-addressing Raman beams with the same wave-vector difference $\vec{k}_a$

along a radial direction, phase differences ϕ_i , and a common beatnote frequency ω_L on the qubit ions. The trapped-ion Hamiltonian is ($\hbar = 1$)

$$H = \sum_i \frac{\omega_0}{2} \sigma_i^z + \sum_v \omega_v a_v^\dagger a_v + \sum_i \frac{\Omega_i}{2} \left[\exp \left\{ \sum_v i\eta_{iv} (a_v + a_v^\dagger) - i\omega_L t - i\phi_i \right\} \sigma_i^+ + \text{H.c.} \right], \quad (\text{G1})$$

where ω_0 is the frequency difference between the internal states of the qubit ions, ω_v is the v th collective mode frequency of the chain associated with the raising (lowering) operator $a_v^\dagger$ (a_v), Ω_i is the Rabi coupling strength to ion i , and $\eta_{iv} = k_a \sqrt{1/2m\omega_v} b_{iv}$ is the Lamb-Dicke parameter with m the qubit mass and b_{iv} the normalized motional eigenvector for ion i in the collective mode v , where the lowest- and highest-frequency modes ω_1 and ω_N are known as the zig-zag and center-of-mass modes, respectively.

Under the transformation with respect to $\sum_i (\omega_0/2) \sigma_i^z + \sum_{v'} \omega_{v'} a_{v'}^\dagger a_{v'} + \mu a_k^\dagger a_k$, Eq. (G1) becomes a resonant interaction Hamiltonian that rotates at $\mu = \omega_L - \omega_0 \equiv \omega_k + \delta_k$, where δ_k is the detuning from the k th motional mode, and $\forall v' = \forall v \setminus \{k\}$ [87]. For $\mu + \omega_k \gg |\mu - \omega_k| = \delta_k$, we neglect the terms that rotate at $\mu + \omega_k$ under the rotating-wave approximation, which results in the transformed Hamiltonian

$$H^{\text{res}} = \sum_i \frac{\Omega_i}{2} \left[\exp \left\{ \sum_{v'} i\eta_{iv'} (a_{v'} e^{-i\omega_{v'} t} + a_{v'}^\dagger e^{i\omega_{v'} t}) \right\} \times e^{i\eta_{ik} (a_k e^{-i\mu t} + a_k^\dagger e^{i\mu t})} e^{-i(\omega_L - \omega_0)t - i\phi_i} \sigma_i^+ + \text{H.c.} \right] - \delta_k a_k^\dagger a_k \equiv H_I^{\text{res}} + H_\delta^{\text{res}}. \quad (\text{G2})$$

The second term, $H_\delta^{\text{res}} = -\delta_k a_k^\dagger a_k$, corresponds to the vibrational energy term in Eq. (1) with $-\delta_k \mapsto \omega$ and $a_k^\dagger(a_k) \mapsto a^\dagger(a)$. To obtain the vibronic coupling term, we apply individual-addressing Raman beams with two symmetric beatnote frequency tones $\omega_r = \omega_0 - \mu$ with controlled phase ϕ_i^r and Rabi frequency Ω_i^r and $\omega_b = \omega_0 + \mu$ with controlled phase ϕ_i^b and Rabi frequency Ω_i^b . Hence, the interaction Hamiltonian becomes

$$H_I^{\text{res}} = \sum_{l=r,b} \sum_i \frac{\Omega_i^l}{2} \left[\exp \left\{ \sum_{v'} i\eta_{iv'} (a_{v'} e^{-i\omega_{v'} t} + a_{v'}^\dagger e^{i\omega_{v'} t}) \right\} \times e^{i\eta_{ik} (a_k e^{-i\mu t} + a_k^\dagger e^{i\mu t})} e^{-i(\omega_l - \omega_0)t - i\phi_i^l} \sigma_i^+ + \text{H.c.} \right]. \quad (\text{G3})$$

In the Lamb-Dicke regime, where $\eta_{i,v}\sqrt{\langle(a_v + a_v^\dagger)^2\rangle} \ll 1$, we can expand the interaction Hamiltonian with respect to η_{iv} to the first order. For $\Omega_i^r = \Omega_i^b \equiv \Omega_i^{\text{sp}}$, we obtain

$$\begin{aligned} H_I^{\text{res}} = & \sum_i \Omega_i^{\text{sp}} \cos(\mu t + \phi_i^m) \sigma_{\phi_i^s + \pi/2} \\ & + \sum_i \frac{\eta_{ik} \Omega_i^{\text{sp}}}{2} [a_k (e^{i\phi_i^m} + e^{-i2\mu t - i\phi_i^m}) \\ & \quad + a_k^\dagger (e^{-i\phi_i^m} + e^{i2\mu t + i\phi_i^m})] \sigma_{\phi_i^s} \\ & + \sum_{v'} \sum_i \eta_{iv'} \Omega_i^{\text{sp}} (a_{v'} e^{-i\omega_{v'} t} + a_{v'}^\dagger e^{i\omega_{v'} t}) \\ & \quad \times \cos(\mu t + \phi_i^m) \sigma_{\phi_i^s}, \end{aligned} \quad (\text{G4})$$

where the motional and spin phases are denoted by $\phi_i^m \equiv (\phi_i^b - \phi_i^r)/2$ and $\phi_i^s \equiv (\phi_i^b + \phi_i^r)/2 - \pi/2$, respectively, and we define the spin operator $\sigma_{\phi_i^s} \equiv \cos(\phi_i^s) \sigma_i^x + \sin(\phi_i^s) \sigma_i^y$ [49]. We note that $\sigma_i^y \rightarrow \sigma_i^z$ in the driven rotating lab frame.

Since the interaction Hamiltonian is time dependent, we can approximate the time-ordering evolution operator, $U(t) = T[\exp[-i \int_0^t dt_1 H_I^{\text{res}}(t_1)]]$, with the Magnus expansion and obtain $U(t) \approx \exp(-i H_I^{\text{eff}} t)$ with the effective Hamiltonian

$$\begin{aligned} H_I^{\text{eff}} = & \sum_i \frac{\eta_{ik} \Omega_i^{\text{sp}}}{2} (a_k e^{i\phi_i^m} + a_k^\dagger e^{-i\phi_i^m}) \sigma_{\phi_i^s} \\ & + \sum_{v'} \sum_{i < j} \frac{\eta_{iv'} \eta_{jv'} \omega_{v'}}{\mu^2 - \omega_{v'}^2} \Omega_i^{\text{sp}} \Omega_j^{\text{sp}} \cos(\phi_i^m - \phi_j^m) \\ & \quad \times \sigma_{\phi_i^s} \sigma_{\phi_j^s} \end{aligned} \quad (\text{G5})$$

for when $\delta_{v'} \equiv \mu - \omega_{v'} \gg \eta_{iv'} \Omega_i^{\text{sp}}$. Here, the first term is the desired vibronic coupling term in Eq. (1) with $\eta_{ik} \Omega_i^{\text{sp}} \mapsto g_i$ and an appropriate choice of optical phases, while the second term accounts for off-resonant couplings to other motional modes, which leads to an unwanted two-body electronic coupling term:

$$\begin{aligned} H_{ij}^{\text{cr}} \equiv & \sum_{v'} \sum_{i < j} \frac{\eta_{iv'} \eta_{jv'} \omega_{v'}}{\mu^2 - \omega_{v'}^2} \Omega_i^{\text{sp}} \Omega_j^{\text{sp}} \cos(\phi_i^m - \phi_j^m) \\ & \quad \times \sigma_{\phi_i^s} \sigma_{\phi_j^s}. \end{aligned} \quad (\text{G6})$$

In the rotated frame that the quantum simulation of our model is performed, the interaction becomes

$$H_{ij}^{\text{cr}} = \sum_{i < j} J_{ij}^{\text{cr}} \sigma_i^z \sigma_j^z, \quad (\text{G7})$$

where

$$J_{ij}^{\text{cr}} \equiv \sum_{v'} \frac{\eta_{iv'} \eta_{jv'} \omega_{v'}}{\mu^2 - \omega_{v'}^2} \Omega_i^{\text{sp}} \Omega_j^{\text{sp}} \cos(\phi_i^m - \phi_j^m).$$

Therefore, in the relevant electronic manifold, where one of the ions is in the spin-up state representing the location of the electronic excitation ($\{|i\rangle\}$), the unwanted two-body electronic coupling Hamiltonian corresponds to a state-dependent shift in energy for the relevant spin configurations:

$$\langle k | H_{ij}^{\text{cr}} | k \rangle = \sum_{i < j} f(k, i, j) J_{ij}^{\text{cr}}, \quad (\text{G8})$$

$$f(k, i, j) = \begin{cases} -1 & \text{if } i = k \text{ or } j = k, \\ +1 & \text{otherwise.} \end{cases} \quad (\text{G9})$$

Similarly, in any basis spanned by $\{|i\rangle\}$, the interaction Hamiltonian also leads to state-dependent energy shifts. For instance, in the $\{|T_D\rangle, |S_D\rangle, |T_A\rangle, |S_A\rangle\}$ basis used to study the two-monomer model in Sec. III, we obtain

$$\begin{aligned} \langle T_D | H_{ij}^{\text{cr}} | T_D \rangle &= -J_{12}^{\text{cr}} + J_{34}^{\text{cr}}, \\ \langle S_D | H_{ij}^{\text{cr}} | S_D \rangle &= -J_{13}^{\text{cr}} - J_{14}^{\text{cr}} + J_{23}^{\text{cr}} + J_{24}^{\text{cr}}, \\ \langle T_A | H_{ij}^{\text{cr}} | T_A \rangle &= +J_{12}^{\text{cr}} - J_{34}^{\text{cr}}, \\ \langle S_A | H_{ij}^{\text{cr}} | S_A \rangle &= -J_{13}^{\text{cr}} + J_{14}^{\text{cr}} - J_{23}^{\text{cr}} + J_{24}^{\text{cr}}. \end{aligned}$$

Hence, this two-body electronic coupling crosstalk term should only induce changes in the energy landscape and, in turn, affect the excitation transfer resonance conditions, while preserving the physical dynamics of the system unless an appropriate energy compensation is introduced.

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- [1] R. Fazio, J. Keeling, L. Mazza, and M. Schirò, Many-body open quantum systems, *SciPost Phys. Lect. Notes*, **99** (2025).
 - [2] T. E. Lee, S. Gopalakrishnan, and M. D. Lukin, Unconventional magnetism via optical pumping of interacting spin systems, *Phys. Rev. Lett.* **110**, 257204 (2013).
 - [3] P. Sierant, G. Chiriacò, F. M. Surace, S. Sharma, X. Turkeshi, M. Dalmonte, R. Fazio, and G. Pagano, Dissipative Floquet dynamics: from steady state to measurement induced criticality in trapped-ion chains, *Quantum* **6**, 638 (2022).
 - [4] B. Debecker, J. Martin, and F. Damanet, Controlling matter phases beyond Markov, *Phys. Rev. Lett.* **133**, 140403 (2024).
 - [5] B. Debecker, J. Martin, and F. Damanet, Spectral theory of non-Markovian dissipative phase transitions, *Phys. Rev. A* **110**, 042201 (2024).
 - [6] S. Diehl, A. Micheli, A. Kantian, B. Kraus, H. Büchler, and P. Zoller, Quantum states and phases in driven open quantum systems with cold atoms, *Nat. Phys.* **4**, 878 (2008).
 - [7] F. Verstraete, M. M. Wolf, and J. Ignacio Cirac, Quantum computation and quantum-state engineering driven by dissipation, *Nat. Phys.* **5**, 633 (2009).

- [8] J. Cho, S. Bose, and M. S. Kim, Optical pumping into many-body entanglement, *Phys. Rev. Lett.* **106**, 020504 (2011).
- [9] S. F. Huelga, A. Rivas, and M. B. Plenio, Non-Markovianity-assisted steady state entanglement, *Phys. Rev. Lett.* **108**, 160402 (2012).
- [10] A. González-Tudela and D. Porras, Mesoscopic entanglement induced by spontaneous emission in solid-state quantum optics, *Phys. Rev. Lett.* **110**, 080502 (2013).
- [11] D. Kienzler, H.-Y. Lo, B. Keitch, L. de Clercq, F. Leupold, F. Lindenfesler, M. Marinelli, V. Negnevitsky, and J. P. Home, Quantum harmonic oscillator state synthesis by reservoir engineering, *Science* **347**, 53 (2015).
- [12] G. Morigi, J. Eschner, C. Cormick, Y. Lin, D. Leibfried, and D. J. Wineland, Dissipative quantum control of a spin chain, *Phys. Rev. Lett.* **115**, 200502 (2015).
- [13] P. M. Harrington, E. J. Mueller, and K. W. Murch, Engineered dissipation for quantum information science, *Nat. Rev. Phys.* **4**, 660 (2022).
- [14] D. C. Cole, S. D. Erickson, G. Zarantonello, K. P. Horn, P.-Y. Hou, J. J. Wu, D. H. Slichter, F. Reiter, C. P. Koch, and D. Leibfried, Resource-efficient dissipative entanglement of two trapped-ion qubits, *Phys. Rev. Lett.* **128**, 080502 (2022).
- [15] A. Piñeiro Orioli, J. K. Thompson, and A. M. Rey, Emergent dark states from superradiant dynamics in multilevel atoms in a cavity, *Phys. Rev. X* **12**, 011054 (2022).
- [16] B. Sundar, D. Barberena, A. M. Rey, and A. P. Orioli, Driven-dissipative four-mode squeezing of multilevel atoms in an optical cavity, *Phys. Rev. A* **109**, 013713 (2024).
- [17] M. Ratner, A brief history of molecular electronics, *Nat. Nanotechnol.* **8**, 378 (2013).
- [18] L. Wang, M. A. Allodi, and G. S. Engel, Quantum coherences reveal excited-state dynamics in biophysical systems, *Nat. Rev. Chem.* **3**, 477 (2019).
- [19] G. D. Scholes, G. R. Fleming, L. X. Chen, A. Aspuru-Guzik, A. Buchleitner, D. F. Coker, G. S. Engel, R. van Grondelle, A. Ishizaki, D. M. Jonas, J. S. Lundeen, J. K. McCusker, S. Mukamel, J. P. Ogilvie, A. Olaya-Castro, M. A. Ratner, F. C. Spano, K. B. Whaley, and X. Zhu, Using coherence to enhance function in chemical and biophysical systems, *Nature* **543**, 647 (2017).
- [20] N. P. Gallop, D. R. Maslennikov, N. Mondal, K. P. Goetz, Z. Dai, A. M. Shankler, W. Sung, S. Nihonyanagi, T. Tahara, M. I. Bodnarchuk *et al.*, Ultrafast vibrational control of organohalide perovskite optoelectronic devices using vibrationally promoted electronic resonance, *Nat. Mater.* **23**, 88 (2024).
- [21] T. G. Park, H. R. Na, S.-H. Chun, W. B. Cho, S. Lee, and F. Rotermund, Coherent control of interlayer vibrations in Bi₂Se₃ van der Waals thin-films, *Nanoscale* **13**, 19264 (2021).
- [22] D. M. Monahan, L. Whaley-Mayda, A. Ishizaki, and G. R. Fleming, Influence of weak vibrational-electronic couplings on 2D electronic spectra and inter-site coherence in weakly coupled photosynthetic complexes, *J. Chem. Phys.* **143**, 065101 (2015).
- [23] Y. Fujihashi, G. R. Fleming, and A. Ishizaki, Impact of environmentally induced fluctuations on quantum mechanically mixed electronic and vibrational pigment states in photosynthetic energy transfer and 2D electronic spectra, *J. Chem. Phys.* **142**, 212403 (2015).
- [24] G. S. Engel, T. R. Calhoun, E. L. Read, T. K. Ahn, T. Mančal, Y. C. Cheng, R. E. Blankenship, and G. R. Fleming, Evidence for wavelike energy transfer through quantum coherence in photosynthetic systems, *Nature* **446**, 782 (2007).
- [25] E. Romero, R. Augulis, V. I. Novoderezhkin, M. Ferretti, J. Thieme, D. Zigmantas, and R. V. Grondelle, Quantum coherence in photosynthesis for efficient solar-energy conversion, *Nat. Phys.* **10**, 676 (2014).
- [26] F. Fassioli, R. Dinshaw, P. C. Arpin, and G. D. Scholes, Photosynthetic light harvesting: Excitons and coherence, *J. R. Soc. Interface* **11**, 20130901 (2014).
- [27] E. A. Arsenaault, Y. Yoneda, M. Iwai, K. K. Niyogi, and G. R. Fleming, Vibronic mixing enables ultrafast energy flow in light-harvesting complex II, *Nat. Commun.* **11**, 1460 (2020).
- [28] A. J. Sneyd, T. Fukui, D. Paleček, S. Prodhon, I. Wagner, Y. Zhang, J. Sung, S. M. Collins, T. J. A. Slater, Z. Andajigarmaroudi, L. R. MacFarlane, J. D. Garcia-Hernandez, L. Wang, G. R. Whittell, J. M. Hodgkiss, K. Chen, D. Beljonne, I. Manners, R. H. Friend, and A. Rao, Efficient energy transport in an organic semiconductor mediated by transient exciton delocalization, *Sci. Adv.* **7**, eabh4232 (2021).
- [29] A. J. Sneyd, D. Beljonne, and A. Rao, A new frontier in exciton transport: Transient delocalization, *J. Phys. Chem. Lett.* **13**, 6820 (2022).
- [30] P. G. Wolynes, Some quantum weirdness in physiology, *Proc. Natl. Acad. Sci.* **106**, 17247 (2009).
- [31] A. Ishizaki and G. R. Fleming, Theoretical examination of quantum coherence in a photosynthetic system at physiological temperature, *Proc. Natl. Acad. Sci.* **106**, 17255 (2009).
- [32] M. B. Plenio, J. Almeida, and S. F. Huelga, Origin of long-lived oscillations in 2D-spectra of a quantum vibronic model: Electronic versus vibrational coherence, *J. Chem. Phys.* **139**, 235102 (2013).
- [33] S. J. Jang and B. Mennucci, Delocalized excitons in natural light-harvesting complexes, *Rev. Mod. Phys.* **90**, 035003 (2018).
- [34] M. Kang, H. Nuomin, S. N. Chowdhury, J. L. Yuly, K. Sun, J. Whitlow, J. Valdiviezo, Z. Zhang, P. Zhang, D. N. Beratan, and K. R. Brown, Seeking a quantum advantage with trapped-ion quantum simulations of condensed-phase chemical dynamics, *Nat. Rev. Chem.* **8**, 340 (2024).
- [35] R. Dutta *et al.*, Simulating chemistry on bosonic quantum devices, *J. Chem. Theory Comput.* **20**, 6426 (2024).
- [36] D. J. Gorman, B. Hemmerling, E. Megidish, S. A. Moeller, P. Schindler, M. Sarovar, and H. Haeflner, Engineering vibrationally assisted energy transfer in a trapped-ion quantum simulator, *Phys. Rev. X* **8**, 011038 (2018).
- [37] K. Sun, C. Fang, M. Kang, Z. Zhang, P. Zhang, D. N. Beratan, K. R. Brown, and J. Kim, Quantum simulation of polarized light-induced electron transfer with a trapped-ion qutrit system, *J. Phys. Chem. Lett.* **14**, 6071 (2023).

- [38] J. Whitlow, Z. Jia, Y. Wang, C. Fang, J. Kim, and K. R. Brown, Quantum simulation of conical intersections using trapped ions, *Nat. Chem.* **15**, 1509 (2023).
- [39] C. H. Valahu, V. C. Olaya-Agudelo, R. J. MacDonell, T. Navickas, A. D. Rao, M. J. Millican, J. B. Pérez-Sánchez, J. Yuen-Zhou, M. J. Biercuk, C. Hempel, T. R. Tan, and I. Kassal, Direct observation of geometric-phase interference in dynamics around a conical intersection, *Nat. Chem.* **15**, 1503 (2023).
- [40] D. G. A. Cabral, P. Khazaei, B. C. Allen, P. E. Videla, M. Schäfer, R. G. Cortiñas, A. C. Carrillo de Albornoz, J. Chávez-Carlos, L. F. Santos, E. Geva, and V. S. Batista, A roadmap for simulating chemical dynamics on a parametrically driven bosonic quantum device, *J. Phys. Chem. Lett.* **15**, 12042 (2024).
- [41] A. C. C. de Albornoz, R. G. Cortiñas, M. Schäfer, N. E. Frattini, B. Allen, D. G. A. Cabral, P. E. Videla, P. Khazaei, E. Geva, V. S. Batista, and M. H. Devoret, Oscillatory dissipative tunneling in an asymmetric double-well potential, [arXiv:2409.13113](https://arxiv.org/abs/2409.13113).
- [42] N. E. Frattini, R. G. Cortiñas, J. Venkatraman, X. Xiao, Q. Su, C. U. Lei, B. J. Chapman, V. R. Joshi, S. M. Girvin, R. J. Schoelkopf, S. Puri, and M. H. Devoret, Observation of pairwise level degeneracies and the quantum regime of the Arrhenius law in a double-well parametric oscillator, *Phys. Rev. X* **14**, 031040 (2024).
- [43] A. Potočník, A. Bargerbos, F. A. Y. N. Schröder, S. A. Khan, M. C. Collodo, S. Gasparinetti, Y. Salathé, C. Creatore, C. Eichler, H. E. Türeci, A. W. Chin, and A. Wallraff, Studying light-harvesting models with superconducting circuits, *Nat. Commun.* **9**, 904 (2018).
- [44] V. So, M. D. Suganthi, A. Menon, M. Zhu, R. Zhuravel, H. Pu, P. G. Wolynes, J. N. Onuchic, and G. Pagano, Trapped-ion quantum simulation of electron transfer models with tunable dissipation, *Sci. Adv.* **10**, eads8011 (2024).
- [45] G.-X. Wang, Y.-K. Wu, R. Yao, W.-Q. Lian, Z.-J. Cheng, Y.-L. Xu, C. Zhang, Y. Jiang, Y.-Z. Xu, B.-X. Qi, P.-Y. Hou, Z.-C. Zhou, L. He, and L.-M. Duan, Simulating the spin-boson model with a controllable reservoir in an ion trap, *Phys. Rev. A* **109**, 062402 (2024).
- [46] K. Sun, M. Kang, H. Nuomin, G. Schwartz, D. N. Beratan, K. R. Brown, and J. Kim, Quantum simulation of spin-boson models with structured bath, *Nat. Commun.* **16**, 4042 (2025).
- [47] A. Mattioni, F. Caycedo-Soler, S. F. Huelga, and M. B. Plenio, Design principles for long-range energy transfer at room temperature, *Phys. Rev. X* **11**, 041003 (2021).
- [48] C. R. Myers, G. J. Milburn, and J. Twamley, Vibrationally assisted quantum energy pumps, *New J. Phys.* **17**, 093030 (2015).
- [49] C. Monroe, W. C. Campbell, L.-M. Duan, Z.-X. Gong, A. V. Gorshkov, P. W. Hess, R. Islam, K. Kim, N. M. Linke, G. Pagano, P. Richerme, C. Senko, and N. Y. Yao, Programmable quantum simulations of spin systems with trapped ions, *Rev. Mod. Phys.* **93**, 025001 (2021).
- [50] G. D. Scholes, Long-range resonance energy transfer in molecular systems, *Annu. Rev. Phys. Chem.* **54**, 57 (2003).
- [51] G. J. Daniels, R. D. Jenkins, D. S. Bradshaw, and D. L. Andrews, Resonance energy transfer: The unified theory revisited, *J. Chem. Phys.* **119**, 2264 (2003).
- [52] G. D. Scholes, Limits of exciton delocalization in molecular aggregates, *Faraday Discuss.* **221**, 265 (2020).
- [53] D. Giovanni, M. Righetto, Q. Zhang, J. W. M. Lim, S. Ramesh, and T. C. Sum, Origins of the long-range exciton diffusion in perovskite nanocrystal films: Photon recycling vs exciton hopping, *Light: Sci. Appl.* **10**, 2 (2021).
- [54] M. Kastner, Diverging equilibration times in long-range quantum spin models, *Phys. Rev. Lett.* **106**, 130601 (2011).
- [55] D.-M. Storch, M. van den Worm, and M. Kastner, Interplay of soundcone and supersonic propagation in lattice models with power law interactions, *New J. Phys.* **17**, 063021 (2015).
- [56] A. Y. Guo, M. C. Tran, A. M. Childs, A. V. Gorshkov, and Z.-X. Gong, Signaling and scrambling with strongly long-range interactions, *Phys. Rev. A* **102**, 010401 (2020).
- [57] M. C. Tran, A. Y. Guo, A. Deshpande, A. Lucas, and A. V. Gorshkov, Optimal state transfer and entanglement generation in power-law interacting systems, *Phys. Rev. X* **11**, 031016 (2021).
- [58] C. Jameson, B. Basyildiz, D. Moore, K. Clark, and Z. Gong, Time optimal quantum state transfer in a fully-connected quantum computer, *Quantum Sci. Technol.* **9**, 015014 (2023).
- [59] F. Schlawin, M. Gessner, A. Buchleitner, T. Schätz, and S. S. Skourtis, Continuously parametrized quantum simulation of molecular electron-transfer reactions, *PRX Quantum* **2**, 010314 (2021).
- [60] A. Garg, J. N. Onuchic, and V. Ambegaokar, Effect of friction on electron transfer in biomolecules, *J. Chem. Phys.* **83**, 4491 (1985).
- [61] A. Lemmer, C. Cormick, D. Tamascelli, T. Schaetz, S. F. Huelga, and M. B. Plenio, A trapped-ion simulator for spin-boson models with structured environments, *New J. Phys.* **20**, 073002 (2018).
- [62] D. Tamascelli, A. Smirne, S. F. Huelga, and M. B. Plenio, Nonperturbative treatment of non-Markovian dynamics of open quantum systems, *Phys. Rev. Lett.* **120**, 030402 (2018).
- [63] M. Yang, A reduced density-matrix theory of absorption line shape of molecular aggregate, *J. Chem. Phys.* **123**, 124705 (2005).
- [64] S. Polyutov, O. Kühn, and T. Pullerits, Exciton-vibrational coupling in molecular aggregates: Electronic versus vibronic dimer, *Chem. Phys.* **394**, 21 (2012).
- [65] V. Tiwari, W. K. Peters, and D. M. Jonas, Electronic resonance with anticorrelated pigment vibrations drives photosynthetic energy transfer outside the adiabatic framework, *Proc. Natl. Acad. Sci.* **110**, 1203 (2013).
- [66] S. S. Skourtis, A. J. R. Da Silva, W. Bialek, and J. N. Onuchic, New look at the primary charge separation in bacterial photosynthesis, *J. Phys. Chem.* **96**, 8034 (1992).
- [67] V. So, M. D. Suganthi, M. Zhu, A. Menon, G. Tomaras, R. Zhuravel, H. Pu, P. G. Wolynes, J. N. Onuchic, and G. Pagano, Quantum simulation of charge and exciton transfer in multi-mode models using engineered reservoirs, [arXiv:2505.22729](https://arxiv.org/abs/2505.22729).

- [68] N. J. Hestand and F. C. Spano, Expanded theory of H- and J-molecular aggregates: The effects of vibronic coupling and intermolecular charge transfer, *Chem. Rev.* **118**, 7069 (2018).
- [69] D. E. Logan and P. G. Wolynes, Dephasing and Anderson localization in topologically disordered systems, *Phys. Rev. B* **36**, 4135 (1987).
- [70] F. Caruso, A. W. Chin, A. Datta, S. F. Huelga, and M. B. Plenio, Highly efficient energy excitation transfer in light-harvesting complexes: The fundamental role of noise-assisted transport, *J. Chem. Phys.* **131**, 105106 (2009).
- [71] C. Maier, T. Brydges, P. Jurcevic, N. Trautmann, C. Hempel, B. P. Lanyon, P. Hauke, R. Blatt, and C. F. Roos, Environment-assisted quantum transport in a 10-qubit network, *Phys. Rev. Lett.* **122**, 050501 (2019).
- [72] D. Dwiputra and F. P. Zen, Environment-assisted quantum transport and mobility edges, *Phys. Rev. A* **104**, 022205 (2021).
- [73] Z.-Z. Li, L. Ko, Z. Yang, M. Sarovar, and K. B. Whaley, Interplay of vibration- and environment-assisted energy transfer, *New J. Phys.* **24**, 033032 (2022).
- [74] M. Mohseni, P. Rebentrost, S. Lloyd, and A. Aspuru-Guzik, Environment-assisted quantum walks in photosynthetic energy transfer, *J. Chem. Phys.* **129**, 174106 (2008).
- [75] M. B. Plenio and S. F. Huelga, Dephasing-assisted transport: Quantum networks and biomolecules, *New J. Phys.* **10**, 113019 (2008).
- [76] A. Chenu, M. Beau, J. Cao, and A. del Campo, Quantum simulation of generic many-body open system dynamics using classical noise, *Phys. Rev. Lett.* **118**, 140403 (2017).
- [77] F. Campaioli and J. H. Cole, Exciton transport in amorphous polymers and the role of morphology and thermalisation, *New J. Phys.* **23**, 113038 (2021).
- [78] E. Scheer and J. C. Cuevas, *Molecular Electronics: An Introduction to Theory and Experiment* (World Scientific, Singapore, 2017), Vol. 15.
- [79] R. Zhuravel, H. Huang, G. Polycarpou, S. Polydorides, P. Motamarri, L. Katrivas, D. Rotem, J. Sperling, L. A. Zotti, A. B. Kotlyar, J. C. Cuevas, V. Gavini, S. S. Skourtis, and D. Porath, Backbone charge transport in double-stranded DNA, *Nat. Nanotechnol.* **15**, 836 (2020).
- [80] F. Levi, S. Mostarda, F. Rao, and F. Mintert, Quantum mechanics of excitation transport in photosynthetic complexes: A key issues review, *Rep. Prog. Phys.* **78**, 082001 (2015).
- [81] V. I. Novoderezhkin, A. B. Doust, C. Curutchet, G. D. Scholes, and R. van Grondelle, Excitation dynamics in phycoerythrin 545: Modeling of steady-state spectra and transient absorption with modified redfield theory, *Biophys. J.* **99**, 344 (2010).
- [82] W. Dür, G. Vidal, and J. I. Cirac, Three qubits can be entangled in two inequivalent ways, *Phys. Rev. A* **62**, 062314 (2000).
- [83] D. Cruz, R. Fournier, F. Gremion, A. Jeannerot, K. Komagata, T. Tosic, J. Thiesbrummel, C. L. Chan, N. Macris, M.-A. Dupertuis, and C. Javerzac-Galy, Efficient quantum algorithms for GHZ and W states, and implementation on the IBM quantum computer, *Adv. Quantum Technol.* **2**, 1900015 (2019).
- [84] C. Monroe, D. M. Meekhof, B. E. King, W. M. Itano, and D. J. Wineland, Demonstration of a fundamental quantum logic gate, *Phys. Rev. Lett.* **75**, 4714 (1995).
- [85] M. Cetina, L. Egan, C. Noel, M. Goldman, D. Biswas, A. Risinger, D. Zhu, and C. Monroe, Control of transverse motion for quantum gates on individually addressed atomic qubits, *PRX Quantum* **3**, 010334 (2022).
- [86] P.-Y. Hou, J. J. Wu, S. D. Erickson, G. Zarantonello, A. D. Brandt, D. C. Cole, A. C. Wilson, D. H. Slichter, and D. Leibfried, Indirect cooling of weakly coupled trapped-ion mechanical oscillators, *Phys. Rev. X* **14**, 021003 (2024).
- [87] C. Schneider, D. Porras, and T. Schaetz, Experimental quantum simulations of many-body physics with trapped ions, *Rep. Prog. Phys.* **75**, 024401 (2012).
- [88] Z. Davoudi, M. Hafezi, C. Monroe, G. Pagano, A. Seif, and A. Shaw, Towards analog quantum simulations of lattice gauge theories with trapped ions, *Phys. Rev. Res.* **2**, 023015 (2020).
- [89] X.-L. Deng, D. Porras, and J. I. Cirac, Effective spin quantum phases in systems of trapped ions, *Phys. Rev. A* **72**, 063407 (2005).
- [90] Y. Zhu, A. M. Green, N. H. Nguyen, C. Huerta Alderete, E. Mossman, and N. M. Linke, Pairwise-parallel entangling gates on orthogonal modes in a trapped-ion chain, *Adv. Quantum Technol.* **6**, 2300056 (2023).
- [91] T. Navickas, R. J. MacDonell, C. H. Valahu, V. C. Olaya-Agudelo, F. Scuccimarra, M. J. Millican, V. G. Matsos, H. L. Nourse, A. D. Rao, M. J. Biercuk, C. Hempel, I. Kassal, and T. R. Tan, Experimental quantum simulation of chemical dynamics, *J. Am. Chem. Soc.* **147**, 23566 (2025).
- [92] R. J. MacDonell, C. E. Dickerson, C. J. T. Birch, A. Kumar, C. L. Edmunds, M. J. Biercuk, C. Hempel, and I. Kassal, Analog quantum simulation of chemical dynamics, *Chem. Sci.* **12**, 9794 (2021).
- [93] I. Vybornyi, L. S. Dreissen, D. Kiesenhofer, H. Hainzer, M. Bock, T. Ollikainen, D. Vadlejch, C. F. Roos, T. E. Mehlstäubler, and K. Hammerer, Sideband thermometry of ion crystals, *PRX Quantum* **4**, 040346 (2023).
- [94] G.-D. Lin, S.-L. Zhu, R. Islam, K. Kim, M.-S. Chang, S. Korenblit, C. Monroe, and L.-M. Duan, Large-scale quantum computation in an anharmonic linear ion trap, *Europhys. Lett.* **86**, 60004 (2009).
- [95] M. Zhu, V. So, G. Pagano, and H. Pu, Dissipation-assisted steady-state entanglement engineering based on electron transfer models, *Phys. Rev. A* **112**, 012617 (2025).
- [96] J. D. Guimarães, J. Lim, M. I. Vasilevskiy, S. F. Huelga, and M. B. Plenio, Accelerating two-dimensional electronic spectroscopy simulations with a probe qubit protocol, *Phys. Rev. Res.* **7**, 023130 (2025).
- [97] C.-K. Lee, C.-Y. Hsieh, S. Zhang, and L. Shi, Simulation of condensed-phase spectroscopy with near-term digital quantum computers, *J. Chem. Theory Comput.* **17**, 7178 (2021).
- [98] A. D. Somoza, O. Marty, J. Lim, S. F. Huelga, and M. B. Plenio, Dissipation-assisted matrix product factorization, *Phys. Rev. Lett.* **123**, 100502 (2019).
- [99] R. H. Goldsmith, M. R. Wasielewski, and M. A. Ratner, Electron transfer in multiply bridged donor-acceptor molecules: Dephasing and quantum coherence, *J. Phys. Chem. B* **110**, 20258 (2006).

- [100] Z.-Z. Li, L. Ko, Z. Yang, M. Sarovar, and K. B. Whaley, Unraveling excitation energy transfer assisted by collective behaviors of vibrations, *New J. Phys.* **23**, 073012 (2021).
- [101] N. Wu, J. Feist, and F. J. Garcia-Vidal, When polarons meet polaritons: Exciton-vibration interactions in organic molecules strongly coupled to confined light fields, *Phys. Rev. B* **94**, 195409 (2016).
- [102] S. Ding, G. Maslennikov, R. Hablützel, and D. Matsukevich, Cross-Kerr nonlinearity for phonon counting, *Phys. Rev. Lett.* **119**, 193602 (2017).
- [103] C. Zhang, M. Gruebele, D. E. Logan, and P. G. Wolynes, Surface crossing and energy flow in many-dimensional quantum systems, *Proc. Natl. Acad. Sci.* **120**, e2221690120 (2023).
- [104] H. Lee, Y.-C. Cheng, and G. R. Fleming, Coherence dynamics in photosynthesis: Protein protection of excitonic coherence, *Science* **316**, 1462 (2007).
- [105] S. Jang, Y.-C. Cheng, D. R. Reichman, and J. D. Eaves, Theory of coherent resonance energy transfer, *J. Chem. Phys.* **129**, 101104 (2008).
- [106] A. Ishizaki and G. R. Fleming, On the adequacy of the Redfield equation and related approaches to the study of quantum dynamics in electronic energy transfer, *J. Chem. Phys.* **130**, 234110 (2009).
- [107] A. Ishizaki and G. R. Fleming, Unified treatment of quantum coherent and incoherent hopping dynamics in electronic energy transfer: Reduced hierarchy equation approach, *J. Chem. Phys.* **130**, 234111 (2009).
- [108] Z.-Z. Li and K. B. Whaley, Enhancement of vibrationally assisted energy transfer by proximity to exceptional points, probed by fluorescence-detected vibrational spectroscopy, *Phys. Rev. Res.* **6**, 023149 (2024).
- [109] V. C. Olaya-Agudelo, B. Stewart, C. H. Valahu, R. J. MacDonell, M. J. Millican, V. G. Matsos, F. Scuccimarra, T. R. Tan, and I. Kassal, Simulating open-system molecular dynamics on analog quantum computers, *Phys. Rev. Res.* **7**, 023215 (2025).
- [110] M. I. Collins, F. Campaioli, M. J. Y. Tayebjee, J. H. Cole, and D. R. McCamey, Quintet formation, exchange fluctuations, and the role of stochastic resonance in singlet fission, *Commun. Phys.* **6**, 64 (2023).
- [111] F. Campaioli, A. Pagano, D. Jaschke, and S. Montangero, Optimization of ultrafast singlet fission in one-dimensional rings towards unit efficiency, *PRX Energy* **3**, 043003 (2024).
- [112] C. Senko, P. Richerme, J. Smith, A. Lee, I. Cohen, A. Retzker, and C. Monroe, Realization of a quantum integer-spin chain with controllable interactions, *Phys. Rev. X* **5**, 021026 (2015).
- [113] C. L. Edmunds, E. Rico, I. Arrazola, G. K. Brennen, M. Meth, R. Blatt, and M. Ringbauer, Symmetry-protected topological Haldane phase on a qudit quantum processor, *PRX Quantum* **6**, 020349 (2025).
- [114] R. Sweke, J. Eisert, and M. Kastner, Lieb–Robinson bounds for open quantum systems with long-ranged interactions, *J. Phys. A: Math. Theor.* **52**, 424003 (2019).
- [115] D. Fallas Padilla, V. So, A. Menon, R. Zhuravel, H. Pu, and G. Pagano, Code: Delocalized excitation transfer in open quantum systems with long-range interaction, <https://doi.org/10.5281/zenodo.16923297> (2025).
- [116] A. Wunsche, Displaced Fock states and their connection to quasiprobabilities, *Quantum Opt.: J. Eur. Opt. Soc. Part B* **3**, 359 (1991).
- [117] C. H. Bennett, D. P. DiVincenzo, J. A. Smolin, and W. K. Wootters, Mixed-state entanglement and quantum error correction, *Phys. Rev. A* **54**, 3824 (1996).
- [118] V. Coffman, J. Kundu, and W. K. Wootters, Distributed entanglement, *Phys. Rev. A* **61**, 052306 (2000).