Quantum simulation of clustered photosynthetic light harvesting in a superconducting quantum circuit

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We propose a method to emulate the exciton energy transfer (EET) of photosynthetic complexes in a quantum superconducting circuit. Our system is composed of two pairs of superconducting charge qubits coupled to two separated high-Q superconducting transmission line resonators (TLRs), respectively. The two TLRs interact with each other capacitively. When the frequencies of the qubits are largely detuned from those of the TLRs, we simulate the process of EET from the first qubit to the fourth qubit. By tuning the couplings between the qubits and the TLRs, as well as the coupling between the two TLRs, we can modify the effective coupling strengths between the qubits and thus study the geometric effects on the EET. It is shown that a moderately-clustered geometry supports optimal EET by using exciton delocalization and an energy matching condition. And the population loss during the EET has been trapped in the two TLRs.

Keywords: photosynthetic light harvesting; exciton energy transfer; superconducting quantum circuit; quantum simulation
0.1. Introduction

Energy plays an important role in modern society. The chemical energy supporting all lives on earth is mainly from the solar energy harvested by photosynthesis [1, 2, 3, 4]. The solar energy can be captured and transferred to the reaction centers of photosynthetic systems in a short time with high efficiency [1, 5, 6]. Therefore, it might be beneficial to learn from the natural photosynthesis to design efficient artificial light-harvesting devices.

In the past few decades, many researches were focused on the study of the exciton energy transfer (EET) process in photosynthesis [6, 7, 8, 9, 10, 11]. Based on the quantum dynamics of open systems [2, 12], much knowledge has been learned about the efficiency of the EET [13, 14, 15, 16, 17], together with the spatial and energetic arrangement of the pigments [18, 19, 20, 21, 22]. The experimentally-observed coherent phenomena in 2D spectroscopy may be attributed to nearly-resonant coupling to an underdamped vibrational mode in the bath [12, 23]. In natural photosynthesis, EET can be accomplished within 100 picoseconds with almost 100% efficiency [24].

Schulten et al. [25] observed that the bacteriochlorophylls involved in the overall excitation transfer are found in a coplanar arrangement. Ishizaki and Fleming [26] showed that, by dimerization, the energy flow in the Fenna-Matthews-Olson (FMO) complex occurs primarily through two EET pathways. Yang et al. [27] found out that the dimerization in light-harvesting complex II (LH2) can effectively speed up the energy transfer between LH2 rings due to symmetry breaking. In a wheel-shaped artificial light-harvesting complex, the energy of the initially-excited antenna can be efficiently directed to the reaction center, with quantum beating lasted for hundreds of femtoseconds [28, 29]. In 2013, Ai et al. [30] revealed that clustered geometry utilizes exciton delocalization and energy matching to optimize EET, which can be utilized to explain the efficient EET in FMO complex [31].

In order to obtain the explicit relation between geometry and efficiency, some researches have explored random networks to discover some interesting findings [32, 33,
Because the EET is sensitively influenced by the interference within the photosynthetic complex, even small changes in the geometry of the complex could turn constructive interference to destructive and thus result in significant drop in the efficiency. Pair sites renders EET properties robust against perturbations [32]. Compact structures tend to display high performance in the transport dynamics [33]. The networks characterized by Hamiltonians with centrosymmetry outperform those with completely-random arrangements [34]. These discoveries seem to suggest that clustered geometries could favour efficient quantum transport.

On the other hand, much progress has been made in quantum information science, inspiring several interesting quantum simulation [35, 36] experiments to verify the design principles for optimal light-harvesting [28 29, 37, 38, 39, 40, 41, 42]. By using bath engineering and the gradient ascent pulse engineering algorithm, Wang et al. [37] performed an experimental quantum simulation of photosynthetic energy transfer by using nuclear magnetic resonance (NMR). It was demonstrated that the open quantum dynamics in an $N$-level system, with arbitrary Hamiltonian and bath spectral density, can be effectively emulated by an NMR system with $\log_2 N$ qubits [38]. Meanwhile, Gorman et al. [43] showed in a trapped-ion system that the long-lived vibrational mode in the bath can assist the energy transfer. Superconducting quantum circuits provide another intriguing platform for quantum simulation [44, 45, 46, 47, 48, 49, 50, 51]. In 2012, Mostame et al. [39] simulated a complicated environment with a given spectral density for the EET in photosynthetic complexes by using inductor-resistor-capacitor oscillators. In 2018, Potočnik et al. [40, 41] experimentally demonstrated that light harvesting for a given geometry can be optimized by tuning the environmental noise.

However, although they have shown the potential of optimizing energy transfer by engineering the bath, none of them have experimentally demonstrated the effect of geometry on the EET efficiency. In Ref. [30], it was shown that, in a linear geometry, moderate dimerization promotes the energy transfer. Therefore, it might be interesting to simulate the EET in different geometries to verify the design principals of optimal geometries. In this paper, we design a system composed of four superconducting charge
qubits and two superconducting transmission line resonators (TLRs). Here, two qubits form a pair and are coupled to one TLR. And the two TLRs are capacitively coupled with each other. Although there are no direct interactions between the qubits, the effective couplings among them can be induced by the simultaneous couplings to the common mode in each TLR [52, 53]. Furthermore, the effective couplings can be tuned by adjusting the level spacings of the qubits, and their interaction strengths with the TLRs [54], and the coupling strength between the two TLRs. In this way, we can investigate the EET for different geometries. Alternatively, for the sake of simplicity, the direct couplings between the qubits can be introduced instead. This alternative setup can simplify the arrangement and approximately simulate the EET dynamics for the parameter regime when the nearest-neighbour couplings dominate the transport.

This paper is organized as follows: In the next section, we briefly introduce the theory for describing the EET in photosynthesis. In Sec. 0.3, we propose a setup consisting of four superconducting charge qubits and two TLRs to simulate the photosynthetic energy transfer. The effective Hamiltonian for the four qubits is obtained by the Fröhlich-Nakajima transformation. In Sec. 0.4, the energy transfer dynamics is numerically simulated by solving the Lindblad master equation, which confirms the previous investigation in Ref. 30. Finally, the experimental feasibility and the main conclusions are discussed in Sec. 0.5.

### 0.2. Photosynthetic Light Harvesting

In the photosynthesis with 4 chromophores, the EET is governed by the Frenkel-exciton Hamiltonian [1 30],

\[
H = \sum_{j=1}^{4} \varepsilon_j |j\rangle \langle j| + \sum_{i \neq j=1}^{4} J_{ij} |i\rangle \langle j| + \text{h.c.,} \tag{1}
\]
where \( \varepsilon_j \) is the site energy when \( j \)-th chromophore is in the excited state, \( |j\rangle \) is the state when \( j \)-th chromophore is in the excited state while all other chromophores are in the ground state, \( J_{ij} \) is the dipole-dipole interaction between \( i \)-th and \( j \)-th chromophores.

Due to the strong coupling \( J_{ij} \gtrsim |\varepsilon_i - \varepsilon_j| \), the exciton energy can coherently oscillate between any two sites \( i \) and \( j \). However, because of the pure-dephasing-form system-bath Hamiltonian,

\[
H_{SB} = \sum_{j,k} g_{jk} |j\rangle\langle j| (a_{jk}^\dagger + a_{jk}),
\]

(2)

where \( g_{jk} \) is the coupling strength between \( j \)-th chromophore and its local bath mode with frequency \( \omega_k \) and creation (annihilation) operator \( a_{jk}^\dagger \) \( (a_{jk}) \), the exciton energy can be irreversibly transferred to the target chromophore. In general, the system-bath coupling is described by the spectral density,

\[
G(\omega) = \sum_k g_{jk}^2 \delta(\omega - \omega_k).
\]

(3)

For a given geometry, the position \( \vec{r}_i \) and transition dipole \( \vec{\mu}_i \) of every chromophore is fixed and thus the dipole-dipole interaction between any pair of two chromophores is determined by

\[
J_{ij} = \frac{1}{4\pi\varepsilon_0|\vec{r}_{ij}|^3} [\vec{\mu}_i \cdot \vec{\mu}_j - 3(\vec{\mu}_i \cdot \vec{r}_{ij})(\vec{\mu}_j \cdot \vec{r}_{ij})],
\]

(4)

where \( \vec{r}_{ij} = r_{ij} \hat{r}_{ij} = \vec{r}_i - \vec{r}_j \) is the displacement vector from site \( j \) to site \( i \), \( \varepsilon_0 \) is the vacuum permittivity. In addition, the spatial distribution of \( \varepsilon_j \) also facilitates the energy transfer by making use of the energy gradient towards the target chromophore, as shown in Fig. 1(b). In Ref. [30], by using the coherent modified Redfield theory, it is shown that, in a clustered geometry, exciton delocalization and energy matching cooperate to optimize EET.
Figure 1: (a) Schematic diagram of the superconducting circuit for simulating photosynthetic energy transfer. The two TLRs are capacitively coupled with each other. Charge qubits \( Q_1 \) and \( Q_2 \) form the donor, while qubits \( Q_3 \) and \( Q_4 \) act as the acceptor. \( Q_1 \) and \( Q_2 \) (\( Q_3 \) and \( Q_4 \)) are capacitively coupled to TLR \( R_a \) \( (R_b) \). (b) In a linear photosynthetic system, sites 1 and 2 form the donor pair, while sites 3 and 4 are the acceptor pair. The total distance between the two ends is fixed at \( R \) and the intra-pair distance is \( r \). (c) Energy configuration of the four qubits for energy transfer, where only the excited states are involved in the EET and the ground states are not shown for simplicity.

0.3. Physical setup

Let us consider a superconducting quantum circuit composed of four superconducting charge qubits and two 1D high-\( Q \) superconducting TLRs, as shown in Fig. 1(a). In order to study the cluster-to-cluster geometric effects, there should be two clusters at least, and two qubits can form a cluster. Therefore, four qubits are the minimum number of qubits to observe this effect. As shown in Fig. 1(b), we investigate the EET efficiency in a linear photosynthetic complex with 4 chromophores. The distance between the two ends is fixed at \( R = 40 \, \text{Å} \), while sites 1 and 2 form the donor pair, and sites 3 and 4 are the acceptor pair, with intra-pair distance \( r < R/3 \). The energy-level diagram of the four qubits is schematically shown in Fig. 1(c). Qubits \( Q_1 \) and \( Q_2 \) (\( Q_3 \) and \( Q_4 \)) are coupled to the TLR \( R_a \) \( (R_b) \) capacitively. Here, we take \( Q_1 \) and \( Q_2 \) as donors because their energies are higher than those of the qubits \( Q_3 \) and \( Q_4 \), acting as acceptors. The effective couplings among these qubits exhibit the geometrical effects in
photosynthetic complexes, because the couplings between pigments sensitively depend on their relative distances and orientations of electric dipoles. The couplings other than the nearest-neighbor couplings subtly modify the energy spectrum and thus the quantum dynamics. Furthermore, instead of direct couplings among qubits, we simplify the quantum circuit by introducing the additional TLRs. In other words, the TLRs play the role as the quantum data bus to induce the indirect couplings between the qubits. The frequencies of TLRs should be much smaller than the qubits to avoid the excitation of the TLRs. The distances between any two qubits are far enough to avoid direct interactions between them. Therefore, the energy is transferred from $Q_1$ to $Q_4$ by the indirect interactions among the qubits induced by simultaneously couplings to the common TLRs.

Under the rotating-wave approximation \[57\], the Hamiltonian of the four qubits and two TLRs can be written as

\[
H_1 = \omega_a a^\dagger a + \sum_{j_1=1}^{2} \left( \frac{\omega_{j_1}}{2} \sigma^z_{j_1} + g_{j_1} (a^\dagger \sigma^-_{j_1} + a \sigma^+_{j_1}) \right) \\
+ \omega_b b^\dagger b + \sum_{j_2=3}^{4} \left( \frac{\omega_{j_2}}{2} \sigma^z_{j_2} + g_{j_2} (b^\dagger \sigma^-_{j_2} + b \sigma^+_{j_2}) \right) \\
+ g^a_b (a^\dagger + a) (b^\dagger + b),
\]

where $\omega_a$, $\omega_b$, $\omega_{j_1}$, and $\omega_{j_2}$ are the transition frequencies of the TLRs $R_a$ and $R_b$, and qubits $Q_{j_s}$ ($s = 1, 2$), respectively. Here, $j_1 = 1, 2$ and $j_2 = 3, 4$. $g_{j_1}$ ($g_{j_2}$) is the coupling strength between the qubit $Q_{j_s}$ ($Q_{j_z}$) and the TLR $R_a$ ($R_b$). $g^a_b$ is the coupling strength between TLRs $R_a$ and $R_b$. $a^\dagger$ and $b^\dagger$ are the creation operators of $R_a$ and $R_b$, respectively. $\sigma^+_{j_s} = |e\rangle_{j_s} \langle g|$ and $\sigma^-_{j_s}$ are the rising and Pauli operator of $Q_{j_s}$, respectively. $|g\rangle_{j_s}$ and $|e\rangle_{j_s}$ are the ground and excited states of $Q_{j_s}$, respectively.

By using the Fröhlich-Nakajima transformation \[46\]

\[
U = \exp \left[ \sum_{j_1=1}^{2} \frac{g_{j_1}}{\delta_{j_1}} (a^\dagger \sigma^-_{j_1} - a \sigma^+_{j_1}) + \sum_{j_2=3}^{4} \frac{g_{j_2}}{\delta_{j_2}} (b^\dagger \sigma^-_{j_2} - b \sigma^+_{j_2}) \right]
\]

(6)
with $\delta_{j_1} = \omega_{j_1} - \omega_a \gg g_{j_1}$ and $\delta_{j_2} = \omega_{j_2} - \omega_b \gg g_{j_2}$, the original Hamiltonian of the system $H_1$ becomes

$$H_2 = U^\dagger H_1 U. \quad (7)$$

In the appendix, we give the detailed expression of $H_2$.

When the TLRs are initially prepared in the vacuum state and the high-order terms of $g_j/\delta_j$ can be omitted, the Hamiltonian $H_2$ can be reduced to

$$H_{\text{eff}} = \sum_{j_1=1}^{2} \left( \omega_{j_1} + \frac{g_{j_1}^2}{\delta_{j_1}} \right) |e\rangle_{j_1} \langle e| + \sum_{j_2=3}^{4} \left( \omega_{j_2} + \frac{g_{j_2}^2}{\delta_{j_2}} \right) |e\rangle_{j_2} \langle e|$$

$$+ J_{12} \left( \sigma_1^+ \sigma_2^- + \sigma_2^+ \sigma_1^- \right) + J_{34} \left( \sigma_3^+ \sigma_4^- + \sigma_4^+ \sigma_3^- \right) + J_{24} \left( \sigma_2^+ \sigma_4^- + \sigma_4^+ \sigma_2^- \right) + J_{14} \left( \sigma_1^+ \sigma_4^- + \sigma_4^+ \sigma_1^- \right), \quad (8)$$

where $J_{12} = \frac{g_{j_1} g_{j_2}}{\delta_1 \delta_2} (\delta_1 + \delta_2)$, $J_{34} = \frac{g_{j_3} g_{j_4}}{\delta_3 \delta_4} (\delta_3 + \delta_4)$, $J_{23} = \frac{g_{j_2} g_{j_3}}{\delta_2 \delta_3}$, $J_{13} = \frac{g_{j_1} g_{j_3}}{\delta_1 \delta_3}$, $J_{24} = \frac{g_{j_2} g_{j_4}}{\delta_2 \delta_4}$, and $J_{14} = \frac{g_{j_1} g_{j_4}}{\delta_1 \delta_4}$ are the indirect coupling strengths between any two qubits, respectively. In order to mimic the geometric effects, the effective couplings $J_{ij}$’s should be tunable. This can be achieved by adjusting detunings $\delta_j$’s, qubit-resonator couplings, and resonator-resonator coupling [54].

When a quantum system interacts with a quantum bath, the states of the quantum system entangle with those of the quantum bath [11]. The correlation function of the classical bath is real-valued and time-symmetric. However, when a quantum system interacts with a quantum bath, the quantum system experiences time-dependent transition frequencies and thus results in a complex-valued time-asymmetric correlation function of the bath [11]. Due to this difference, the energy generally prefers to transfer from the higher-energy level to the lower-energy level of the quantum system interacting with a quantum bath [11]. Furthermore, the populations of all levels tend to be equal at the steady state in the case of a classical bath. The latter has been recently observed in the experimental NMR simulation [37]. In our configuration, the transi-
tion frequencies of the four qubits are assumed to satisfy the following relation, i.e.,
\[
\left(\omega_1 + \frac{g_1^2}{\delta_1}\right) > \cdots > \left(\omega_4 + \frac{g_4^2}{\delta_4}\right),
\]
as shown in Fig. 1(b). Moreover, coupling strengths \(J_{12}\) and \(J_{34}\) are assumed to be larger than \(J_{23}\) to indicate that qubits \(Q_1\) and \(Q_2\) form the donor pair, while qubits \(Q_3\) and \(Q_4\) are the acceptor pair. For simplicity, we take \(g_1 = g_4, g_2 = g_3\) and \(\delta_1 \approx \delta_2 \approx \delta_3 \approx \delta_4\) to achieve \(J_{12} = J_{34} > J_{23}\) [30]. Thus we use the following parameters \(\omega_a/2\pi = \omega_b/2\pi = 3\) GHz, \(\omega_1/2\pi = 13.115\) GHz, \(\omega_2/2\pi = 13.009\) GHz, \(\omega_3/2\pi = 12.991\) GHz, \(\omega_4/2\pi = 13.078\) GHz [44, 46] in our numerical simulations. The proposed parameters in the superconducting circuit model can be obtained from those found in biological systems by scaling down a factor \(3 \times 10^4\).

0.4. simulation of EET process

In the previous section, we obtain the effective Hamiltonian for the 4 qubits by the Fröhlich-Nakajima transformation and the rotating-wave approximation. It is reasonable to question the validity of rotating-wave approximation since there is strong coupling between the TLRs and the qubit-TLR couplings are relative large as compared to the frequencies of the TLRs. Hereafter, we shall numerically simulate quantum dynamics of the master equation under the exact Hamiltonian without the rotating-wave approximation.

In the interaction picture with respect to

\[
H_0 = \omega_a a^\dagger a + \sum_{j_1=1}^{2} \frac{\omega_{j_1}}{2} \sigma_{j_1}^z + \omega_b b^\dagger b + \sum_{j_2=3}^{4} \frac{\omega_{j_2}}{2} \sigma_{j_2}^z,
\] (9)
we can derive the Lindblad-form master equation as \[58, 16\]

\[
\dot{\rho} = -i [H_I, \rho] + \sum_{r=a,b} \kappa_r (N_r + 1) D[r] \rho \\
+ \sum_{r=a,b} \kappa_r N_r D[r^\dagger] \rho + \sum_{l=1}^4 \Gamma_l^\gamma (N_l + 1) D[\sigma^\gamma_l] \rho \\
+ \sum_{l=1}^4 \Gamma_l^\gamma N_l D[\sigma^\gamma^\dagger_l] \rho + \sum_{l=1}^4 \Gamma_l^\gamma D[\sigma^\gamma^\dagger_l] \rho, \tag{10}
\]

where the interaction Hamiltonian reads

\[
H_I = \sum_{j=1}^2 g_j \left( a\sigma_j^+ e^{i\delta_j t} + a^{\dagger}\sigma_j^+ e^{i(\omega_a + \omega_j)t} \right) \\
+ \sum_{j=3}^4 g_j \left( b\sigma_j^+ e^{i\delta_j t} + b^{\dagger}\sigma_j^+ e^{i(\omega_b + \omega_j)t} \right) \\
+ g_a \left( ab e^{-i\Delta_a t} + a^{\dagger}b^{\dagger} e^{i\delta_a t} \right) + \text{h.c.}, \tag{11}
\]

\[
\delta_a^b = \omega_b - \omega_a, \tag{12}
\]

\[
\Delta_a^b = \omega_b + \omega_a, \tag{13}
\]

\[
D[A] \rho = \left( 2A\rho A^\dagger - A^\dagger A\rho - \rho A^\dagger A \right) / 2, \tag{14}
\]

\[
N_r = \frac{1}{\exp(h\omega_r/k_B T) - 1}, \tag{15}
\]

\[
N_l = \frac{1}{\exp(h\omega_l/k_B T)}, \tag{16}
\]

\(\kappa_r (r = a, b)\) is the leakage rate of TLR \(r\), \(\Gamma_l^\gamma\) and \(\Gamma_l^\gamma (l = 1, 2, 3, 4)\) are the spontaneous emission and pure-dephasing rates of the \(l\)-th qubit, respectively.

In natural photosynthesis, the energy transfer is generally restricted in the single-excitation subspace. For simplicity, we label the bases as \(|1\rangle = |e\rangle_1|g\rangle_2|g\rangle_3|g\rangle_4|0_a0_b\rangle, \n\]

\(|2\rangle = |g\rangle_1|e\rangle_2|g\rangle_3|g\rangle_4|0_a0_b\rangle, |3\rangle = |g\rangle_1|g\rangle_2|e\rangle_3|g\rangle_4|0_a0_b\rangle, \text{ and } |4\rangle = |g\rangle_1|g\rangle_2|g\rangle_3|e\rangle_4|0_a0_b\rangle. \n\]

Here, \(|n_an_b\rangle\) is the Fock state of TLR\(_a\) and TLR\(_b\). In addition, \(|a\rangle = |g\rangle_1|g\rangle_2|g\rangle_3|g\rangle_4|1_a0_b\rangle\) and \(|b\rangle = |g\rangle_1|g\rangle_2|g\rangle_3|g\rangle_4|0_a1_b\rangle\) indicate single-excitation in one of the TLRs. In our
simulation, the system composed of four superconducting qubits and two TLRs is initially prepared at the state $|1\rangle$. In Fig. 2, we show the time evolution of the populations of single-excitation states of the four qubits $P_m = \langle m | \rho | m \rangle$ ($m = 1, 2, 3, 4$). In a realistic experiment, each qubit can be dispersively coupled to a TLR. The qubit’s population can be extracted by measuring the phase of the signal in the TLR [59]. And the qubit’s decay time will not be significantly modified by Purcell-like couplings to the TLRs, which will be discussed in detail in Sec. 0.5. Here, we take $1/\Gamma_j^\gamma = 3 \mu s$ and $1/\Gamma_j^\phi = 70 \text{ ns}$ ($j = 1, 2, 3, 4$) [59], which meet the requirement $\Gamma_j^\phi \gg \Gamma_j^\gamma$ [60, 61, 62].

The leakage rates of two TLRs are $\kappa_a^{-1} = \kappa_b^{-1} = 10 \mu s$ [63].

In Fig. 2, we demonstrate the population dynamics of the four qubits for three different geometries, corresponding to three different sets of nearest-neighbor couplings $J_{12}, J_{34}$, and $J_{23}$. In Ref. [30], it has been proven that the next-nearest-neighbor couplings $J_{13}$ and $J_{24}$, and the end-to-end coupling $J_{14}$ plays a minor role in the EET. First of all, we would like to simulate the energy transfer for an equal-coupling geometry. Therefore, we investigate the energy transfer dynamics of the system with approximately equal-coupling strengths between adjacent qubits in Fig. 2(a). It corresponds to the equally-spaced geometry in Ref. [30]. In order to achieve $J_{12} = J_{34} \approx J_{23}$, coupling strengths are assumed to be $g_1/2\pi = g_4/2\pi = 100 \text{ MHz}$, $g_2/2\pi = g_3/2\pi = 990 \text{ MHz}$, and $g_a^\circ/2\pi = 980 \text{ MHz}$. In this case, the energy transfer can be accomplished within about 300 ns. Figure 2(b) simulates the energy transfer dynamics in the moderately-clustered geometry with $J_{12} = J_{34} = 1.62 J_{23}$. Here, we adopt $g_1/2\pi = g_4/2\pi = 150 \text{ MHz}$, $g_2/2\pi = g_3/2\pi = 990 \text{ MHz}$, and $g_a^\circ = 930 \text{ MHz}$. Compared to Fig. 2(a), the energy transfer from the first qubit to the last one can be completed within a shorter time, i.e., approximately 150 ns. The moderately-clustered geometry supports a faster energy transfer because the enhanced couplings within the cluster enlarge the intra-cluster energy gap and reduce the inter-cluster energy gap. Both the strong coherent hopping within the cluster and the resonant energy transfer between the two clusters accelerate the overall energy transfer. Moreover, when the ratio $J_{12}/J_{23}$ increases to a larger value, e.g., $J_{12} = J_{34} = 3.11 J_{23}$ for $g_1/2\pi = g_4/2\pi = 230 \text{ MHz}$,
Figure 2: The curves (symbol) show the propagation of the populations of excitation on each of the four qubits by $H_1 \ (H_{\text{eff}})$ for three different geometries: (a) equally-spaced geometry with $J_{12} = J_{34} \approx J_{23}$ and $r \approx R/3$, (b) moderately-clustered geometry with $J_{12} = J_{34} = 1.62 J_{23} \text{ and } r < R/3$, and (c) over-clustered geometry with $J_{12} = J_{34} = 3.11 J_{23}$ and $r < R/3$. The red solid line $(\cdot)$ is for $P_1 \ (P_1^e)$, blue dotted line $(\cdot)$ for $P_2 \ (P_2^e)$, green dashed line $(\cdot)$ for $P_3 \ (P_3^e)$, and black dash-dotted line $(\times)$ for $P_4 \ (P_4^e)$.

$g_2/2\pi = g_3/2\pi = 920 \text{ MHz, and } g_b^e = 800 \text{ MHz, the energy transfer becomes extremely slow and it does not finish even at 300 ns, as shown in Fig. 2(c). That’s because the strong intra-cluster couplings enlarge the intra-cluster energy gap excessively and thus increase the inter-cluster energy gap. In order to find out the optimal parameters for the EET efficiency, we simulate the EET dynamics of our circuit for a broad range of the parameters $g_1, g_2,$ and $g_b^e$ with $g_1 = g_4$ and $g_2 = g_3$ and keeping other parame-
ters unchanged. The parameters are changed from 10 MHz to 990 MHz with 10 MHz step. According to the numerical simulations, we find that the optimal energy transfer occurs at $J_{12} = J_{44} = 1.62J_{23}$, as shown in Fig. 2(b).

$$J_{12} = J_{44} = 1.62J_{23}.$$  

**Figure 3:** The energy-level diagrams corresponding to quantum dynamics in Fig. 2 (a) equally-spaced geometry with $J_{12} = J_{44} \approx J_{23}$ and $r \approx R/3$, (b) moderately-clustered geometry with $J_{12} = J_{44} = 1.62J_{23}$ and $r < R/3$, (c) over-clustered geometry with $J_{12} = J_{44} = 3.11J_{23}$ and $r \ll R/3$. $r$ and $R$ are respectively the intra-pair distance and the distance between the two ends in Fig. 1(b).

In Fig. 2, there are coherent oscillations in the short-time regime, which correspond to the quantum coherence phenomena discovered in 2D spectroscopy experiments 1 64. Because there is strong coupling between the two donor (acceptor) qubits, the energy gap between their eigen-states will be opened up, i.e., $|\varepsilon_1\rangle$ and $|\varepsilon_2\rangle$, as shown in Fig. 1(b). And the energy coherently moves within the donor cluster. However, since the couplings between the donor and acceptor clusters are relatively weak, the energy will incoherently hop between the clusters. According to Förster theory 65, 66, the energy transfer rate from the lower donors’ eigen-state $|\varepsilon_2\rangle$ to the higher acceptors’ eigen-state $|\varepsilon_3\rangle$ is integral of donor’s emission spectrum and acceptor’s absorption spectrum. The former is centered at the lower donors’ eigen-state, and the latter is centered at the higher acceptors’ eigen-state. When the coupling within the donor (acceptor) pair is moderately enlarged, cf. Fig. 3(a) vs (b), the energy gap between $|\varepsilon_2\rangle$ and $|\varepsilon_3\rangle$ has been reduced, and thus results in an enhanced transfer rate. In this way, the clustered geometries utilize energy matching to optimize energy transfer.
between two clusters. However, if the coupling within the donor (acceptor) pair is over-enlarged, cf. Fig. 3(c), this may suppress the transfer rate as the energies of two states mismatch [11]. Moreover, strong coupling between a charge qubit and a TLR can be achieved in superconducting circuit [67–69].

In Fig. 2, we also show the quantum simulations by $H_{\text{eff}}$. The simulations by $H_{\text{eff}}$ coincide with those by $H_1$ in the aspects of oscillation frequency and energy transfer rate. We notice that in Fig. 2(b), the population of each qubit is about 24%, when the circuit is in the steady-state. To explore the reason why the summation of all the populations of the four qubits can not reach 100%, we plot the time evolution of populations in the TLRs $P_a = |1_a\rangle\langle 1_a|$ and $P_b = |1_b\rangle\langle 1_b|$ in Fig. 4. It is shown that after some oscillations in the short-time regime, $P_a$ and $P_b$ increase linearly in time. And both of them reach about 2% at 150 ns. Therefore, since only a small portion of the population, which has not been transferred to the target qubit, has been trapped in the two TLRs, our simulation approach can effectively mimic the EET in the 4-chromophore linear photosynthetic complex, as shown in Fig. 1(b).

![Figure 4: The populations trapped in the TLRs $P_a$ ($P_b$) vs time.](image)

Figures 2 and 3 clearly show the dependence of transfer time $\tau$ on the detuning $\Delta = \varepsilon_2 - \varepsilon_3$ between the lower eigen-state $|\varepsilon_2\rangle$ of $|1\rangle$ and $|2\rangle$, and the higher eigen-state $|\varepsilon_3\rangle$ of $|3\rangle$ and $|4\rangle$. And the previous discoveries [39, 43] suggested the design of bath can also optimize the energy transfer. Therefore, we investigate the energy transfer.
for a broad range of $\Delta$ and the dephasing time $\Gamma_\phi^{-1}$, as shown in Fig. 5. By using a two-exponential-decay model in Ref. [30],

$$P_4(t) = a_L(1 - e^{-t/\tau}) + a_S(1 - e^{-t/\tau'}),$$ \hspace{1cm} (17)

where $a_L \gg a_S$, we numerically fit the population dynamics of qubit 4, and effectively obtain the transfer time $\tau$. We may identify the optimal transfer time $\tau_{\text{opt}} = 35.03$ ns at $\Delta = 0$ and $\Gamma_\phi^{-1} = 60$ ns. First of all, we explore the relation between $\tau$ and $\Delta$, for a fixed $\Gamma_\phi^{-1}$. When $\Delta$ is decreased from a large positive detuning, the transfer time experiences a decrease as $|\varepsilon_2\rangle$ and $|\varepsilon_3\rangle$ approach the energy matching, i.e., $\Delta = 0$. However, if $\Delta$ further decreases, the transfer time increases since the two states become more and more mismatched. On the other hand, we also explore the relation between $\tau$ and $\Gamma_\phi^{-1}$, for a fixed $\Delta$, i.e., in a given geometry. By tuning $\Gamma_\phi^{-1}$, the system gradually reaches optimal transfer time at $\Gamma_\phi \simeq 16.7$ MHz, which is comparable to the effective coupling $J_{23} = 8.87$ MHz between $|2\rangle$ and $|3\rangle$. In the coherent-dynamics limit, i.e., $\Gamma_\phi^{-1} \rightarrow \infty$, because the population will oscillate backwards and forwards, the energy can not be effectively transferred. In the opposite case, when $\Gamma_\phi^{-1} \rightarrow 0$, since the
strong system-bath couplings frequently probe the qubits’ populations, the dynamic localization freezes the population dynamics \cite{60, 61, 62}. In other words, the quantum Zeno effect \cite{70, 71} prohibits the effective energy transfer.

0.5. DISCUSSION AND SUMMARY

In this paper, instead of direct couplings among two neighbouring qubits, we explore two additional TLRs to induce the couplings among any two qubits. In linear geometries, there are next-nearest-neighbor couplings besides the nearest-neighbor couplings. This architecture also enables us to simulate photosynthetic complexes beyond the linear geometries in Ref. \cite{30}, since natural photosynthetic complexes possess more interesting geometries, such as ring-shape LH1(LH2), FMO, and photosystem I(II). Furthermore, by using TLRs, we can simplify the quantum circuit for non-linear geometries beyond the nearest-neighbor couplings.

In our simulations, we select the charge qubits with the dissipation time $T_1 = 200 \ \mu s$ \cite{59}, which is much longer than their pure-dephasing time $T_2 = 0.07 \ \mu s$ \cite{59}, because in dephasing-assisted photosynthetic energy transport the spontaneous fluorescence can be ignored \cite{60, 61, 62}. However, since the energy transfer generally completes within 0.3 $\mu s$, e.g. in Figs. 2(a,b), a much shorter dissipation time, e.g. $T_1 = 3 \ \mu s$, is enough to obtain the same simulations in realistic experiments. This has been confirmed by our numerical simulations which are not shown here. Furthermore, due to couplings to the TLRs, the qubits’ decay times may be shortened due to Purcell effect. In Ref. \cite{72}, the Purcell decay rate is analytically estimated as

$$\Gamma = \frac{\kappa}{2} - \frac{\sqrt{2}}{2} \sqrt{-A + \sqrt{A^2 + (\kappa\Delta)^2}},$$  \quad (18)

where

$$A^2 = \Delta^2 + 4g^2 - \kappa^2/4,$$  \quad (19)
\( \kappa^{-1} \) is the decay time of the TLR, \( \Delta \) is the detuning between the qubit and TLR, \( g \) is the coupling strength. Because the qubits dispersively couple with the TLRs, i.e., \( g/\Delta \ll 1 \), \( \Gamma \simeq \kappa g^2/\Delta^2 \) to the lowest order of \( g/\Delta \). For the present parameters, the Purcell decay time is about 2 ms, which is much longer than 4 \( \mu \)s. Therefore, the Purcell effect will not significantly modify the qubits’ decay times.

Parameters used here have been realized in experiments. The quality factor of a superconducting TLR can reach \( 10^5 \) \[73\]. The frequency of the fundamental mode of the TLR can be designed from 1 GHz to 10 GHz \[46, 74, 75\]. The frequency of a superconducting charge qubit can be effectively tuned from 5 GHz to 15 GHz, by varying the flux that applied though the loop of the qubit \[46\]. In addition, the ultra-strong coupling between a charge qubit and a TLR is achieved when \( \min\{g_j\} \gg \sqrt{\kappa_\alpha \Gamma_j} \) \( (j = 1, 2, 3, 4, \alpha = a, b) \) \[76\]. Although the coupling strength reaches the ultra-strong regime, i.e., \( g_j/\omega_a \simeq 1/3 \), the Jaynes-Cumming model is still valid as the large-detuning condition still holds \[77\]. As a result, the population dynamics will not be essentially modified.

In Ref. \[37\], the NMR experimental simulation was compared to the numerically-exact simulation using the hierarchical equation of motion (HEOM) \[26, 78\]. Here, in our configuration, there are 2 TLRs in addition to 4 qubits. The presence of extra modes in the TLRs results in the increasing complexity of the quantum master equation, which can be solved by using QuTiP \[79, 80\], as compared to the HEOM for 4 chlorophylls.

In natural photosynthetic complexes, the energy is transferred from the outer antenna to the reaction center across tens of nanometers \[1, 30, 37\]. There is a large energy gap between the lowest eigen-state of the outer antenna and the reaction center, which can prevent the back transfer of energy. As a result, the transfer rate from the lowest-energy state to the reaction center is much smaller than the transfer rate within the outer antenna \[37, 81\]. In this work, it is thus reasonable to simulate the energy transfer without the reaction center. However, a TLR can be utilized to effectively simulate the reaction center. Coupled to some specific qubit, the TLR can act
as a reaction center through Purcell-like coupling [40, 41]. By tuning the TLR’s decay time and the qubit-TLR detuning, we can effectively modified the qubit’s decay time through Purcell-like coupling, according to Eq. (18).

In summary, we have proposed a simulation scheme for demonstrating geometric effects on the photosynthetic EET in four superconducting charge qubits plus two separated high-$Q$ TLRs. The loss of population during the EET is trapped in the TLRs. In the future work, it might be interesting to demonstrate the effect of fluorescence on the EET by varying the couplings between the qubits and the TLRs.

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**Appendix: Complete Expression of $H_2$**

Here, we give the detailed expression of $H_2$ used in Sec. 0.3. We can omit the high-order terms of $g_{j}/\delta_{j}$, and simplify

$$H_2 = U^\dagger H_1 U$$

(A1)
\[ H_2 = \omega_0 a^\dagger a + \omega_b b^\dagger b + g_0^b (a^\dagger + a) (b^\dagger + b) \]
\[ + \sum_{j_1=1}^2 \frac{g_0^b g_{j_1}^2}{2 \delta_{j_1}} (a^\dagger + a) (b^\dagger + b) \sigma_{j_1}^z \]
\[ + \sum_{j_2=3}^4 \frac{g_0^b g_{j_2}^2}{2 \delta_{j_2}} (a^\dagger + a) (b^\dagger + b) \sigma_{j_2}^z \]
\[ + \sum_{j_1=1}^2 \left[ \frac{\omega_{j_1}}{2} \sigma_{j_1}^z + \frac{g_{j_1}^2}{\delta_{j_1}} (a a^\dagger \sigma_{j_1}^+ \sigma_{j_1}^- - a^\dagger a \sigma_{j_1}^+ \sigma_{j_1}^-) \right] + \frac{g_1 g_2}{2 \delta_1 \delta_2} (\delta_1 + \delta_2) (\sigma_{j_1}^- \sigma_{j_2}^+ + \sigma_{j_1}^+ \sigma_{j_2}^-) \]
\[ + \sum_{j_2=3}^4 \left[ \frac{\omega_{j_2}}{2} \sigma_{j_2}^z + \frac{g_{j_2}^2}{\delta_{j_2}} (b b^\dagger \sigma_{j_2}^+ \sigma_{j_2}^- - b^\dagger b \sigma_{j_2}^- \sigma_{j_2}^+) \right] + \frac{g_3 g_4}{2 \delta_3 \delta_4} (\delta_3 + \delta_4) (\sigma_{j_2}^- \sigma_{j_3}^+ + \sigma_{j_2}^+ \sigma_{j_3}^-) \]
\[ + \frac{g_0^b g_1 g_3}{\delta_1 \delta_3} (\sigma_{j_1}^- \sigma_{j_3}^+ + \sigma_{j_1}^+ \sigma_{j_3}^-) + \frac{g_0^b g_1 g_4}{\delta_1 \delta_4} (\sigma_{j_1}^- \sigma_{j_4}^+ + \sigma_{j_1}^+ \sigma_{j_4}^-) \]
\[ + \frac{g_0^b g_2 g_3}{\delta_2 \delta_3} (\sigma_{j_2}^- \sigma_{j_3}^+ + \sigma_{j_2}^+ \sigma_{j_3}^-) + \frac{g_0^b g_2 g_4}{\delta_2 \delta_4} (\sigma_{j_2}^- \sigma_{j_4}^+ + \sigma_{j_2}^+ \sigma_{j_4}^-). \]