Optical Non-Reciprocity in Coupled Resonators by Detailed Balance

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Inspired by the photosynthetic energy transfer process, a method to realize non-reciprocal optical transmission in an array of coupled resonators is theoretically proposed. The optical non-reciprocity of the coupled resonators arises from the frequency gradient between adjacent cavities and the interaction with the environment, which is similar to photosynthetic energy transfer. An increase in the frequency gradient or the number of the cavities can lead to better non-reciprocity. However, although a higher environment temperature will increase the total photon number in the coupled cavities, non-reciprocity will be weakened. All these findings can be well described by the detailed balance. The similarity between the noise-induced optical non-reciprocity and exciton energy transfer in natural photosynthesis is revealed by the discovery.

1. Introduction

Structures in living beings may have significant effects on physiological functions and thus enlighten inventions of bionic devices, for example, honeycomb and light-harvesting antenna in photosynthesis. Natural photosynthesis, generally includes three steps.^[1] First of all, the solar photon is absorbed by the outer antenna. It follows that the absorbed energy is delivered to the reaction center. Finally, the chemical reaction in which the energy is stored in the chemical products is activated. In the first two steps, the outer antenna is made up of pigments, for example, chlorophylls and carotenes and protein skeleton. The chlorophyll molecule consists of a chromophore, which can be modeled as a two-level atom and a long tail.^[2] When the chlorophyll molecule absorbs the solar photon, the chromophore is excited from the ground state to the excited state. During the energy transfer process, the chromophore in the excited state will excite a nearby chromophore in the ground state and return to the ground state. In this way, the excitation state and thus the excitation energy are delivered from the outer antenna to the reaction center. In Figure 1a, the spatial arrangement of seven chromophores in the Fenna-Matthews-Olson (FMO) complex and energy transfer pathways are schematically illustrated.^[3-5] In order to make

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ple, funnel, have been widely used in daily life when pouring oil into a bottle in order to avoid waste, cf. Figure 1b. However, it is known that the energy difference between two states alone will not induce the irreversible and unidirectional transfer of population from one state to the other, as the quantum dynamics are coherent.^[6] Many studies have shown that noise, that is, the interaction with the environment, plays a prominent role in this process of energy transfer.^[7–11] It is the unavoidable environmental noise

the energy transfer unidirectional toward

the reaction center, an energy gradient

in the site energies of the chromophores is employed. Similar devices, for exam-

in combination with the energy gradient in the site energies that lead to an optimal energy transfer efficiency.

The coupled chromophores in photosynthesis form a network of chromophores and it is analogous to an array of coupled resonators. A resonator localizes the eigen-modes of the electromagnetic field through a "mirror-cavity-mirror" structure.^[12] In a closed resonator, the wavefunction of each mode $\Psi(\mathbf{r}, t) = \psi(t)\xi(\mathbf{r})$ includes the time-domain $\psi(t)$, with the resonant frequency ω , and the spatial structure of the field $\xi(\mathbf{r})$. Resonators can be coupled by the fields penetrating through the barriers and thus form the coupled resonators^[12] (cf. Figure 1c). Interestingly, by adding one atom in one of the coupled resonators, perfect refraction can be switched on/off by tuning the detuning between the atomic transition frequency and the resonant frequency of the resonator.^[13]

The coupled resonators have been shown to demonstrate numerous interesting phenomena, for example, quantum routers^[14] and non-reciprocal optical transmission.^[15,16] According to the Lorentz reciprocity theorem, the transmittance of the light will not be modified when the emission port and the reception port are exchanged.^[17] However, by magnetic-field-induced breaking of time-reversal symmetry, spatio-temporal modulation of system permittivity, and nonlinearity, optical non-reciprocity can be realized.^[18,19] Non-reciprocal devices, for example, optical isolators, optical circulators, and directional amplifiers, play a crucial role in communication and quantum information processing. Interestingly, a levitated rotating nano-diamond with nitrogen-vacancy centers can demonstrate non-reciprocal optical transmission because of electromagnetic-induced transparency and Doppler effect.^[20]

In this paper, inspired by the rapid progress in photosynthetic energy transfer and optical non-reciprocity, we theoretically

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Figure 1. Schematic illustration of the similarity between excitation energy transfer in photosynthesis and artificial non-reciprocal transmission. a) Spatial arrangement of chlorophylls and excitation energy transfer pathways in the monomeric subunit of the FMO complex. b) Illustration of an energy funnel. c) Structure of coupled cavities and d) the energy levels of the cavities.

propose a method to realize non-reciprocal optical transmission in an array of coupled resonators. Analogous to the photosynthetic light harvesting, the optical non-reciprocity in the coupled resonators is achieved by a gradient in the resonant frequencies of the resonators (Figure 1d) and the system-bath interaction. Impressively, the side-coupled resonators with the enclosed synthetic magnetic flux can induce the non-reciprocal transport in the coupled-resonator array by introducing the environmentinduced gain or loss,^[21–24] which is similar to the physical mechanism of non-reciprocity in this work. In contrast to the above three criteria,^[18,19] the optical non-reciprocity in this work is due to the breaking of time-reversal symmetry induced by the systembath entanglement.

This paper is organized as follows: In the next section, the model for optical non-reciprocal transmission and the quantum master equation approach is introduced. In Section 3, the effects of the frequency gradient, the temperature, and the number of coupled resonators on the non-reciprocal transmission are analyzed by the numerical simulations. Finally, the main discoveries have been summarized in Section 4.

2. Model

As shown in Figure 1c, the Hamiltonian of the coupled resonators reads

$$H_{\rm S} = \sum_{j} \omega_{j} a_{j}^{\dagger} a_{j} - \xi \sum_{j} \left(a_{j}^{\dagger} a_{j+1} + a_{j+1}^{\dagger} a_{j} \right) \tag{1}$$

where $a_j^{\dagger}(a_j)$ is the creation (annihilation) operator of the *j*th cavity with frequency ω_j , ξ is the coupling constant between two adjacent resonators, and we have assumed $\hbar = 1$. Here, we assume an energy gradient in the frequencies of resonators, as illustrated in Figure 1d.

When there are interactions with the environments, the open quantum dynamics is determined by the quantum master equation^[25]

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -i[H_{\mathrm{S}},\rho] + \mathcal{L}[\rho] \tag{2}$$

$$\mathcal{L}[\rho] = \frac{\gamma}{2} \sum_{j} \left[(N_{j}^{\text{th}} + 1)(2a_{j}\rho a_{j}^{\dagger} - a_{j}^{\dagger}a_{j}\rho - \rho a_{j}^{\dagger}a_{j}) + N_{j}^{\text{th}}(2a_{j}^{\dagger}\rho a_{j} - a_{j}a_{j}^{\dagger}\rho - \rho a_{j}a_{j}^{\dagger}) \right]$$
(3)

where ρ is the reduced density matrix of the coupled resonators, $N_i^{\text{th}} = (e^{\omega_j/k_{\text{B}}T} - 1)^{-1}$ is the average photon number in the bath with $k_{\rm B}$ and T being the Boltzmann constant and the temperature, respectively, γ is the relaxation rate at zero temperature. We use QuTiP^[26,27] to solve the quantum master equation numerically. In deriving the quantum master equation, several approximations have been applied, for example, Born approximation, Markovian approximation, and secular approximation.^[28,29] Generally speaking, the generators in the Lindblad operator (3) are the raising and lowering operators between the eigen states of the system Hamiltonian $H_{\rm S}$.^[28] Here, we approximate them as the creation and annihilation operators of the bare cavities in order to better illustrate the underlying physical mechanism without being trapped by sophisticated mathematics. We also remark that the exact open quantum dynamics can be reproduced by the hierarchical equation of motion and a recently-proposed quantum simulation approach in the high-temperature limit.[30-33]

3. Results

First of all, we adopt the system containing two cavities as a demonstration and the eigenfrequency of the first cavity is lower than the second. **Figure 2** shows the results of the case with



Figure 2. The non-reciprocal transmission in two-coupled resonators with single photon incident from the left to the right (solid lines) and from the right to the left (dashed lines). The photon number in the first (second) cavity is labeled by the blue (red) lines. Here, $n_j = \langle a_j^{\dagger} a_j \rangle$ is the average photon number in the *j*th cavity. The parameters used in the calculation are $\omega_2 = 2\omega_1$, $\gamma = \xi = 0.3\omega_1$, and $k_BT = \omega_1$.

 $\omega_2 = 2\omega_1$. If we place one photon in the first cavity at the initial moment, the photon number in the first cavity, n_1 , will decline as depicted by the blue solid line and the photon number in the second cavity, n_2 , will rise as labeled by the red solid line, with $n_i = \langle a_i^{\dagger} a_i \rangle$ being the average photon number in the *j*th cavity. In this case, these two lines will not intersect before they reach their respective steady states. Moreover, at the short-time regime, they oscillate with a small amplitude due to the coherent term in the master equation (Equation (2)) and the small inter-resonator coupling in $H_{\rm S}$ with respect to the large energy gap between the frequencies of two cavities. If we place one photon in the second cavity at the initial moment, the photon number in the first cavity, n_1 , will rise as denoted by the blue dashed line, and the photon number in the second cavity, n_2 , will decline as shown by the red dashed line. In this case, these two lines will intersect and reach the same steady states as in the previous case. Here, the photon number is always distributed more in the cavity with a lower frequency no matter what the initial state is. The ratio between the photon numbers in the first and second cavities reads $\exp(-\Delta\omega/k_{\rm B}T)$ with $\Delta\omega = \omega_2 - \omega_1$ being the detuning between the two cavities, as determined by the detailed balance.^[28,34]

In open quantum systems, the detailed balance subtly governs the instantaneous open quantum dynamics as well as the thermal equilibrium at the steady state. In order to explicitly reveal its effect, we resort to the quantum master Equation (2). Notice that the Hamiltonian $H_{\rm S}$ conserves the total number of the photons in all cavities. For the sake of simplicity, we restrict our consideration in the subspace with no more than one photon in either cavity, that is, $a_j^{\dagger}|0\rangle$ (j = 1, 2) and $|0\rangle$ with $|0\rangle$ being the vacuum state for all cavities, and thus obtain

$$\dot{\rho}_{00} = \gamma \sum_{j} [(N_{j}^{\text{th}} + 1)\rho_{jj} - N_{j}^{\text{th}}\rho_{00}]$$
(4)

$$\dot{\rho}_{jj} = -\gamma (N_j^{\text{th}} + 1)\rho_{jj} + \gamma N_j^{\text{th}}\rho_{00}$$
(5)

where $\rho_{jj} = \langle 0 | a_j \rho a_j^{\dagger} | 0 \rangle$ is the probability of one photon in the *j*th cavity, $\rho_{00} = \langle 0 | \rho | 0 \rangle$ is the probability of all cavities in the vacuum state. In Equations (4) and (5) the coherent terms are neglected because of the large detuning condition $\Delta \omega \gg \xi$. At the steady state, since $\dot{\rho}_{jj} = \dot{\rho}_{00} = 0$, we have $\rho_{jj} / \rho_{00} = N_j^{\text{th}} / (N_j^{\text{th}} + 1) = \exp(-\omega_j / k_B T)$. In the case of two cavities, the ratio between the probabilities of one photon in the two cavities reads

$$\rho_{22}/\rho_{11} = \exp(-\Delta\omega/k_{\rm B}T) \tag{6}$$

In **Figure 3**, we compare n_2/n_1 calculated by Equation (2) with ρ_{22}/ρ_{11} by Equation (6). It can be seen that the approximated analytic result reproduces the exact solution to the quantum master equation fairly well in the whole parameter regime, especially $\Delta \omega = 0$ and $\Delta \omega \gg k_{\rm B}T$. The small difference between them in the intermediate regime may be attributed to the truncation of three states in the above over-simplified model.

Transmission efficiencies denoted by the photon number are different in the cases of left-to-right and right-to-left propagation. Note that in the left-to-right propagation case, the photon number in the cavity at the exit end is n_2 , that is, the red solid line, while





Figure 3. The ratio between the average photon numbers in the second and the first cavities n_2/n_1 as a function of frequency gradient $\Delta\omega$. The dots are obtained by the quantum master equation (Equation (2)), while the solid line is obtained by Equation (6). Other parameters are the same as Figure 2.

in the right-to-left propagation case, the photon number in the cavity at the exit end is n_1 , that is, the blue dashed line, which is higher than the former. In other words, the transmission of photons in the coupled-cavities array is non-reciprocal.

We further investigate the effect of the noise on the nonreciprocity. In Equation (3), when $\gamma = 0$, that is, without the interaction with the environment, the time evolution of the density matrix is equivalent to that of an isolated system. It can be seen in **Figure 4** that without dissipation the photon number in the cavity at the incident end coherently oscillates around the initial value 1, while the photon number in the cavity at the exit end oscillates around 0. The sum of the photon numbers in both cavities is always unity, which is required by the conservation of probability. The small amplitude of oscillations is due to the relatively-large detuning, as denoted by $\xi < \Delta \omega$ in H_s . The red solid line and



Figure 4. The transmission in 2-coupled resonators when $\gamma = 0$. Other parameters are the same as Figure 2. When there is no interaction with the environment, the non-reciprocal transmission disappears.





Figure 5. The photon number *n* in the terminal cavity at steady state and the transmission contrast η as a function of frequency gradient $\Delta \omega / \omega_1$ in the case of 2-coupled resonators. Other parameters are the same as Figure 2.

the blue dashed line are completely coincident with each other, which imply that the system is reciprocal.

To quantitatively evaluate the non-reciprocity of the system, we define the transmission contrast ${\rm as}^{[19]}$

$$\eta = \frac{|T_+ - T_-|}{T_+ + T_-} = \frac{|n_+ - n_-|}{n_+ + n_-} \tag{7}$$

where T_+ (T_-) is forward (backward) transmission coefficient, n_+ (n_-) is the steady-state photon number in the cavity at the exit end in the case of forward (backward) transmission. Here, $\eta = 1$ implies the maximum non-reciprocity because the optical transmission is allowed unidirectionally, while $\eta = 0$ suggests no non-reciprocity, as there is no difference between the optical transmissions in both directions.

We investigate the effect of the frequency gradient $\Delta \omega$ on the transmission contrast η in **Figure 5**. When there is no difference between the frequencies of the two cavities, the transmissions in both directions are the same and thus reciprocal. If we gradually increase $\Delta \omega$, the change of the transmission from the right to the left is negligibly small, while the transmission from the left to the right decreases dramatically. In other words, the transmission becomes more and more non-reciprocal as $\Delta \omega$ is enlarged. Alternatively, we calculate the transmission contrast η by the detailed balance,^[28,34] that is,

$$\eta = \frac{\left|\frac{n_{+}}{n_{-}} - 1\right|}{\frac{n_{+}}{n_{-}} + 1} = \frac{\left|e^{\Delta\omega/k_{\rm B}T} - 1\right|}{e^{\Delta\omega/k_{\rm B}T} + 1}$$
(8)

Similar to Figure 3, the results by both methods coincide with each other in Figure 5. Therefore, we may safely arrive at the conclusion that the detailed balance accounts for the non-reciprocal transmission observed in the coupled resonators. The black solid line in Figure 5 is symmetric with respect to 0, while the black dotted line is not strictly symmetric with respect to 0. The asymmetry of the black dotted line reflects the effect of the states with more than one photon in the resonators and the coherence between the states. Furthermore, we investigate the dependence of





Figure 6. The dependence of the transmission contrast η on the frequency gradient $\Delta \omega / \omega_1$ and the relaxation rate γ . Other parameters are the same as Figure 2.

 η on both $\Delta \omega$ and γ in **Figure 6**. The transmission contrast η is generally not influenced by γ , which is consistent with the prediction by the detailed balance. The non-reciprocal transmission can be effectively tuned by the frequency gradient.

In the above discussions, we assume that the environment is in thermal equilibrium with a fixed temperature $T = \omega_1/k_{\rm B}$. The effect of temperature on transmission is next observed in **Figure 7**. It can be seen that when the temperature is absolute zero, there are no photons in the cavities at the final state because the photons initially in the cavity leak to the environment. Despite the small *n*, the transmission contrast is close to unity and non-reciprocity occurs. We remark that this nonreciprocal transmission is unfavorable, as the transmission has been greatly depressed as compared to the input. As the temperature increases, in both cases, the photon number in the terminal cavity increases. When the temperature is high to a certain extent, the sum of the photon number in the coupled cavities will be greater than 1 due to the heating effect of the environment.



Figure 7. The photon number, *n*, in the terminal cavity at steady state and the transmission contrast, η , as a function of the temperature *T* in the case of 2-coupled resonators. Other parameters are the same as Figure 2.





Figure 8. The photon number, *n*, in the terminal cavity at steady state as a function of the number of cavities, *N*, when the frequency gradient equals to ω_1 . The dots are obtained by solving the quantum master equation, while the solid lines are obtained by the detailed balance. Other parameters are the same as Figure 2.

However, as the temperature increases, the transmission contrast η decreases, and the non-reciprocity eventually disappear for $k_{\rm B}T \gg \Delta \omega$. To conclude, there exists an optimal temperature for the non-reciprocity at which both *n* and η are of the order of unity, that is, $k_{\rm B}T \approx \omega_1$.

We further study the case with more cavities. We keep the frequency gradient of adjacent cavities $\omega_{j+1} - \omega_j$ and the eigenfrequency of the leftmost cavity ω_1 constant and only vary the number of coupled cavities *N*. In **Figure 8**, it can be seen that as the number of cavities *N* increases, although it does not influence the photon number in the leftmost cavity, the photon number in the cavity at the right end decreases significantly. This is because more cavities will enlarge the gap between the eigenfrequencies of the cavities at both ends. As a result, the non-reciprocal transmission is enhanced for a larger *N*.

4. Conclusion

In this paper, we discuss the non-reciprocal optical transmission in an array of coupled resonators. We show that the optical non-reciprocity is akin to photosynthetic energy transfer. In both cases, the non-reciprocal transmissions result from the frequency (energy) gradient and the interaction with the environment. The open quantum dynamics and thus the non-reciprocity are analyzed by numerically solving the quantum master equation. It is demonstrated that an increase in the frequency gradient leads to better non-reciprocity. We also observe that there exists an optimal temperature at which both the transmission contrast and the absolute transmission are sufficiently high to ensure a favorable non-reciprocity. When the frequency gradient of adjacent cavities and the eigenfrequency of the first cavity are kept constant, the more cavities coupled, the better non-reciprocity of the system is.

In the quantum master equation (Equation (2)), we assume a time-independent relaxation rate γ . However, if the Markovian approximation is canceled, we would expect non-Markovian effects might emerge.^[35,36] For example, the entanglement and

the quantum mutual information may not decay monotonically. Since the array of coupled resonators involves a network of multiresonators, it may lose information to the environment in a cavity while it regains information from the environment in another cavity.^[37]

To summarize, our findings bridge the gap between nonreciprocal optical transmission and energy transfer in photosynthesis. And it may enrich the understanding of the underlying physical mechanisms for both issues.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

coupled resonators, detailed balance, non-reciprocity, photosynthesis, system-bath entanglement

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- [1] G. R. Fleming, R. van Grondelle, Phys. Today 1994, 47, 48.
- [2] R. E. Blankenship, Molecular Mechanisms of Photosynthesis, Wiley Blackwell, Hoboken, NJ 2014.
- [3] J. Moix, J. Wu, P. Huo, D. Coker, J. Cao, J. Phys. Chem. Lett. 2011, 2, 3045.
- [4] J. Wu, F. Liu, J. Ma, R. J. Silbey, J. Cao, J. Chem. Phys. 2012, 137, 174111.
- [5] Q. Ai, T.-C. Yen, B.-Y. Jin, Y.-C. Cheng, J. Phys. Chem. Lett. 2013, 4, 2577.
- [6] M. O. Scully, M. S. Zubairy, *Quantum Optics*, Cambridge University Press, Cambridge 1997.
- [7] M. Mohseni, P. Rebentrost, S. Lloyd, A. Aspuru-Guzik, J. Chem. Phys. 2008, 129, 174106.
- [8] F. Caruso, A. W. Chin, A. Datta, S. F. Huelga, M. B. Plenio, J. Chem. Phys. 2009, 131, 105106.
- [9] M. S. am Busch, F. Müh, M. El-Amine Madjet, T. Renger, J. Phys. Chem. Lett. 2011, 2, 93.
- [10] M. B. Plenio, S. F. Huelga, New J. Phys. 2008, 10, 113019.
- [11] A. W. Chin, A. Datta, F. Caruso, S. F. Huelga, M. B. Plenio, *New J. Phys.* 2010, 12, 065002.

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- [12] K. Y. Bliokh, Y. P. Bliokh, V. Freilikher, S. Savel'ev, F. Nori, *Rev. Mod. Phys.* 2008, 80, 1201.
- [13] L. Zhou, Z. R. Gong, Y.-X. Liu, C. P. Sun, F. Nori, Phys. Rev. Lett. 2008, 101, 100501.
- [14] Lan Zhou, Li-Ping Yang, Yong Li, C. P. Sun, Phys. Rev. Lett. 2013, 111, 103604.
- [15] X. Y. Huang, C. C. Lu, C. Liang, H. G. Tao, Y.-C. Liu, *Light-Sci. Appl.* 2021, 10, 30.
- [16] H.-Y. Zhu, X.-Y. Hu, J.-J. Lin, J.-Y. Wu, S. Li, Y.-X. Wang, F.-G. Deng, N.-N. Zhang, Ann. Phys. (Berlin) 2021, 533, 2100297.
- [17] H. A. Lorentz, Amsterdammer Akad. Wet. 1896, 4, 176.
- [18] Dirk. Jalas, A. Petrov, M. Eich, W. Freude, S. H. Fan, Z. F. Yu, R. Baets, M. Popović, A. Melloni, J. D. Joannopoulos, M. Vanwolleghem, C. R. Doerr, H. Renner, *Nat. Photonics* **2013**, *7*, 579.
- [19] C. Caloz, A. Alù, S. Tretyakov, D. Sounas, K. Achouri, Z. Deck-Léger, Phys. Rev. Appl. 2018, 10, 047001.
- [20] H.-B. Huang, J.-J. Lin, Y.-X. Yao, K.-Y. Xia, Z.-Q. Yin, Q. Ai, Ann. Phys. (Berlin) 2022, 534, 2200157.
- [21] L. Jin, P. Wang, Z. Song, *Sci. Rep.* **2016**, *6*, 32919.
- [22] L. Du, Y. Zhang, J.-H. Wu, Sci. Rep. 2020, 10, 1113.
- [23] L. Jin, Z. Song, Phys. Rev. Lett. 2018, 121, 073901.
- [24] L. Jin, Phys. Rev. A 2018, 97, 033840.
- [25] H. Carmichael, An Open Systems Approach to Quantum Optics, Springer-Verlag, Germany 2009.

[26] J. R. Johansson, P. D. Nation, F. Nori, Comput. Phys. Commun. 2012, 183, 1760.

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- [27] J. R. Johansson, P. D. Nation, F. Nori, Comput. Phys. Commun. 2013, 184, 1234.
- [28] H.-P. Breuer, F. Petruccione, The Theory of Open Quantum Systems, Oxford University Press, Oxford 2007.
- [29] M.-J. Tao, N.-N. Zhang, P.-Y. Wen, F.-G. Deng, Q. Ai, G.-L. Long, Sci. Bull. 2020, 65, 318.
- [30] I. Buluta, F. Nori, Science 2009, 326, 108.
- [31] I. M. Georgescu, S. Ashhab, F. Nori, Rev. Mod. Phys. 2014, 86, 153.
- [32] B.-X. Wang, M.-J. Tao, Q. Ai, T. Xin, N. Lambert, D. Ruan, Y.-C. Cheng, F. Nori, F.-G. Deng, G.-L. Long, *npj Quantum Inf.* 2018, 4, 52.
- [33] N.-N. Zhang, M.-J. Tao, W.-T. He, X.-Y. Chen, X.-Y. Kong, F.-G. Deng, N. Lambert, Q. Ai, Front. Phys. 2021, 16, 51501.
- [34] S. Mukamel, Principles of Nonlinear Optics and Spectroscopy, Oxford University Press, New York 1995.
- [35] A. Ishizaki, T. R. Calhoun, G. S. Schlau-Cohen, G. R. Fleming, *Phys. Chem. Chem. Phys.* 2010, 12, 7319.
- [36] F. Caruso, A. W. Chin, A. Datta, S. F. Huelga, M. B. Plenio, *Phys. Rev.* A 2010, 81, 062346.
- [37] X.-Y. Chen, N.-N. Zhang, W.-T. He, X.-Y. Kong, F.-G. Deng, Q. Ai, G.-L. Long, *npj Quantum Inf.* **2022**, *8*, 22.