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Autonomy in excitation transfer via optical near-field interactions and its implications for information networking

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ABSTRACT

We demonstrate optical excitation transfer in a mixture composed of quantum dots of two different sizes (larger and smaller) networked via optical near-field interactions. For the optical near-field interaction network based on a density matrix formalism, we introduce an optimal mixture that agrees with experimental results. Based on these findings, we theoretically examine the topology-dependent efficiency of optical excitation transfer, which clearly exhibits autonomous, energy-efficient networking behavior occurring at the nanometer scale. We discuss what we can learn from this optical excitation transfer and its implications for information and communications applications.

1. Introduction

Optics is expected to play a crucial role in enhancing system performance to handle the continuously growing amount of digital data and new requirements demanded by industry and society [24]. However, there are some fundamental difficulties impeding the adoption of optical technologies in information processing and communication systems [17,16,8,1,2]. One problem is the poor integrability of optical devices in systems due to the diffraction limit of light. This is because the optical wavelength used in a given system is typically around 1 μm, which is about 100 times larger than the gate length of present silicon VLSI hardware.

Nanophotonics, on the other hand, is based on local interactions between nanometer-scale materials via optical near-fields, which are not restricted by conventional diffraction of light, allowing ultrahigh-density integration [17,16,8]. Optical excitation transfer between quantum dots via optical near-field interactions will be one of the most important mechanisms for realizing novel devices and systems [17,16,25,9,5]. Moreover, qualitatively novel features that are unavailable in conventional optics and electronics will be made possible by such optical excitation transfer [14,12].

In this paper, we demonstrate optical excitation transfer in a mixture composed of different-sized (larger and smaller) quantum dots networked via optical near-fields in their vicinities. We introduce a theoretical model of a mixture of two different-sized quantum dots that is optimal in terms of the optical near-field interaction network. This model is based on a density matrix formalism in which the mixture agrees well with experimental results using CdSe/ZnS quantum dot mixtures with diameters of 2.0 and 2.8 nm. Based on these findings, we theoretically examine the topology-dependent efficiency of

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optical excitation transfer, which clearly exhibits autonomous, energy-efficient networking behavior at nanometer scales. Such autonomous behavior can also give us valuable insights when we look at network architectures in general or at communication networks in particular. At the stage of designing future communication networks, the operational conditions are usually unknown, so these networks must be planned in such a way that they can adapt to changing environments, for example, caused by link failures, changes in traffic patterns, etc. We hope that our discussion on the implications of this optical excitation transfer will also lead to some new aspects in the design of autonomous, robust, and energy-efficient communications architectures.

This paper is organized as follows. In Section 2, we describe the physical fundamentals of optical excitation transfer between quantum dots, followed by the introduction of a network of optical near-field interactions, whereby we analyze the mixture-dependent optical excitation transfer theoretically and experimentally. In Section 3, we investigate the topology-dependency and autonomy of the excitation transfer. Section 4 reflects on what can be learned from these principles and phenomena physically existing at nanometer scales and discusses their implications for information and communications applications.

2. Network of optical near-field interactions

2.1. Theoretical background

We briefly review the fundamental principles of optical excitation transfer involving optical near-field interactions [17,16]. The interaction Hamiltonian between an electron and an electric field is given by

$$\hat{H}_{\text{int}} = -\int \hat{\psi}^{\dagger}(\vec{r})\vec{\mu}\hat{\psi}(\vec{r}) \bullet \hat{\vec{D}}(\vec{r})d\vec{r}, \qquad (1)$$

where $\vec{\mu}$ is the dipole moment, $\hat{\psi}^{\dagger}(\vec{r})$ and $\hat{\psi}(\vec{r})$ are respectively the creation and annihilation operators of the electron at $\vec{r}$, and $\hat{\vec{D}}(\vec{r})$ is the operator of electric flux density. In usual light–matter interactions, the operator $\hat{\vec{D}}(\vec{r})$ is a constant since the electric field of propagating light is considered to be constant at nanometer scales. Therefore, one can derive optical selection rules by calculating a transfer matrix of an electric dipole. In the case of cubic quantum dots, for instance, transitions to states described by quantum numbers containing an even number are prohibited [17]. Contrast this with optical near-field interactions, where due to the steep electric field of optical near-fields in the vicinity of a nanometer-scale structure, such as a quantum dot, an optical transition is allowed that would otherwise violate conventional optical selection rules [17,16].

Optical excitations in nanostructures can be transferred to neighboring nanostructures via optical near-field interactions [17,16,25,9,5,14,12,13,15]. Assume that two cubic quantum dots with side lengths a and $\sqrt{2}a$, which are called QD_S and QD_L, respectively, are located near to each other, as shown in Fig. 1(a). The energy eigenvalues for the quantized exciton energy level specified by quantum numbers (n_x, n_y, n_z) in the quantum dot with side length a (QD_S) are given by

$$E_{(n_x,n_y,n_z)} = E_B + \frac{\hbar^2\pi^2}{2Ma^2}(n_x^2 + n_y^2 + n_z^2), \qquad (2)$$

where E_B is the energy of the bulk exciton and M is the effective mass of the exciton. According to Eq. (2), there exists a resonance between the level of quantum number (1, 1, 1) in QD_S and that of quantum number (2, 1, 1) in the quantum dot with side length $\sqrt{2}a$ (QD_L). There is an optical near-field interaction, which is denoted by U_{SL}, due to the steep electric field in the vicinity of quantum dots. It is known that the inter-dot optical near-field interaction is given by a Yukawa-type potential [17]. Therefore, excitations in QD_S can move to the (2, 1, 1)-level in QD_L. Note that such a transfer is prohibited for propagating light since the (2, 1, 1)-level in QD_L contains an even number. In QD_L, the excitation sees a sublevel energy relaxation, denoted by Γ, which is faster than the near-field interaction, and so the excitation goes to the (1, 1, 1)-level in QD_L. In Section 3, we apply these theoretical arguments to systems composed of multiple quantum dots and investigate their impact on fundamental features of optical excitation transfer occurring at nanometer scales.

2.2. Network of optical near-field interactions

Consider the quantum dot system in Fig. 1(b), where multiple smaller dots (denoted by S_i) can be coupled with one larger dot, denoted by L. We assume inter-dot interactions between adjacent smaller quantum dots (Fig. 1(c)); that is, (i) S_i interacts with S_{i+1} ($i = 1, \dots, N-1$) and (ii) S_N interacts with S_1, where N is the number of smaller quantum dots. For instance the system shown in Fig. 1(d) consists of two smaller quantum dots and one larger quantum dot, denoted by **S2-L1**. Similarly, **S3-L1**, **S4-L1**, **S5-L1** systems are composed of three, four, and five smaller quantum dots in addition to one large quantum dot, which are respectively shown in Fig. 1(e)–(g).

Now, what is of interest is to calculate the flow of excitations from the smaller dots to the larger one. The theoretical and experimental details can be found in Ref. [13]; here we introduce the information necessary for discussing the topology-dependency and autonomy in optical excitation transfer in Section 3.

We deal with the problem theoretically based on a density matrix formalism. In the case of the **S2-L1** system, which is composed of two smaller quantum dots and one larger quantum dot, the inter-dot interactions between the smaller dots and the larger one are denoted by U_{S_iL}, and the interaction between the smaller dots is denoted by $U_{S_1S_2}$, as schematically shown in Fig. 2(a). The radiations from S_1, S_2, and L are respectively represented by the relaxation constants γ_{S_1}, γ_{S_2}, and γ_L. We suppose that the system initially has two excitations in S_1 and S_2. With such an initial state, we can prepare a total of eleven bases where zero, one, or two excitation(s) occupy the energy levels; the state transitions are schematically shown in Fig. 2(b). In the numerical calculation, we assume $U_{S_iL}^{-1} = 200$ ps, $U_{S_1S_2}^{-1} = 100$ ps, $\gamma_L^{-1} = 1$ ns, $\gamma_{S_1}^{-1} = 2.92$ ns, $\Gamma^{-1} = 10$

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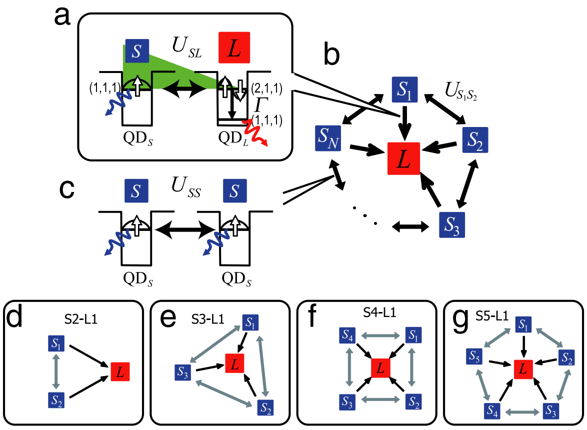

Fig. 1. (a) Optical excitation transfer from smaller quantum dot (QD_S) to larger quantum dot (QD_L) via optical near-field interactions. (b) Multiple-quantum-dot system composed of multiple smaller quantum dots and one large quantum dot networked via optical near-field interactions. (c) Interactions between smaller quantum dots. (d–g) Example systems composed of multiple smaller quantum dots and one large quantum dot.

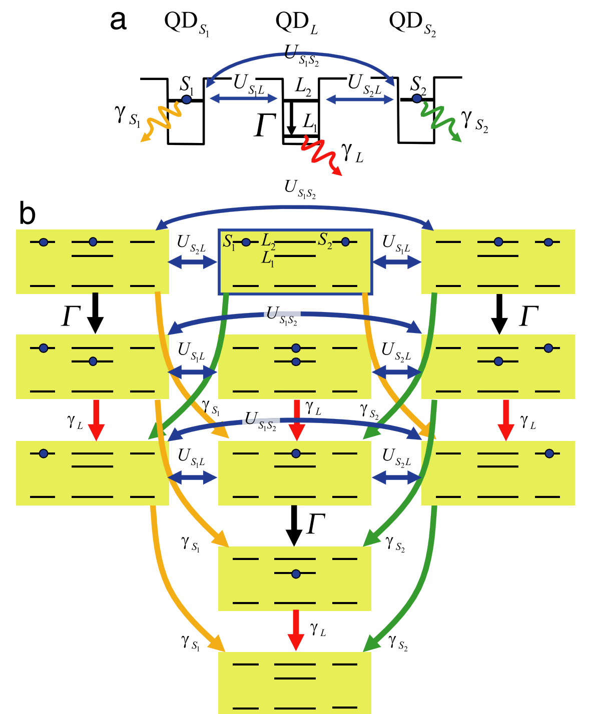

Fig. 2. Example of system modeling based on a density matrix formalism. (a) Parameterizations for inter-dot near-field interactions, radiative relaxations, and non-radiative relaxations in the **S2-L1** system. (b) Schematic representation of state transitions in the **S2-L1** system.

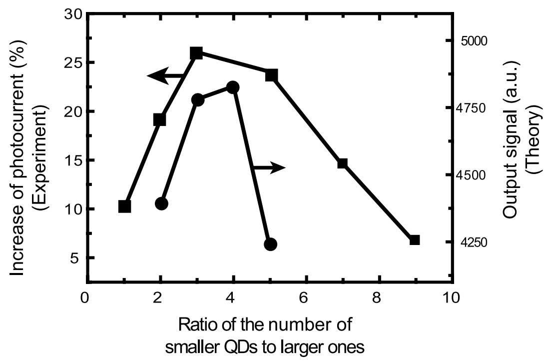

Fig. 3. Optimal ratio of the number of smaller quantum dots to larger quantum dots so that the optical excitation transfer is most efficiently induced.

ps as parameter values. Following the same procedure, we also derive quantum master equations for the **S3-L1**, **S4-L1**, and **S5-L1** systems that have initial states in which all smaller quantum dots are excited. Finally, we calculate the population of the lower level of a larger quantum dot, of which we regard the time integral as the output signal.

We compare the output signal as a function of the *ratio* of the *number* of smaller dots to the number of larger dots assuming that the total number of quantum dots in a given unit area is the same, regardless of their sizes (smaller or larger). As shown by the circles in Fig. 3, the most efficient transfer is obtained when the ratio of the number of smaller dots to the number of larger dots is 4. In other words, increasing the number of smaller quantum dots beyond a certain level does not necessarily contribute to increased output signals. Because of the limited radiation lifetime of large quantum dots, not all of the initial excitations can be successfully transferred to the large quantum dots due to the states occupying the lower excitation levels of the large quantum dots. Therefore, part of the input populations of smaller quantum dots must decay, which results in a loss in the transfer from the smaller quantum dots to the large quantum dots when there are too many excitations in the smaller quantum dots surrounding one large quantum dot.

An optimal mixture of smaller and larger quantum dots was experimentally demonstrated by using two kinds of CdSe/ZnS core/shell quantum dots whose diameters were 2.0 and 2.8 nm [13,15]. In the experimental details in Ref. [13], the increase of the photocurrent used in the output signal was measured. As shown by the squares in Fig. 3, the maximum increase was obtained when the ratio of the number of smaller quantum dots to larger dots was 3:1, which agrees well with the theoretical optimal ratio discussed above.

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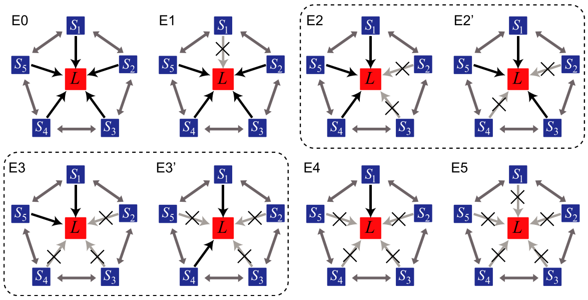

Fig. 4. Eight different network topologies in the **S5-L1** system, where some of the interactions between the smaller quantum dots QD_S and the large quantum dot QD_L are degraded, or lost. (Degraded interactions are indicated by "X".) The notation *EN* indicates that the system contains *N* degraded interactions.

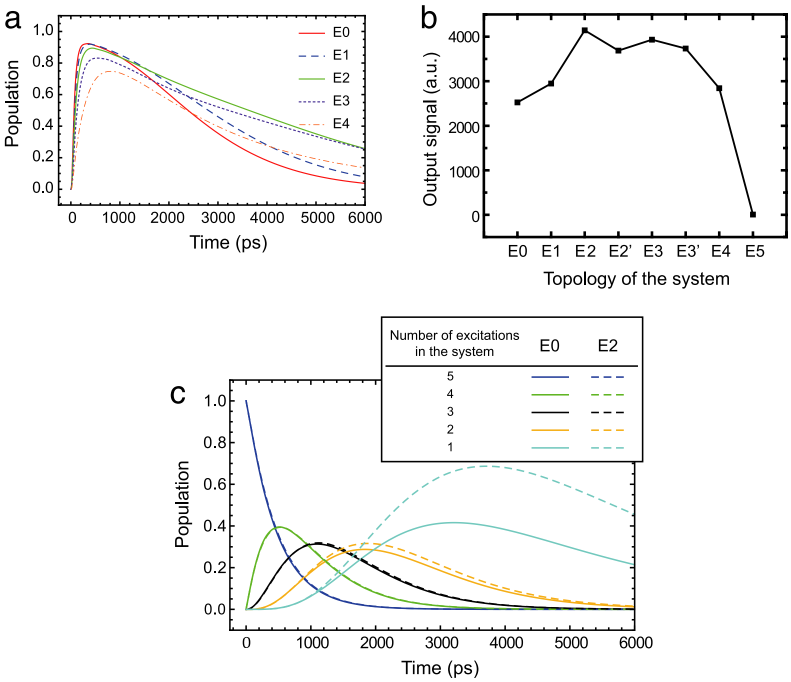

Fig. 5. (a) The evolution of the populations associated with the large quantum dot QD_L in systems **E0**, **E1**, **E2**,**E3**, and **E4** in Fig. 4. (b) Time-integrated populations for the systems in Fig. 4, where systems with certain negligible, or essentially nonexistent, links result in higher output signal levels. (c) The evolution of the populations associated with the number of excitations (ranging from 1 to 5) in systems **E0** and **E2**.

3. Topology-dependent, autonomous, efficient optical excitation transfer

In the previous section, we observe that the amount of optical excitation transferred from smaller quantum dots to larger quantum dots depends on the ratio of their numbers. This suggests that we could increase the output by engineering the network structure of the quantum dots. This section takes the **S5-L1** system in Fig. 1(g) as an example, and demonstrates that it is possible to increase the output signal by appropriately configuring the network of quantum dots. We set all of the inter-dot interaction times to 100 ps, while keeping all other parameter values the same as those in Section 2.

Fig. 4 shows the original **S5-L1** system, denoted by **E0**, which is the same as the system shown in Fig. 1(g). Assume that some of the interactions between the smaller quantum dots (denoted by S_1 to S_5) and the large quantum dot surrounded by them are degraded, or lost, due to, for instance, material disorders, such as a violation of the condition represented by Eq. (1). In total there are eight such configurations when symmetries are taken into account; for instance, when one of the five links between the smaller quantum dots on one hand and the large quantum dot on the other hand is degraded, we obtain the system **E1** in Fig. 4. The mark "X" indicates a degraded interaction between S_1 and L. Similarly, when there are two degraded links, the system should be represented either by the system **E2** or the system **E2'** shown in Fig. 4.

Fig. 5(a) demonstrates the time evolutions of the populations associated with radiation from the large quantum dots. Fig. 5(b) summarizes the integrated populations as a function of the network configurations in Fig. 4. Interestingly, except for the system **E5**, which has no valid links

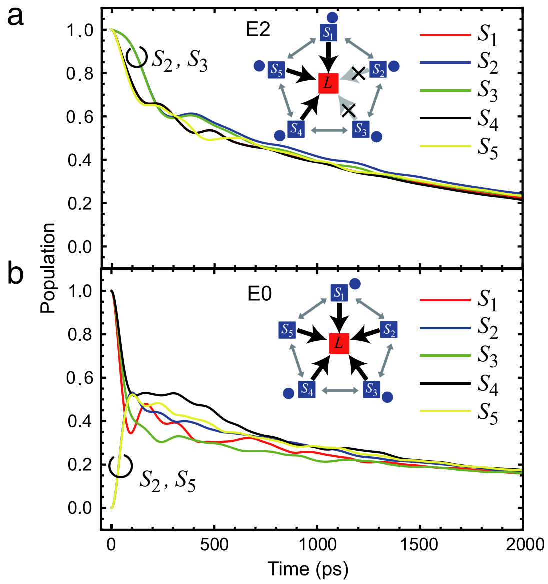

Fig. 6. (a) Time evolutions of the populations associated with the smaller quantum dots (S_1 to S_5) in system **E2** in Fig. 4 while assuming that all smaller quantum dots contain excitations in the initial setup. (b) Time evolutions of the populations associated with the smaller quantum dots (S_1 to S_5) in system **E0** in Fig. 4 while assuming three excitations at S_1, S_3, and S_4.

between the smaller quantum dots and the large quantum dot, systems with degraded interactions exhibit a higher output signal than the system **E0** without the link defects. System **E2** exhibits an output signal that is about 1.64 times higher than system **E0**. This corresponds to the results described in Section 2, where the output is maximized when the ratio of the number of smaller dots to large dots is 4, meaning that the excessively high number of excitations in the smaller dots cannot be transferred to the large dot they surround. Due to the "limited" interactions between the smaller dots and the large dot, such as in the case of systems **E2** and **E2'**, the excitations located in the smaller dots have a higher probability to be transferred to the larger dot. Fig. 5(c) demonstrates the evolution of populations associated with the total number of excitations contained in the system, ranging from 1 to 5. The solid and dashed curves in Fig. 5(c) respectively refer to systems **E0** and **E2**. The populations containing one excitation increase dramatically in **E2** as compared with **E0**, which is another indication that the excitations can be kept in the system until they are successfully transferred to the destination, exhibiting a topology-dependent efficiency increase.

The autonomous behavior of optical excitation transfer is emphasized by Fig. 6(a), which summarizes the evolutions of populations associated with S_1 to S_5 in system **E2,** where both the interaction between S_2 and L and the interaction between S_3 and L are negligible. Initially, all of the smaller dots contain excitations. Note that the populations associated with S_2 and S_3 remain at a higher level for a short initial time, indicating that the excitations in S_2 and S_3 are effectively "waiting" in the smaller dots until they have the opportunity to be transferred to a large dot. Such an autonomous transport is also observed in a "redundant" situation. Fig. 6(b) characterizes the excitation transfer in system **E0** which contains only the three excitations at S_1, S_3, and S_4. We can observe that the populations associated with S_2 and S_5 grow instantaneously, whereas they are zero at the start. In other words, we can see that the excitations autonomously exploit the free, usable resources in the system in order to yield efficient transport.

4. Discussion and implications for information and communications systems

Finally, we make a few remarks about how we can apply these findings in nanometer-scale light-matter interaction networks to information and communications technologies (ICTs).

4.1. Autonomous behavior of optical excitations

The first point we should highlight is the autonomous behavior observed in the optical excitations. As we saw from the experiments, there is no "central controller" in the systems, and yet, efficient transport of the optical excitations is realized. Such an intrinsic, seemingly intelligent behavior of the nanometer-scale physical system may also provide valuable lessons for designing self-organizing, distributed, complex ICT systems on the Internet scale.

Currently, great efforts are being made towards designing a new Internet architecture that is capable of supporting the heterogeneity and cooperation among various types of devices and services anticipated in the future [19,7]. While traditional communication technology follows the *client/server* paradigm, new proposals suggest shifting towards fully distributed control mechanisms and network topologies. In a client/server system, clients (e.g., web browsers) make requests for services to servers (e.g., requests for a web page), and the server tightly controls the content and its delivery. On the other hand, distributed network architectures with simple units that exhibit capabilities such as self-organization, self-adaptation, and self-healing have shown benefits in the past when it comes to scalability of the number of requests and robustness to server failures [10]. One popular example is peer-to-peer (P2P) networks, where all network nodes, being equal peers, may simultaneously play the roles of client and server. P2P networks are currently used as application layer overlay networks for content distribution or distributed directory services on the Internet, but it is foreseeable that more and more network architectures will be based on the P2P concept in the future rather than client/server. The benefits of such distributed topologies are that (i) a single point of failure at the server is avoided, (ii) the overall complexity of the system is reduced, and (iii) distributed topologies are more suitable in handling suddenly appearing overload conditions or balancing traffic load and energy consumption.

A recent trend in communication networks also shows that distributed and cooperative methods inspired by biological [3,4] or physical [23] phenomena have gained much attention as flexible and robust mechanisms for autonomous network management and control. Although these nature-inspired autonomous mechanisms often

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show inferior performance compared with tightly controlled systems in a static environment, they have great benefits in sustainability and reliability under unknown or changing conditions similar to the autonomous and efficient optical excitation transfer in systems involving missing or failing links discussed in Section 3.

4.2. Robustness against errors

The second important observation is that the increase in the output signal induced by degraded interactions indicates robustness against errors occurring in the system. Such behavior is also of great importance for future communication networks. Since new generation networks are expected to accommodate a large number of heterogeneous end-devices, access technologies, network protocols/services, and traffic characteristics, the consideration of failures or sudden fluctuations in performance seems inevitable. Designing robust mechanisms is, therefore, a key issue, and utilizing such an intrinsic robustness of nanophotonics, which has the potential to provide superior behavior in the presence of errors, while requiring less hardware redundancy than current proposals with redundancies of the order of 10–100 [21], may provide helpful guidelines and principles for constructing efficient future ICT systems.

Let us consider the following simple analogy of a sink node in an arbitrary (wired or wireless) communication network, receiving packets from neighboring source nodes. The node layout follows that of the star topology in Fig. 4, where the sink node corresponds to the large dot and the source nodes resemble the small dots. Our results in Section 3 imply the following. If a source node attempts to transmit a packet, but finds the direct channel to the sink busy or inaccessible, it is promising for this node to attempt a retransmission via a neighboring node since that node may have available resources to forward the packet. Particularly in the case where links may suddenly fail (e.g., due to instantaneous fading on a wireless communication channel), the additional diversity achieved by relaying over a neighboring node results in a higher delivery rate of packets. Furthermore, from the discussion of the best ratios of small quantum dots to large dots, we can see that there is an optimal number of neighboring nodes. Such a value may also exist in communications. There are numerous studies on network topologies using complex network theory, studying the theoretical properties of network connectivity by means of node degree distribution or clustering coefficient. If each node sets its connectivity degree to the ideal number of neighbors depending on their transmission and processing rates, efficient management of distributed ad hoc or P2P networks with less overheads can be achieved.

4.3. Energy-efficiency of optical excitation transfer

Finally, it was demonstrated that a single process of optical excitation transfer is about 10^4 times more energy-efficient compared with the single bit flip energy required in current electrical devices [11]. A system-level, comprehensive comparison of energy efficiency is an important and timely subject that should be explored in the future. On the other hand, energy transfer in light harvesting antennas exhibits superior efficiency [18,22], and these structures have similarities with nanostructures networked via optical near-field interactions. These studies will be extremely helpful for developing energy-efficient strategies to assist in handling the tremendous growth in traffic and required processing energy anticipated in future communication networks [6,20].

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