PAPER

Long-lived quantum speedup based on plasmonic hot spot systems

Jun Ren1,2, Tian Chen3 ☭ and Xiangdong Zhang2 ☭

1 College of Physics and Information Engineering, Hebei Advanced Thin Films Laboratory, Hebei Normal University, 050024, Shijiazhuang, Hebei, People’s Republic of China
2 Beijing Key Laboratory of Nanophotonics & Ultrafine Optoelectronic Systems, School of Physics, Beijing Institute of Technology, 100081, Beijing, People’s Republic of China
E-mail: chentian@bit.edu.cn

Keywords: surface plasmon polaritons, hot spots, quantum speedup, quantum walk, Green’s tensor technique

Abstract

Long-lived quantum speedup serves as a fundamental component for quantum algorithms. The quantum walk is identified as an ideal scheme to realize the long-lived quantum speedup. However, one finds that the duration of quantum speedup is too short in real systems to implement the quantum algorithms, for instance the speedup in the photosynthetic light-harvesting systems can last only dozens of femtoseconds. Here, we construct one plasmonic system with two-level molecules embodied in the hot spots of one-dimensional nanoparticle chains to realize the long-lived quantum speedup. The coherent and incoherent coupling parameters in the system are obtained by means of Green’s tensor technique. Our results reveal that the duration of quantum speedup in our scheme can exceed 500 fs under strong coherent coupling conditions. Moreover, our plasmonic system has far prospect in realizing high-dimensional quantum walk, which is very beneficial for quantum algorithms.

1. Introduction

Quantum information exhibits the advantage over its classical counterpart due to the appearance of the quantum speedup [1–12]. Long-lived quantum speedup has been widely applied in the quantum information processing, e.g. quantum algorithms. One ideal theoretical scheme involving the long-lived quantum speedup is the quantum walk [2–17]. In the quantum walk [2–15], a walker initially located at one site of a graph has a probability of reaching two adjacent sites, and then reaches each site of the graph as time passes. The walker’s state is described by a superposition of positions in the quantum walk, which is different from a probability distribution over positions in the classical walk [3, 4]. The intrinsic property of quantum speedup in the quantum walk is reflected by the relation between the mean squared displacement $\langle (\Delta x)^2 \rangle$ of walker and the time $t$ [3, 4]. The operator $\Delta x$ represents the displacement of the walker away from its initial position. The ideal quantum walk on a line features a long-lived ballistic spreading in which the mean squared displacement of the walker grows quadratically with time, $\langle (\Delta x)^2 \rangle \propto t^2$. In comparison, the classical walk on a line features a diffusive spreading where the mean squared displacement of the walker grows linearly with time, $\langle (\Delta x)^2 \rangle \propto t$. Compared to the spreading of the classical walk, such enhanced rate of spreading in the quantum walk can be regarded as a generic indicator of quantum speedup, which could be used for an efficient quantum search algorithm [5, 6]. However, in a practical system, e.g. photosynthetic light-harvesting systems [18–21], due to the existence of dissipations, weak coherent couplings and the other factors, although the quantum coherence in these systems can last tens of picoseconds, the duration of quantum speedup is only about 70 fs, which is too short to realize quantum speedup algorithm [22].

In recent years, the quantum coherence has also been found in nanophotonic systems such as nanocavities [23, 24], photonic crystal [25] and plasmonic system [26]. As addressed in [26–29], the coupling resonances among nanoparticle (NP) trimer could induce the strong couplings between the molecules, and the strong quantum coherence has been revealed between two molecules in a symmetrical NP trimer system [26]. Given that the quantum coherence is the key factor for constructing the quantum walk, these works motive us to study...
how to realize the ideal quantum walk based on strong coupling plasmonic nanostructures and explore the quantum speedup in these nanostructures.

In this work, we demonstrate that one-dimensional (1D) continuous-time quantum walk can be constructed within a scheme of the NP chain involving plasmonic hot spots. The decoherence from the ambient environment has been taken into account. The dynamics for such a system is obtained by means of the Lindblad master equation approach and the electromagnetic (EM) Green’s tensor technique. We find that when fitting the mean squared displacement of excitation with the power law \( \langle (\Delta x)^2 \rangle \propto t^b \), the duration with the power \( b > 1 \) which indicates the quantum speedup can reach 500 fs. The longer duration of quantum speedup can be realized in our system with a longer NP chain. Although the study on photonic waveguides has reported that the duration of running time reaches several picoseconds \([30]\), such coupled waveguides are unable to realize three or higher-dimensional continuous-time quantum walk for one of the spatial dimensions is used as propagation space (time). In comparison, due to the flexible construction within high-dimensional space \([31–33]\), such plasmonic nanostructures can be designed to realize three or higher-dimensional continuous-time quantum walk, which can exhibit the quantum advantage over its correspondingly classical walk in executing the search algorithms \([34–36]\).

2. General descriptions in 1D plasmonic np chain

The 1D NP chain is depicted in figure 1, in which the separation distances between the particles are assumed as the same (denoted by \( d \)). The two-level molecules (marked by numbers in figure 1) are inserted into the gaps of the chain, and the orientations of electric dipole moments of these molecules are assumed along the axis of the chain. Here, we consider one excitation in these two-level molecules. The molecules can be mapped to the chain, and the orientations of electric dipole moments of these molecules are assumed along the axis of the environment has been taken into account. The dynamics for such a system is obtained by means of the Lindblad master equation approach and the electromagnetic (EM) Green’s tensor technique. We find that when fitting the mean squared displacement of excitation with the power law \( \langle (\Delta x)^2 \rangle \propto t^b \), the duration with the power \( b > 1 \) which indicates the quantum speedup can reach 500 fs. The longer duration of quantum speedup can be realized in our system with a longer NP chain. Although the study on photonic waveguides has reported that the duration of running time reaches several picoseconds \([30]\), such coupled waveguides are unable to realize three or higher-dimensional continuous-time quantum walk for one of the spatial dimensions is used as propagation space (time). In comparison, due to the flexible construction within high-dimensional space \([31–33]\), such plasmonic nanostructures can be designed to realize three or higher-dimensional continuous-time quantum walk, which can exhibit the quantum advantage over its correspondingly classical walk in executing the search algorithms \([34–36]\).

The inevitable dissipations from metallic NPs and radiations into the free space should be considered for studying the dynamics of molecules. The spreading speed and distance of excitation relate closely to the dynamics of the molecules. For such a system composed of molecules and NPs, the environment consisting of EM modes surrounding the molecules should be included in the dynamics of excitation. These EM modes give rise to the decoherence and dephasing in the molecular system. In our study, we apply the Markov approximation to the study of molecules in which all effects of EM modes are treated as Lindblad terms. This approximation demands the condition that the couplings between single molecule and EM modes are much weaker than the couplings between molecules through EM modes. This requirement is consistent with \([37, 38]\), where the Markov approximation is justified from the perspective of time and Green tensor respectively. In this case, the relative rates of fluctuations between the molecules and environment are much smaller than those among molecules. Some studies have verified that the dynamics of molecular system can be appropriately obtained under the Markov approximation \([37, 39, 40]\). In the following section, we will show that this approximation is satisfied in our plasmonic hot spot system when discussing the quantum speedup. Under the Markov approximation, the dynamics of \( N \) molecules inserted in 1D NP chain can be described in the form of the Lindblad master equation \([39]\).
\[
\partial_t \rho = \frac{i}{\hbar} [\rho, H] \\
+ \sum_{i=1}^{N} \frac{\gamma_i}{2} (2\sigma_i \rho \sigma_i^\dagger - \sigma_i^\dagger \sigma_i \rho - \rho \sigma_i \sigma_i^\dagger) \\
+ \sum_{i=j}^{N} \frac{\gamma_{ij}}{2} (2\sigma_i \rho \sigma_j^\dagger - \sigma_j^\dagger \sigma_i \rho - \rho \sigma_i \sigma_j^\dagger),
\]
(2)

where \(\sigma_i^\dagger\) and \(\sigma_i\) are the creation and annihilation operators of the \(i\)th molecule, respectively. The coupling strength \(\gamma_i\) represents the interference term (incoherent coupling) between the \(i\)th and \(j\)th molecules and could lead to the decay of the off-diagonal elements of density matrix \(\rho\). It could be easily demonstrated that \(\gamma_i = \gamma_j\) according to the symmetry of electric Green’s tensor (see equation (6)). The strength \(\gamma_{ij}\) represents the dissipation of the \(i\)th molecule, which includes the dissipation in the environment and non-radiation transition. In the metallic NP cluster environment, this dissipation arises mainly from the loss in the metal. In this work, all the dissipations and interactions among molecules have been included in the calculations. The Hamiltonian of these molecules is described as

\[
H = \sum_{i=1}^{N} \hbar \omega_i \sigma_i^\dagger \sigma_i + \sum_{i>j}^N \gamma_{ij} (\sigma_i^\dagger \sigma_j + \sigma_j^\dagger \sigma_i).
\]
(3)

Here, \(N\) is the number of molecules. The parameter \(\omega\) is the transition frequency of molecules, and here for simplicity, we assume they have the same transition frequency. The dipole–dipole interaction term \(g_{ij}\) between the \(i\)th and \(j\)th molecules can be written as \([41]\)

\[
g_{ij} = \frac{1}{\pi \varepsilon_0 \hbar} \mathcal{P} \int_0^\infty \frac{\omega^2 \text{Im}[\tilde{\mu}_i^* \cdot \tilde{G}(\vec{r}_i, \vec{r}_j, \omega') \cdot \tilde{\mu}_j]}{c^2(\omega' - \omega)} d\omega',
\]
(4)

where \(\mathcal{P}\) stands for the principal integral, and \(\varepsilon_0\) is the relative dielectric constant of the background medium, in this work the background is taken as water \((\varepsilon_0 = 1.77)\). Here \(\tilde{G}\) is the total Green’s tensor. To avoid the sophisticated principle integral, the \(g_{ij}\) can be simplified as \([26, 41]\)

\[
g_{ij} = \frac{\omega^2}{\varepsilon_0 \hbar c^2} \text{Re} [\tilde{\mu}_i^* \cdot \tilde{G}^s(\vec{r}_i, \vec{r}_j, \omega) \cdot \tilde{\mu}_j],
\]
(5)

where \(\tilde{G}^s\) represents the scattered Green’s tensor of the NP chain, the detailed calculations about \(g_{ij}\) can be found in appendix A.

The incoherent coupling strength \(\gamma_{ij}\) and spontaneous decay rate \(\gamma_{ii}\) appeared in equation (2) can be obtained as

\[
\gamma_{ij} = \frac{2\omega^2}{\varepsilon_0 \hbar c^2} \text{Im}[\tilde{\mu}_i^* \cdot \tilde{G}^s(\vec{r}_i, \vec{r}_j, \omega) \cdot \tilde{\mu}_j],
\]
(6)

where the detailed calculations can also be found in appendix A.

Now we will investigate the correspondence between the dynamics of molecules in hot spot systems and the standard quantum walk. We need to emphasize that when we deal with the dynamics of molecules, we still employ the master equation (equation (2)). The effective master equation as addressed below is just to show the formal correspondence between the dynamics of molecules in our hot spot systems and the standard quantum walk. By employing the quantum trajectory method introduced in \([20]\), we can re-express the dynamics of the excitation in molecules as

\[
\partial_t \rho = \frac{i}{\hbar} (\rho H^\text{eff} - H^\text{eff} \rho) + \sum_{i=1}^{N} \gamma_i \sigma_i \rho \sigma_i^\dagger + \sum_{i<j}^{N} \gamma_{ij} \sigma_i \rho \sigma_j^\dagger,
\]
(7)

where the effective Hamiltonian can be written as

\[
H^\text{eff} = H - \frac{1}{2} \sum_{i=1}^{N} \gamma_{ii} \sigma_i^\dagger \sigma_i - \frac{1}{2} \sum_{i=1}^{N} \gamma_{ii} \sigma_i \sigma_i^\dagger.
\]
(8)

Notice that \(H^\text{eff}\) is a non-Hermitian Hamiltonian, and it commutes with excitation number operator \(\sum_{i=1}^{N} \sigma_i^\dagger \sigma_i\), thus it preserves the number of excitation, and can only give rise to the jump of excitation between different molecules. The second term in the right of equation (7) originates from the incoherent interaction between the two-level molecules and plasmons, it can induce dephasing in these molecules without changing the number of excitations. These two terms mentioned above induce the jumps in the single-excitation manifold and no jump to the other excitation manifolds. The last term in the right of equation (7) originates from the dissipation in the plasmonic environment and the radiation to the free space, and it can change the number of
excitations and generate jumps between excitation manifolds. When neglecting the jumps to other excitation manifolds, we have the master equation in the case of a no jump trajectory as

$$\partial_t \rho_{\text{eff}} = \frac{i}{\hbar} (\rho_{\text{eff}} H_{\text{eff}} - H_{\text{eff}} \rho_{\text{eff}}) + \sum_{i,j} \gamma_{ij} \sigma_i \rho_{\text{eff}} \sigma_j,$$

(9)

where $\rho_{\text{eff}}$ is the effective density operator describing the density matrix of all molecules. This equation is often considered as a directed quantum walk on the single-excitation manifold described by the density operator $\rho_{\text{eff}}$. In our discussion below, we will focus on the dynamics of density operator $\rho$ including the jumps between other excitation manifolds (i.e. equation (2)). We will show that even when influenced by the ambient environment, our system can still possess long-lived quantum speedup.

3. The quantum walk dynamics in 1D plasmonic hot spot systems

In this section, we show the dynamics of molecules in 1D plasmonic hot spot systems by using the master equation method. We take the system that composed of six molecules ($N = 6$) and seven NPs as an example. We find that the dynamic properties with other numbers of molecules (e.g. $N = 5, N = 7$ and etc) are similar with the case of $N = 6$, the results can be addressed in appendix B.

According to the theory in section 2, the dynamics of our system involving one excitation depends closely on the coherent and incoherent coupling strengths among molecules. In our study, the NPs are taken as the Ag spheres. For the dielectric functions of Ag, the Drude model ($\omega_p = 9.01 \text{ eV}$ and $\gamma = 0.05 \text{ eV}$ for Ag) describing a widely accepted metallic permittivity model is adopted in this work. When the separation distance between the NPs is very small (several nanometers), the strong couplings formed among the NPs can bring about many resonance surface plasmon polariton modes in the system. These surface plasmon polariton modes can induce coherent or incoherent couplings between molecules located in the gaps. Here in figure 2, by taking the radii of the Ag spheres as $R = 12 \text{ nm}$, and the separation distances between two nearest-neighbor spheres as $d = 2 \text{ nm}$, we present the coherent and incoherent coupling strengths and molecule decay rates with the transition frequency of molecules.

According to the symmetry of the system, the nearest-neighbor interactions (coherent and incoherent coupling) between the 1st and 2nd molecules are the same as the interactions between the 5th and 6th molecules, namely $g_{12} = g_{56}$ and $\gamma_{12} = \gamma_{56}$, and for the other nearest-neighbor interactions, there is $g_{23} = g_{45}$ and $\gamma_{23} = \gamma_{45}$. Similarly, for the non-nearest-neighbor interactions we have $g_{34} = g_{45}$ and $\gamma_{34} = \gamma_{45}, g_{23} = g_{35} = g_{56}$ and $\gamma_{23} = \gamma_{35}$, $g_{43} = g_{45}$ and $\gamma_{43} = \gamma_{45}$. In figure 2(a), the nearest-neighbor coherent couplings $g_{12}/\gamma_{11}, g_{23}/\gamma_{22}$ and $g_{34}/\gamma_{33}$ are showed by black, red and blue lines, respectively. The three lines in figure 2(b) correspond to the decay rates of the first, second and third molecules ($\gamma_{11}, \gamma_{22}$ and $\gamma_{33}$). As for figure 2(c), the three lines correspond to the nearest-neighbor incoherent couplings $\gamma_{12}/\gamma_{11}, \gamma_{13}/\gamma_{12}$ and $\gamma_{24}/\gamma_{33}$, respectively. In comparison, if there are no NPs in our system, the coupling between two molecules with a distance $d = 26 \text{ nm}$ is very small and the energy transfer efficiency between them will be reduced to 0.055% [43], which means the initially excited molecule has a high probability of emitting energy into free space rather than the other molecule.

It is seen clearly that, there exists multiple resonances in three panels of figure 2. These resonances can affect the dynamics of the excitation of molecules if the transition frequency of the two-level molecules is resonant with the EM mode. To clarify the properties of the resonances, we focus on some special cases. These special frequencies are marked by $\omega_{1} - \omega_{6}$ in figure 2, in which $\omega_{1} - \omega_{4}$ are the cases that the system possesses the large coherent couplings and small incoherent couplings. For example, when $\omega = 3.25 \text{ eV}$ (marked by $\omega_{1}$), one of the nearest-neighbor coherent couplings ($g_{12}/\gamma_{11}$) can reach to 3, however the other two are small. And when $\omega = 3.72$ and $3.78 \text{ eV}$ (marked by $\omega_{2}$ and $\omega_{3}$), the nearest-neighbor coherent couplings ($g_{23}/\gamma_{22}, g_{34}/\gamma_{33}$) can reach to 2 simultaneously. When $\omega = 4.64 \text{ eV}$ (marked by $\omega_{4}$), the three coherent couplings are 0.8 simultaneously. Under the frequencies $\omega = \omega_{1}-\omega_{4}$, the coherent couplings are in peak values (figure 2(a)) and the decay rates induced by EM modes are small and flat (figure 2(b)). The coherent coupling strengths between the nearest-neighbor molecules induced by EM modes are larger than the coupling strengths between a single molecule and EM modes. The coupling strength between the single molecule and EM field is inversely proportional to the width at half maximum of the peak [37], while the frequencies $\omega = \omega_{1}-\omega_{4}$ deviate from the peak frequency (figure 2(b)), namely the coupling between the molecule and EM field is small in these cases, thus the relative rate of fluctuations between the molecules and environment is much smaller than those among the molecules. This justifies the reasonable use of the Markov approximation, and the main conclusions of this work are obtained based on these frequencies. While, when the frequencies of molecules are $\omega = \omega_{5}$ or $\omega_{6}$, the large dissipation $\gamma_{45}$ in the 3th and 4th molecules appears and the interaction between the 3th and 4th molecules is small. These two frequencies are only provided as the comparisons, in which the coherent couplings are very small and incoherent couplings are not very large simultaneously.
To further clarify the properties of the resonances, in figure 3 we plot the corresponding normalized electric field intensity distribution in XY plane with the plane wave incidence in the Y direction. Here the incident frequencies \( \omega_1 - \omega_6 \) correspond to six frequencies marked in figure 2. It is shown that when the couplings (coherent or incoherent) are maximums (\( \omega = \omega_1 - \omega_3 \) and \( \omega_5 \)), the electric field patterns are similar, and the fields in hot spots are much larger than other regions. This stems from the coupling resonances among the NP chain as has been discussed in [26, 27]. In comparison, we also plot the electric field pattern under the single scattering frequency (\( \omega = 5.25 \) eV as marked by arrow in figure 2(b) and refers to the resonant frequency of a single, isolated NP), which is showed in the last panel of figure 3. In this panel, the intensities of the fields in the gaps are nearly the same as that around the particles. That means the fields in the hot spots cannot be confined. When \( \omega = \omega_4 \) and \( \omega_6 \), the fields in hot spots are very weak due to the weak coupling resonances in the system. Therefore in these cases (\( \omega = \omega_4 \) and \( \omega_6 \)), the electric field patterns are similar to the case of single scattering resonance (last panel in figure 3).

The electric field patterns in figure 3 show that under the condition of strong resonance couplings, the large electric field can be confined within the positions of hot spots. Such strong confinement of fields results in the strong couplings between the molecules that located at the hot spots. In addition, comparing figure 2 (c) with figure 2(b), we find that, incoherent couplings between the molecules correspond very well to the resonance peaks, which are resonant couplings; while coherent couplings have a shift compared to resonance peaks, which are off-resonant couplings [26].

Various resonance couplings can lead to different populations of the excitation of molecules. The time evolution of the populations of six molecules (denoted by \( P_{1}-P_{6} \)) are shown in figure 4 with six different frequencies (marked by \( \omega_1 - \omega_6 \) as shown in figures 2 and 3). It has to be pointed out that, in the calculations of populations (figure 4), all the non-nearest-neighbor couplings among molecules have also been included. The results of the non-nearest-neighbor coherent and incoherent coupling strengths between the molecules have been shown in appendix C, see figure C1. In figure 4, the dashed black line denoted by \( P_0 \) is the time evolution of population in all molecules. As addressed in figure 4, when \( \omega = 3.25 \) eV (\( \omega_1 \)), only one nearest-neighbor coupling strength \( \frac{g_{34}}{\gamma_3} \) can reach to -3, the other nearest-neighbor coupling strengths \( \frac{g_{12}}{\gamma_1} \) and \( \frac{g_{23}}{\gamma_2} \) are...
very small, which leads to small populations at the 4th and 5th molecules ($P_4 = 0.049$, $P_5 = 0.087$). In comparison, when the transition frequency is taken as $\omega = 3.72$ eV ($\omega_2$) or $3.78$ eV ($\omega_3$), all the nearest-neighbor coherent couplings showed in figure 2 can reach 2 simultaneously; while the nearest-neighbor coherent couplings at these frequencies, with the plane wave incidence in the $Y$ direction.

Figure 3. The normalized electric field intensity distributions in $XY$ plane with the plane wave incidence in the $Y$ direction. The incident frequencies $\omega_1=\omega_2=\omega_3=\omega_4=\omega_5=\omega_6$ correspond to six frequencies marked in figures 2, and 5.25 eV is a reference frequency. The parameters of the system are the same as that in figure 2.

Figure 4. The log–log plot of the time evolution of the populations of six molecules when the first molecule is excited and other five molecules are in their ground states initially. Where the solid lines $P_1$–$P_6$ denote the populations of the six molecules, and the dashed black line $P_0$ is the time evolution of population residing in all molecules. The parameters of the system are the same as figure 2.
incoherent couplings and decay rates are small enough. In these cases, the molecules inserted in the gaps of the NP cluster can be treated as an ideal 1D chain with the same nearest-neighbor couplings. As revealed in figure 4, the populations on all molecules at these two transition frequencies ($\omega_2$ and $\omega_3$) can reach about 0.2. However, when the frequency for molecule takes $\omega = \omega_1$, $\omega_5$ or $\omega_6$ the strong coupling between molecules cannot be formed. For these three frequencies, only some of molecules can be excited during the evolution (figure 4).

4. The quantum speedup in plasmonic hot spot systems

In this section, we explore under what conditions our hot spot system holds the long-lived quantum speedup. A natural way to evaluate the quantum speedup is the exponent $b$, which relates to the spreading of the excitation in the 1D chain by the power law $\langle (\Delta x)^2 \rangle \propto t^b$. Here, $b = 2$ corresponds to the ideal quantum speedup in the quantum walk and $1 < b < 2$ displays the non-ideal quantum speedup. While $b = 1$ corresponds to the diffusive transport of classical random walks and $b < 1$ corresponds to the sub-diffusive transport, in which the dynamics does not belong to quantum realms [22]. The long-lived quantum speedup is one of the most important property in the quantum walk, which makes it exhibit advantages over the classical walk in search algorithms and other information processing. However, in one realistic system, the ideal quantum speedup could turn to the diffusive transport gradually due to the decoherence in the system, and then to the sub-diffusive transport. Such transition can be reflected by the change of the power $b$ from 2 to 1, and then less than 1. Thus, the duration of $b > 1$ determines the quality of the quantum speedup in quantum walks. In figure 5(a), we present the mean squared displacement of excitation $\langle (\Delta x)^2 \rangle$ as a function of time, in which the parameters of the system are the same as those in figure 2. In figure 5(b), we plot the fitted power $b$ as a function of time at four frequencies. In figures 5(a) and (b), the frequencies $\omega_1 - \omega_6$ correspond to the six frequencies showed in figure 4.

According to figures 2 and 4, when the frequency is taken as $\omega = \omega_1$ or $\omega_6$, the nearest-neighbor coherent couplings between any two adjacent molecules in 1D chain are strong, and the decay for each molecule is rather small. In these two cases, these molecules along 1D chain have the similar configuration as that in the continuous-time quantum walk, and populations at all molecules can reach the nearly same value in sequence. Our calculations in figure 5(b) show that the duration of quantum speedup under $\omega_1$ or $\omega_6$ can reach about 400–500 fs. Such duration of quantum speedup is larger than those in photosynthetic light-harvesting systems (about 70 fs) [22]. While when the transition frequencies take other values ($\omega_1$ and $\omega_1 - \omega_6$), the weak coherent couplings between molecules and the strong decays for molecules make the durations of quantum speedup smaller than 100 fs.
In the following, we explore how the separation distance between adjacent NPs affects the quantum speedup in the system. When the separation distance changes, the competition between couplings and dissipations in the system could largely affect the duration of quantum speedup. We investigate two cases with \(d = 1\) nm and \(d = 4\) nm, respectively. In both cases, the radii of NPs is same, \(R = 12\) nm.

The solid lines in figures 6(a) and (b) show that the time evolution of the populations of six molecules at different frequencies for \(d = 1\) nm and \(4\) nm, respectively. And the dashed black lines in figure 6 denote the time evolution of population in all molecules. It is clear that the energy dissipates much more quickly when \(d = 1\) nm than the case with \(d = 4\) nm due to the much larger dissipation. Similar to figure 4 (the populations for \(d = 2\) nm), we choose frequencies \(\omega_1 - \omega_3\) in figure 6 corresponding to the cases that nearest-neighbor coherent coupling strengths are in their peak values and the incoherent couplings are almost zero. We first focus on the case with \(d = 1\) nm (figure 6(a)). When the transition frequency is \(\omega_2\) or \(\omega_3\), the nearest-neighbor coherent coupling strengths between any two adjacent molecules can reach 1.8 simultaneously, which are a little smaller than the case with \(d = 2\) nm (the nearest-neighbor coherent coupling strengths reach 2.0 in figure 2(a)). While, due to large dissipations for the case with \(d = 1\) nm, populations on all molecules are dissipated in a very short-time. Under the other transition frequencies (\(\omega_1, \omega_4, \omega_5\) or \(\omega_6\)), our calculation reveals that the nearest-neighbor coupling (coherent/incoherent) strengths between some molecules are small. When the separation distance is \(d = 4\) nm (figure 6(b)), nearest-neighbor coherent coupling strengths between adjacent molecules only reach about 1.1 simultaneously, that is much smaller than the nearest-neighbor coherent coupling strengths with \(d = 1\) nm and \(d = 2\) nm. In this case with \(d = 4\) nm, the dissipation strengths are one order of magnitude smaller than those with \(d = 2\) nm. Such small dissipations make populations survive with longer time (as shown in figure 6(b)). However, when \(d = 4\) nm and \(\omega = \omega_5\), some of the nearest-neighbor coherent coupling strengths between two adjacent molecules are small, which makes the short-time quantum speedup. When \(d = 4\) nm and \(\omega = \omega_2 - \omega_3\), due to the small coherent couplings, the excitation can only reach the third molecule at most and cannot transfer any more.

Figure 7 shows the mean squared displacement of excitation \(\langle (\Delta x)^2 \rangle\) (a) and the fitted power \(b\) (b) as a function of time when \(d = 1\) nm (left column) and \(d = 4\) nm (right column). The quantum speedup with different frequencies are presented with different curves of colors. When the separation distance \(d = 1\) nm, we find that the duration of quantum speedup is no longer than 150 fs (see the case with the transition frequency chosen as \(\omega_2\) or \(\omega_3\) in figure 7(b)), which is much smaller than the duration of quantum speedup with \(d = 2\) nm (about 500 fs in figure 4(b)). While when \(d = 4\) nm, the duration of quantum speedup can reach about 350 fs under \(\omega = \omega_4\) (figures 7(c) and (d)), which is shorter than the case with \(d = 2\) nm. However, although the curves with \(\omega_2\) and \(\omega_3\) in figure 7(d) appear to have a long-lived quantum speedup (in the order of picosecond), the excitation can only be limited within the first two or three molecules (populations in figure 6(b)). Since the realization of quantum search algorithm needs to search in a large space [34–36], such confinement of excitation in some of molecules with \(\omega_2\) and \(\omega_3\) make our system do not suitable for realizing quantum search algorithm.

![Figure 6](image_url)
Considering the discussion above, in the case with the radii of NP \( R = 12 \text{ nm} \), the separation distance \( d = 2 \text{ nm} \) is an optimal solution for the ideal quantum walk with long-lived quantum speedup (compare figures 5 and 7). In our plasmonic hot spot systems, we are able to find an appropriate separation distance for the fixed radii of NPs. In this way, the relatively large nearest-neighbor coherent coupling strengths between any two adjacent molecules are obtained and the long-lived quantum speedup is realized. Similarly, when the radii of the NP is changed, the competition between the coherent/incoherent couplings and dissipations still exists in our scheme, and the optimal separation distance to construct quantum walk with long-lived quantum speedup should be changed. We have also studied the coherent/incoherent coupling strengths and populations for the case with \( R = 10 \text{ nm} \) and \( d = 4 \text{ nm} \). Comparing to the case with \( R = 12 \text{ nm} \) and \( d = 4 \text{ nm} \), we find that the coherent coupling strengths decrease about 10% when the radii of spheres decreases to \( R = 10 \text{ nm} \). This directly leads to the decrease of populations on molecules. Therefore, when we decrease the radii of the spheres, we need to decrease the separation distance to ensure the large enough nearest-neighbor coherent couplings between molecules. On the contrary, when we increase the radii of the spheres in our scheme, dissipations in these molecules and coherent couplings between adjacent molecules increase simultaneously. In this case, the optimal separation distance also increases to implement a long-lived quantum speedup.

Based on the study above, to achieve long-lived quantum speedup in 1D plasmonic hot spot system, we need the large nearest-neighbor coherent couplings between adjacent molecules and small dissipation for each molecule. When these two requirements are satisfied, we can realize the time duration of quantum speedup about 420 fs, 500 fs and 580 fs for plasmonic hot spot system with 5, 6 and 7 molecules, respectively.

### 5. Discussions and conclusions

In our discussion above, we do not consider the effects from imperfections in our system. Here, we study two primary imperfections (random gap distance and nonlocal effect) and show the quantum speedup still exists. Firstly, we take the case of \( N = 5, R = 12 \text{ nm} \) and \( d = 2 \text{ nm} \) as an example to investigate the effect of random gap distance. We present the calculated coupling parameters with the random separation distances.
and $\tau$ (dashed lines) and $2.0 \pm 0.2$ nm (dotted lines) in figure C2 of appendix C. We find these curves for coupling strengths only present a shift in frequency and the maximums have no obvious decrease. Therefore, the quantum speedup still exists in our system. Secondly, we consider the nonlocal effect in the study of quantum speedup. In fact, when the radii of spheres decreases to 10 nm and separation distances are smaller than 1 nm, the nonlocal effect cannot be ignored [44–47]. In these cases, the plasmon-enhanced fluorescence could be decreased by a small value [48], which leads to a small decrease in the dissipation strengths of molecules. In addition, previous researches have shown that coherent coupling strengths in a trimer system ($R = 10$ nm and $d = 1$ nm) only present small blue shifts due to the nonlocal effect [26], and the values of coupling strengths keep the same. In our plasmonic hot spot system, we also investigate the nonlocal effect (see figure C3 in appendix C). We find that the nearest-neighbor couplings and dissipations only have blue shifts and have no obvious change in values. Thus we can obtain the similar quantum speedup behavior by shifting the molecular frequencies. It means that the random gap distance and nonlocal effect affects dissipations and couplings among molecules little, and does not influence quantum speedup and the formation of quantum walk in our discussion.

In conclusion, we theoretically investigate the long-lived quantum speedup within molecules inserted in the gaps of 1D NP chain in this work. In our study, the crucial requirement to realize the long-lived quantum speedup suggests that all nearest-neighbor coupling strengths between molecules should be strong enough and the losses of molecules are small. Our results verify that this requirement can be satisfied in our plasmonic hot spot system with some frequencies. In this way, the duration of quantum speedup in plasmonic hot spots system under room temperature can reach hundreds of femtoseconds, which is much longer than that in photosynthetic light harvesting systems. Although this duration of quantum speedup is shorter than that reported in coupled waveguides [30], our plasmonic hot spots system can be easily extended to three or high-dimensional continuous-time quantum walk based on nowadays technologies in nanostructure constructions [31–33]. Within these realizations of three or high-dimensional quantum walks, we can display the quantum advantage in search algorithms, which is the main goal of our future research.

Acknowledgments

This work was supported by the National Key R and D Program of China under Grant No. 2017YFA0303800 and the National Natural Science Foundation of China (11574031 and 11604014). J R was also supported by the Hebei NSF under Grant No. A2016205215 and the National Natural Science Foundation of China (11847060 and 11575051).

Appendix A: The detailed calculations of coupling strengths among molecules and electric fields in the NP chain

Here we present the detailed calculation methods of the coherent and incoherent terms ($g_{ii}$, $\gamma_i$ and $\gamma_i^0$), as shown in equations (5) and (6), and the electric field intensity distributions showed in figure 3. In equations (5) and (6), $G^i(\vec{r}_i, \vec{r}_j, \omega)$ and $G(\vec{r}_i, \vec{r}_j, \omega)$ represent the scattered and total Green’s tensors of the NP chain, respectively. The vectors $\vec{r}_i$ and $\vec{r}_j$ are the positions of the $i$th and $j$th molecules, respectively. The Green’s tensor $G(\vec{r}_i, \vec{r}_j, \omega)$ has the meaning of the total electric field in the position of $\vec{r}_i$ caused by a unit dipole at the location of $\vec{r}_j$ in the presence of NP chain, similar to $\tilde{G}^i(\vec{r}_i, \vec{r}_j, \omega)$ [49].

In this work, the Green’s tensors of the NP system are all calculated with the multiple scattering $T$-matrix technique [50–52]. The total and scattered Green’s tensors have the following relation

$$\tilde{G}(\vec{r}_i, \vec{r}_j, \omega) = G^i(\vec{r}_i, \vec{r}_j, \omega) + G^\text{vac}(\vec{r}_i, \vec{r}_j, \omega),$$  \hspace{1cm} (A1)

where $G^\text{vac}(\vec{r}_i, \vec{r}_j, \omega)$ is the Green’s tensor in the vacuum, which can be readily known in [49]. In the following, we introduce the multiple scattering $T$-matrix method to calculate the scattered and total Green’s tensors mentioned above.

The incident field $E^\text{inc}$ and the scattered field of the $i$th NP $\tilde{E}^i_\text{inc}$ can be expanded with the vector spherical functions [50–52]

$$E^\text{inc}(\vec{r}) = \sum_{\nu=1}^{\infty} a^\nu_i M^\nu_i(k\vec{r}) + b^\nu_i N^\nu_i(k\vec{r}),$$  \hspace{1cm} (A2)

$$\tilde{E}^i_\text{inc}(\vec{r}) = \sum_{\nu=1}^{\infty} f^\nu_i \tilde{M}^\nu_i(k\vec{r}) + g^\nu_i \tilde{N}^\nu_i(k\vec{r}),$$  \hspace{1cm} (A3)

where $\tilde{M}^\nu_i, \tilde{N}^\nu_i, \tilde{M}^\nu_i$ and $\tilde{N}^\nu_i$ are the well-known vector spherical functions, and $\vec{r}$ is the position vector in the coordinate of the $i$th NP.
In equations (A2) and (A3), the coefficients $a_i^s$, $b_i^s$, $f_i^s$ and $g_i^s$ can be easily solved as soon as the form of the incident wave is given. The subscript $\nu$ stands for $(m, n)$ which are the indices of spherical harmonic functions. At the same time the internal field of the $i$th NP can be written as

$$E_{\text{int}}^{(i)}(r_i) = \sum_{r=1}^{\infty} c_i^r \mathcal{M}_r^i(kr_i) + d_i^r N_i^r(kr_i). \quad (A4)$$

According to the T-matrix method [50–52], $c_i^r$ and $d_i^r$ are related to $a_i^s$ and $b_i^s$ by the following matrix equation:

$$\begin{bmatrix} Q_i^{11} & Q_i^{12} \\ Q_i^{21} & Q_i^{22} \end{bmatrix} \begin{bmatrix} c_i^r \\ d_i^r \end{bmatrix} = \sum_{j=1}^{\infty} \begin{bmatrix} T_{ij}^{11} & T_{ij}^{12} \\ T_{ij}^{21} & T_{ij}^{22} \end{bmatrix} \begin{bmatrix} R_g \quad Q_j^{11} & R_g \quad Q_j^{12} \\ R_g \quad Q_j^{21} & R_g \quad Q_j^{22} \end{bmatrix} \begin{bmatrix} c_j^r \\ d_j^r \end{bmatrix} + \begin{bmatrix} a_i^s \\ b_i^s \end{bmatrix}. \quad (A5)$$

where $Q_i^{pq}$ and $R_g \quad Q_j^{pq}$ are the $T$-matrix blocks for the $i$th and $j$th NPs, and $T_{ij}^{pq}$ is the block of the transition matrix between the $i$th and $j$th NPs [50–52]. By solving these equations, expansion coefficients of the inner field for each NP can be obtained. And according to the matrix equation

$$\begin{bmatrix} f_i^s \\ g_i^s \end{bmatrix} = \begin{bmatrix} R_g \quad Q_i^{11} & R_g \quad Q_i^{12} \\ R_g \quad Q_i^{21} & R_g \quad Q_i^{22} \end{bmatrix} \begin{bmatrix} c_i^r \\ d_i^r \end{bmatrix}, \quad (A6)$$

the scattered expansion coefficients $f_i^s$ and $g_i^s$ of each NP can be easily calculated. The field outside the NPs then can be obtained using the following equation:

$$E_{\text{ext}} = E_{\text{inc}} + \sum_{i=1}^{N} E_i^s, \quad (A7)$$

where $N$ is the number of NPs. Obviously, we can calculate the total electric filed intensity distribution according to the method above once we have known the incident field.

In the following we calculate the external scattered field $E_i^s$ induced by a dipole source. We take the exciting source as a dipole $\vec{M}_i^0$ located in $\vec{r}_i$, the incident electric field can be expressed as

$$E_{\text{inc}} = \frac{-\nabla \times \nabla \times \vec{A}_{p}^{(s)}}{4\pi|\vec{r} - \vec{r}_i|}, \quad (A8)$$

where

$$\vec{A}_{p}^{(s)} = -\frac{\omega}{4\pi} \frac{\vec{r} - \vec{r}_i}{|\vec{r} - \vec{r}_i|} \vec{M}_i^0. \quad (A9)$$

Expanding equation (A8) to the same form of equation (A2), from equations (A3)–(A7) we can obtain the external scattered field $E_i^s = \sum_{i=1}^{N} E_i^s$ caused by the dipole source. Then, the scattered Green’s tensor $\vec{G}(\vec{r}_i, \vec{r}_j, \omega)$ can be obtained with

$$\vec{h}_i \cdot \vec{G}(\vec{r}_i, \vec{r}_j, \omega) \cdot \vec{h}_j = -\vec{h}_i \cdot \vec{E}_d^s(\vec{r}_i)|_{\vec{r}_j}, \quad (A10)$$

where $\vec{h}_i$ and $\vec{h}_j$ are the unit vectors of the field and source dipole moments, respectively. $E_d^s(\vec{r}_i)|_{\vec{r}_j}$ represents the field in the position of $\vec{r} = \vec{r}_i$ induced by the source dipole that locates at $\vec{r} = \vec{r}_j$ in the presence of NP chain. For the decay rate of the $i$th molecule $\gamma_i$, the Green’s tensor $\vec{G}(\vec{r}_i, \vec{r}_j, \omega)$ represents the field generated by the dipole in its own position in the presence of NP chain, which can be calculated with

$$\vec{h}_i \cdot \vec{G}(\vec{r}_i, \vec{r}_j, \omega) \cdot \vec{h}_j = -\vec{h}_i \cdot \vec{E}_d^s(\vec{r}_i)|_{\vec{r}_i}, \quad (A11)$$

where $E_d^s(\vec{r}_i)|_{\vec{r}_i}$ represents the field induced by the source dipole that locates at $\vec{r} = \vec{r}_i$ in its own position in the presence of NP chain.

**Appendix B: The dynamics of molecular system with different numbers of molecules**

In the main text, we focus on the dynamics of molecules with the number of molecules $N = 6$. Here, we change the number of molecules and explore the dynamics of molecular system with different numbers of molecules. Firstly, we calculate the coupling strength of system with five molecules ($N = 5$, as shown in the top inset of figure B1), and provide the dynamics with different frequencies (figure B2); then, we change to the system
Figure B1. Top inset: 1D NP chain with five molecules in the gaps. (a) The nearest-neighbor coherent coupling strengths $g_{12}/\gamma_{11}$ (black line) and $g_{23}/\gamma_{22}$ (red line). (b) The decay rates of single molecule $\gamma_{11}$ (black line), $\gamma_{22}$ (red line) and $\gamma_{33}$ (blue line). (c) The nearest-neighbor incoherent coupling strengths $\gamma_{12}/\gamma_{11}$ (black line) and $\gamma_{23}/\gamma_{22}$ (red line). The number of molecules is 5. The parameters of the system are $R = 12$ nm, $d = 2$ nm, the dielectric function of Ag spheres is taken as Drude model ($\omega_p = 9.01$ eV, $\gamma = 0.05$ eV).

Figure B2. The log–log plot of the time evolution of the populations of five molecules when the first molecule is excited and other four molecules are in their ground states initially. Where the solid lines $P_1$–$P_5$ denote the populations of the five molecules, and the dashed black line $P_0$ is the time evolution of the percentage of energy residing in the molecules. The parameters of the system are the same as figure B1.
containing seven molecules ($N = 7$, as shown in the top inset of figure B4). In all discussions, the radii of sphere is set as $R = 12$ nm and the separation distance between two adjacent sphere is $d = 2$ nm.

In figure 8, we show the nearest-neighbor coupling strengths between molecules. With the obtained coupling strengths (the non-nearest-neighbor couplings strengths have been included in the calculation of dynamics, and are not addressed in this paper) in the hot spot system containing five molecules, we apply the master equation method presented in section 2 of main text to solve the dynamics of molecules.

In figure B2 we provide the time evolution of the populations of the five molecules. The six frequencies ($\omega_1, \omega_2$) in the figures correspond to the peak values of nearest-neighbor coherent coupling strengths in figure B1. As explained in the main text, the simultaneously large nearest-neighbor coherent coupling strengths can lead to the long duration of quantum speedup. For these six frequencies, we can find that the percentage of energy residing in the molecules can last hundreds of femtoseconds.

We present the quantum speedup properties in figure B3 with $N = 5$. We fix the parameters of the hot spot system with the radii of sphere $R = 12$ nm, and the distance between two adjacent spheres is $d = 2$ nm. We find that the quantum walk properties in these systems are similar to the case with $N = 6$, that is simultaneously large nearest-neighbor coherent couplings lead to long-lived quantum speedup. We obtain from figure B3 that the optimal duration of quantum speedup is 420 fs with the number of molecules $N = 5$.

In figure 11 we show the nearest-neighbor coupling strengths between molecules. With obtained coupling strengths in the hot spot system containing seven molecules, we apply the master equation method presented in section 2 of main text to solve the dynamics of molecules.
In figure B5 we provide the time evolution of the populations of the seven molecules. The six frequencies \((\omega_1 - \omega_6)\) in the figures correspond to the peak values of nearest-neighbor coherent coupling strengths in figure B5. As explained in the main text, the simultaneously large nearest-neighbor coherent coupling strengths can lead to the long duration of quantum speedup. For these six frequencies, we can find that the percentage of energy residing in the molecules can last hundreds of femtoseconds, and even thousands of femtoseconds.

We present the quantum speedup properties in figure B6 with \(N = 7\). The parameters of the hot spot system are taken as \(R = 12 \text{ nm}, d = 2 \text{ nm}\). We obtain from figure B6 that the optimal duration of quantum speedup is 580 fs with \(N = 7\). Considering that the quantum speedup has been addressed with the number of molecules \(N = 6\) in our main text, we can realize the long-live quantum speedup in the plasmonic hot spot system with different numbers of molecules.
Figure B5. The log–log plot of the time evolution of the populations of seven molecules when the first molecule is excited and other six molecules are in their ground states initially. Where the solid lines $P_1$–$P_7$ denote the populations of the seven molecules, and the dashed black line $P_0$ is the time evolution of the percentage of energy residing in the molecular system. The parameters of the system are the same as figure B4.

Figure B6. The mean squared displacement of excitation $\langle (\Delta x)^2 \rangle$ and (b) the fitted power $b$ as a function of time when $N = 7$. The cases are presented with black ($\omega = \omega_1$), red ($\omega = \omega_2$), green ($\omega = \omega_3$) and blue ($\omega = \omega_5$) lines respectively. The parameters of the system are $R = 12$ nm, $d = 2$ nm.
Appendix C: The non-nearest-neighbor coupling strengths and robustness

Figure C1 shows the results of the non-nearest-neighbor coherent and incoherent coupling strengths between the molecules. The radii of spheres is $R = 12$ nm, and the distance between two adjacent spheres is $d = 2$ nm.

The six lines in the left column of figure C1 represent the non-nearest-neighbor coherent couplings $g_{13}/\gamma_{11}$, $g_{24}/\gamma_{11}$, $g_{14}/\gamma_{11}$, $g_{25}/\gamma_{11}$, $g_{15}/\gamma_{11}$, and $g_{16}/\gamma_{11}$ respectively, and the right column describes the non-nearest-neighbor incoherent couplings.

In figure C2, we show the random gap distance effect with the case of $N = 5$, $R = 12$ nm and $d = 2$ nm, in which the solid curves are the same with figure B1 and the dashed and dotted curves represent the calculations with the random separation distances $2.0 \pm 0.2$ nm and $2.0 \pm 0.4$ nm respectively.

Figure C3 shows the nonlocal effect on the coupling strengths and decay rates. From figure C2, compared with the study without considering nonlocal effect (dashed lines), we find that the nearest-neighbor couplings and dissipations only have blue shifts and have no obvious change in values if we take the nonlocal effect into account. It means that the nonlocal effect affects dissipations and couplings among molecules little, and we can obtain the similar quantum speedup behavior with the blue shift of the molecular frequencies.
Figure C2. Random gap distance effect. The parameters of the system are $N = 5$, $R = 12$ nm, $d = 2$ nm, the dielectric function of Ag spheres is taken as Drude model ($\omega_p = 9.01$ eV, $\gamma = 0.05$ eV). The dashed and dotted curves represent the calculated coupling parameters with the random separation distances $2.0 \pm 0.2$ nm and $2.0 \pm 0.4$ nm respectively under the same system parameters with figure B1.

Figure C3. Solid and dashed lines represent local and nonlocal (NL) results respectively: (a) The nearest-neighbor coherent coupling strengths $g_{12}/\gamma_{11}$ (black lines), $g_{23}/\gamma_{22}$ (red lines) and $g_{34}/\gamma_{33}$ (blue lines). (b) The decay rates of single molecule $\gamma_{11}$ (black lines), $\gamma_{22}$ (red lines) and $\gamma_{33}$ (blue lines). (c) The nearest-neighbor incoherent coupling strengths $g_{12}/\gamma_{11}$ (black lines), $g_{23}/\gamma_{22}$ (red lines) and $g_{34}/\gamma_{33}$ (blue lines). The number of molecules is 6. The parameters of the system are $R = 12$ nm, $d = 2$ nm, the local dielectric function of Ag spheres is taken as Drude model ($\omega_p = 9.01$ eV, $\gamma = 0.05$ eV). For the nonlocal parameters, the Fermi velocity is taken as $V_F = 1.39 \times 10^6$ m s$^{-1}$, and the transverse response is $\varepsilon(\omega) = \varepsilon_{\infty} + \sigma(\omega)/i\omega$, where $\varepsilon_{\infty} = 1.0$, Drude conductivity is $\sigma(\omega) = i\omega\sigma_0/\omega + i\eta$ with plasma frequency $\omega_p = 9.01$ eV, $\eta = 0.05$ eV.
ORCID iDs
Tian Chen e https://orcid.org/0000-0003-1655-5249
Xiangdong Zhang e https://orcid.org/0000-0002-7725-8814

References
[1] Motwani R and Raghavan P 1995 Randomized Algorithms (Cambridge: Cambridge University Press)
[2] Aharonov Y, Davidovich L and Zagury N 1993 Quantum random walks Phys. Rev. A 48 1667–90
[3] Du J, Li H, Xu X, Shi M, Wu J, Zhou X and Han R 2003 Experimental implementation of the quantum random-walk algorithm Phys. Rev. A 67 042316
[4] Lu D et al 2016 Chiral quantum walks Phys. Rev. A 93 042302
[5] Kempe J 2003 Quantum random walks: an introductory overview Contemp. Phys. 44 307–27
[6] Venegas-Andraca SE 2012 Quantum walks: a comprehensive review Quantum Inf. Process. 11 1015–106
[7] Underwood MS and Feder DL 2010 Universal quantum computation by discontinuous quantum walk Phys. Rev. A 82 042304
[8] Chen T and Zhang X 2016 Extraordinary behaviors in a two-dimensional decoherent alternative quantum walk Phys. Rev. A 94 012316
[9] Chen T and Zhang X 2016 The defect-induced localization in many positions of the quantum random walk Sci. Rep. 6 25767
[10] Solenov D and Fedichkin L 2006 Continuous-time quantum walks on a cycle graph Phys. Rev. A 73 1–5
[11] Farhi E and Gutmann S 1998 Quantum computation and decision trees Phys. Rev. A 58 915–28
[12] Perets HB, Lahini Y, Pozzi F, Sorel M, Morandotti R and Silberberg Y 2008 Realization of quantum walks with negligible decoherence in waveguide lattices Phys. Rev. Lett. 100 170506
[13] Mülken O, Bierbaum V and Blumen A 2006 Coherent exciton transport in dendrimers and continuous-time quantum walks J. Chem. Phys. 124 124905
[14] Schreiber A, Cassemiro KN, Potoček V, Gabris A, Mosley PJ, Andersson E, Jex I and Silberhorn C 2010 Photons walking the line: a quantum walk with adjustable coin operations Phys. Rev. Lett. 104 050502
[15] Manouchehri K and Wang J 2014 Physical Implementation of Quantum Walks (Berlin: Springer)
[16] Wang B, Chen T and Zhang X 2018 Experimental observation of topologically protected bound states with vanishing Chern numbers in a two-dimensional quantum walk Phys. Rev. Lett. 121 100503
[17] Chen C et al 2018 Observation of topologically protected edge states in a photonic two-dimensional quantum walk Phys. Rev. Lett. 121 100502
[18] Engel GS, Calhoun TR, Read EL, Ahn TK, Mančal T, Cheng YC, Blankenship RE and Fleming GR 2007 Evidence for wavelike energy transfer through quantum coherence in photosynthetic systems Nature 446 782–6
[19] Thyrrhaug E, Tempelhar R, Alcocer MJP, Zalek K, Bina D, Knoester J, Jansen TLC and Zigmantas D 2018 Identification and characterization of diverse coherences in the Fenna–Matthews–Olson complex Nat. Chem. 10 780–6
[20] Mohseni M, Rebentrost P, Lloyd S and Aspuru-Guzik A 2008 Environment-assisted quantum walks in photosynthetic energy transfer J. Chem. Phys. 129 174106
[21] Fassioli F and Olaya-Castro A 2010 Distribution of entanglement in light-harvesting complexes and their quantum efficiency New J. Phys. 12 085006
[22] Hoyer S, Sarovar M and Birgitta Whaley K 2010 Limits of quantum speedup in photosynthetic light harvesting New J. Phys. 12 065041
[23] Laucht A, Villas-Boas J M, Stobbe S, Hauke N, Hofbauer F, Böhm G, Lodahl P, Amann M C, Kaniber M and Finley J 2010 Mutual coupling of two semiconductor quantum dots via an optical nanocavity Phys. Rev. B 82 1–6
[24] Shalabney A, George J, Hutchinson J, Papillo G, Genet C and Ebbesen T W 2018 Identification and characterization of diverse coherences in the Fenna–Matthews–Olson complex Nat. Chem. 10 780–6
[25] Mohseni M, Rebentrost P, Lloyd S and Aspuru-Guzik A 2008 Environment-assisted quantum walks in photosynthetic energy transfer J. Phys. Chem. A 129 174106
[26] Fassioli F and Olaya-Castro A 2010 Distribution of entanglement in light-harvesting complexes and their quantum efficiency New J. Phys. 12 085006
[27] Hoyer S, Sarovar M and Birgitta Whaley K 2010 Limits of quantum speedup in photosynthetic light harvesting New J. Phys. 12 065041
[28] Laucht A, Villas-Boas J M, Stobbe S, Hauke N, Hofbauer F, Böhm G, Lodahl P, Amann M C, Kaniber M and Finley J 2010 Mutual coupling of two semiconductor quantum dots via an optical nanocavity Phys. Rev. B 82 1–6
[29] Shalabney A, George J, Hutchinson J, Papillo G, Genet C and Ebbesen T W 2018 Coherent coupling of molecular resonators with a nanocavity mode Nat. Commun. 9 5981
[30] Gallardo E et al 2010 Optical coupling of two distant InAs/GaAs quantum dots by a photonic-crystal microcavity Phys. Rev. B 81 2–5
[31] Ren J, Chen T, Wang B and Zhang X 2017 Ultrafast coherent energy transfer with high efficiency based on plasmonic nanostructures J. Chem. Phys. 146 144101
[32] Ren J, Wu T, Yang B and Zhang X 2016 Simultaneously giant enhancement of Förster resonance energy transfer rate and efficiency based on plasmonic excitations Phys. Rev. B 94 125416
[33] Wang B et al 2014 Experimental observation of giant chiroptical amplification of small chiral molecules by gold nanosphere clusters J. Phys. Chem. C 118 9090–5
[34] Ren J, Wu T and Zhang X 2015 Multifrequency multi-qubit entanglement based on plasmonic hot spots Sci. Rep. 5 13941
[35] Tang H et al 2018 Experimental two-dimensional quantum walk on a photonic chip Sci. Adv. 4 eaat3174
[36] Li J F et al 2010 Shell-isolated nanoparticle-enhanced Raman spectroscopy Nature 494 392–3
[37] Asapu R et al 2017 Plasmonic near-field localization of silver core–shell nanoparticle assemblies via wet chemistry nanogap engineering ACS Appl. Mater. Interfaces 9 41577–85
[38] Lin P-Y, Le G-Y, Chiu W-J, Jian R-S and Lu C-J 2019 A single light spot GC detector employing localized surface plasmon resonance of porous Au/SiO2 nanoparticle multilayer Analyst 144 698–706
[39] Ambainis A, Kempe J and Rivosh A 2003 Coins make quantum walks faster Proc. 16th ACM-SIAM Symp. Discrete Algorithms (Society for Industrial and Applied Mathematics, Philadelphia, PA) pp 1099–108
[40] Childs A M and Goldstone J 2004 Spatial search by quantum walk Phys. Rev. A 70 022314
[41] Meyer D A and Wong T G 2015 Connectivity is a poor indicator of fast quantum search Phys. Rev. Lett. 114 110503
[42] Dzsotjan D, Sørensen A S and Fleischhauer M 2010 Quantum emitters coupled to surface plasmons of a nanowire Phys. Rev. B 82 075427
[43] Asenojo-Garcia A, Moreno-Cardoner M, Albrecht A, Kimble H J and Chang D E 2017 Exponential improvement in photon storage fidelities using subradiance and ‘Selective Radiance’ in atomic arrays Phys. Rev. X 7 031024
[44] González-Tudela A, Martín-Cano D, Moreno E, Martín-Moreno L, Tejedor C and García-Vidal F J 2011 Entanglement of two qubits mediated by one-dimensional plasmonic waveguides Phys. Rev. Lett. 106 023021
[45] González-Tudela A, Ruiz-Moreno L, Martín-Cano D, García-Vidal F J 2013 Theory of strong coupling between quantum emitters and propagating surface plasmons Phys. Rev. Lett. 110 126801
[46] Dzsotjan D, Kastel J and Fleischhauer M 2011 Dipole–dipole shift of quantum emitters coupled to surface plasmons of a nanowire Phys. Rev. B 84 075419
[42] Palik E. D. 1985 *Handbook of Optical Constants of Solids* (USA: Academic Press)
[43] Gonzaga-Galeana J A and Zurita-Sánchez J R 2013 A revisitation of the Förster energy transfer near a metallic spherical nanoparticle: (1) efficiency enhancement or reduction? (2) The control of the Förster radius of the unbounded medium. (3) The impact of the local density of states *J. Chem. Phys.* 139 244302
[44] Christensen T, Yan W, Raza S, Jauho A P, Mortensen N A and Wubs M 2014 Nonlocal response of metallic nanospheres probed by light, electrons, and atoms *ACS Nano* 8 1745–58
[45] Mortensen N A, Raza S, Wubs M, Søndergaard T and Bozhevolnyi S I 2014 A generalized non-local optical response theory for plasmonic nanostructures *Nat. Commun.* 5 3809
[46] Scholl J A, Koh A L and Dionne J A 2012 Quantum plasmon resonances of individual metallic nanoparticles *Nature* 483 421–7
[47] David C and García de Abajo F J 2011 Spatial nonlocality in the optical response of metal nanoparticles *J. Phys. Chem. C* 115 19470–5
[48] Tserkezis C, Mortensen N A and Wubs M 2017 How nonlocal damping reduces plasmon-enhanced fluorescence in ultranarrow gaps *Phys. Rev. B* 96 085413
[49] Tai C-T 1993 *Dyadic Green Functions in Electromagnetic Theory* (Piscataway, NJ: IEEE)
[50] Doicu A, Wriedt T and Eremin Y A 2006 *Light Scattering by Systems of Particles* (Berlin: Springer)
[51] Mishchenko M I, Travis L D and Mackowski D W 1998 T-matrix computations of light scattering by nonspherical particles: a review *J. Quant. Spectrosc. Radiat. Transfer* 55 535–75
[52] Mishchenko M I and Travis L D 1998 Capabilities and limitations of a current FORTRAN implementation of the T-matrix method for randomly oriented, rotationally symmetric scatterers *J. Quant. Spectrosc. Radiat. Transfer* 60 309–24