Supplementary Information for 'Decision Maker Based on Nanoscale Photo-excitation Transfer'

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1 Excitation Transfer Mediated by Optical Near-field Interactions

The interaction Hamiltonian between an electron and an electric field is given by

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

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 an 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 because the electric field of propagating light is considered to be constant on the nanometer scale. Therefore, one can derive optical selection rules by calculating the transfer matrix of an electric dipole. Consequently, in the case of cubic quantum dots, transitions to states described by quantum numbers containing an even number are prohibited. On the other hand, in the case of optical near-field interactions, there is a steep electric gradient of optical near-fields in the vicinity of nanometer-scale matter, and this facilitates optical transitions that violate conventional optical selection rules. Such an energy transfer to a conventionally dipole-forbidden energy level cannot be explained by carrier tunnelling [1, 2, 3] or dipole interactions [4, 5].

Optical excitations in nanostructures, such as quantum dots, can be transferred to neighbouring structures via optical near-field interactions [6]. Assume that two cubic quantum dots $\mathbf{QD}_S$ and $\mathbf{QD}_M$, whose side lengths are a and $\sqrt{2}a$, respectively, are located close to each other [Fig. 1(a)]. Suppose that the energy eigenvalues for the quantised exciton energy level specified by quantum numbers (n_x, n_y, n_z) in the QD with side length a ($\mathbf{QD}_S$) are given by

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

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 $(1, 1, 1)$ for $\mathbf{QD}_S$ [denoted as S_1 in Fig. 1(a)] and $(2, 1, 1)$ for $\mathbf{QD}_M$ (M_2). An optical near-field interaction, denoted as $U_{S_1M_2}$, results from the steep gradient of the electric field in the vicinity of $\mathbf{QD}_S$. It is known that the inter-dot optical near-field interaction is given by a Yukawa-type potential denoted by

$$U(r) = Aexp(-r/a)/r, \tag{3}$$

where r is the inter-dot distance, and A and a are constants [6]. Therefore, excitons in $\mathbf{QD}_S$ can move to the $(2, 1, 1)$-level in $\mathbf{QD}_M$. Note that an optical excitation of

the $(2,1,1)$-level in $\mathbf{QD}_M$ corresponds to an electric dipole-forbidden transition. However, the optical near-field allows this level to be populated because of the steep electric field in the vicinity of the $\mathbf{QD}_S$. In $\mathbf{QD}_M$, the exciton sees a sublevel energy relaxation, denoted by Γ, which is faster than the near-field interaction, and so the exciton goes to the $(1,1,1)$-level of $\mathbf{QD}_M$ (M_1).

In the optical excitation transfer discussed above, the energy dissipation occurring in the destination quantum dot, $\mathbf{QD}_M$, determines the uni-directionality of signal transfer [7]. Therefore, when the lower energy level of the destination quantum dot is filled with another excitation (called "state-filling"), the optical excitation occurring in a smaller QD cannot move to a larger one. As a result, the optical excitation will go back and forth between these dots (optical nutation).

This suggests that the flow of optical excitation can be controlled differently in systems composed of multiple quantum dots by inducing state-filling therein. For example, Fig. 1(b) schematically represents a system consisting of five QDs denoted as $\mathbf{QD}_{LL}$, $\mathbf{QD}_{ML}$, $\mathbf{QD}_S$, $\mathbf{QD}_{MR}$ and $\mathbf{QD}_{LR}$. The sizes of $\mathbf{QD}_S$, $\mathbf{QD}_{Mi}$ $(i = L, R)$ and $\mathbf{QD}_{Li}$ $(i = L, R)$ are respectively given by a, $\sqrt{2}a$ and $2a$. The energy levels in the system are summarised as follows.

Energy Level	QD	Designation
(1,1,1)	$\mathbf{QD}_{ML}$	ML_1
	$\mathbf{QD}_{MR}$	MR_1
	$\mathbf{QD}_{LL}$	LL_1
	$\mathbf{QD}_{LR}$	LR_1
(2,1,1)	$\mathbf{QD}_{ML}$	ML_2
	$\mathbf{QD}_{MR}$	MR_2
	$\mathbf{QD}_{LL}$	LL_2
	$\mathbf{QD}_{LR}$	LR_2
(2,2,2)	$\mathbf{QD}_{LL}$	LL_3
	$\mathbf{QD}_{LR}$	LR_3

In such a system, the optical near-field interactions, shown schematically in Fig. 1(b), are as follows $(i = L, R)$.

$U_{S_1 Mi_2}$	Between $(1,1,1)$-level of $\mathbf{QD}_S$ (S_1) and $(2,1,1)$-level of $\mathbf{QD}_{Mi}$ (Mi_2)
$U_{S_1 Li_3}$	Between $(1,1,1)$-level of $\mathbf{QD}_S$ (S_1) and $(2,2,2)$-level of $\mathbf{QD}_{Li}$ (Li_3)
$U_{Mi_1 Li_2}$	Between $(1,1,1)$-level of $\mathbf{QD}_{Mi}$ (Mi_1) and $(2,1,1)$-level of $\mathbf{QD}_{Li}$ (Li_2)
$U_{Mi_2 Li_3}$	Between $(2,1,1)$-level of $\mathbf{QD}_{Mi}$ (Mi_2) and $(2,2,2)$-level of $\mathbf{QD}_{Li}$ (Li_3)

Through these interactions, an optical excitation generated at $\mathbf{QD}_S$ is transferred to the lowest energy levels in the largest-size QD, namely the energy level

LL_1 or LR_1. However, when LL_1 and LR_1 are occupied by other excitations, the input excitation generated at S_1 should be relaxed from the middle-sized QD, that is, ML_1 and MR_1. The idea of the QD-based decision maker is to induce state-filling at LL_1 and/or LR_1 while observing radiations from ML_1 and MR_1. In Sec. 2, we describe the detailed dynamics of such a system.

2 Dynamics of Excitation Transfer in a Five Quantum Dot System

Here we analyse the dynamic behaviour of the system based on a density matrix formalism assuming optical near-field interactions between these five dots. For simplicity, we assume one excitation system. There are in total 11 energy levels in the system: S_1 in $\mathbf{QD}_S$; ML_1 and ML_2 in $\mathbf{QD}_{ML}$; LL_1, LL_2 and LL_3 in $\mathbf{QD}_{LL}$; MR_1 and MR_2 in $\mathbf{QD}_{MR}$; LR_1, LR_2 and LR_3 in $\mathbf{QD}_{LR}$. Therefore, the number of different states occupying these energy levels is 12 including the vacancy state, as schematically shown in Fig. 1(c).

The optical near-field interaction between energy levels E_1 and E_2 is represented by $U_{E_1E_2}$; for instance, the interaction between the $(1,1,1)$-level of $\mathbf{QD}_S$ (S_1) and the $(2,1,1)$-level of $\mathbf{QD}_{ML}$ (ML_2) is denoted by $U_{S_1ML_2}$. The radiative relaxation rates from S_1, Mi_1, and Li_1 are given by γ_{S_1}, γ_{Mi_1} and γ_{Li_1}, respectively. The quantum master equation of the total system is then given by [8]

$$\begin{aligned}\frac{d\rho(t)}{dt} &= -\frac{\mathrm{i}}{\hbar}[H_{int},\rho(t)] \\ &+ \sum_{i=S_1,ML_1,MR_1,LL_1,LR_1} \frac{\gamma_i}{2}\Big(2R_i\rho(t)R_i^\dagger - R_i^\dagger R_i\rho(t) - \rho(t)R_i^\dagger R_i\Big) \\ &+ \sum_{i=ML_2,LL_3,LL_2,MR_2,LR_3,LR_2} \frac{\Gamma_i}{2}\Big(2S_i\rho(t)S_i^\dagger - S_i^\dagger S_i\rho(t) - \rho(t)S_i^\dagger S_i\Big),\end{aligned} \tag{4}$$

where the interaction Hamiltonian is given by H_{int}. Let the (i,i) and (j,j) elements of $\rho(t)$ be the probabilities of the two states that are transformable between each other via an optical near-field interaction denoted by U_{ij}. Then, the (i,j) and (j,i) elements of the interaction Hamiltonian are given by U_{ij}. The matrices R_i ($i = S_1$, ML_1, MR_1, LL_1, LR_1) are annihilation operators that respectively annihilate excitations in S_1, ML_1, MR_1, LL_1 and LR_1 via radiative relaxations. The matrices $R_i^\dagger$ ($i = S_1$, ML_1, MR_1, LL_1, LR_1) are creation operators given by the transposes of the matrices of R_i. The radiative decay times $\gamma_{LL_1}^{-1} = \gamma_{LR_1}^{-1} = 1$ ns, $\gamma_{ML_1}^{-1} = \gamma_{MR_1}^{-1} = 2^{\frac{3}{2}} \times 1 \sim 2.83$ ns and $\gamma_{S_1}^{-1} = 2^3 \times 1 \sim 8$ ns are inversely proportional to the volumes of the $\mathbf{QD}_S$ [9]. The matrices S_i ($i = ML_2, LL_3, LL_2, MR_2, LR_3, LR_2$) are annihilation

operators that respectively annihilate excitations in ML_2, LL_3, LL_2, MR_2, LR_3 and LR_2 via sublevel relaxations Γ_i and populate excitations in the corresponding lower energy levels. The diagram shown in Fig. 1(c) schematically summarises the state transitions.

When there are no state-filling induced at LL_1 and LR_1, the sublevel relaxations are assumed to be equally fast, $\Gamma_i = 10$ ps. On the other hand, when control lights are irradiated to induce state-filling at LL_1 and/or LR_1, we have to consider a multi-excitation system in order to calculate the exact dynamics of the system. Another way of describing the effect of state-filling is that the sublevel relaxation time increases, for example by a factor of 10, that is, $\Gamma_{LL_2}^{-1} = \Gamma_{LR_2}^{-1} = 10 \times 10$ ps. The validity of such an approach has been verified in Ref. [10].

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