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Optically Controlled Energy Transfer between Two Molecules Near a MoS_2 Nanodisk †

Natalia Domenikou [1], Ioannis Thanopulos [1], Vasilios Karanikolas [2] and Emmanuel Paspalakis [1,*]

[1] Materials Science Department, School of Natural Science, University of Patras, 26504 Patras, Greece; domenikou.n@gmail.com (N.D.); ithano@upatras.gr (I.T.)
[2] International Center for Young Scientists (ICYS), National Institute for Materials Science (NIMS) 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan; KARANIKOLAS.Vasileios@nims.go.jp
* Correspondence: paspalak@upatras.gr
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Abstract: We present a theoretical investigation of the effects of quantum coherence on the energy transfer efficiency between a donor and an acceptor molecule located at opposite sides of a MoS_2 nanodisk. We use the Lindblad master equation, incorporating the electromagnetic Green's function approach, taking into consideration the dissipation in the molecules. We focus on the transfer dynamics of the two molecules with and without quantum coherence by varying different parameters of our system. The presence of the MoS_2 nanodisk enhances the relaxation rate of the donor, as well as the coherent and the incoherent terms of the coupling coefficients between the molecules, thus affecting their dynamic response. Our results show high energy transfer efficiency between the two molecules in the presence of strong coherence and for small separation distances from the MoS_2 nanodisk. Also, for specific parameters, when quantum coherence is accounted for, we observe ultrafast and largely improved energy transfer process compared to the case without coherence.

Keywords: energy transfer; two-level quantum systems; transition metal dichalcogenide nanostructures; molybdenum disulfide nanodisk

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1. Introduction

Recent studies have shown that transition metal dichalcogenides (TMD) 2D semiconducting materials are utilized for efficient control of the light-matter interactions at the nanoscale [1]. This study considers a 2D MoS_2 nanodisk as the photonic element to facilitate long range interactions and energy transfer between a donor and an acceptor molecule. Up to now the MoS_2 nanodisk geometry has been used to present an enhanced radiative emission of a nearby placed quantum system (QS), which can be used for a light emitting device [2]. Moreover, at short QS-MoS_2 nanodisk separation distances, the interaction strength between them lies in the strong coupling regime, where reversible dynamics is observed in the population probability of the excited state of the QS [3]. In this work, we extent the previous studies to investigate the energy transfer rate between a pair of molecular QSs interacting through the MoS_2 nanodisk. The MoS_2 nanodisk supports two sets of localized excitons modes at the visible part of the electromagnetic spectrum, that can be excited by the two different transition dipole moment of the QSs.

Here, we use the density matrix theory, where the Purcell factor, of the single molecule, and the coupling terms, between the pair of molecules, are calculated through the electromagnetic Green's tensor theory, taking into consideration the dissipation in the donor and the charge separation rate [4–6]. We study the transfer dynamics of the molecules in the absence and presence of quantum coherence by varying various parameters that affect our system, such as the dipole moment orientations and the distance between the

molecules and the surface of the MoS_2 nanodisk. The energy transfer efficiency n and the transfer time t_f are introduced to quantify the energy transfer process [6]. We find that highly efficient and ultrafast energy transfer can be achieved between the donor and the acceptor near the MoS_2 nanodisk with coherent couplings for specific dipole moment orientations and suitable distances between the molecules and the MoS_2 nanodisk. Also, we show that this process is highly efficient when strong coherence is combined with weak relaxation rate of the donor molecule and weak incoherent coupling.

2. Theoretical Model

We consider a hybrid system which contains two identical molecules, a donor and an acceptor, modeled as two-level QSs, which they interact with a two-dimensional MoS_2 nanodisk. Each molecule fulfills the point-dipole approximation and is located at equal distance d away from the center of the MoS_2 nanodisk, on opposite sides, as depicted in Figure 1. We focus on a MoS_2 disk with radius R = 30 nm.

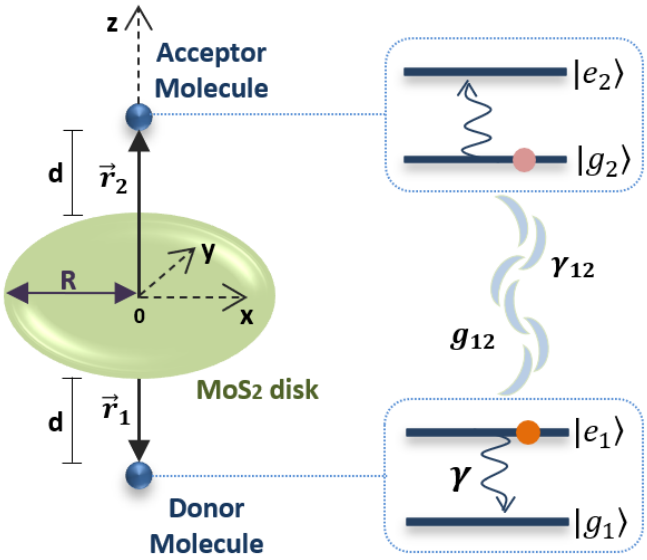

Figure 1. Schematic representation of the donor and acceptor molecules, modeled as two-level quantum systems, at opposite distances d of the MoS_2 nanodisk with radius R.

The energy transfer dynamics of the two molecules, in this quantum open system, is obtained by using the Lindblad master equation considering nonzero dissipation in the donor, Γ, and charge separation rate, κ [6]. We set different transition dipole moment orientations, either perpendicular (z axis) or parallel (x axis) to the surface of the MoS_2 nanodisk. The presence of the MoS_2 nanodisk influences the relaxation process of the donor molecule as well as the coherent and incoherent coupling terms, which are calculated via the electromagnetic Green's tensor technique [2,3]. For small distance d, these three parameters are enhanced due to the interaction of the molecules with the localized exciton polariton resonances of the MoS_2 nanodisk at specific energies, at which the Purcell factor of the molecules takes its highest value. We use coherent energy transfer theory, solve the derived master equation and obtain the energy transfer efficiency n and the transfer time t_f [6].

3. Results and Discussion

We present numerically obtained results from the analytical expressions of n and t_f as a function of the transition energy with and without quantum coherence for two dipole orientations and for distances d = 15 nm, 10 nm, and 5 nm, as depicted in the inset of the panels with $\Gamma = 20\ ns^{-1}$ and $\kappa = 4\ ps^{-1}$.

In Figure 2, we observe that in the absence of the coherent couplings in both dipole moment orientations, as d decreases the efficiency of energy transfer increases but remain under 40%. New peaks arise as the molecules approach the MoS_2 nanodisk at the two energy ranges that the localized exciton modes emerge, namely 1.91–2 eV and 2.1–2.3 eV. On the other hand, the presence of the quantum coherence modifies the shape of n. We obtain high efficiency energy transfer, above 80% for the shortest distance, for energies away from the exciton energies of the MoS_2 nanodisk. Also, strong peaks of highly efficient transfer appear between 1.9 eV and 2 eV. In addition, better energy transfer among the molecules is achieved in the case of perpendicular dipole orientations, compared with the parallel ones, due to the presence of the MoS_2 nanodisk which gives an enhanced Purcell factor along this orientation.

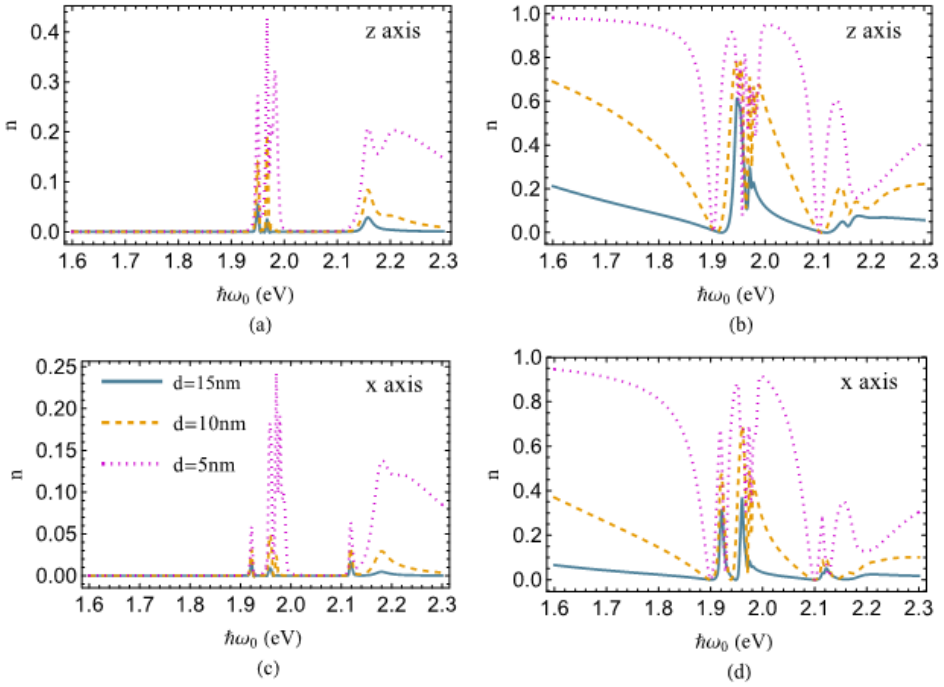

Figure 2. The energy transfer efficiency n as function of the transition energy for different molecules/MoS_2 separation distances d (**a**), (**c**) without and (**b**), (**d**) with coherent couplings. The transition dipole moment orientation of the molecules is shown in the panels.

In Figure 3, in which the energy transfer time is presented, we find that the transfer time is reduced as the separation distance d decreases and more peaks in the plots appears, similar to Figure 2. For nonzero coherent coupling terms, the energy transfer time t_f significantly decreases sharply in almost all energy range for distances below 10 nm. In this case, the energy transfer time with coherence is almost two orders of magnitude smaller compared with the case without coherence. For d = 10 nm and above, we observe that the presence of the coherence has little effect on the energy transfer dynamical response of the molecules and the transfer time is almost the same. Again, for dipole orientations along the z axis, we obtain smaller transfer times.

4. Summary

In summary, in this work, we have studied the influence of quantum coherence in the energy transfer process from a donor to an acceptor molecule which are located at opposite sites of a MoS_2 nanodisk. The two molecules are considered as two-level quantum systems. We study the energy transfer dynamical response with and without coherence either for perpendicular or for parallel molecular dipole moment orientations with respect to the surface of the MoS_2 nanodisk for different separation distances. We use

coherent energy transfer theory which is combined with an electromagnetic Green's tensor approach. We observe that very close to the MoS_2 nanodisk the relaxation of the donor molecule, the coherent and incoherent coupling terms are enhanced affecting the dynamical behavior of the system. We obtain highly efficient and ultrafast, in the scale of picosecond, energy transfer between the molecules for strong coherent coupling terms, weak incoherent coupling terms and weak decay rates of the donor molecule as well as for small separation distances. The results are expected to have a positive impact on the coherent techniques of energy transfer for the design of new, improved, and useful harvesting light devices in nanotechnology.

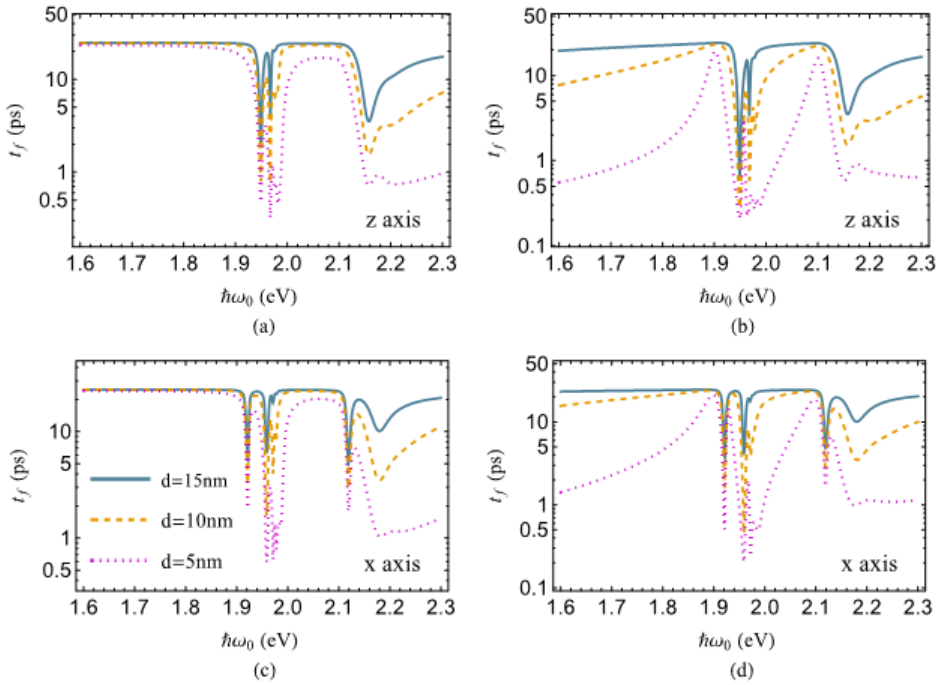

Figure 3. The energy transfer time t_f as function of the transition energy for different molecules/MoS_2 separation distances d (**a**), (**c**) without and (**b**), (**d**) with coherent couplings. The transition dipole moment orientation of the molecules is shown in the panels.

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