Inhomogeneous light quantum dot solar cell

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A new model of quantum dot solar cell based on inhomogeneous intensity of light has been studied theoretically. In symmetrically structured system of conductor-quantum dot-conductor configuration, the inhomogeneous light provides asymmetric potential creating polarization of electron number distribution in the neighboring quantum dots and furthermore gives rise to net current. Current density and efficiency of such kind solar cells are estimated to be comparable to the traditional p-n type material based solar cells. Motion of electron is described with quantum master equation at room temperature. The model of inhomogeneous field solar cell has potential applications for the gain of more economical photocurrent.

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Photovoltaic effect is one of the important ways to obtain green energy, which highlights the researches and development of solar cells. Solar cell technologies are classified into three generations[1]. First generation solar cells are based on crystalline wafer of silicon[2, 3]. They have advantage of higher conversion efficiency compared to the other types of cells, but have disadvantage of high costs at the same time[4]. Second generation solar cells are made from layers only a few micrometers thick films which is much thinner than crystalline silicon based cells[5, 6]. Therefore, they are also called thin film solar cells. The advantages of thin film cells are integrable, flexible and economical, however, they have drawback like the feature of low efficiency or toxicity[7]. Third generation solar cells do not rely on traditional p-n junctions, they are made from new developed sensitizing materials, such as dye molecules[8, 9], quantum dots (QDs)[10, 11] and organic polymers[12]. These nano materials could help us to harvest light energy at low cost and low heat emission compared with the two generation solar cells.

Actually, the third generation solar cells inherit some significant features of the first and second generation cells. For example, p-n type bilayer structure, integrability, thin film designs still play important role in recent research and development of nano material based solar cells[13, 14]. p-type and n-type materials are significant since the p-n junctions could provide potential difference to the cell system for the separation and collection of charge careers, such as electrons (negative), holes (positive)[17]. Task of light is just excite electrons into higher levels of nano crystals or molecules, then electrons and holes would be separated naturally due to the asymmetric potential of p-type and n-type bilayer.

In this paper, we proposed a new model of solar cell in which inhomogeneous light field is used to generate photocurrent in symmetrically designed conductor-QD-conductor system without the p-n type materials. In this model, p-n type bilayer with asymmetric potential in a traditional solar cell is replaced by the inhomogeneous light with asymmetric field intensity. For a definite air mass (AM), power of light transferred to the earth surface is uniform. How to obtain spatially nonuniform light field is the key of the inhomogeneous light solar cell. The model of solar cell is conceptually shown in Fig. 1. A non-transparent material is sandwiched between the QDs film and the film is separated into two parts. The non-transparent material constructs light intensity difference in the QDs film.

The characteristic of such solar cell is very simply structured. It can be made more economically since the p-type and n-type materials are neglected here. Furthermore, there is not particular toxic material is necessary for the cell. The new technology used here is just to create light intensity difference on neighboring QDs in the film.

In the following, based on a one dimensional neighboring double QDs system, the solar cell model would be quantitatively described. Motion of electrons in the double-QD opened system under external light is illustrated in Fig. 2 (a)-(d). The double-QD system is coupled to left (L) and right (R) electrodes with the same Fermi level $\varepsilon_F$. As both of the two electrodes are conductors, $\varepsilon_F > \varepsilon_C, \varepsilon_V$, where $\varepsilon_C$ is the bottom level of conductance band and $\varepsilon_V$ is the top level of valence band. The ground and excited levels of the QDs are denoted by $\varepsilon_1$ and $\varepsilon_2$, respectively. They are required to satisfy $\varepsilon_1 < \varepsilon_F < \varepsilon_2$ here as shown in Fig. 2. Intensities of

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to be insensitive to the electronic states for the left and right electrodes. Sunlight excitation can be derived under the assumption that coupled to external light could be described in the Hamiltonian and initial condition.

The first part of (2) describes energy of free electrons in QDs. The second and third term indicate the exchange of electrons between QDs and electrodes. The Lindblad super operators acting on the density matrix $\rho$ can be written as $L_{\rho} = \frac{1}{2} \sum \{ f(\varepsilon_s) c_{LS}^\dagger \rho c_{LS} - c_{LS}^\dagger c_{LS} \rho + (1 - f(\varepsilon_s)) (c_{LS}^\dagger \rho c_{LS} - c_{LS}^\dagger c_{LS} \rho) + H.c. \}$ and $L_{\rho} = \frac{1}{2} \sum \{ f(\varepsilon_s) (c_{RS}^\dagger \rho c_{RS} - c_{RS}^\dagger c_{RS} \rho) + (1 - f(\varepsilon_s)) (c_{RS}^\dagger \rho c_{RS} - c_{RS}^\dagger c_{RS} \rho) + H.c. \}$. Single electron tunneling rate between a dot and an electrode is given by the energy independent coefficient $\Gamma = \frac{1}{2} | g |^2 N D(\varepsilon_s)$ under adiabatic approximation ($D(\varepsilon_s)$ is density of states of electrons in any electrode at energy level $\varepsilon_s$). Mean occupation number of the single-electron state with level $\varepsilon_s$ in any electrode is given by the Fermi-Dirac distribution function $f(\varepsilon_s) = \frac{1}{1 + e^{(\varepsilon_s - \varepsilon_F)/k_B T} + 1}$ at equilibrium temperature $T$ and Fermi level $\varepsilon_F$.

Considering single-electron transit in a QD, three states would be involved in each dot, empty state $|0\rangle$, occupation of a ground state electron $|1\rangle$, occupation of an excited state electron $|2\rangle$. Mean occupation number of the single-electron state $\langle s | \rho \rangle$ can be defined as $P_{s0} = \text{tr} \{ c_{s0}^\dagger c_{s0} \rho \}$, $P_{s1} = \text{tr} \{ c_{s1}^\dagger c_{s0} \rho \}$, $P_{s2} = \text{tr} \{ c_{s2}^\dagger c_{s0} \rho \}$, respectively. They are determined by the equation of motion (3). tr here represents trace over the 9 double-QD states $\{ |s\rangle_L |r\rangle_R \} \ (s, r = 0, 1, 2)$.

Defining the total occupation number of electrons in the double-QD at time $t$ by $N(t)$ and $n_{s0}(t)$, current in the unit component could be calculated using the continuity equation

$\frac{dN(t)}{dt} = j_L - j_R.$

where $j_L$ and $j_R$ denotes left and right sides currents of the double-QD system. According to Kirchhoff’s Laws, left and right currents should satisfy $j_L - j_R = 0$ in stationary circuit. Therefore, current through system can be written as $j = (j_L + j_R) / 2$. Here, direction of the positive current is defined to be from left to right. Substituting Eq. (3) into the continuity equation (4), expressions of the current $j$ can be reached in the form

$j = \frac{e \Gamma}{2} \sum_s \{ f(\varepsilon_s) (P_{s0} - P_{sR}) - (1 - f(\varepsilon_s)) (P_{sL} - P_{sR}) \}.$

Eq. (5) indicates that net current $j$ in the cell is directly proportional to the polarization (the difference of mean occupation number) of the electron number distribution in the neighboring QDs. To understand the current formula (4), we may write it in the conceptual relation as

$\frac{\partial \rho}{\partial t} = -i \hbar [V_{dot}, \rho] + L_{LP} \rho + L_{RP} \rho,$
input light intensity states in left and right QD are shown as a function of \( \lambda \). This work is \( \mu \phi \) a figure. In resonant absorption (\( \Delta = 0 \)), \( \tau \) the double-QD system and \( \Delta \) larger than that in the right QD, namely \( \mu_1 \) left QD is increasing, at the same time, light intensity in both the two QDs are fully occupied by the ground state. \( \epsilon \) for different material and dimensions. In this way, the Rabi frequency can be connected to the light intensity as \( \Omega_\alpha = \frac{\| \alpha \| E_\alpha}{\hbar} \) for \( \alpha = L \) and \( \alpha = R \). According to electromagnetic field theory, light intensity \( I_\alpha \) in dot \( \alpha \) is proportional to square of electric field amplitude \( E_\alpha \), namely \( I_\alpha = \frac{n}{2 \mu_0} |E_\alpha|^2 \), where \( n \) is refractive index, \( c \) is speed of light, and \( \mu_0 \) is permeability of vacuum. After sunlight travels through the earth’s atmosphere to the earth’s surface, it’s intensity is reduced to nearly 1000 W/m² which is called 1 sun. 

Current \( J \) can be calculated associated with the solutions of Eq. \( 3 \) under the normalization constraint \( tr \rho = 1 \). The Rabi frequency \( \Omega_\alpha \) is proportional to electric field amplitude \( E_\alpha \) and the absolute value of dipole moment \( \varphi_{12} \). For the semiconductor QD, dipole moment is around \( \varphi_{12} = 30 \sim 90 \) D [24, 31] for different material and dimensions. In this way, the Rabi frequency can be connected to the light intensity as \( \Omega_\alpha = \frac{|\varphi_{12}| E_\alpha}{\hbar} \) for \( \alpha = L \) and \( \alpha = R \). According to

ej = \frac{Q}{\tau} \frac{2 \mu_0 |E_\alpha|}{n}, \quad \text{where } Q = -e \sum_j f(\varepsilon_j)(P_{L0} - P_{R0}) - \left( 1 - f(\varepsilon_j) \right) (P_{Ls} - P_{Rs}) \right) \}

\text{is the inversion charge difference in the double-QD system and } \tau = 2/\Gamma \text{ is the characteristic time of an electron transit from one electrode to the other electrode through QDs.}

The whole cell is composed of a QDs film sandwiched between two conductors as illustrated in Fig. 1. Then, current density \( J \) of the solar cell could be calculated as

\[ J = \sigma_\alpha, \]

where \( j \) represents current through the single unit component and \( \sigma \) denotes surface density of the unit component. Here, \( \sigma \) would be taken as the sheet density of QDs. The sheet QD density commonly ranged from \( 10^9 \) cm\(^{-2} \) to \( 10^{11} \) cm\(^{-2} \) which can be controlled by QD growth techniques [34, 36]. Here, a typical value of \( \sigma = 8 \times 10^9 \) cm\(^{-2} \) has been taken in the numerical treatments.

Considering the whole cell, current densities for different parameters have been plotted in Fig. 4. Fig. 4 (a) reveals the bigger light intensity difference, the larger current can be obtained for a given input light. The rates of electron tunneling \( \Gamma \) and \( \Delta \) play different roles in the system as shown in Fig. 4 (b). When \( \Gamma \) is very small, electrons are hard to exchange between electrodes and QDs, which would depress current. When \( \Gamma \) is too large, polarization of electron number distribution in the neighboring QDs hard to be constructed. As a result, currents have top values for the change of \( \Gamma \). In contrast, the inter-QD coupling strength \( \Delta \) always improves current. Although our results are achieved with the model using coherent light, the inhomogeneous field solar cell should work under sun light which has much short coherent time. It is demonstrated in Fig. 4 (c) that current in the system is insensitive to the phase \( \phi \) of light. Fig. 4 (c) also reflects the dipole moment \( \mu \) improves photocurrent due to its connection with the Rabi frequency. As manifested in the figures, current densities in the inho-
inhomogeneous field solar cell through the formula, \( P = \text{energy level difference} \times \text{related to the energy change of electron in the cell, namely} \)
could be calculated as \( P\left(\varepsilon_2 - \varepsilon_1\right) \) where the voltage \( U_e \) should be related to the energy change of electron in the cell, namely the energy level difference \( \varepsilon_2 - \varepsilon_1 \) of a QD. Therefore, we have \( U_e = \varepsilon_2 - \varepsilon_1 \). Power of light \( P_L \) on the area \( S \) is equal to \( P_L = I_L S \). From the electronic current power and light power, we may calculate the efficiency of the inhomogeneous field solar cell through the formula,
\[
\eta = \frac{P_e}{P_L} \times 100\%.
\] (7)

Next, let us try to calculate the photocurrent efficiency. Current of electron through an area \( S \) of the solar cell could be calculated as \( I_e = J S \). Corresponding power is equal to \( P_e = I_e U_e \), where the voltage \( U_e \) should be related to the energy change of electron in the cell, namely the energy level difference \( \varepsilon_2 - \varepsilon_1 \) of a QD. Therefore, we have \( U_e = \varepsilon_2 - \varepsilon_1 \). Power of light \( P_L \) on the area \( S \) is equal to \( P_L = I_L S \). From the electronic current power and light power, we may calculate the efficiency of the inhomogeneous field solar cell through the formula,
\[
\eta = \frac{P_e}{P_L} \times 100\%.
\] (7)

FIG. 5: (Color on line) (a) Light difference dependent cell efficiencies. (b) Cell efficiencies versus electron tunneling rate. (c) Dipole moment dependent cell efficiencies. In (b) and (c), \( I_L = 1000 \text{ W/m}^2 \).

mogeneous field cell can be comparable to the traditional p-n type material based solar cells [10, 11, 37, 38].

In our model, we just considered single transition energy of such kind device may further decrease the cost of photovoltaic system should be very small as manifested in the above figure. Fortunately, there are some direct methods to supplement our present model for higher current and efficiency. Firstly, practical QDs are usually characterized by quantized energy of multi levels, which reveals more than one transitions are allowed actually for the excitation of more electrons. Secondly, multiple exciton effect in QDs can increase photocurrent. In the multiple exciton process, a single photon with large energy can excite two or more electrons in a QD [39, 40]. Thirdly, density of QDs considered in our model can be raised by at least 10 or 10² times, which could increase the absorption ability of the QD films.

In conclusions, we theoretically proposed a new type QD solar cell based on intensity difference of light. It is pointed out that light with inhomogeneous intensity in neighboring QDs can lead to polarization of electron number distribution and induces net current. As a result, materials which built such solar cell can be symmetrically structured, such as conductor-QD-conductor as considered here. This configuration is substantially different from the traditional solar cells which usually rely on p-n type materials for the creation of potential difference. The polarization of electron number distribution due to the light intensity difference is at the heart of the mechanism of this cell. Considering large array of QDs on a film, current densities and cell efficiencies are estimated numerically. Preliminary results show that current density and efficiency of this kind of solar cell could be comparable with the traditional solar cells and have large flexibility to be tuned in practice. Structure simplicity of such kind device may further decrease the cost of photocurrent in future.

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