Suppressed radiative recombination rate dependent temperatures in the quantum photocell with three electron donors

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The radiative recombination of electron-hole pairs represents a great challenge of the photon-to-charge conversion efficiency in the photocell. In this paper, we investigate the radiative recombination rate (RRR) dependent temperatures in a proposed quantum photocell with three electron donors. The results show the increasing temperatures and energy gap of the donors can inhibit the RRR, while the gaps between the donors and acceptor enhance the RRR in the quantum photocell with three uncoupled donors. However, the suppressed RRR was obtained by the increasing energy gaps of the donors but the RRR was heightened by the gaps between the donors and acceptor, although the increasing temperatures incur much intense radiative recombination in the quantum photocell with three dipole-dipole coupled donors. And at room temperature, the RRR is always less than those in the three uncoupling donors case. What’s more, the RRR can be restrained to a minimum value but can’t be eliminated when the photocell system adjusted by the electrostatic dipole-dipole coupling strength $J$ at room temperature. The features of regulation performance but irremovability demonstrate the detailed balance limits and the possibility of increased efficiency in this quantum photocell, and suggest some encouraging trends for higher efficiency and deserve to prove experimentally.

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I. INTRODUCTION

The photon-to-charge conversion efficiency[1] is an important aspect of photocell. However, the photon-to-charge conversion efficiency cannot exceed the detailed balance limit[2] because the radiative upward transition to generate the excitation and its reversal, the radiative downward transition coexist simultaneously. And the radiative recombination has been considered as the fundamental limits[2] on the conversion efficiency, as has been widely accepted in these artificial light-harvesting systems. Apart from the radiative recombination, other energy loss processes still exit in the photocell, such as surface reflection, internal resistance, thermalization losses, unabsorbed photons with energy less than band gap[3]. And many of these unessential energy loss processes can be minimized by appropriately designed structures[4–8], such as multi-junction[9–11] and intermediate band photocells [12–16].

Recently, theoretical and experimental studies[17–23] have demonstrated that the quantum coherence can alter the conditions of the detailed balance between the radiative recombination and absorption, and thereby suppress radiative recombination which in turn leads to the
increasing the power conversion efficiency in photocells. One of the possible ways suggested by Scully\cite{17} is possible via the coherent drive where the quantum coherence between two levels induced by external source, can cancel the emission processes\cite{18}. As a matter of fact, the coherence can be generated by the mission and absorption of solar photons and thermal phonons without using external fields\cite{19}, which is associated with the Fano effect due to interference between several quantum-mechanical paths to the same final level. Consequently, the presence of quantum coherence of the delocalized donor states alters the conditions for the thermodynamic detailed balance, and then brings out the improvement of the efficiency of the photocell\cite{20–23}.

Considering a higher conversion efficiency currently achieved in the triple-junction photocells\cite{24, 25}, in the following, we focus on RRR dependent temperatures in a proposed quantum photocell with three electron donors, and the three electron donors were simulated to the similar three p-n junctions in the quantum photocell. And we expect to suppress radiative recombination which in turn leads to the increasing the photon-to-charge conversion efficiency in the photocell. The results indicated that RRR could be adjusted to suppression but couldn’t be eliminated, which means the breaking detailed balance but not be eliminated. And some encouraging trends associated these results deserve the further experimental investigation.

The work is organized as follows: in section 2, we describe the photocell model with three electron donors. In section 3, we present results and corresponding discussions regarding the RRR dependent temperature and possible experimental realization. A concise summary is given in the final section.

FIG. 1. Schematic diagram: quantum photocell models with the acceptor and three uncoupled donors (a) and three dipole-dipole coupled donors (b). Solar radiation drives electron transport between the valence band state $|b\rangle$ and the conduction band state $|i\rangle$ (i = 1, 2, 3) in Fig.(a). Transitions between levels $|i\rangle$ (i = 1, 2, 3) and $|\alpha\rangle$, $|\beta\rangle$ and $|b\rangle$ are driven by ambient thermal phonons. Levels $|\alpha\rangle$ and $|\beta\rangle$ are connected to a load. In (a) the three degenerate excited levels split into (b) because of the couplings between three donors, and the dark level ($|a_2\rangle$) is optically forbidden and has no electron transfer path to the acceptor $|\alpha\rangle$.

II. PHOTOCELL MODEL WITH THREE ELECTRON DONORS

Proceeding with the analysis, we consider a photocell model with the conduction band states $|i\rangle$ (i = 1, 2, 3) and the valence band state $|b\rangle$ [depicted in Fig.1(a)] as the donors. And level $|\alpha\rangle$ and level $|\beta\rangle$ connecting to a load are assumed the acceptor molecule. The excitation of a molecule is simply modeled as a two-level system with
the excited state $|i\rangle_{(i=1,2,3)}$ and the ground state $|b\rangle$. Then the excited electrons driven by solar radiation can be transferred to the acceptor molecule, the conduction reservoir state $|\alpha\rangle$, with any excess energy radiated as a phonon into the ambient thermal phonons reservoirs. The excited electron is then assumed to be used to perform work, leaving the conduction reservoir state $|\alpha\rangle$ decaying to the sub-stable state $|\beta\rangle$ at a rate $\Gamma$. The recombination between the acceptor and the donor is modeled as $\chi_1\Gamma$ in Fig.1(a). Where $\chi_1$ is the RRR, a dimensionless fraction. The recombination process brings the system back into the valence state $|b\rangle$ without producing a work current, which could be a significant source of inefficiency. Finally, the state $|\beta\rangle$ decays back to the valence state $|b\rangle$ at a rate $\Gamma_c$ and the cycle terminates. In Fig.1(a), the three donors are assumed to be identical and degenerate, and their three unoccupied excited states $|i\rangle_{(i=1,2,3)}$ have the same excitation levels $E_1=E_2=E_3=E$, and their transition dipole moments are aligned in the same direction, i.e., $\mu_i=e\langle \bar{r}|\bar{r}|\bar{r}\rangle\mu_i$, where $\bar{r}=\bar{r}_b-\bar{r}_i$, and $\mu_i$ is located at $\bar{r}_i$. The dipole-dipole interaction only exists in the nearest neighbors and the dipole-dipole couplings are denoted by $J=\frac{1}{4\pi\varepsilon_0}<\hat{\vec{m}}_i\hat{\vec{m}}_j>/r^3$ between $|a_1\rangle$ and $|a_2\rangle$, and $|a_2\rangle$ and $|a_3\rangle$ in Fig.1(b), but there is no coupling between $|a_1\rangle$ and $|a_3\rangle$. The strength of the dipole-dipole coupling $J$ is much weaker than the excitation energy $E-E_b=\hbar\omega$. The Hamiltonian of the three coupling donors can be written as

$$\hat{H} = \sum_{i=1}^{3} \hbar\omega_\alpha\hat{\sigma}_i^+\hat{\sigma}_i + J(\hat{\sigma}_1^+\hat{\sigma}_2^- + \hat{\sigma}_2^+\hat{\sigma}_3^- + H.c.) \quad (eq\ 1)$$

where H.c. means Hermitian conjugation, $\hat{\sigma}_i^+$ and $\hat{\sigma}_i^-$ are the Pauli raising and lowering operators, respectively. The three single-excitation states of the above Hamiltonian are $|a_1\rangle = \frac{1}{\sqrt{2}}(|1\rangle + \sqrt{2}|2\rangle + |3\rangle)$, $|a_2\rangle = \frac{1}{\sqrt{2}}(|1\rangle - \sqrt{2}|3\rangle)$, and $|a_3\rangle = \frac{1}{2}(|1\rangle + \sqrt{2}|2\rangle + |3\rangle)$, and their eigenvalues are obtained as $E_{a_1}=E+\sqrt{2}J$, $E_{a_2}=E$, $E_{a_3}=E-\sqrt{2}J$. The dynamics behavior of the donors-acceptor system can describe via the master equations for the uncoupled case in eqn(2) (shown in Fig.1(a)) and the coupled case in eqn(3) (shown in Fig.1(b)) as follows, respectively.

$$\rho_{i1} = \gamma_\alpha[n_b\rho_{bb} - (1 + n_b)\rho_{i1}] + \gamma_c[\rho_{aa} - (1 + n_c)\rho_{i1}],$$

$$\rho_{i2} = \gamma_\alpha[n_b\rho_{bb} - (1 + n_b)\rho_{i2}] + \gamma_c[\rho_{aa} - (1 + n_c)\rho_{i2}],$$

$$\rho_{i3} = \gamma_\alpha[n_b\rho_{bb} - (1 + n_b)\rho_{i3}] + \gamma_c[\rho_{aa} - (1 + n_c)\rho_{i3}],$$

$$\rho_{\alpha\alpha} = \gamma_c[(1 + n_c)(\rho_{i1} + \rho_{i2} + \rho_{i3}) - 3\gamma_c\rho_{aa} - \Gamma(1 + \chi_1)\rho_{aa}],$$

$$\rho_{\beta\beta} = \Gamma_c[\rho_{bb} - (1 + N_c)\rho_{\beta\beta}] + \Gamma\rho_{aa},$$

and

$$\dot{\rho}_{a_1} = \gamma_\alpha[n_b\rho_{bb} - (1 + n_b)\rho_{a_1}] + \gamma_\alpha [n_{12}\rho_{a_2} - (1 + n_{12})\rho_{a_1} + \gamma_\alpha[\rho_{a_1} - (1 + n_{12})\rho_{a_1}]] + \gamma_\alpha[\rho_{a_1} - (1 + n_{12})\rho_{a_1}],$$

$$\dot{\rho}_{a_2} = \gamma_\alpha[(1 + n_{12})\rho_{a_1} - n_{12}\rho_{a_2}] + \gamma_\alpha[\rho_{a_2} - n_{12}\rho_{a_2}],$$

$$\dot{\rho}_{a_3} = \gamma_\alpha[n_b\rho_{bb} - (1 + n_b)\rho_{a_3}] + \gamma_\alpha[\rho_{a_3} - n_{23}\rho_{a_3}] + \gamma_\alpha[\rho_{a_3} - n_{32}\rho_{a_3}] - \Gamma(1 + \chi_2)\rho_{aa},$$

where $\rho_{a_i}$ and $\rho_{\alpha\alpha}$ describe the average number of photons with frequencies matching the transition energies from the valence band state $|b\rangle$ to the conduction band states $|i\rangle_{(i=1,2,3)}$ in Fig.1(a), and $\rho_{a_i}$ in Fig.1(b) at the temperature $T_s=(300 + \Delta)K$, where $\Delta$ stands for the temperature difference. $n_c=\frac{1}{\exp(\frac{E_{a_1} - E_b}{k_B T_s}) - 1}$ and $n_{ic}(i=1,3)=\frac{1}{\exp(\frac{E_{a_i} - E_b}{k_B T_s}) - 1}$ are the thermal occupation numbers of ambient phonons at temperature $T_s$. $N_c=\frac{1}{\exp(\frac{E_{a_1} - E_b}{k_B T_s}) - 1}$ is the corresponding thermal occupation at the ambient temperature $T_s$ with the energy $(E_{\beta} - E_b)$. $n_{12}$, $n_{13}$, and $n_{23}$ represent the corresponding thermal occupations at the ambient temperature $T_s$ with energy gaps $(E_{a_1} - E_{a_2})$, $(E_{a_1} - E_{a_3})$, and $(E_{a_2} - E_{a_3})$, respectively. The rates in eqn (2) and (3) lead to a Boltzmann distribution for the level population $|\alpha\rangle$ ($p_\alpha = \exp(-\frac{E_{a_i} - E_b}{k_B T_s})$). $\mu_\alpha$ is defined as the
chemical potential of lead α) when the thermal averages for the photon and phonon reservoirs are in a common temperature. We consider the initial condition to be a fully occupied ground state\textsuperscript{[22]}, i.e., ρ\textsubscript{bb} = 1.

III. SUMMARY AND DISCUSSION

In what follows, we calculate the steady solutions of Eq.(2) and Eq.(3) for the RRRs, χ\textsubscript{1} and χ\textsubscript{2}. In fig.1(b), the dipole moment between the state |α\textsubscript{i}\rangle (i=1,3) and the ground state |b\rangle is enhanced or weakened by constructive and destructive interferences between the individual transition dipole matrix elements. While the dipole moment of the state |α\textsubscript{2}\rangle cancels out due to destructive interference. This means the state |α\textsubscript{2}\rangle, comprised of the antisymmetric combination of the uncoupled |1\rangle and |3\rangle states from fig.1(a), describes an optically dark state which leads charge transfer matrix element to equal to zero.

To discuss the RRR χ\textsubscript{1} in Fig.1, we use the following parameters\textsuperscript{[21, 22]}, E\textsubscript{α} − μ\textsubscript{α} = 0.10 ev, E − E\textsubscript{α} = E\textsubscript{β} − E\textsubscript{b} = 0.20 ev, γ\textsubscript{e} = 6 Mev, Γ = 0.40 ev, Γ\textsubscript{c} = 0.15 ev. And other parameters are γ\textsubscript{h} = 0.62 ev in Fig.1(a), and γ\textsubscript{1h} = 0.62 ev, γ\textsubscript{3h} = 0.45 ev, γ\textsubscript{1e} = γ\textsubscript{3e} = 0.15*\((\frac{3}{2} + \sqrt{2})\) ev, J = 0.10 ev, γ\textsubscript{12} = γ\textsubscript{23} = \(\frac{1}{2} + \frac{1}{2}\sqrt{2}\) = 0.15*\(\sqrt{2}\) ev in Fig.1(b).

Fig.2 shows the RRR, χ\textsubscript{1} as a function of the temperature difference Δ(K) in which the energy gaps (E − E\textsubscript{b}) driven by the solar radiation are set as the control-parameters. In the case of three uncoupled donors in Fig.2(a), it notes that a higher temperature can block the radiative recombination between the donors and acceptor, and the increasing energy gaps (E − E\textsubscript{b}) incur the decreasing efficiency of radiative recombination, which can be obviously shown at the room temperature (Δ = 0) in Fig.2(a). These features suggest that electron transport process becomes intense in a higher temperature when the electron transfer is individually driven by the three uncoupled donors, which leads to more excited electrons to perform work. So, in the donors, the transferred electrons for radiative recombination to valence band becomes weak. In spite of the lower recombination of electron can be obtained in the higher temperature condition as shown in Fig.1(a), the room temperature is still regarded as the optimal operating environment for the quantum photovoltaic because of the real environment.

However, a contrary result appears in the case of three dipole-dipole coupled donors in Fig.2(b). The radiative recombination becomes intense in the increasing temperature condition, although the increasing energy gaps (E − E\textsubscript{b}) can also incur the decreasing efficiency of ra-
radiative recombination. The reason for this interesting phenomenon comes from the higher temperature bringing out the intense quantum coherence between the different electron transfer channels, which blocks more electrons to perform useful work but direct to the radiative recombination between the donors and acceptor. This means that quantum effect caused by the three dipole-dipole coupled donors accelerate the electron recombining from the charge-separated state $|\alpha\rangle$ to the valence band state $|b\rangle$ in the donor molecules. What’s more, the larger control-parameters, i.e., the large values of energy gap ($E - E_b$), the larger suppressive RRR. The result demonstrates the p-n junction in the photocell with the moderately larger energy gap will harvest the suppressed radiative recombination of electron. It also notes that at the room condition ($\Delta=0$) the values of RRR are much smaller than those as compared to the case with three uncoupled donors in Fig.2(a), which means the suppressed RRR is achieved in the three dipole-dipole coupled donors case in Fig.2.

In this paper, another energy gap is the gap between the donors and the acceptor molecular, and the energy gaps are $(E - E_a)$ and $(E_{a_i} - E_a)$ in the three uncoupled donors case and the three dipole-dipole coupled donors case, respectively. In Fig.3, the energy gaps between the conduction band states and the valence state state $|b\rangle$ are set $(E - E_a)=0.38$ ev, and 0.7 ev in (a) and (b), respectively. Other parameters are the same to the corresponding parameters in Fig.2. Fig.3 shows the RRR are much smaller in this quantum photocell with three dipole-dipole coupled donors than those in the case of three uncoupled donors in Fig.3(a) at the room condition ($\Delta=0$). And the energy gap between the donors and the acceptor molecular shows the entirely different manipulating features on the RRR comparing to Fig.2, i.e., the increasing energy gaps enhance the RRR in both Fig.3(a) and Fig.3(b). Not only that, but the RRR in Fig.3(a) is much larger than those Fig.3(b). Which demonstrates that the larger energy gaps do harm to the efficiency of the quantum photocell and that the electron transfer closely relates to the energy gaps between the donors and the acceptor molecular. The reason for this is that more excited electrons can’t be transported to the external load but radiative downward to the electrical neutral state $|b\rangle$ when there is a larger energy gap between the donors and acceptor, which brings out the increasing radiative recombination in Fig.3. However, the radiative recombination dependent of temperature difference is similar to Fig.2. The radiative recombination decreases when it’s in the increasing ambient temperature condition in

![FIG. 3. (Color online) The RRR with three uncoupled donors (a) and three dipole-dipole coupled donors (b) as a function of the temperature difference $\Delta (K)$ with different energy gaps $(E - E_a)$.](image-url)
Fig. 3(a) while it increases with an increasing ambient temperature in Fig. 3(b). It concludes that the laws of RRR dependent temperature are identical in this photocell, but the RRR is differently mediated by two different types of energy gaps. Considering the real environment, we think the room temperature ($\Delta=0$) is still the optimal condition for the quantum photocell regulated by the control-parameter ($E - E_{\alpha}$), in spite of the suppressed RRR is achieved by the increasing temperature condition in this quantum photocell with three uncoupled donors case.

To deeply investigate the effects of electrostatic dipole-dipole coupling strength $J$ on the RRR, we plot the RRR, $\chi^2$ versus the coupling strength $J$ in Fig.4, while setting ($E - E_{b}$)=1.6 ev in Fig.4(a) and ($E - E_{a}$)=0.05 ev in Fig.4(b) at the room temperature ($\Delta=0$). And other parameters are the same to those in Fig.2. The increasing electrostatic dipole-dipole coupling strength $J$ indicates the stepped-up quantum interference between the donors. It notes that $\chi^2$ monotonically decreases with the increasing $J$, but that the intense RRRs are generated both by the two different increasing energy gaps in Fig.4(a) and (b), respectively. But we also notice that RRRs slowly fall down in the range $0 < J < 1$ in Fig.4(a), while the sharp decrease of $\chi^2$ appears about in the range $0.3 < J < 0.5$ in in Fig.4(b). The distinct difference between the two types of energy gaps, $(E - E_{a})$ in Fig.4(a) and $(E - E_{b})$ in Fig.4(b) is the minimal values of RRR during the process of the electrostatic dipole-dipole coupling strength $J$ approaching to 1 gradually. In Fig.4(a) the final values of RRRs are in close proximity to 0.05, while their corresponding values infinitesimally approach to 0.2 in Fig.4(b). First of all, it shows that the RRRs can be suppressed furthest by the electrostatic dipole-dipole coupling strength $J$, and that the energy gaps $(E - E_{a})$ have a better inhibition effect on the RRR than the energy gaps $(E - E_{b})$ in this quantum photocell. Secondly, it demonstrates that the RRR can be adjusted and kept to a minimum but can't be canceled out[17] in this photocell model. In other word, the radiative upward transition to generate the excitation is allowed, and the radiative downward transition must be allowed as well but it can be controlled by the energy gaps $(E - E_{a})$ as far as possible.

IV. POSSIBLE EXPERIMENTAL REALIZATION

Up to now, we have studied the features of RRR dependent the temperature, energy gap and the coupling strength of the dipole-dipole coupling $J$ in the quantum photocell with three electron donors. Now let us suggest some potential experimental researches about the
quantum photocell. First of all, two type of energy gaps discussed here display some significant results about the radiative recombination. It manifests that the semiconductor materials with appropriate energy gaps can effectively inhibit radiative recombination, so seeking a semiconductor material with suitable band gap in experiment may be an interesting direction to suppress the radiative recombination. Secondly, the energy gap between the donors and acceptor may be another experimental investigation according our results, and best-effort to reduce this gap is a possible experimental realization for the suppressed RRR. How to align the donors of photocells for an intense electrostatic dipole-dipole coupling strength \( J \) in the manufacturing process may be another research field. The scenario proposed here may be a different approach for the efficient photon-to-charge conversion and deserve further experimental investigation.

V. CONCLUSION

To summarize, in this work we explored the RRR dependent ambient temperature in a quantum photocell system with three electron donors. The RRR is reduced by the increasing energy gaps \( (E - E_b) \) and the increasing ambient temperatures in the quantum photocell system with three uncoupling donors case. In the meantime, but the RRR is enhanced by the increasing gaps between the donors and acceptor \( (E - E_\alpha) \). However, the increasing temperature incurred much intense radiative recombination with the increasing energy gaps \( (E - E_b) \) but the RRR was still suppressed by the the increasing energy gaps \( (E - E_b) \) of the donors in the quantum photocell with three dipole-dipole coupled donors case. And in the condition of room temperature, the quantum photocell system possesses the smaller RRR comparing to those in the three uncoupling donors case. What’s more, the RRR can be restrained to a minimum value but can’t be canceled out when the photocell system adjusted by the electrostatic dipole-dipole coupling strength \( J \) between the donors at room temperature. As demonstrates the breaking detailed balance limits and a way of high photon-to-charge conversion efficiency in this quantum photocell. These results suggest some encouraging trends of the suppressed RRR, such as seeking a semiconductor material with suitable band gap, minimizing the gap between the donors and acceptor, and aligning properly the donors for an intense electrostatic dipole-dipole coupling strength \( J \). And these theoretical results in this work deserve the further experimental confirmation.

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