Tunneling Current Spectra of a Metal Core/Semiconductor Shell Quantum Dot Molecule

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1. Introduction

Although many of the interesting transport phenomena related to quantum dots (QDs) including the Coulomb blockade, Kondo effect, Fano resonance, Pauli spin blockade, photon (phonon) assisted tunneling process and negative differential conductance, have already been extensively studied, there has been little reported on the effects of electron plasmon interactions (EPIs) on the tunneling current spectra of QDs. Recently, EPIs have received considerable attention for their applications in nanophotonics, biology, and the harvesting of solar energy. Much effort has been focused on the effects of EPIs on the optical properties of nanostructures. Experiments can observe the strong effect of EPIs on the exciton spectrum of individual semiconductor QDs arising from adjacently metallic nanostructures.

In the application of QD-based biosensor, the metal core/semiconductor shell QDs play an crucial role. Because this type QDs can be readily coupled to detected proteins through electrostatic interactions. Despite the measurement of optical spectra can resolve molecules, tunneling current spectra provide a high efficient means to identify molecules and address electrically a single nano-object. This inspires us to study the tunneling current through individual metal core/semiconductor shell QDs for the application of nanoscale biosensor. However, it is difficult to avoid the proximity effect between such type QDs since QDs are randomly distributed. Therefore, we consider a metal core/semiconductor shell QD molecule (QDM) embedded in a matrix connected to metallic electrodes shown in Fig. 1 to clarify how the EPIs to influence the proximity effect resulting from interdot electron Coulomb interactions, electron hopping process, and plasmon hopping between two QDs in the absence of detected proteins.

2. Formalism

Here, we consider nanoscale semiconductor QDs, the energy levels separation of each QD is much larger than their on-site Coulomb interactions and thermal energies. One energy level for each quantum dot is considered in this study. The two-level Anderson model including EPIs is employed to simulate the QDM junction system as shown in Fig. 1. The Hamiltonian of the QDM junction is given by $H = H_0 + H_{QDM} + H_T$.

$$H_0 = \sum_{k,l} \epsilon_{kl} a_{kl}^+ a_{kl} + \sum_{k,l} \epsilon_{kl} b_{kl}^+ b_{kl} + \sum_{k,l} V_{kl} d_{kl}^+ d_{kl} + c.c., \quad (1)$$

where the first two terms describe the free electron gas at the left and right metallic electrodes. $a_{kl}^+$ ($b_{kl}^+$) creates an electron of momentum $k$ and spin $\sigma$ with energy $\epsilon_k$ at the left (right) metallic electrode. $V_{kl}$ ($l = 1, 2$) describes the coupling between the metal electrodes and the QDs. $d_{kl}^+$ ($d_{kl}$) creates (destroys) an electron in the $l$-th dot:

$$H_{QDM} = \sum_{l\sigma} E_l n_{l\sigma} + \sum_{l\sigma} U_l n_{l\sigma} n_{l\bar{\sigma}}$$

$$+ \frac{1}{2} \sum_{l,j,\sigma,\sigma'} U_{lj} n_{l\sigma} n_{j\sigma'} + \sum_{l,j} t_{lj} d_{lj}^+ d_{lj}, \quad (2)$$

where $E_l$ is the spin-independent QD energy level, and $n_{l\sigma} = d_{lj}^+ d_{lj}$.

Notations $U_l$ and $U_{lj}$ describe the intradot and interdot Coulomb interactions, respectively. $t_{lj}$ describes the electron interdot hopping. The Hamiltonian of QD molecule described by Eqs. (1) and (2) has already been extensively considered for studying the transport properties of nanostructures. The Hamiltonian of EPIs arising from the metallic nanostructure of each QDs can be written as $H_T$:

$$H_T = \sum_{l=1,2} \Omega_l c_{lj}^+ c_{lj} + \sum_{l\sigma} \Omega_l n_{l\sigma} (c_{lj}^+ + c_{lj}) - \sum_{l\sigma} \Omega_{l\sigma} c_{lj}^+ c_{lj}, \quad (3)$$

where $\Omega_l$ is the plasmon frequency of the metallic nanostructures shown in Fig. 1, and $\Omega_l$ is the coupling strength of EPIs. The last term involving $\Omega_{l\sigma}$ describes the plasmon hopping between two metallic nanostructures. When metal nanostructures are close enough, the plasmonic modes on...
one nanostructure can couple with that on another nanostructures so that hopping of plasmons from one metallic nanostructure to other becomes possible.

A canonical transformation can be used to remove the on-site EPIS from Eq. (3), that is $H_{\text{new}} = e^2 \hbar \epsilon \lambda$, where $\lambda = - \sum_{i,j} \Omega_{ij} (c_j^\dagger - c_j)$. In the new Hamiltonian, we have the following effective physical parameters: 

$$V_{k,j,a} = V_{k,j,a} e^{-1/(\epsilon \omega_{ij})}, \quad \tilde{t}_j = t_j - \sqrt{\omega_{ij}^2 - \omega_{ij}^2 \lambda_i}, \quad \tilde{t}_j^\dagger = \tilde{t}_j^\dagger - \sqrt{\omega_{ij}^2 - \omega_{ij}^2 \lambda_i^2},$$

and interdot hopping strength

$$t \exp \left( \frac{1}{2} (A - A_2) \coth^2 \left( \frac{\hbar \omega_0}{2k_B T} \right) \right) = t_c X_{i,j} = t_c.$$ 

This approximation is valid when the tunneling rate arising from the coupling between the QDs and the electrodes is smaller than the EPIS, which is our condition of interest. Based on such an approximation, the $\omega_0$-dependent tunneling rates are neglected in this study. Meanwhile, we assume that there is no voltage difference between two dots. This implies that the tunneling currents directly involving $t_c$ can be ignored.

Using the Keldish–Green’s function technique, (23) the tunneling currents of the QDMs in the Coulomb blockade regime are given by

$$J = -\frac{e^2}{\hbar} \sum_{i,j} \int d\epsilon \frac{\Gamma_{i,j}(\epsilon) \Gamma_{j,i}(\epsilon)}{\tilde{t}_j(\epsilon) + \tilde{t}_j^\dagger(\epsilon)} \Im G_{i,j}(\epsilon)[f_{\epsilon}(\epsilon) - f_{\epsilon}(\epsilon)]$$

(4)

where

$$f_{\epsilon}(\epsilon) = \left[ \exp \left( \frac{\epsilon - \mu_{i,j}(\epsilon)}{k_B T} \right) + 1 \right]^{-1}$$

denotes the Fermi distribution function of the left (right) electrode. The left (right) chemical potential is given by $\mu_{i,j}(\epsilon) = \mu_{i,j} + eV_0$, where $V_0$ denotes the applied bias. Notation $T$ denotes the equilibrium temperature of the left and right electrodes. $e$ and $h$ denote the electron charge and Planck’s constant, respectively. Notation $\Gamma_{i,j}(\epsilon) = \sum_{a} V_{k,i,a}^2 \delta(\epsilon - \epsilon_k)$ denotes the tunneling rate from the left (right) electrode to dot 1 and dot 2, which is assumed to be energy- and bias-independent. The retarded Green function of the QD density of states ($-\Im G_{i,j}(\epsilon)$) has the following expression

$$G_{i,j}(\epsilon) = \sum_{n=-\infty}^{\infty} L_n [f(\epsilon + n\hbar \omega_0) G_{i,j}(\epsilon + n\hbar \omega_0) + f(\epsilon - n\hbar \omega_0) G_{i,j}(\epsilon - n\hbar \omega_0)],$$

(5)

where $L_n$ is given by

$$L_n(\epsilon) = \exp[-g^2(1 + N_n)] \exp \left( \frac{n\hbar \omega_0}{2k_B T} \right).$$

(6)

with a boson distribution function of $N_n = 1/|\exp[\hbar \omega_0/(k_B T)] - 1|$, and a Bessel function of $L_n(\epsilon)$. The expressions of $f(\epsilon + n\hbar \omega_0)$ and $f(\epsilon - n\hbar \omega_0)$ are

$$f(\epsilon + n\hbar \omega_0) = \frac{\Gamma_{i,j}(\epsilon + n\hbar \omega_0) + \Gamma_{j,i}(\epsilon + n\hbar \omega_0)}{\Gamma_{i,j}(\epsilon) + \Gamma_{j,i}(\epsilon)}$$

(7)

$$f(\epsilon - n\hbar \omega_0) = 1 - \frac{\Gamma_{i,j}(\epsilon - n\hbar \omega_0) + \Gamma_{j,i}(\epsilon - n\hbar \omega_0)}{\Gamma_{i,j}(\epsilon) + \Gamma_{j,i}(\epsilon)},$$

(8)

where the effective tunneling rate of $\Gamma_{i,j,\text{LR}}(\epsilon) = \Gamma_{i,j,\text{LR}}(\epsilon + X)^2$ with the reduction factor of $X = \exp[-(1/2g^2) \coth[\hbar \omega_0/(2k_B T)]$). The dressed electron retarded Green function under $H_{\text{new}}$ can be obtained following the procedure introduced in our previous work. (20) We have the expression of $G_{i,j}(\epsilon)$

$$G_{i,j}(\epsilon) = \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \Im \left[ \frac{\hbar \omega_0}{2k_B T} \right]$$

(9)

where $\mu_0 = -e^2/\hbar t_c$, $\mu_1 = -e^2/\hbar t_c$. $\Gamma_{i,j}(\epsilon)$ is given by

$$\Gamma_{i,j}(\epsilon) = \sum_{\text{occupied}} \Gamma_{i,j}(\epsilon) + \sum_{\text{empty}} \Gamma_{i,j}(\epsilon),$$

(10)

and $c_1$ can be obtained by solving the on-site lesser Green’s functions, (20) their expression is

$$N_{i,j} = -\frac{\hbar \omega_0}{2k_B T}$$

(11)
Note that $t \neq j$ in Eqs. (9)–(11), which are valid in the condition of $t_{ij}/U_i \ll 1$. In addition, the limitation of $0 \leq N_{\alpha}(c_I) \leq 1$ should be satisfied.

3. Results and Discussion

Bulk metals have very large plasmon frequencies, which can be 10 times larger than the on-site Coulomb interactions of QDs. Therefore, the effects of EPIs on the tunneling current were ignored in the previous studies.\cite{18,19,20} The plasmon frequency of metallic nanostructures is the same order of magnitude as the on-site Coulomb interactions and the EPIs is strong.\cite{13} One can expect to observe the plasmon assisted tunneling processes in the tunneling current spectra. To reveal the effects of EPIs on the tunneling current spectra of QDs, we initially consider the case without EPIs. The tunneling current and differential conductance are plotted in Fig. 2 with $U_j = U_0 = 60\Gamma_1$, $U_{ij} = U_1 = 20\Gamma_0$, and $E_i = E_0 + \eta eV_a$. There is a large voltage across the junction, therefore the shift of energy level $E_j$ arising from the applied bias is considered by $\eta eV_a$. We have adopted $\eta = 0.5$ based on the assumption that QDs are located in the central position between two electrodes. The black lines show the typical staircase structure of the tunneling current and an oscillatory differential conductance with respect to the applied bias arising from the intradot Coulomb interactions. These structures will be washed out with increasing temperature. We will focus on the transport behavior throughout at the low temperature of $k_B T = 1\Gamma_0$. In the presence of interdot Coulomb interactions (red lines), new staircase structures appear in the tunneling current. Five peaks in differential conductance labeled from $V_1$ to $V_5$ result from electrons of the left electrode through the resonant channels of $\epsilon_1 = E_0$, $\epsilon_2 = E_0 + U_1$, $\epsilon_3 = E_0 + U_0$, $\epsilon_4 = E_0 + U_0 + U_1$, and $\epsilon_5 = E_0 + U_0 + 2U_1$. In the presence of $t_0$ (see the blue lines), each peaks ($V_1$, $V_2$, $V_4$ and $V_5$) split into the bonding (BD) and antibonding (ABD) states. Such structures can be depicted by using a single molecule with $E_0 - t_0$ and $E_0 + t_0$ states filled with one, two, three, and four electrons. The electron filling of such a QDM satisfies Hund's rule. In addition, peaks $V_2$ and $V_3$ have extra peaks, which correspond to the spin singlet states (two electrons and three electrons). For instance the two electron singlet state has resonant pole $\epsilon = E_0 + U_1 - \frac{t_0^2}{U_0 - U_1}$ resulting from the $p_2$ in Eq. (9), which is different from the electronic triplet state with pole $\epsilon = E_0 + U_1 + \frac{t_0^2}{U_0 - U_1}$ from the $p_3$ of Eq. (9). The differential conductance structure resulting from three electrons can also be analyzed from the $p_3$ and $p_7$ of Eq. (9). On the basis of results in Fig. 2, we find that the interdot Coulomb interactions play an important role in distinguishing between the configurations of one electron, two electrons, three electrons, and four electrons. Many theoretical works have been devoted to investigate the tunneling current through parallel QDs for the applications of quantum computing.\cite{24} Nevertheless, there still lacks a comprehensive theory to reveal the spin states of parallel QDs. The retarded Green function of Eq. (9) provides a closed form expression to distinguish eight configurations in the parallel QDs.

Figure 3 shows the tunneling current ($J$) and differential conductance ($dG$) for different strengths of EPIs at $U_0 = 60\Gamma_0$, $U_1 = 20\Gamma_0$, $t_0 = 0$, $\theta_{ij} = \theta_0 = 20\Gamma_0$. With increasing the strength of EPIs, the energy levels of the QDs, intradot Coulomb interactions, and interdot Coulomb interactions are renormalized by EPIs such as $E_j = E_l - g^2\theta_0$, $U_j = U_l - 2g^2\theta_0$, and $U_{ij} = U_{i,j} - 2g^2\theta_0$. The current spectra and

Fig. 2. (Color online) (a) Tunneling current $J$ and (b) differential conductance $dG = dJ/dV_a$ as a function of the applied bias for $U_l = U_0 = 60\Gamma_0$, $E_l = E_0 + 30\Gamma_0$, $k_B T = 1\Gamma_0$, and $\Gamma_{1,l} = \Gamma_{1,R} = 0.5\Gamma_0$ in the absence of electron plasmon interactions. Black lines ($U_{ij} = 0$), red lines ($U_{ij} = 20\Gamma_0$ and $t_0 = 0$), and blue lines ($U_{ij} = 20\Gamma_0$ and $t_0 = 6\Gamma_0$). Note that the tunneling current $J$ is in units of $J_0 = e^2\Gamma_0/h$.

Fig. 3. (Color online) (a) Tunneling current $J$ and (b) differential conductance $dG$ as a function of the applied bias in the presence of electron plasmon interactions. Black lines ($g = 0.0$), red lines ($g = 0.25$), and blue lines ($g = 0.5$). We have a plasmon frequency $\omega_0 = 20\Gamma_0$. The other physical parameters are the same as those for the red lines in Fig. 2.
differential conductance change considerably. Five peaks of \( dG \) labeled from \( V_1 \) to \( V_5 \) at \( g = 0 \) are shifted to the low bias regime. The magnitude and width of these peaks become smaller and narrower with increasing \( g \). This is attributed to the current reduction factor of \( e^{-x^2(1+h_0)} \) [see Eq. (6)] and reduction of the tunneling rates \( \Delta \gamma = \Gamma \chi^2 \). In addition to these five peaks, there is a satellite peak labeled \( n = 1 \), which arises from one plasmon assisted tunneling process. For the blue line, we see two one-plasmon assisted tunneling peaks corresponding to \( \varepsilon = E_0^s + \omega_0 \) and \( \varepsilon = E_0^t + U_1^s + \omega_0 \). As a consequence of the very low temperatures, the structures arising from \( \varepsilon = E_0^s - \omega_0 \) and \( \varepsilon = E_0^t + U_1^s - \omega_0 \), which are contributed from the first term of Eq. (5), correspond to electrons of the QDM with energy levels \( E_0^s \) and \( E_0^t + U_1^s \) to have one plasmon emission process to escape out the QDM. This processes are obviously suppressed in the small bias regime resulting from small electron population of the QDM.

Since we consider \( t_c = 0 \), the spin degree of freedom cannot be resolved in Fig. 3.\(^a\) To further understand the EPIs on the current spectra of QDMs with finite \( t_c \), we plot the tunneling current (\( J \)) and differential conductance (\( dG \)) as a function of the applied bias for the case of \( U_l = 100 \Gamma_0 \), \( U_{ij} = 40 \Gamma_0 \), and \( t_c = 6 \Gamma_0 \) in Fig. 4. The first two peaks indicated by the black line for \( g = 1 \) correspond to the BD and ABD states. The following three peaks are similar to those indicated by the blue line in Fig. 2. They result from the spin singlet and triplet states. For \( g = 0.5 \), these spin-dependent spectra of \( dG \) just shift to the low bias regimes, whereas the separation between the BD and ABD peaks in the triplet state is not changed. The exchange energy of the singlet state \(-t_c^2/(U_1^s - U_1^t)\) is also invariant. For \( g = 1 \), the tunneling currents are enhanced resulting from the absence of the interdot Coulomb blockade. The spectra from the two particle states merge into that of one particle at \( g = 1 \). Consequently, the spin-dependent spectra of \( dG \) is suppressed. In addition, we observe multiple plasmon assisted tunneling processes (\( n = 2 \)).

4. Summary and Conclusions
In this study we analyzed the effects of homogeneous EPIs on the tunneling current spectra of QDMs in the absence of detected proteins. As a result of the renormalization of the energy levels of QDs, intradot and interdot Coulomb interactions, and the tunneling rates, there is a significant change in the tunneling current spectra for strong EPIs coupling. Because of the hopping of plasmons between two metal nanostructures, the indirect interdot plasmon mediated electron electron Coulomb interactions appear. We predict that the multiple plasmon assisted tunneling processes can be observed in the tunneling current spectra of metal core/shell semiconductor QDs for strong EPIs. In the presence of detected proteins, which will glue to the QDMs, the DOS of QDMs is changed. Therefore, the measured tunneling current spectra are tilted to judge the identity of detected proteins.

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