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Enhancement of the thermoelectric figure of merit in a quantum dot due to the Coulomb blockade effect

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We investigate the figure of merit of a quantum dot (QD) in the Coulomb blockade regime. It is found that the figure of merit *ZT* may be quite high if only single-energy level in the QD is considered. On the other hand, with two or multienergy levels in the QD and without the Coulomb interaction, the *ZT* is strongly suppressed by the bipolar effect due to small level spacing. However, in the presence of the Coulomb interaction, the effective level spacing is enlarged and the bipolar effect is weakened, resulting in *ZT* to be considerably high. Thus, it is more likely to find a high efficient thermoelectric QDs with large Coulomb interaction. By using the parameters for a typical QD, the *ZT* can reach over 5.

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I. INTRODUCTION

Thermoelectric materials are such materials that can directly convert the thermal energy into the electrical energy. Thus the thermoelectric energy conversion technology has been recognized as the most feasible energy-conversion technology. However, due to its low efficiency, this technology has not been widely used. Thus it is important to find high efficient thermoelectric materials. The efficiency of thermoelectrical materials is measured by the dimensionless-thermoelectrical figure of merit ZT while $ZT=\sigma S^2 T/\kappa$. Here S is the Seebeck coefficient, σ is the electric conductivity, and κ is the total thermal conductivity which contains the lattice-thermal conductivity κ_l and electric (carrier) thermal conductivity κ_e, and T is the operating temperature of the device.[1] For a material to be a good thermoelectric material, it must have a large ZT, which means in order to achieve a large ZT, one must increase the Seebeck coefficient S and electric conductivity σ and decrease the thermal conductivity κ. However, it seems difficult to have a high ZT in nature materials. Several reasons hinder the rising of ZT. First, in conventional solids the Wiedemann-Franz law $[\kappa_e/\sigma T=(k_B\pi)^2/3e^2]$ is obeyed,[2] which means that an increase in the electric conductivity also leads to an increase in the thermal conductivity. Second, according to the Mott relation,[3] an increase in the electric conductivity is apt to lead to an decrease in Seebeck coefficient. Thus in the past 50 years, the maximum ZT is holding at about 1. This largely affect the industrial applications.

Recently, the advances in nanostructure materials have largely stimulated the development in thermoelectric materials. Due to the quantum phenomena emerged in nanostructure materials, the classical results such as the Mott relation and the Wiedemann-Franz law may not hold.[4] What is more, the thermoelectric properties of the nanostructure materials can be modulated by changing the gate voltage. Thus it opens a new and wide road to find efficient thermoelectric materials. The idea of using low-dimensional structure materials to gain high ZT was first introduced by Hicks and Dresselhaus in 1993.[5] They theoretically show that ZT increases swiftly as the dimensions decrease, far beyond the value obtainable in bulk materials. Following this suggestion and with the development in nanotechnology, various groups were able to fabricate nanostructures and measure their thermoelectric properties.[6–10] For example, Harman *et al.*[7] have measured the thermoelectric properties of quantum dot and a maximum value $ZT\approx 2$ was obtained. Venkatasubramanian *et al.*[8] have measured a thin-film thermoelectric device and have observed a maximum ZT of ~ 2.4 at room temperature. Apart from the experimental efforts, many theoretical studies have been carried out on low-dimensional structures such as quantum dots, nanowires, and superlattices.[5,11–14] For example, Venkatasubramanian and Chen[13] have concluded that the main reason of high ZT in low-dimensional materials is due to a significant reduction in lattice thermal conductivity. A giant figure of merit in single-molecule device is obtained by Finch *et al.*[14] All these efforts show that a high figure of merit may exist in nanomaterials. However, due to the complexity and expensiveness of the nanomaterials, there is still a long way to go for the commercial applications of nanostructure-thermoelectric materials. At present the most promising nanostructure-thermoelectric material is nanocomposite-thermoelectric material.[2,15,16]

In this paper, we study the thermoelectric properties of lead-QD-lead system with the QD in the Coulomb-blockade regime. The thermoelectric properties of QD have been widely studied. For example, Beenakker[17] have investigated the thermal properties of QD with multiple energy levels[4] but they only considered the situation that temperature (k_BT) is much bigger than the level width (Γ). On the other hand, Turek and Matveev[18] and Murphy *et al.*[19] have studied the thermal properties of the QD in the situation $k_BT\sim\Gamma$ but the QD considered contains only a single-energy level. Here we consider the QD having the multiple-energy levels and in the situation of the temperature smaller than the interaction U. In this regime, some new phenomena emerge. By using the Landauer-Bütticker formalism combining the nonequilibrium Green's functions,[20] the electronic conductivity, Seebeck coefficient, and thermal conductivity are obtained. Due to the electron-hole symmetry, the Seebeck coefficient is always antisymmetric. If only a single-energy level in the QD is considered, the *ZT* increases monotonously with temperature *T*, and *ZT* can be very large at high temperature, consistent with previous results. However, with two or multilevels and the temperature is on the order of energy gap, the *ZT* is strongly suppressed by the bipolar effect, mainly caused by the antisymmetric property of the Seebeck coefficient. On the other hand, when the Coulomb interaction *U* is considered, the energy spacings are enlarged due to the Coulomb-blockade effect. The bipolar effect is greatly reduced and high value of *ZT* may again be achieved. In a typical QD, the Coulomb interaction *U* is usually larger by an order than the linewidth Γ and the single-particle energy spacing. Under these conditions, the *ZT* can be quite high with its maximum value reaching over 5.

II. MODEL AND FORMALISM

The system of the lead-QD-lead can be described by the following Hamiltonian:

$$H = \sum_{\alpha,k,\sigma} \varepsilon_{\alpha k}\hat{c}^{\dagger}_{\alpha k\sigma}\hat{c}_{\alpha k\sigma} + \sum_{\alpha,k,i,\sigma} t_{\alpha k}(\hat{d}^{\dagger}_{i\sigma}\hat{c}_{\alpha k\sigma} + \text{H.c.}) + \sum_{i=1,2;\sigma} \varepsilon_i \hat{n}_{i\sigma} + \frac{U}{2}\sum_{i,\sigma,j,\sigma'(i\sigma\neq j\sigma')} \hat{n}_{i\sigma}\hat{n}_{j\sigma'}, \tag{1}$$

where $\hat{n}_{i\sigma}=\hat{d}^{\dagger}_{i\sigma}\hat{d}_{i\sigma}$ and $\alpha=L,R$ represent the left and right leads. $\hat{c}^{\dagger}_{\alpha k\sigma}$ and $\hat{d}^{\dagger}_{i\sigma}$ create an electron with spin σ in the α lead and the *i*th energy level of QD, respectively. Here the intradot electron-electron Coulomb interaction is considered with the interaction strength *U*. The second term in Eq. (1) describes the tunneling coupling between the QD and the two leads and $t_{\alpha k}$ is the hopping-matrix element.

By using nonequilibrium Green's-function methods, the electronic current and electric-thermal current from the left lead flowing into the QD can be written in the forms[20,21]

$$\begin{pmatrix} I \\ Q \end{pmatrix} = \frac{2}{h}\int d\omega \begin{pmatrix} -e \\ \omega-\mu_L \end{pmatrix} (f_L - f_R)T(\omega), \tag{2}$$

where $f_\alpha = f(\omega-\mu_\alpha) = 1/\{exp[(\omega-\mu_\alpha)/k_BT]+1\}$ is the Fermi distribution of the α lead and $T(\omega)$ is the transmission coefficient. $T(\omega)$ can be expressed by the following expression:

$$T(\omega) = \mathrm{Tr}\left[\frac{\Gamma_L\Gamma_R}{\Gamma_L+\Gamma_R}(\mathbf{G}^r - \mathbf{G}^a)\right]. \tag{3}$$

Here $\mathbf{G}^r(\mathbf{G}^a)$ is the standard retarded (advanced) Green's function of the QD (Refs. 20 and 21) and $\Gamma_{\alpha,ij} = \Sigma_k 2\pi |t_{\alpha k}|^2 \delta(\omega-\epsilon_{\alpha k})$ is the linewidth functions which assume to be independent of the energy ω. We introduce the following integrals $I_n(T)$ $(n=0,1,2,\ldots)$: $I_n(T) = -(2/h)\int \omega^n(\partial f/\partial\omega)T(\omega)d\omega$. By using the quantities $I_n(T)$, the linear-electric conductance *G*, thermopower *S*, and thermal conductance κ can be expressed in very simple forms[22]

$$G = e^2 I_0(T), \tag{4a}$$

$$S = -I_1(T)/[eTI_0(T)], \tag{4b}$$

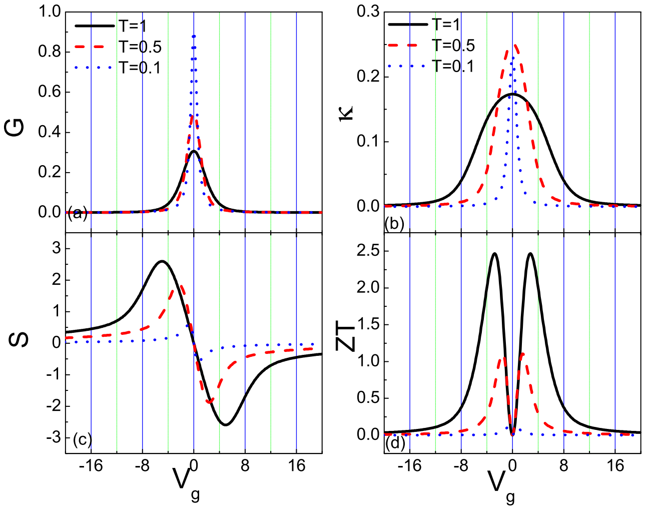

FIG. 1. (Color online) $G(2e^2/h)$, $k(2k_B/h)$, $S(k_B/e)$, and *ZT* vs the level ε_1 for the different temperature *T* for the single-level QD and $U=0$.

$$\kappa = (1/T)[I_2(T) - I_1^2(T)/I_0(T)]. \tag{4c}$$

Therefore, the only question left is to calculate the Green's functions of the QD.

III. NUMERICAL RESULT

In the numerical investigation, we consider the symmetric barriers with $\Gamma_{L,ij}=\Gamma_{R,ij}=\Gamma$, and set $\Gamma=0.5$ and $\mu_R=0$ as the energy-zero point. We consider the linear regime, then $\mu_L = \mu_R \equiv \mu = 0$. First, we study the case with QD possessing only one energy level ε_1 and in the absence of the Coulomb interaction $(U=0)$. In this case, the Green's function of QD can be easily obtained as $\mathbf{G}^r_{1\sigma}(\omega)=\mathbf{G}^{a*}_{1\sigma}(\omega)=1/(\omega-\varepsilon_1+i\Gamma)$. Inserting this into Eq. (3), the transmission coefficient $T(\omega)$ can be obtained and the thermoelectric properties can then be calculated straightforwardly. Figure 1 shows the electric conductance *G*, the thermal conductance κ, the thermopower *S*, and *ZT* versus the level ε_1 for the different temperature *T*. Variation in ε_1 is equivalent to variation of the gate voltage in an experimental setting. The electric conductivity *G* and the thermal conductance κ exhibit a single-resonant peak at the position $\varepsilon_1=0$. The peak height of the thermal conductance κ first increases and then decreases whith the increase of the temperature k_BT [see Fig. 1(b)]. The reason is as follows. The thermal conductance is determined by two aspects: the heat transferred by each electron and the tunneling probability of each electron. When temperature increases, the average tunneling probability decreases but the heat transferred by each electron increases, leading to a nonmonotonic relation of κ and k_BT. The property of thermopower *S* is described in Fig. 1(c). Here we can see that the curves are antisymmetric due to the electron-hole symmetry. The reason is as follows. The thermoelectric effect is caused by the temperature difference. There are more electrons being excited above the chemical potential μ in the hotter region and correspondingly more holes being generated below μ. When the energy level of QD is below μ, the main carriers are holes and then the thermal power is positive. When the energy level is above μ, the main carriers are electrons and thus the thermal power is negative. So one can adjust the gate voltage or equivalently ε_1 and obtain the optimized thermal power. Once the thermal power, the electron conductivity and the thermal conductivity are known, *ZT* can be calculated. Figure 1(d) describes *ZT* as a function of QD's level ε_1. The optimized *ZT* can be obtained by modulating ε_1 when the system is kept at a fixed temperature. With increase in temperature, the value of optimized *ZT* also increases and it goes to infinity as *T* approaches infinity. This is consistent with the previous result.[24] Of course, this is a nonsensible result due to only single level being considered here. As we can see, at temperature $T=1$ (i.e., $T=2\Gamma$), the optimized *ZT* is about 2.5.

In a realistic situation, depending on temperature, dot size, etc., one normally has to consider multilevels. For a multilevel dot, the spacing between neighboring levels is an important quantity. For that purpose, investigating a two-level dot will capture essential physics due to the level spacing. In the following we study the thermoelectric properties of a QD containing two energy levels. The Green's function of QD with two energy levels is

$$\mathbf{G}^r_\sigma(\omega) \equiv \begin{pmatrix} \mathbf{G}^r_{11\sigma} & \mathbf{G}^r_{12\sigma} \\ \mathbf{G}^r_{21\sigma} & \mathbf{G}^r_{22\sigma} \end{pmatrix} = \mathbf{g}^r_\sigma(\omega) + \mathbf{g}^r_\sigma(\omega)\mathbf{\Sigma}^r\mathbf{G}^r_\sigma(\omega), \quad (5)$$

$$\mathbf{G}^<_\sigma(\omega) \equiv \begin{pmatrix} \mathbf{G}^<_{11\sigma} & \mathbf{G}^<_{12\sigma} \\ \mathbf{G}^<_{21\sigma} & \mathbf{G}^<_{22\sigma} \end{pmatrix} = \mathbf{G}^r_\sigma(\omega)\Sigma^<\mathbf{G}^a_\sigma(\omega). \quad (6)$$

Here, the boldface letters (**G**, **g**, and **Σ**) represent the 2×2 matrix. $\mathbf{g}^r_\sigma$ is the Green Function of QD without coupling to the leads. $\mathbf{g}^r_\sigma$ can be obtained from the equation of motion technique (the detailed deduction can be seen in Appendix) and $\mathbf{\Sigma}^{r,<}$ are obtained from Dyson equations (Here we just consider the first order of self-energy correction and have neglected the higher order of self-energy correction that is due to the e-e interaction)[23]

$$\mathbf{g}^r_{ij\sigma}(\omega) = \left\{ \frac{1-\{N_{i\sigma}\}}{\omega-\varepsilon_i-[N_{i\sigma}]U} + \frac{\{N_{i\sigma}\}}{\omega-\varepsilon_i-([N_{i\sigma}]+1)U} \right\}\delta_{ij}, \quad (7)$$

$$\mathbf{\Sigma}^r_\sigma = \begin{pmatrix} -i\Gamma & -i\Gamma \\ -i\Gamma & -i\Gamma \end{pmatrix}, \quad (8)$$

$$\mathbf{\Sigma}^<_\sigma = \begin{pmatrix} i[\Gamma_L f_L + \Gamma_R f_R] & 0 \\ 0 & i[\Gamma_L f_L + \Gamma_R f_R] \end{pmatrix}, \quad (9)$$

where $N_{i\sigma}=n_{i\bar{\sigma}}+n_{\bar{i}\sigma}+n_{\bar{i}\bar{\sigma}}$, $n_{i\sigma}$ is the electron occupation number in the *i*th energy level with the spin state σ, $\bar{\sigma}=\downarrow$ while $\sigma=\uparrow$ and $\bar{\sigma}=\uparrow$ while $\sigma=\downarrow$, and $\bar{i}=1$ while $i=2$ and $\bar{i}=2$ while $i=1$. In Eq. (7), $[N_{i\sigma}]$ means the integer part of $N_{i\sigma}$, $\{N_{i\sigma}\}=N_{i\sigma}-[N_{i\sigma}]$, namely, the decimal part of $N_{i\sigma}$. In addition, the electron-occupation numbers $n_{i\sigma}$ need to be self-consistently calculated with the self-consistent equation $n_\sigma=-i\int(d\omega/2\pi)\mathbf{G}^<_\sigma(\omega)$.

Figure 2 shows the conductance *G*, the thermal conductivity κ, the thermopower *S*, and *ZT* versus the gate voltage V_g in the absence of the Coulomb interaction ($U=0$). Here the energy levels and the gate voltage are related by: $\varepsilon_1=V_g$ and $\varepsilon_2=V_g+\Delta\varepsilon$, where $\Delta\varepsilon$ is the spacing between the two levels. At the low temperature, *G* and κ in Figs. 2(a) and 2(b) exhibit two peaks due to the two energy levels. There seems no great change in *G* in comparison with that for the single-level QD. However, behavior of κ in Fig. 2(b) is more sensitive to temperature. When temperature is on the order of $\Delta\varepsilon$, the peaks are broadened to a degree to give rise to a huge peak. Meanwhile, the thermopower *S* and the *ZT* are largely suppressed when the lead's chemical potential μ is between the two energy levels ($\varepsilon_2>\mu>\varepsilon_1$). On the other hand, the optimized *ZT* remains considerably large when μ is outside the two energy levels ($\mu<\varepsilon_1,\varepsilon_2$ or $\mu>\varepsilon_1,\varepsilon_2$). In a real system there are many energy levels in a QD. The *ZT* with the chemical potential outside of the two levels is influenced by other levels. So we only focus on the *ZT* for the case with $\varepsilon_2>\mu>\varepsilon_1$, in which the optimized *ZT* is rather low. This is because the electron and holes are excited in the range of k_BT, then the carriers can tunnel through the *i*th energy level in QD when $\mu-k_BT<\varepsilon_i<\mu+k_BT$. When temperature is significantly lower than the level spacing $\Delta\varepsilon$, the carriers can only choose one energy level to tunnel, which is similar to the single-level QD. When temperature is on the order of $\Delta\varepsilon$, the carriers can choose both levels to tunnel. Due to the temperature difference between the two leads, the electrons above (below) the chemical potential μ in the left lead are more (less) than the electrons in the right lead. While $\varepsilon_1<\mu<\varepsilon_2$, electrons tunnel from the left lead to the right lead through the level ε_2, at the same time electrons tunnel from the right lead to the left lead through the level ε_1, in other words the hole tunnels from the left lead to the right lead through ε_1. This is a bipolar effect: a nonzero heat conduction emerges even when the net electrical current is zero. Furthermore, the Seebeck coefficient *S* is significantly suppressed since the carriers are tunneling through the QD in both channels via opposite directions. The above reasons cause *ZT* to be very small.

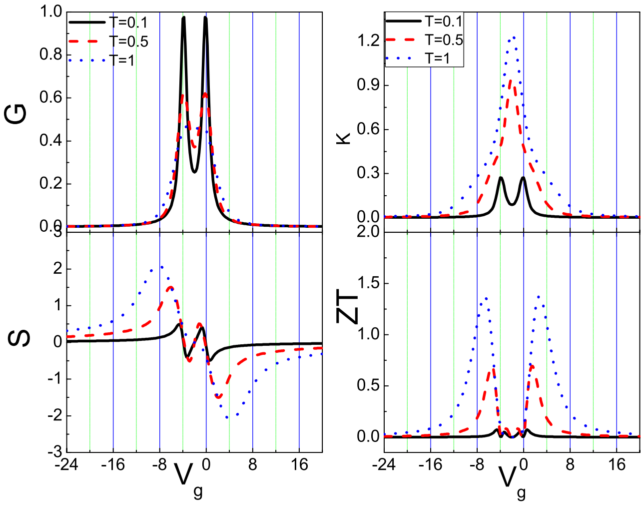

FIG. 2. (Color online) $G(2e^2/h)$, $k(2k_B/h)$, $S(k_B/e)$, and *ZT* vs V_g for the different temperature *T* for the two-levels QD and with the parameters $U=0$ and level interval $\Delta\varepsilon=4$.

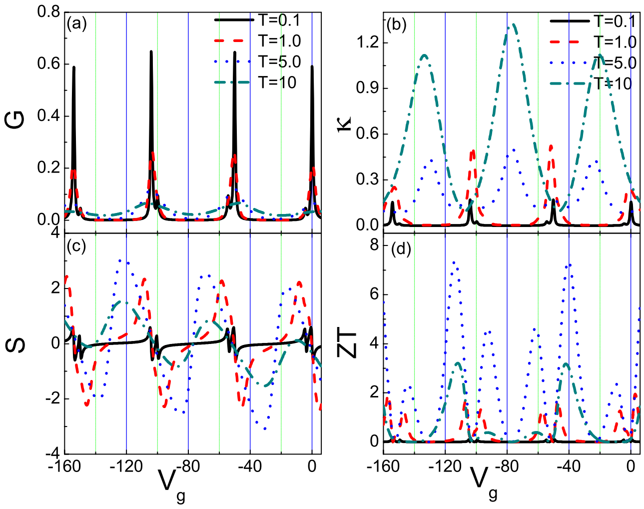

FIG. 3. (Color online) $G(2e^2/h)$, $k(2k_B/h)$, $S(k_B/e)$, and ZT vs V_g for the different temperature T for the two-levels QD with the parameters $U=50$ and level interval $\Delta\varepsilon=4$.

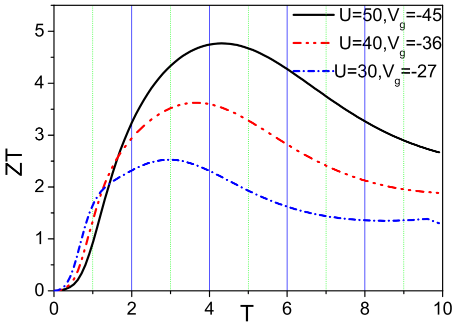

FIG. 4. (Color online) ZT vs temperature T for the different coulomb interaction U with $\Delta\varepsilon=4$. For each situation we choose a suitable gate voltage.

From Fig. 2(d), we can see that the optimized ZT is arising with temperature when temperature is much lower than the level spacing $\Delta\varepsilon$. However, when T is on the order of $\Delta\varepsilon$, the bipolar effect is enhanced and ZT is decreased. In order to increase ZT, one needs to find a QD with large enough level spacing $\Delta\varepsilon$ such that to reduce the bipolar effect. In a typical QD, the bare value of $\Delta\varepsilon$ is not big enough. An alternative way is to choose a QD with strong Coulomb interaction U. Though the Coulomb interaction may suppress ZT,[24] at the same time it can broaden the energy level. The effective level spacings can be widened to $U+\Delta\varepsilon$, which can give rise to a large ZT. In a typical QD, U can be quite large. For example, U in C_{60} is on the order of 0.1 eV.[25] In addition, the region that reduces the bipolar effect is $U > k_BT/10$. In this situation, the suppression of ZT due to Coulomb interaction can be neglected.

Now we consider that the QD contains two energy levels and both interlevel and intralevel Coulomb interactions exist. For convenience, we set the interlevel interaction to be equal to intralevel interaction. The interaction U is about one order larger than the level spacing $\Delta\varepsilon$, as for a typical QD. In the existence of the interaction U, the levels are located at ε_1, ε_1+U, $\varepsilon_1+\Delta\varepsilon+2U$, and $\varepsilon_1+\Delta\varepsilon+3U$ due to the Coulomb-blockade effect. Thus the linear-electric conductivity G exhibits four main resonant peaks at the positions of $V_g=0, -U$, $-2U-\Delta\varepsilon$, and $-3U-\Delta\varepsilon$ [see Fig. 3(a)]. In addition, there are also some smaller peaks at low temperature due to the tunneling through the excite states. Figure 3(b) shows the thermal conductivity κ versus the gate voltage. We can see that at high temperature ($k_BT\sim U$) the bipolar effect occurs and huge peaks of the thermal conductivity emerge at the valleys between the two main adjacent peaks of the conductivity.

The thermopower S shown in Fig. 3(c) is sensitive to the slope of conductivity. It is clearly seen that at low temperature the thermal power changes from positive to negative when the gate voltage moves across each peak of the conductivity (e.g., $T=0.1$). With temperature rising, the small peaks in the conductivity are absorbed to the main peaks and accordingly the transitions from positive to negative of the thermal power are reduced to four. The peak and valley values of the thermal power are enhanced with rising temperature. However, due to the bipolar effect, the peaks and valleys reach their maximum values at about $T=5$ (i.e., $U/10$). Further increasing of T will decrease these values. After G, κ, and S are calculated, the ZT can be determined, shown in Fig. 3(d). Since the Coulomb interaction U broadens the level spacing, the bipolar effect is greatly suppressed and ZT is enhanced. The optimized ZT can be over 5, much larger than the value in Fig. 2 without interaction.

To further investigate the effect of temperature and Coulomb interaction, we numerically calculate the ZT versus temperature at different Coulomb interaction U (see Fig. 4). Notice that the bipolar effect can be enhanced with increasing temperature but weakened with broadening of the energy spacing. Thus when $k_BT < U/10$, ZT is enhanced with rising temperature because of the weak bipolar effect. When $k_BT > U/10$, ZT saturates with further increasing of temperature. Moreover, with increase of U, the optimized ZT also increases.

IV. CONCLUSION

We investigate the thermoelectric properties of a QD that contains one or two levels and is in the Coulomb blockade regime. The results exhibit that in the absence of the Coulomb interaction, the ZT can be very high if only one level in the QD is considered but the ZT is greatly suppressed with multilevels due to the bipolar effect. When the Coulomb interaction U is considered in the QD, the spacings of energy levels are increased, and the bipolar effect is weakened, thus the ZT can be considerably high. For an actual QD in which its Coulomb interaction is one order larger than the level spacing, the optimized ZT can be over 5, much larger than the values from natural materials.

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APPENDIX: THE GREEN'S FUNCTION OF QD

In this appendix we give a detailed deduction on how to get the Green's function of the QD with two energy levels. First, a single QD without coupling to the leads can be described by the following Hamiltonian:

$$H_D = \sum_{i=1,2;\sigma} \varepsilon_i \hat{n}_{i\sigma} + \frac{U}{2} \sum_{i,\sigma,j,\sigma'(i\sigma \neq j\sigma')} \hat{n}_{i\sigma}\hat{n}_{j\sigma'} . \tag{A1}$$

For an isolated QD, the exact Green's function can be obtained by the equation of motion technique

$$\begin{aligned} \mathbf{g}^r_{ij\sigma} = \Bigg\{ & \frac{(\omega - \varepsilon_i - U) + U(\langle n_{i\bar{\sigma}}\rangle + \langle n_{i'\sigma}\rangle + \langle n_{i'\bar{\sigma}}\rangle)}{(\omega - \varepsilon_i)(\omega - \varepsilon_i - U)} \\ & + \frac{2U^2(\langle n_{i\bar{\sigma}} n_{i'\sigma}\rangle + \langle n_{i'\sigma} n_{i'\bar{\sigma}}\rangle + \langle n_{i\bar{\sigma}} n_{i'\bar{\sigma}}\rangle)}{(\omega - \varepsilon_i)(\omega - \varepsilon_i - U)(\omega - \varepsilon_i - 2U)} \\ & + \frac{6U^3\langle n_{i\bar{\sigma}} n_{i'\sigma} n_{i'\bar{\sigma}}\rangle}{(\omega - \varepsilon_i)(\omega - \varepsilon_i - U)(\omega - \varepsilon_i - 2U)(\omega - \varepsilon_i - 3U)} \Bigg\} \delta_{ij} . \end{aligned} \tag{A2}$$

This is an exact solution without any approximation. However, it is difficult to self-consistently calculate $\langle n_{i\sigma} n_{i'\sigma'}\rangle$ and $\langle n_{i\bar{\sigma}} n_{i'\sigma} n_{i'\bar{\sigma}}\rangle$ through numerical means, and some approximations are needed. Here, we make the approximation $\langle n_{i\sigma} n_{i'\sigma'}\rangle = 0$ while $\langle n_{i\sigma}\rangle + \langle n_{i'\sigma'}\rangle < 1$ and $\langle n_{i\sigma} n_{i'\sigma'}\rangle = \langle n_{i\sigma}\rangle + \langle n_{i'\sigma'}\rangle - 1$ while $\langle n_{i\sigma}\rangle + \langle n_{i'\sigma'}\rangle > 1$. In addition, we make another approximation $\langle n_{i\bar{\sigma}} n_{i'\sigma} n_{i'\bar{\sigma}}\rangle = 0$ while $N_{i\sigma} < 2$ and $\langle n_{i\bar{\sigma}} n_{i'\sigma} n_{i'\bar{\sigma}}\rangle = \{N_{i\sigma}\}$ while $N_{i\sigma} > 2$. These approximations are reasonable since the fluctuation of the occupation number in the QD is less than one at zero bias and the temperature $k_B T < U$.[23] Thus, when $N_{i\sigma} < 1$, the Green function can be simplified as

$$\mathbf{g}^r_{ij\sigma} = \left[\frac{1 - \{N_{i\sigma}\}}{\omega - \varepsilon_i} + \frac{\{N_{i\sigma}\}}{\omega - \varepsilon_i - U} \right] \delta_{ij} , \tag{A3}$$

when $1 < N_{i\sigma} < 2$, the Green's function can be simplified as

$$\mathbf{g}^r_{ij\sigma} = \left[\frac{1 - \{N_{i\sigma}\}}{\omega - \varepsilon_i - U} + \frac{\{N_{i\sigma}\}}{\omega - \varepsilon_i - 2U} \right] \delta_{ij} , \tag{A4}$$

when $2 < N_{i\sigma} < 3$, the Green's function can be simplified as

$$\mathbf{g}^r_{ij\sigma} = \left[\frac{1 - \{N_{i\sigma}\}}{\omega - \varepsilon_i - 2U} + \frac{\{N_{i\sigma}\}}{\omega - \varepsilon_i - 3U} \right] \delta_{ij} , \tag{A5}$$

then the final form of Green's function can be written as

$$\mathbf{g}^r_{ij\sigma}(\omega) = \left\{ \frac{1 - \{N_{i\sigma}\}}{\omega - \varepsilon_i - \{N_{i\sigma}\}U} + \frac{\{N_{i\sigma}\}}{\omega - \varepsilon_i - [\{N_{i\sigma}\} + 1]U} \right\} \delta_{ij} . \tag{A6}$$

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