Effects of interdot hopping and Coulomb blockade on the thermoelectric properties of serially coupled quantum dots

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Abstract
We have theoretically studied the thermoelectric properties of serially coupled quantum dots (SCQDs) embedded in an insulator connected to metallic electrodes. In the framework of Keldysh Green's function technique, the Landauer formula of transmission factor is obtained using the equation of motion method. Based on such analytical expressions of charge and heat currents, we calculate the electrical conductance, Seebeck coefficient, electron thermal conductance, and figure of merit (ZT) of SCQDs in the linear response regime. The effects of interdot hopping and electron Coulomb interactions on ZT are analyzed. We demonstrate that ZT is not a monotonic increasing function of interdot electron hopping strength (t_c). We also show that in the absence of phonon thermal conductance, SCQD can reach the Carnot efficiency as t_c approaches zero.

Review
Introduction
Recently, many considerable studies have been devoted to seeking efficient thermoelectric materials with the figure of merit (ZT) larger than 3 because there exist potential applications of solid-state thermal devices such as coolers and power generators [1-6]. Some theoretical efforts have pointed out that a single quantum dot (QD) junction system can have a very impressive ZT in the absence of phonon conductance [7-9]. However, in practice, it is difficult to maintain a large temperature gradient which is needed to produce sufficient temperature difference across the nanoscale junction. To reduce the temperature gradient across the QD junction, it is essential to consider many serially coupled quantum dots (SCQDs) [1,5]. The transport property of a junction involving N serially coupled QDs with strong electron Coulomb interactions is one of the most challenging topics of condensed matter physics. To gain some insight, we investigate in the present paper the thermoelectric effect of serially coupled quantum dots (SCQDs) as shown in the inset of Figure 1a.

Theoretical model
Using Keldysh-Green's function technique [13], the charge and heat currents of SCQD connected to metallic electrodes are given by

\[ J = \frac{2e}{h} \int d\epsilon \tau(\epsilon) \left[ f_L(\epsilon) - f_R(\epsilon) \right], \]

where \( \tau(\epsilon) \) is the relaxation time and \( f_L(\epsilon) \) and \( f_R(\epsilon) \) are the Fermi-Dirac distributions.

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\[ Q = \frac{2}{\hbar} \int d\epsilon \mathcal{T}(\epsilon)(\epsilon - E_F - e\Delta V) \left[f_L(\epsilon) - f_R(\epsilon)\right], \]

where \( \mathcal{T}(\epsilon) \equiv (\mathcal{T}_{12}(\epsilon) + \mathcal{T}_{21}(\epsilon))/2 \) is the transmission factor. \( f_L(\epsilon) = 1 / \left[ e^{(\epsilon - \mu_L(\epsilon))/k_B T_e} + 1 \right] \) denotes the Fermi distribution function for the left (right) electrode. The left (right) chemical potential is given by \( \mu_L(\mu_R) \). \( T_e(\epsilon) \) denotes the equilibrium temperature of the left (right) electrode. \( e \) and \( \hbar \) denote the electron charge and Planck’s constant, respectively. \( \mathcal{T}_{\ell,j}(\epsilon) \) denotes the transmission function, which can be calculated by evaluating the on-site retarded Green’s function (GF) and lesser GF [13]. The indices \( \ell \) and \( j \) denote the \( \ell \)th QD and the \( j \)th QD, respectively. Based on the equation of motion method, we can obtain analytical expressions of all GFs in the Coulomb blockade regime. Details are provided in [13]. The transmission function in the weak interdot limit \( (t_{\ell}/U_\ell \ll 1) \), where \( t_{\ell} \) and \( U_\ell \) denote the electron interdot hopping strength and on-site Coulomb interaction, respectively, can be recast into the following form:

\[ \mathcal{T}_{\ell,j}(\epsilon) = -2 \sum_{m=1}^{8} \Gamma_{\ell}(\epsilon) \Gamma_j^{m}(\epsilon) \text{Im} \mathcal{G}_{\ell,j,m,\sigma}^{\ell}(\epsilon), \]

where Im means taking the imaginary part of the function that follows and

\[ \mathcal{G}^{\ell}_{\ell,m,\sigma}(\epsilon) = p_m/(\mu_\ell - \Sigma_m). \]

\( \Gamma_{\ell}(\epsilon) = \epsilon - E_\ell + \Gamma_{\ell}/2 \) denotes the tunnel rate from the left electrode to dot A \( (E_1) \) and the right electrode to dot B \( (E_2) \), which is assumed to be energy- and bias-independent for simplicity. \( \mu_\ell = \epsilon - E_\ell + \Gamma_{\ell}/2 \). We can assign the following physical meaning to Equation 3. The sum in Equation 3 is over eight possible configurations labeled by \( m \). We consider an electron (of spin \( \sigma \)) entering level \( \ell \), which can be either occupied (with probability \( N_{\ell,\sigma} \)) or empty (with probability \( 1 - N_{\ell,\sigma} \)). For each case, the electron can hop to level \( j \), which can be empty (with probability \( a_j = 1 - N_{j,\sigma} - N_{j,\bar{\sigma}} + c_j \)), singly occupied in a spin \( \bar{\sigma} \) state (with probability \( b_{j,\bar{\sigma}} = N_{j,\bar{\sigma}} - c_j \) or spin \( \sigma \) state (with probability \( b_{j,\sigma} = N_{j,\sigma} - c_j \)), or a double-occupied state (with probability \( c_j \)). Thus, the probability factors associated with the eight configurations appearing in Equation 4 become \( p_1 = (1 - N_{\ell,\sigma}) a_\ell \), \( p_2 = (1 - N_{\ell,\sigma}) b_{\ell,\bar{\sigma}} \), \( p_3 = (1 - N_{\ell,\sigma}) b_{\ell,\sigma} \), \( p_4 = (1 - N_{\ell,\sigma}) c_\ell \), \( p_5 = N_{\ell,\sigma} a_\ell \), \( p_6 = N_{\ell,\sigma} b_{\ell,\bar{\sigma}} \), \( p_7 = N_{\ell,\sigma} b_{\ell,\sigma} \), and \( p_8 = N_{\ell,\sigma} c_\ell \). \( \Sigma_m \) in the denominator of Equation 4 denotes the self-energy correction due to Coulomb interactions and coupling with level \( j \) (which couples with the other electrode) in configuration \( m \). We have \( \Sigma_1 = \frac{t_{\ell}^2}{\mu_\ell}, \Sigma_2 = U_{\ell,j} + \frac{t_{\ell}^2}{(\mu_\ell - U_{\ell,j})}, \Sigma_3 = U_{\ell,j} + \frac{t_{\ell}^2}{(\mu_\ell - U_{\ell,j})}, \Sigma_4 = 2U_{\ell,j} + \frac{t_{\ell}^2}{(\mu_\ell - U_{\ell,j})}, \Sigma_5 = U_{\ell,j} + \frac{t_{\ell}^2}{(\mu_\ell - U_{\ell,j})}, \Sigma_6 = U_{\ell,j} + \frac{t_{\ell}^2}{(\mu_\ell - U_{\ell,j})}, \Sigma_7 = U_{\ell,j} + \frac{t_{\ell}^2}{(\mu_\ell - U_{\ell,j})}, \Sigma_8 = U_{\ell,j} + \frac{t_{\ell}^2}{(\mu_\ell - U_{\ell,j})} \). \( \mathcal{E}_\ell, \mathcal{U}_\ell, \) and \( \mathcal{U}_{\ell,j} \) denote, respectively, the energy levels of dots, intradot Coulomb interactions, and interdot Coulomb interactions. Here, \( \Gamma_j^{m} = -2\text{Im} \Sigma_0 \) denotes the effective tunneling rate from level \( \ell \) to the other electrode through level \( j \) in configuration \( m \). For example, \( \Gamma_j^1 = -2\text{Im} \frac{t_{\ell}^2}{\mu_\ell} = \frac{t_{\ell}^2}{\mu_\ell} = \frac{t_{\ell}^2}{\mu_\ell} \). It is noted that \( \Gamma_j^{m} \) has a numerator \( \Gamma_j^{m} \) for all configurations. Furthermore, \( \mathcal{G}_{\ell,j,m,\sigma}(\epsilon) = \sum_{m=1}^{8} \mathcal{G}^{\ell}_{\ell,m,\sigma}(\epsilon) \) is just the on-site single-particle retarded GF for level \( \ell \) as given in Equation (A16) of [13], and \( \mathcal{G}^{\ell}_{\ell,j,m,\sigma}(\epsilon) \) corresponds to its partial GF in configuration \( m \). The transmission function written this
way has the same form as Landauer’s formula for a single QD with multiple energy levels including intralevel and interlevel electron Coulomb interactions [14,15].

The probability factors of Equation 3 are determined by the thermally averaged one-particle occupation number and two-particle correlation functions, which can be obtained by solving the on-site lesser Green’s functions [13]:

\[ N_{L,m} = -\frac{e}{\pi} \sum_{m=1}^{8} \frac{\Gamma_{m,f}(\epsilon) + \Gamma_{m,f}(\epsilon)}{\Gamma_{L} + \Gamma_{m}} \text{Im} G_{L,m}(\epsilon) , \]  

...and

\[ c_{L} = -\frac{e}{\pi} \sum_{m=5}^{8} \frac{\Gamma_{m,f}(\epsilon) + \Gamma_{m,f}(\epsilon)}{\Gamma_{L} + \Gamma_{m}} \text{Im} G'_{L,m}(\epsilon) . \]  

Note that \( \ell \neq j \) in Equations 3, 5, and 6. In the linear response regime, Equations 1 and 2 can be rewritten as follows:

\[ J = \mathcal{L}_{11} \frac{\Delta V}{T} + \mathcal{L}_{12} \frac{\Delta T}{T^2} \]  

\[ Q = \mathcal{L}_{21} \frac{\Delta V}{T} + \mathcal{L}_{22} \frac{\Delta T}{T^2} , \]

where \( \Delta V = \mu_L - \mu_R \) and \( \Delta T = T_L - T_R \) are the voltage and temperature differences across the junction, respectively. Thermoelectric response functions in Equations 7 and 8 are given by

\[ \mathcal{L}_{11} = \frac{2e^2T}{h} \int d\epsilon \mathcal{T}(\epsilon) \left( \frac{\partial f(\epsilon)}{\partial E_F} \right)_{T} , \]  

\[ \mathcal{L}_{12} = \frac{2eT^2}{h} \int d\epsilon \mathcal{T}(\epsilon) \left( \frac{\partial f(\epsilon)}{\partial T} \right)_{E_F} , \]  

\[ \mathcal{L}_{21} = \frac{2eT}{h} \int d\epsilon \mathcal{T}(\epsilon)(\epsilon - E_F) \left( \frac{\partial f(\epsilon)}{\partial E_F} \right)_{T} , \]  

\[ \mathcal{L}_{22} = \frac{2T^2}{h} \int d\epsilon \mathcal{T}(\epsilon)(\epsilon - E_F) \left( \frac{\partial f(\epsilon)}{\partial T} \right)_{E_F} . \]

Here, \( \mathcal{T}(\epsilon) \) and \( f(\epsilon) = 1/[e^{(\epsilon - E_F)/k_B T} + 1] \) are evaluated in the equilibrium condition. It can be shown that the Onsager relation \( \mathcal{L}_{12} = \mathcal{L}_{21} \) is preserved. These thermoelectric response functions can also be found in [7], where authors investigated the thermoelectric properties of a single QD.

If the system is in an open circuit, the electrochemical potential will form in response to a temperature gradient; this electrochemical potential is known as the Seebeck voltage (Seebeck effect). The Seebeck coefficient (amount of voltage generated per unit temperature gradient) is defined as \( S = \Delta V/\Delta T = -\mathcal{L}_{12}/(T\mathcal{L}_{11}) \). To judge whether the system is able to generate power or refrigerate efficiently, we need to consider the figure of merit, which is given by

\[ ZT = \frac{S^2G_e T}{\kappa_e + \kappa_{ph}} = \frac{(ZT)_0}{1 + \kappa_{ph}/\kappa_e} . \]  

Here, \( G_e = \mathcal{L}_{11}/T \) is the electrical conductance, and \( \kappa_e = ((\mathcal{L}_{22}/T^2) - \mathcal{L}_{11}S^2) \) is the electron thermal conductance. \( (ZT)_0 \) represents the ZT value in the absence of phonon thermal conductance, \( \kappa_{ph} \). For simplicity, we assume \( \kappa_{ph} = \kappa_{ph,0}F_s \) [16-18], \( \kappa_{ph,0} = \frac{\pi^2k_B^2 T}{3h} \) is the universal phonon thermal conductance arising from acoustic phonon confinement in a nanowire [16-18], which was confirmed in the phonon wave guide [19]. The expression of \( \kappa_{ph} = \kappa_{ph,0}F_s \) with \( F_s = 0.1 \) can explain well the phonon thermal conductance of silicon nanowire with surface states calculated by the first-principles method [16]. The dimensionless scattering factor \( F_s \) arises from phonon scattering with surface impurities or surface defects of quantum dots [1,16]. Here, we adopt \( F_s = 0.02 \), which is smaller than \( F_s = 0.1 \) because QDs can enhance the phonon scattering rates and reduce phonon thermal conductance as pointed out in [1].

**Results and discussion**

Here, we consider the case of identical QDs in the optimization of ZT, although it is understood that the size fluctuation of QDs can suppress ZT [13]. In Figure 1a,b we plot \( (ZT)_0 \) and ZT as a function of temperature for various electron hopping strengths. We adopt the following physical parameters: \( \epsilon_L = EF + 30T_0, \lambda_L = 30T_0, \lambda_{L,j} = 10T_0, \) and \( \Gamma_L = \Gamma_R = \Gamma = 1\Gamma_0 \). All energy scales are in the units of the characteristic energy, \( \Gamma_0 \). In Figure 1a, we see that \( (ZT)_0 \) increases with decreasing \( \tau_c \) and diverges as \( \tau_c \to 0 \). This behavior can be proved rigorously as we shall illustrate below. It implies that SCQD can reach the Carnot efficiency in the limit of extremely weak interdot coupling, if one can fully suppress \( \kappa_{ph} \), for example, by inserting a nanoscale vacuum layer to block the phonon heat current. Although it would be a challenging task to implement a vacuum layer between one of the electrodes and SCQD, it may be possible to test this idea out via a scanning tunneling microscopic experiment using a setup as shown in the inset of Figure 1a. In Figure 1b, we see that ZT is enhanced with increasing \( \tau_c \) until \( \tau_c \) reaches \( 3\Gamma_0 \), and it becomes reduced for higher \( \tau_c \). The diverging behavior of \( (ZT)_0 \) with respect to \( \tau_c \) is further illustrated in Figure 2d. The maximum ZT is suppressed in the presence of \( \kappa_{ph} \), which is much larger than \( \kappa_e \) for small \( \tau_c \). The behaviors of ZT shown in Figure 1b are mostly determined by the power factor \( (S^2G_e) \). Once \( \tau_c \) is larger than \( 3\Gamma_0 \), the reduction of \( S^2 \) is faster than the increase of \( G_e \). This explains why the maximum ZT at
Coulomb interactions are negligible when $\Delta = \Delta_0$ and $G_0 \Delta T_1$. Such a behavior is similar to that of a single QD with multiple energy levels \cite{7,8}. The effect of electron Coulomb interactions is significant only for temperature below $6\Gamma_0$ and $50\Gamma_0$. Namely, the electron Coulomb interactions are negligible when $U/(k_B T) \gg 1$ or $U/(k_B T) \ll 1$.

To further understand the behavior of $ZT$ with respect to $t_c$, we plot the electrical conductance ($G_0$), Seebeck coefficient ($S$), electrical conductance $\kappa_e$, and $(ZT)_0$ as functions of $t_c$ in Figure 2 for various detuning energies, $\Delta \equiv E_i - E_F$. When $E_i$ is close to the Fermi energy, $G_0$ and $\kappa_e$ are enhanced, whereas $S$ and $(ZT)_0$ are suppressed. The behavior of $(ZT)_0$ at $\Delta = 30\Gamma_0$ in the absence of Coulomb interactions is also shown by the curve with triangles, which has a similar trend as the solid line. Thus, it is instructive to analyze $(ZT)_0$ in the absence of Coulomb interactions. Keeping the leading order of $t_c^2$, we have $L_{11} = \frac{2e^2}{\hbar k_B} \frac{\ell_s^2}{\gamma_1/2} \frac{1}{\cosh^2(\Delta/2k_B T)}$, $L_{12} = L_{21} = \frac{2e^2}{\hbar k_B} \frac{\ell_s^2}{\gamma_1/2} \frac{1}{\cosh^2(\Delta/2k_B T)}$, and $L_{22} = \frac{2e^2}{\hbar k_B} \frac{\ell_s^2}{\gamma_1/2} \frac{\Delta^2}{\cosh^2(\Delta/2k_B T)}$. Therefore, $G_0 \propto t_c^2$, $S = -\Delta/eT$ is independent on $t_c$, and $\kappa_e = (L_{22} - L_{12}^2/L_{11})/T^2$ vanishes up to $t_c^2$. Thus, the leading order of $\kappa_e$ is $t_c^3$. This indicates that $(ZT)_0 \propto 1/t_c^2$ in the limit of weak interdot hopping.

Figure 3 shows $ZT$ as a function of $\Delta = E_i - E_F$ for various electron hoping strengths at $k_B T = 10\Gamma_0$. Other physical parameters are kept the same as those for Figure 1. When $t_c = 0.1\Gamma_0$, the maximum $ZT$ $(ZT_{\text{max}})$ occurs near $\Delta = 27\Gamma_0$. The peak position only shifts slightly to higher $\Delta$ with increasing $t_c$. We have $ZT_{\text{max}} = 2.79$ and $3.18$ for $t_c = 10\Gamma_0$ and $30\Gamma_0$, respectively. However, at $t_c = 4\Gamma_0$, we have $ZT_{\text{max}} = 3.07$, which is smaller than $ZT_{\text{max}}$ for $t_c = 30\Gamma_0$. Thus, it also illustrates that $ZT$ is not a monotonically increasing function of $t_c$. We further calculated $ZT$ as a function of $t_c$ for $\Delta = 10, 20, 30\Gamma_0$ and $k_B T = 10\Gamma_0$ in the presence of $\kappa_{ph}$ and found that again, $ZT$ is not a monotonically increasing function of $t_c$ (not shown here). We conclude that as long as $\kappa_{ph}$ dominates over $\kappa_e$, the $t_c$ dependence of $ZT$ is mainly determined by the power factor $S^2 G_e$, where the behaviors of $G_e$ and $S$ are similar to the results shown in Figure 2a,b. When $t_c/\Gamma_0 \leq 1$, $G_e$ increases much faster than the reduction of $S^2$ for increasing $t_c$, and the power factor slowly reaches the maximum when $t_c$ approaches $3\Gamma_0$. When $t_c > 3\Gamma_0$, the power factor decreases due to the fast reduction of $S^2$ which prevails over the increase of $G_e$. The curve with triangle marks is for $t_c = 30\Gamma_0$ in the absence of Coulomb interaction. We see that $ZT_{\text{max}}$ is maximal when $U_i = U_{ij} = 0$. Based on the results of Figure 3, we conclude that it is important to control the detuning energy $\Delta$ for the optimization of $ZT$.

In Figures 1, 2, and 3 we have considered the case with $E_F$ below QD energy levels. It would be interesting to investigate the case with $E_F$ above the energy levels of QDs. Figure 4 shows $G_e$, $S$, $\kappa_e$, and $ZT$ of an SCQD with $t_c = 31\Gamma_0$ as functions of applied gate voltage for various temperatures. Once $t_c > (\Gamma_i + \Gamma_j) = 2\Gamma_0$, the eight peaks for $G_e$ can be resolved at $k_B T = 1\Gamma_0$. These eight peaks
Figure 3 ZT as a function of $\Delta$ for different electron hopping strength at $k_B T = 10 \Gamma_0$. Other parameters are the same as those of Figure 1.

Figure 4 Function of applied gate voltage. (a) $G_e$, (b) $S$, (c) $\kappa_e$, and (d) ZT as a function of applied gate voltage for $k_B T = 1 \Gamma_0$ (solid), $2 \Gamma_0$ (dashed), and $3 \Gamma_0$ (dotted). $E_\ell = E_F + 10 \Gamma_0$ and $t_c = 3 \Gamma_0$. Other parameters are the same as those of Figure 1. The curves with triangle marks are for the case without electron Coulomb interactions for $k_B T = 3 \Gamma_0$.

correspond to the following resonant channels: $E_\ell - t_c$, $E_\ell + t_c$, $E_\ell + U_{ij} - t_c$, $E_\ell + U_{ij} + t_c$, $E_\ell + U_{ij} + U_{ij} - t_c$, $E_\ell + U_{ij} + U_{ij} + t_c$, $E_\ell + 2U_{ij} + U_{ij} - t_c$, and $E_\ell + 2U_{ij} + U_{ij} + t_c$, which are tuned by the gate voltage to be aligned with $E_F$. These eight channels result from the four configurations of $p_1$, $p_3$, $p_6$, and $p_8$ in Equation 4. Such a result implies that SCQD with identical QDs acts as a QD with effective two levels of $E_\ell - t_c$ and $E_\ell + t_c$ and satisfying Hund’s rule.

These eight peaks are smeared out with increasing temperature. The sign changes of $S$ with respect to the gate voltage result from the bipolar effect, i.e., the competition between electrons and holes, where holes are defined as the unoccupied states below $E_F$ [13]. The electronic thermal conductance ($\kappa_e$) also exhibits eight peaks, and we noticed that the local maxima of the $\kappa_e$ curve nearly coincide with the local minima of the $G_e$ curve. We see that ZT...
values are still larger than 3 even when \( E_F \) is deeply below \( E_F \) (say, at \( eV_g = 70\Gamma_0 \)). This is attributed to the electron Coulomb interaction. To illustrate that, we also show the results with \( U_j = U_{ij} = 0 \) at \( k_BT = 3\Gamma_0 \) (see the curve with triangle marks). The oscillation of ZT in the case of \( U_j = U_{ij} = 0 \) is attributed to the sign change of \( S \) at \( V_g = 10\Gamma_0 \). Note that \( S \) goes to zero at \( V_g = 10\Gamma_0 \), which results from the electron-hole symmetry (with \( E_i + t_c \) and \( E_i - t_c \) straddling \( E_F \) symmetrically). We see that ZT vanishes for \( eV_g > 40\Gamma_0 \) in the absence of electron Coulomb interactions. Unlike the case of \( E_F < E_R \), where the finite \( U \) causes reduction of ZT, here, the electron Coulomb interaction leads to enhancement of ZT when \( E_F > E_R \).

Conclusions

In summary, the thermoelectric properties including \( G_s \), \( S \), \( k_r \), and ZT of the SCQD junction system are investigated theoretically. We demonstrate that the Carnot efficiency can be reached when \( t_c \) approaches zero in the absence of phonon thermal conductance. When the phonon contribution dominates the thermal conductance of the SCQD junction, the optimization of ZT can be obtained by the thermal power defined as \( S^2G_r \). We also found that the presence of electron Coulomb interactions can lead to either reduction or enhancement of ZT, depending on whether the Fermi level is below or above the QD level.

Competing interests

The authors declare that they have no competing interests.

Authors’ contributions

DMTK and Y-CC established the theoretical formalism. DMTK carried out the numerical calculations and drafted the manuscript. Y-CC conceived this study and participated in its coordination. All authors read and approved the final manuscript.

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