A Strongly Correlated Quantum Dot Heat Engine with Optimal Performance: A Nonequilibrium Green’s Function Approach

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Herein, an analytical study of a strongly correlated quantum dot-based thermoelectric particle-exchange heat engine for both finite and infinite on-dot Coulomb interaction is presented. Employing Keldysh’s nonequilibrium Green’s function formalism for different decoupling schemes in the equation of motion, the thermoelectric properties within the nonlinear transport regime have been analyzed. Initially, Hubbard-I approximation has been used to study the quantum dot level position (ε_d), thermal gradient (∆T), and on-dot Coulomb interaction (U) dependence of the thermoelectric properties. Furthermore, as a natural extension, a decoupling beyond Hubbard-I (Lacroix approximation) with infinite-U limit (strong on-dot Coulomb repulsion) has been used to provide additional insight into the operation of a more practical quantum dot heat engine. Within this infinite-U limit, the role of the symmetric dot-reservoir tunneling (I) and external serial load resistance (R) in optimizing the performance of the strongly correlated quantum dot heat engine is examined. The infinite-U results show a good quantitative agreement with recent experimental data for a quantum dot coupled to two metallic reservoirs.

1. Introduction

The heat and thermoelectric transport properties of hybrid quantum dot(s) nanostructures are recently attracting great attention experimentally and theoretically due to their potential applications in solid-state thermal devices.[1–9] One such hybrid nanostructure is a quantum dot (QD) particle-exchange heat engines. The particle-exchange heat engines consist of quantum dot(s) connected to metallic source and drain reservoirs by tunnel junctions.[10–18] Due to quantum confinement and Coulomb blockade (CB) effects on QD, these low-dimensional heat engines are more efficient at converting thermal energy into electrical energy than their bulk counterparts.[19–24] The enhancement of thermoelectric efficiency in QD heat engines is due to the strong violation of the Wiedemann–Franz law[25–31] and the significant drop in lattice thermal conductivity.[32–35]

In QD heat engines, electrons tunnel from the left hot reservoir to the QD and then to the right cold reservoir, resulting in a thermovoltage, or voltage caused by the temperature difference between the reservoirs due to the Seebeck effect. Thus, the temperature difference can drive an electric current in the external circuit. The power output generated by the heat engine is available for consumption in an external serial load R (Figure 1). This thermoelectric power conversion could be useful for future energy harvesting in nanoelectronic devices and quantum technologies.[4,6] Apart from their potential practical uses, these hybrid QD systems can also be used as a testing ground for various theoretical techniques to further analyze the transport properties of novel strongly correlated materials.

Thermovoltage, thermopower, and thermoelectric performance of hybrid normal-metal QD heat engines have been widely studied using various theoretical techniques such as equation of motion (EOM),[36–44] slave Boson,[27,45] perturbation theory,[25,26,28,46] master equation approach,[10,30,47–52] Wilson’s numerical renormalization-group (NRG),[53–55] and functional renormalization-group (FRG) method.[56] The majority of these papers focused on the linear response regime, i.e., for small thermal gradients (δT → 0) and small voltage biases (eV → 0). In the linear response regime, the thermoelectric conversion efficiency is characterized by a dimensionless figure of merit $\eta = S^2GT/B$, where $T$ is the absolute temperature, $S$ is the Seebeck coefficient (or thermopower), $G$ is electrical conductance, and $K$ is thermal conductance (includes both electron and phonon contributions). For larger $ZT$, the thermoelectric conversion efficiency ($\eta$) approaches its upper bound, i.e., the Carnot efficiency ($\eta_c$). The efficiency for short-lived quantum states ($\Gamma/k_B T >> 1$, where $k_B$ is Boltzmann constant) is relatively low, while it approaches the Carnot efficiency for long-lived quantum states (i.e., for $\Gamma/k_B T << 1$). The enhancement in efficiency is caused by sequential tunneling events (i.e., lowest order incoherent tunneling processes), which dominate transport at relatively higher temperatures (but $U >> k_B T$) and violate the Wiedemann–Franz law by suppressing thermal conductance in comparison to charge conductance.

The influence of asymmetric dot-reservoir coupling,[43] electron–electron interactions,[26–30,40–42,47–57] Kondo effect,[25,26,31,44,54,55] and small voltage biases (eV → 0) in the linear response regime.

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energy spectrum of the QD,[43,47,52] electron–phonon interactions,[7,48,58,59] and the Fano effect[60–62] on the thermoelectric properties of QD heat engines has also been investigated in the linear and nonlinear regime. Krawiec et al.[45] studied the influence of asymmetric dot–reservoir coupling ($\Gamma_L \neq \Gamma_R$) on thermoelectric properties and showed that in the linear regime, the thermopower $S$ and figure of merit $ZT$ are unaffected by asymmetry. On the other hand, the nonlinear $S$ strongly depends on asymmetry. The effects of electron–electron Coulomb interaction on thermoelectric properties were analyzed in several works. However, in the nonlinear regime, the effect of Coulomb interaction on the thermovoltage or $S$, thermocurrent, and thermoelectric power has been studied in a few works.[37,40,42] At very low temperatures ($T \leq T_K$, where $T_K$ is the Kondo temperature), where the Kondo effects develop and the ground state shows a Fermi liquid behavior, the Wiedemann–Franz law is recovered.[26,31,54] Both $S$ and $ZT$ for $T \leq T_K$ are rather small and can be enhanced in the relatively high-temperature regime ($T \gg T_K$). The effect of multilevels of QD on the thermoelectric properties has been investigated in refs. [43,47,52] within the sequential tunneling regime. Also, at finite electron–phonon coupling, the decrease in the power factor $S^2 GT$ and increase in $K$ can significantly reduce $ZT$.[58] However, the electron–electron–phonon coupling is small at low temperatures and can be effectively suppressed in the experiments. Recently, Taniguchi[62] showed that in the nonlinear regime, even if the temperature is much lower than the dot-reservoir tunneling rate ($\Gamma/k_B T \gg 1$), which is unfavorable for good thermoelectric conversion efficiency, one can still achieve reasonably good thermoelectric performance by regulating quantum coherence via the Fano effect. Other theoretical studies have also shown that optimal thermoelectric operation is possible in the nonlinear transport regime.[63–68]

On the other hand, the experimental studies on thermoelectric transport properties of QD heat engine are limited.[69–82] Only a couple of these experiments have focused on measuring power output and corresponding thermoelectric efficiency.[78–80] Recently, Josefsson et al.[78] demonstrated that for $\Gamma \ll k_B T$ with strong on-set Coulomb interaction ($U \gg k_B T$), a QD heat engine can achieve a thermoelectric conversion efficiency close to 70% of the Carnot limit. These experiments also required additional theoretical research into the practical optimization of QD-based heat engines, particularly the impact of nonlinear effects and external circuit elements on the performance of these nanodevices.

In the present work, we contribute to previous studies by analyzing the nonlinear transport regime as well as the effect of the external load resistance on the practical optimization of the performance of QD-based heat engines (Figure 1). The present study not only quantitatively reproduces the recent experimental results[78] in the weak dot-reservoir coupling and strong Coulomb blockade regime ($U \gg k_B T \gg \Gamma$) but also discusses the issues which have not received much attention in the previous studies such as the variation of the nonlinear thermoelectric transport properties with the thermal gradient $\Delta T$ for both weak and strong Coulomb interaction. Also, in the previous studies, the thermoelectric performance of the QD-based particle-exchange heat engines was calculated using the open-circuit condition, i.e., $I_C = 0$. However, in practical QD-based heat engines, the presence of the external serial load $R$ also affects the optimal power output and corresponding thermoelectric efficiency. Therefore in the present study, we investigate the effect of external load resistance $R$ on the thermoelectric performance of the single QD heat engine. Further, we also extend the parameter regime considered in the experimental study and analyze the effect of $R$ on the thermoelectric performance in both weak and strong coupling regimes.

The rest of the article is organized as follows. The preceding Section 2 contains the model Hamiltonian and theoretical description. In Section 3, the numerical results and discussion of the nonlinear transport regimes are provided. In Section 4, we conclude the present work.

2. Model Hamiltonian and Theoretical Description

The system under consideration consists of a single-level QD coupled between the normal metallic source and drain reservoirs. Such a system is modeled by a single-impurity Anderson Hamiltonian in second quantization formalism[83,84]

$$\hat{H} = \hat{H}_{\text{reservoirs}} + \hat{H}_{\text{QD}} + \hat{H}_{\text{tunnel}}$$

where

$$\hat{H}_{\text{reservoirs}} = \sum_{k \sigma, \alpha \in L,R} (\epsilon_k \hat{c}^\dagger_{k \sigma \alpha} \hat{c}_{k \sigma \alpha})$$

represents the Hamiltonian of the metallic reservoirs, where $\epsilon_k$ is the kinetic energy of the electrons in the reservoirs, and $c_{k \sigma} (c_{k \sigma}^\dagger)$ is the annihilation (creation) operator of an electron with spin $\sigma$ and wave vector $k$.

$$\hat{H}_{\text{QD}} = \sum_{\sigma} \epsilon_d n_{d\sigma} + U n_{d\uparrow} n_{d\downarrow}$$

represents the Hamiltonian of the QD with $n_{d\sigma} = \hat{d}_{\sigma}^\dagger \hat{d}_{\sigma}$ as the number operator and $\epsilon_d$ is the annihilation (creation) operator of electron with spin $\sigma$. The QD consists of a single electronic level of energy $\epsilon_d$ and can be occupied by up to two electrons, i.e., $\sigma \in \uparrow, \downarrow$. We also
consider the intradot or on-dot electron–electron Coulomb repulsion with the interaction strength $U$.

$$F_{\text{tunnel}} = \sum_{k\sigma}(V_{k,\sigma}d_{k\sigma}^{\dagger}c_{k\sigma} + V_{k,\sigma}^{*}c_{k\sigma}^{\dagger}d_{k\sigma})$$ represents the tunneling Hamiltonian between the QD and reservoirs with coupling strength $V_{k\sigma}$.

Using the nonequilibrium Green’s function formalism, the electric (charge) current $I_C$ and heat current $J_Q$ from left to right reservoir across the QD for symmetric tunneling rate ($f_L = f_R = \Gamma$) can be expressed as\textsuperscript{[86,87]}

$$I_C = \frac{2e}{\hbar} \int \left[ f_L(\omega - \mu_I) - f_R(\omega - \mu_R) \right] T(\omega) d\omega$$  \hspace{1cm} (2)

$$J_Q = \frac{2}{\hbar} \int \left[ (\omega - \mu_I)f_L(\omega - \mu_I) - f_R(\omega - \mu_R) \right] T(\omega) d\omega$$  \hspace{1cm} (3)

where $T(\omega) = \Gamma^2|G_{\text{d},\alpha}(\omega)|^2$ is the tunneling amplitude or transmission function and $f_{\omega}(\omega \pm \mu_I) = \exp[\exp(\omega \pm \mu_I)/k_B T_r]^{-1}$ is the Fermi–Dirac distribution function of $\alpha$-reservoir with chemical potential $\pm \mu_I$ and temperature $T_r$.

$$G_{\text{d},\alpha}(\omega) = \langle \langle d_{\sigma}\rangle \rangle_{\alpha}(\omega)$$

is the Fourier transformation of the single-particle retarded Green’s function for the QD in time domain (i.e., $G_{\text{d},\alpha}(t - t') = \pi \delta(t - t') \langle \langle d_{\sigma}(t), d_{\sigma}^{\dagger}(t') \rangle \rangle$).

To solve the single-impurity Anderson model Hamiltonian (Equation (1)), we apply the Green’s function EOM method in Fourier space with Zubarev notation\textsuperscript{[86,87]}. For a nonzero Coulomb interaction, an EOM for a given Green’s function involves higher order coupled Green’s functions or correlation functions, thus creating a hierarchy of equations. In order to truncate the hierarchy of equations, one needs a decoupling scheme or approximation for higher order correlation functions.

In the present work, we use Hubbard-I\textsuperscript{[18,88,89]} and Lacroix\textsuperscript{[90,91]} approximations to study the CB regime ranging from weak to strong Coulomb correlation. The Hubbard-I decoupling scheme takes into account the Coulomb interaction by using a mean-field treatment. The Lacroix approximation, on the other hand, is capable of accounting for the Coulomb interaction more accurately and is particularly useful for studying the strong Coulomb blockade effects.

It is important to note that the Green’s function EOM technique used in the present study is reliable and quantitatively accurate for studying the Coulomb blockade effect at $T \gg T_K$. This is because the lowest order tunneling processes are accurately captured in approximations used to truncate a hierarchy of coupled equations, allowing for a precise determination of the Coulomb blockade effect. However, because of its approximation nature and inability to handle higher order processes including spin flip, this method is restricted to a qualitative description of the Kondo effect at $T \ll T_K$\textsuperscript{[92]}

For simplification we assume that the coupling strength is $k$ independent, i.e., $V_{k,\sigma} = V_\sigma$ for $V_{k,\sigma} < D$ (wide band) where $-D \leq \epsilon_{k\sigma} \leq D$ and the tunneling rate from dot to the $\alpha$-leads is defined by $\Gamma_{\alpha} = 2\pi V_{\alpha}^2 \rho_{\text{tot}}$, where density of states of metallic reservoir $\rho_{\text{tot}}$ is constant in the range of energy around Fermi level (flat band).

The expression for the single-particle retarded Green’s function of an electron with spin $\sigma$ on the QD within the Hubbard-I approximation is given by

$$G_{\text{d},\alpha}(\omega) = \left( \omega - \epsilon_d - U(1 - n_{\text{d},\alpha}) \right) / \left( (\omega - \epsilon_d)(\omega - \epsilon_d - U) - \Sigma_0(\omega - \epsilon_d - U(1 - n_{\text{d},\alpha})) \right)$$  \hspace{1cm} (4)

Also, the expression for single-particle retarded Green’s function of an electron with spin $\sigma$ on the QD within the Lacroix approximation is given by

$$G_{\text{d},\alpha}(\omega) = \left[ 1 + \frac{U(n_{\text{d},\alpha})}{\omega - \epsilon_d - \Sigma_0 + \omega - 2\epsilon_d - U + \epsilon_{k,\sigma}} \right]$$  \hspace{1cm} (5)

where

$$\Sigma_0 = \sum_{k\sigma} V_{k\sigma}^2 \left[ \frac{1}{\omega - \epsilon_{k\sigma}} \right]$$ & $$\Sigma_i = \sum_{k\sigma} V_{k\sigma}^2 \left[ \frac{1}{\omega - \epsilon_{k\sigma}} + \frac{1}{\omega - 2\epsilon_d - U + \epsilon_{k,\sigma}} \right]$$  \hspace{1cm} (6)

with $A_{k,\sigma} = f_{\alpha}(\epsilon_{k,\sigma})$ and $A_{k,\alpha}^{(2)} = 1$ for $i = 1, 2$.

Expressions in Equation (6) can be simplified further by replacing $\sum_{k\sigma} f_{\alpha}(\epsilon_{k,\sigma}) d_{k,\sigma}$ and then solving these equations analytically using the complex integration for the flat $f_{\alpha}(\epsilon_{k,\sigma}) \rightarrow \rho_{\text{tot}}$ and wide band limit ($D \rightarrow \infty$).

For strong Coulomb blockade limit (i.e., $U \rightarrow \infty$) above Green’s function (Equation (5)) become

$$G_{\text{d},\alpha}(\omega) = \frac{1 - \langle n_{\text{d},\alpha} \rangle}{\omega - \epsilon_d - \Sigma_0}$$  \hspace{1cm} (7)

with $\Sigma_0 = -\sum_{\alpha} \frac{\pi}{2} \Gamma_{\alpha}$, and $\Sigma_1 = -\sum_{e} \frac{\pi}{2} \left[ \frac{\pi}{2} + \psi \left( \frac{1}{2} + \frac{\pi}{2\epsilon_{k,\sigma}} \right) \right]$ where $\psi(\ldots)$ is the digamma function.

Once the single-particle retarded Green’s function of QD is known, the average occupation on the QD ($\langle n_{\text{d},\alpha} \rangle = \langle n_{\text{d},\alpha} \rangle$ for nonmagnetic system) is calculated by using the self-consistent integral equation of the form

$$\langle n_{\text{d},\alpha} \rangle = -\frac{i}{2\pi} \int_{-\infty}^{\infty} G_{\text{d},\alpha}(\omega) d\omega$$

where $G_{\text{d},\alpha}(\omega)$ is the Fourier transformation of the single particle lesser Green’s function for the QD in time domain (i.e., $G_{\text{d},\alpha}(t - t') = i\theta(t - t') \langle \langle d_{\sigma}(t), d_{\sigma}^{\dagger}(t') \rangle \rangle$) and obeys the Keldysh equation\textsuperscript{[86,91]}

$$G_{\text{d},\alpha}^{(\rangle)}(\omega) = G_{\text{d},\alpha}(\omega) \Sigma_{\alpha}\Sigma_{\alpha}^*$$  \hspace{1cm} (8)

where $G_{\text{d},\alpha}^{(\rangle)}(\omega) = \left[ G_{\text{d},\alpha}(\omega) \right]^*$ is the advanced Green’s function and $\Sigma_{\alpha}(\omega) = -\sum_{\sigma} \Sigma_{\alpha} - \Sigma_{\alpha}\Sigma_{\alpha}^* \Sigma_{\alpha}$ is the lesser self-energy.

Thus, the lesser Green’s function for single particles with spin $\sigma$ on the QD is given by

$$G_{\text{d},\alpha}^{(\langle)}(\omega) = \frac{i}{2} \left[ \Gamma_{\alpha}\Sigma_{\alpha} + \Gamma_{\alpha}\Sigma_{\alpha}^* \right] G_{\text{d},\alpha}^{(\rangle)}(\omega)$$  \hspace{1cm} (9)

For the QD-based particle-exchange heat engine, the temperature of the left reservoir is considered as $T_\text{L} = T + \Delta T$ while the
right reservoir remains at the background temperature \( T_R = T \). Due to this temperature difference, electrons move from left hot reservoir to the right cold reservoir and create a potential difference \( (V_{th} = (\mu_l - \mu_R) / e) \) due to accumulation of electron on the right reservoir and positive charge to the left reservoir (see Figure 1). Thus, a thermally induced charge current flows through the external circuit. Further, to achieve optimal thermoelectric conversion we consider symmetric dot-reservoir tunneling rate, i.e., \( \Gamma_L = \Gamma_R = \Gamma \).64\]

Usually, the thermovoltage \( (V_{th}) \) is determined from the open-circuit condition \( I_C(V_{th}, \Delta T) = 0 \), i.e., external reverse bias voltage \( V_{ext} = (\mu_R - \mu_L) / e \) is applied to counteract the thermally induced charge current. The thermopower \( S = V_{th}/\Delta T \) and thermal conductance \( K = J_Q/\Delta T \) are then calculated. In open-circuit condition, the maximal power generated by the heat engine is \( P_{max} = -(I_C)_{\text{max}} V_{max} = -((I_C)_{\text{max}}/2)(V_{th}/2) \). However, if the external voltage source is replaced by the load resistance \( R \), then the current has to self-consistently satisfy the equation

\[
I_C(V_{th}, \Delta T) + V_{th}/R = 0 \quad (11)
\]

Equation (11) is solved numerically to obtain thermovoltage \( V_{th} \).

The finite power output \( P = -I_C V_{th} = J_Q R \) generated by the QD heat engine dissipates across the load resistance \( R \). Thus, only optimizing \( V_{th} \) and \( e_d \) are not sufficient to reach the maximum power because \( P \) also depends on external load resistance \( R \). The efficiency of the heat engine at maximal power output is calculated by using \( \eta_{P_{max}} = P_{max}/J_Q \), which can be normalized by the Carnot efficiency \( \eta_C = 1 - T_R/T_L \). It is also important to note that we only consider the thermal contribution by electrons and neglected the phonon contribution which is small at low temperatures and can also be suppressed effectively in hybrid QD devices.

3. Results and Discussion

The equations derived in the previous section within nonlinear transport regimes are numerically solved using MATLAB, with \( \Gamma_0 \) (in meV) as the energy unit. In this section, we study the effect of 1) thermal gradient \( \Delta T \), on-dot Coulomb interaction \( U \), and 2) external load resistance \( R \) on the thermoelectric transport properties of a QD heat engine through the linear response regime. We have used both Hubbard-I (for finite- \( U \) limit) and beyond Hubbard-I approximation (for infinite- \( U \) limit). Finally, the theoretical results for a QD heat engine connected with external serial load resistance are compared with the recent experimental data.

3.1. Effect of Thermal Gradient and Coulomb Interaction

In Figure 2, we have shown the nonlinear thermopower \( S \), electronic thermal conductance \( K \), and thermoelectric efficiency at maximum power output \( \eta_{P_{max}} \) as a function of QD energy level \( e_d \) and applied thermal gradient \( \Delta T \) for different on-dot Coulomb interaction \( U \). Here, we have used the open-circuit condition, i.e., \( I_C(V_{th}, \Delta T) = 0 \), to calculate the thermoelectric properties of the QD heat engine.

The thermopower in Figure 2a–c is antisymmetric around \( e_d = -U/2 \) due to electron–hole symmetry, and positive (negative) thermopower indicates that holes (electrons) are the majority charge carriers. Due to finite on-dot Coulomb repulsion \( U \), two effective levels lie at \( e_d = e_d + U \) and resonance energies (i.e., for \( e_d = 0 \) and \( e_d + U = 0 \), the electron and hole charge current compensate each other, and thus no thermopower is observed at these points (indicated by dashed lines). If the QD energy level lies above the Fermi resonance energy, i.e., \( e_d > 0 \), more electrons tunnel from the hot reservoir to the cold reservoir. This net flow of electrons from the left to right reservoir gives rise to the negative thermopower (due to negative electron charge \( e \)) under open-circuit conditions. However, if \( -U/2 < e_d < 0 \), then the effective level at \( e_d \) lies close to the Fermi resonance energy from below, and more electrons flow from the right to left reservoir, and thermopower becomes positive. The magnitude of thermopower for \( -U/2 < e_d < 0 \) decreases with increasing thermal gradient \( \Delta T \) as a result of the reduced Coulomb blockade effect due to enhancement in the tunneling of thermally excited electrons from left to the right reservoir. Similarly, one can explain the origin of negative and positive thermopower for \( e_d < -U/2 \). The \( S \) versus \( e_d \) curves for \( e_d > 0 \) and \( e_d < -U \) become flat with increasing \( \Delta T \) due to thermal broadening of the Fermi–Dirac distribution function.

The plots for electronic contribution to the thermal conductance \( K \) are shown in Figure 2d–f. It is clear from these plots that \( K \) shows minima at the resonance energies \( (e_d = 0 \) and \( e_d + U = 0) \) for all values of finite \( \Delta T \) and maxima at the electron–hole symmetry point \( (e_d = -U/2) \) for moderate or relatively larger \( \Delta T \) (depending on \( U \)). This behavior differs from the linear response results, which show that at low temperatures, thermal conductance behaves similarly to electric conductance, i.e., it shows maxima at the resonance energies and minima at the electron–hole symmetry point. However, at a relatively larger temperature or thermal gradient, the resonant tunneling is significantly reduced, and thus, the electrical conductance and, eventually, \( K \) is also suppressed at \( e_d = 0 \) and \( e_d + U = 0 \). For \( e_d > 0 \) and \( e_d < -U \), the thermal conductance is relatively large due to thermally excited electron tunneling and becomes flat as \( \Delta T \) increases due to the thermal broadening of the Fermi–Dirac distribution function. On the other hand, the electron and hole contributions to thermal conductance add constructively near the electron–hole symmetry point, leading to enhanced \( K \) for moderate or relatively larger \( \Delta T \) if \( -U < e_d < 0 \). With increasing on-dot Coulomb interaction \( U \), the effective QD energy levels \( e_d = -U/2 \) and \( e_d + U = U/2 \) move further away from resonance and thus require higher \( T \) or \( \Delta T \) for the tunneling of thermally excited electrons.

The normalized thermoelectric conversion efficiency at maximum power output \( \eta_{P_{max}} \) in Figure 2g–i exhibits similar behavior to that of thermopower, i.e., \( \eta_{P_{max}} \) becomes minimal at resonances, and the electron–hole symmetry point. The CB effect significantly increases \( \eta_{P_{max}} \) between these points but decreases as \( \Delta T \) and, eventually, \( K \) increases further. However, in experimental studies, \( \Delta T \leq T \) and on-dot Coulomb interaction \( U \) is the largest energy parameter. Therefore, from now on, we consider these parameter values when calculating \( \eta_{P_{max}} \) and other thermoelectric properties.
Nex, we take into account very strong Coulomb repulsion on the QD energy level, i.e., infinite-$U$ limit. The variation of thermoelectric transport quantities as a function of QD energy level $\varepsilon_d$ for different thermal gradients $\Delta T$ is shown in Figure 3. For strong on-dot Coulomb repulsion, transport occurs via one single effective energy level $\varepsilon_d$. As a result, electrons tunnel sequentially one at a time, and the number of electrons $N$ on the QD is either 1 or 0 depending on the position $\varepsilon_d$ w.r.t. Fermi resonance energy (see inset in Figure 3b). The thermoelectric transport quantities are minimal at the resonance energy $\varepsilon_d = 0$, as in the noninteracting QD case. However, the peaks in these thermoelectric transport quantities across $\varepsilon_d$ are asymmetric due to asymmetry in the tunneling amplitude for even ($N = 0$) and odd ($N = 1$) electrons on the strongly interacting QD energy level. When $\varepsilon_d > 0$, the average number of electrons on QD is zero ($N = 0$). As a result, electrons with both up and down spin can tunnel to the QD from the reservoir. For $\varepsilon_d < 0$, the QD energy level is already occupied by a single electron ($N = 1$), and only an electron with opposite spin can tunnel to the QD from the reservoir, which leads to a relatively smaller tunneling probability and current. Thus, due to this reason, the peaks in thermal conductance $K$ and maximum power output $P_{\text{max}}$ are low when $\varepsilon_d < 0$ and relatively high when $\varepsilon_d > 0$. On the other hand, low thermal conductance and large power output are required for optimal $S$ and $\eta_{\text{Pmax}}$. Due to the significant asymmetry in $K$ relative to $P_{\text{max}}$, the peaks in $S$ and $\eta_{\text{Pmax}}$ are relatively high when $\varepsilon_d < 0$. Furthermore, for $\Delta T < T$, thermopower $S$ is independent of $\Delta T$ in the range $-\Gamma_0 < \varepsilon_d < \Gamma_0$ due to linear increase in thermovoltage $V_{\text{th}}$ (inset in Figure 3a). Also, the magnitude of thermal conductance $K$ in Figure 3b is not very sensitive to the variation in $\Delta T$. The maximum power output $P_{\text{max}}$ and normalized efficiency $\eta_{\text{Pmax}}$ are increased by approximately 300 times and 14%, respectively, due to a significant increase in thermovoltage and thermocurrent when $k_B\Delta T$ is increased from $0.01\Gamma_0$ to $0.2\Gamma_0$. 

Figure 2. Variation of nonlinear a–c) thermopower ($S$), d–f) thermal conductance ($K$), and g–i) normalized efficiency at maximum power output ($\eta_{\text{Pmax}}$) as a function of temperature difference ($\Delta T$) between the reservoirs and the QD energy level ($\varepsilon_d$) for different on-dot Coulomb repulsion $U$ with $k_B T = 0.2\Gamma_0$, $\Gamma = 0.1\Gamma_0$, and $\Gamma_0$ is the energy unit. The results are obtained within the Hubbard-I approximation for finite $U$ by using Green’s function EOM technique.
Equation (11). For $R$, the external voltage source with a serial load resistance is used in two limiting cases, i.e., for short circuit ($R = 0$) and open circuit ($R = \infty$). The results are obtained within the decoupling scheme beyond Hubbard-I for $U \rightarrow \infty$ limit. The other parameters are: $k_B T = 0.2 \Gamma_{B}$ and $\Gamma = 0.1 \Gamma_{B}$. Inset in (a) shows the variation of thermovoltage $V_{\text{th}}$ with $\epsilon_d$, inset in (b) shows the variation of average QD occupancy $\langle n_{hi} \rangle$ with $\epsilon_d$, and inset in (c) shows the close-up view of $P_{\text{max}}$ for very small $\Delta T$.

### 3.2. Effect of External Load Resistance ($R$) on the Performance of QD Heat Engine

In the previous subsection, the thermoelectric performance of a QD heat engine was calculated using an open circuit condition, i.e., an external reverse bias voltage $V_{\text{ext}} = (\mu_R - \mu_L)/e$ is applied to counteract the thermally induced charge current. Let us now investigate the effect of external load resistance $R$ on the thermoelectric performance of the single QD heat engine by replacing the external voltage source with a serial load resistance $R$. For finite $R$, the current must self-consistently satisfy Equation (11). In Figure 4, maximum power output $P_{\text{max}}$ and corresponding efficiency $\eta_{\text{max}}$ are plotted as a function of $\Gamma/k_B T$ for various values of $R$. These thermoelectric quantities are initially optimized by varying the QD energy level above Fermi energy for each $\Gamma/k_B T$ value. When the dot-reservoir tunneling rate $\Gamma$ is increased, the transmission function becomes broadened, and more electron flows from the hot reservoir to the cold reservoir, generating large thermovoltage. The magnitude of thermovoltage also depends on external serial load $R$ because any voltage drop across $R$ again acts back on the QD. This can be easily understood in two limiting cases, i.e., for short circuit ($R \approx 0$) and open circuit ($R \approx \infty$). For a short circuit, there is no voltage drop in the external circuit, i.e., the thermally generated charge current across the QD is instantaneously compensated by the current flowing in the external circuit. Thus, no net thermovoltage is generated. On the other hand, for an open circuit ($I_p = 0$), the voltage drop is maximum, which causes large thermovoltage. However, power output vanishes in both limits (see Figure 4a). Thus, finding the external load $R$ which gives optimal power output at a finite thermoelectric efficiency is necessary for practical applications.

The broadening of transmission function with increasing $\Gamma$ allows more electrons to contribute to power generation. As a result, as illustrated in Figure 4a, $P_{\text{max}}$ increases until the current caused by the temperature difference between the left and right reservoir becomes saturated and reaches a peak value determined by $R$. If the width of the transmission function is further broadened, electrons may start flowing from the right to the left reservoir, which would result in a decrease in $P_{\text{max}}$. The shift in the peak value of $P_{\text{max}}$ toward a weak dot-reservoir tunneling rate or narrow transmission function with increasing external load $R$ is a manifestation of the maximum power transfer theorem which states that the power transferred from a QD to an external load $R$ is maximal when $R = R_i$ (where $R_i$ is the internal resistance of QD heat engine). For a narrow Dirac delta-like transmission function, $R_i$ is high and requires a large external load to produce the maximum power output. In the present case, the optimal power output can be achieved at either $R \approx 10^3 \text{k}\Omega$ or $R \approx 10^5 \text{k}\Omega$ depending on the $\Gamma/k_B T$. On the other hand, as demonstrated in earlier studies, for a Dirac delta-like narrow transmission function, $\eta_{\text{max}}$ approaches the Carnot efficiency, as shown in Figure 4b. Due to the narrow energy range with an open circuit ($R \approx \infty$), thermal conductance vanishes faster than electrical conductance, which violates the Wiedemann–Franz law and causes significant enhancement in $\eta_{\text{max}}$. For a...
Figure 4. Variation of a) maximum power output $P_{\text{max}}$ and b) corresponding normalized efficiency $\eta_{P_{\text{max}}}$ of a strongly interacting (i.e., $U \rightarrow \infty$) single QD particle-exchange heat engine as a function of tunneling rate ($\Gamma$) for several values of external load resistance $R$. Both $P_{\text{max}}$ and $\eta_{P_{\text{max}}}$ are first optimized with respect to the QD energy level ($\varepsilon_d$). Inset in (a) shows the close-up view of $P_{\text{max}}$ for $\Gamma/k_B T$. The narrow region indicated by a double arrow shows the range of $\Gamma/k_B T$ considered in the experimental study. The other parameters are: $k_B T = 0.1 \Gamma_0$ and $k_B \Delta T = 0.05 \Gamma_0$.

Figure 5. The variation of power output ($P$) versus normalized conversion efficiency ($\eta$) for several values of external load resistance $R$ when QD energy level $\varepsilon_d$ or gate voltage $V_G$ is varied (indicated by black arrows). The results are obtained within the decoupling scheme beyond Hubbard-I for $U \rightarrow \infty$ limit by using Green’s function EOM technique. Black dashed lines at $\eta = 0.56 \eta_C$ indicate the Curzon–Ahlborn efficiency ($\eta_C$), i.e., efficiency corresponding to the maximum power output (in present case at $R = 1.5M\Omega$). The values of the other parameters are similar as considered in the experiment, i.e., $T = 0.9 K$ ($k_B T = 0.078 \text{ meV}$), $\Delta T = 0.6 K$ ($k_B T = 0.052 \text{ meV}$), $\Gamma = 5.8 \mu\text{eV}$, and $\varepsilon_d$ varies from 0 to 1 meV.
transmission function with finite width, thermal conductance increases rapidly, and the power output cannot compensate for the heat loss, which results in relatively low thermoelectric conversion efficiency. For example, when \( R = 10^4 \Omega \), the value of \( \eta_{\text{max}} \) drops by 40% of Carnot efficiency as \( \Gamma \) is increased from 0.01 \( k_B T \) to 0.1 \( k_B T \). Further, when the external load resistance \( R = 10^5 \Omega \) is replaced by \( R \leq 10^3 \Omega \), the efficiency \( \eta_{\text{max}} \) for \( \Gamma < k_B T \) (say at \( \Gamma = 0.01k_B T \)) significantly reduced from 0.75\( \eta_C \) to less then 0.1\( \eta_C \). This drastic reduction in \( \eta_{\text{max}} \) is due to vanishing power output or a sharp decrease in power output as compared to the thermal conductance for \( R \leq 10^2 \Omega \). Also, as the magnitude of external load resistance is reduced, the peak value of \( \eta_{\text{max}} \) shifts toward finite \( \Gamma \), similar to \( P_{\text{max}} \).

For \( R = 10^2 \Omega \), optimal power output with a finite efficiency \( (\eta_{\text{max}} \approx 0.25\eta_C) \) can be achieved at \( \Gamma \approx k_B T \).

Finally, we compare our infinite-U results with the experiment and real-time diagrammatic (RTD) theory results from Ref. \[78\]. Since \( U \) is the largest energy parameter in the experimental device, thus the infinite-U limit is a valid approach. Further, the dot-reservoir tunneling rate is very low, i.e., \( \Gamma < k_B T \). Thus, experimental results are limited to a narrow region (i.e., \( 0.05 \leq \Gamma/k_B T < 0.09 \)), as indicated by a double arrow in the inset of Figure 4a. Figure 5 shows that the power output \( P \) and corresponding thermoelectric conversion efficiency \( \eta \) can be optimized for any given load \( R \) by varying the QD energy level \( \epsilon_d \) (applying gate voltage \( V_G \). However, for \( \Gamma \ll k_B T \), there is always a finite tradeoff between the maximum values of \( P \) and \( \eta \) because these quantities are not maximized by the same \( \epsilon_d \) and \( R \). The power output \( P \) is maximum for \( R = 1.5M\Omega \) with thermoelectric conversion efficiency approximately equal to the Curzon–Ahlborn efficiency, i.e., \( \eta \approx \eta_{\text{CA}} = 0.5\eta_C \). On the other hand, for \( R = 3.3M\Omega \) and \( 9.4M\Omega \), the efficiency \( \eta \) reaches its maximum value, which is approximately equal to 70% of the Carnot efficiency, i.e., \( \eta_{\text{max}} = 0.7\eta_C \).

Figure 6 shows the plot for (a) \( P_{\text{max}} \) and \( \eta_{\text{max}} \) as a function of external load resistance \( R \) for different background temperatures \( T \) and thermal gradients \( \Delta T \). First, for each \( R \), the maximum power output \( P_{\text{max}} \) is calculated by varying \( \epsilon_d \) from 0 to 1 meV, as shown in Figure 5. The load resistance \( R \) is then varied between 1 \( \Omega \) and 1 \( M\Omega \) to find its optimal value \( R_p \), i.e., load resistance corresponding to highest \( P_{\text{max}} \) value. It is important to note that \( R_p \) can be calculated directly from the open-circuit condition by using \( R_p = (V_G/2)/I_{\text{th},2} \). However, because we are interested in analyzing the effect of \( R \) on the performance of the QD heat engine, we calculated \( R_p \) by varying \( R \). Figure 6a shows that \( P_{\text{max}} \) peaked for \( R \) between 1.0 to 2.0 \( M\Omega \) depending on \( T \) and \( \Delta T \). The peak value of \( P_{\text{max}} \) nearly doubles as \( (T, \Delta T) \) increases from \((0.69, 0.33 \text{ K})\) to \((1.13 \text{ K}, 0.7 \text{ K})\). The optimal load \( R_p \) also depends on the ratio \( \Gamma/k_B T \), and as previously noted in Figure 4, \( R_p \) is close to 0.1\( M\Omega \) for \( \Gamma/k_B T \leq 0.3 \). Figure 6b shows that thermoelectric efficiency at maximum power output \( \eta_{\text{max}} \) is peaked for \( R \) between 4 and 10 \( M\Omega \) depending on \( T \) and \( \Delta T \). As \( (T, \Delta T) \) increases from \((0.69, 0.33 \text{ K})\) to \((1.13 \text{ K}, 0.7 \text{ K})\), the peak value of \( \eta_{\text{max}} \) enhanced by 6% of Carnot efficiency. Further, for \( R \approx 10M\Omega \), the efficiency of strongly interacting single QD heat engine can reach near 70% of Carnot efficiency with a finite power output \( P_{\text{max}} \approx 2fW \) and also \( \eta_{\text{max}} \approx \eta_{\text{CA}} \) at the optimal load resistance \( R_p \). Our theoretical estimation for the load resistance necessary to achieve the optimal power output and corresponding thermoelectric efficiency is in good agreement with the recent experimental and RTD theory results.

![Figure 6](image-url)
4. Conclusion

In summary, we have analyzed a strongly correlated single QD-based thermoelectric particle-exchange heat engine using Keldysh’s nonequilibrium Green’s function formalism for various order of decoupling schemes in the EOM technique. We employed Hubbard-I approximation for a finite on-dot Coulomb interaction and beyond Hubbard-I for strongly interacting QD, i.e., $U \to \infty$ limit, to study the thermoelectric transport properties in the Coulomb blockade regime. For finite-$U$, the thermopower and thermoelectric efficiency are significantly enhanced beyond the electron–hole symmetry and resonance points at a relatively small thermal gradient. When the Coulomb interaction on the QD is very strong, i.e., infinite-$U$, the thermoelectric quantities become asymmetric due to asymmetry in the tunneling amplitude for odd and even electrons, and the maximum power output and corresponding efficiency increases significantly with the thermal gradient. For practical applications, however, it is essential to understand the relationship between power output and corresponding efficiency in the presence of a finite external load. Therefore, using the infinite-$U$ limit, the effect of dot-reservoir tunneling rate and external load resistance on the optimization of the power output and corresponding thermoelectric efficiency are investigated. It has been demonstrated that $P_{\text{max}}$ and $\eta_{\text{max}}$ are peaked at different external load resistance, which differs by approximately one order of magnitude. Due to this finite tradeoff, it is essential to make an adjustment between $P_{\text{max}}$ and $\eta_{\text{max}}$ to achieve the best performance in practical applications. These results for the infinite-$U$ limit are in good agreement with recent experimental data and real-time diagrammatic theory results. Thus, Green’s function EOM technique, which is a computationally inexpensive and straightforward analytical method, gives reliable results in the Coulomb blockade regime. The present analysis can be extended to examine the optimal performance of other realistic low-dimensional heat engines based on multiple QDs and multiple reservoirs within the strong Coulomb blockade regime.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

Keywords

Coulomb blockade, nanoelectronics, optimal power, quantum dots, Seebeck effect, thermoelectric heat engine

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