Non-Monotonic Thermoelectric Currents and Energy Harvesting in Interacting Double Quantum-Dots

Yair Mazal and Yigal Meir
Department of Physics and the Ilse Katz Center for nanoscale Science and Technology, Ben-Gurion University of the Negev, Beer Sheva, Israel

Yonatan Dubi
Department of Chemistry and the Ilse Katz Center for nanoscale Science and Technology, Ben-Gurion University of the Negev, Beer Sheva, Israel

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A theoretical study of the thermoelectric current and energy harvesting in an interacting double quantum dot system, connected to reservoirs held at different chemical potentials and temperatures is presented. Using a rate-equation approach, the current is evaluated for different energetic configurations of the double quantum dot. We discuss in detail the current-temperature gradient relations (the thermoelectric analog to current-voltage relations), and demonstrate that, due to interactions, the current is non-monotonically dependent on thermal bias. This interaction-induced non-monotonicity influences the possibility of harvesting thermal energy from the double-quantum dot, and it is shown that in some configurations, energy cannot be harvested at all. We analyze the conditions under which energy can indeed be harvested and converted to useful electrical power, and the optimal conditions for thermoelectric energy conversion.

I. INTRODUCTION

In bulk systems, the efficiency of thermoelectric harvesting - generation of an electrical current and voltage from a temperature gradient - is limited by the material properties. The prospect of using nano-scale systems to enhance thermoelectric performance\textsuperscript{1-3}, along with the huge advances in constructing and manipulating nanoscale structures and devices, now make "nanoscale thermoelectrics" a large and interdisciplinary field of research. Indeed, nanoscale thermoelectric energy harvesting has now been demonstrated in systems such as nano-composite materials, carbon and silicon nanowires, Graphene nanostructures, molecular junctions, nanoparticles and quantum dots (see, e.g., Refs. 4 and 5 and references therein).

Besides the obvious applicative interest, in recent years it was realized that studying thermoelectric effects in nanoscale systems can shed light on the transport mechanisms dominating nanoscale structures. For instance, in molecular junctions, the thermoelectric voltage and thermopower can distinguish between different transport mechanisms\textsuperscript{6-8}. Thermopower in quantum dots was studied over two decades ago, both theoretically and experimentally\textsuperscript{9,10}, with recent resurging interest focusing on role of interactions and non-linearity on thermoelectric efficiency\textsuperscript{11-19} (see review in, e.g., Ref. 20).

Double quantum dots (DQDs) are an excellent platform for studying the interplay between interactions, quantum effects (i.e. interference), charge and heat transport and thermoelectricity\textsuperscript{21-34}. The possibility of arranging them in parallel or series, to couple them either by tunneling or capacitively, and to tune each quantum dot energy separately, gives rise to a large spectrum of parameters which can be tuned, leading to a broad spectrum of thermoelectric phenomena. Here, we study the effect of DQD parameters (level spacing, interaction strength etc.) on thermoelectric energy harvesting. We focus on the non-linear regime, with finite voltages and finite temperature difference. We show that at given temperature difference and finite bias, one cannot always harvest the thermal bias into electric power, and find the conditions for energy harvesting under different DQD parameters. We show that, surprisingly, in the presence of strong interactions, there is a minimal temperature difference under which no harvesting is possible, and provide the mechanism for this effect.

The paper is organized as follows. In Sec. II the method and model are detailed. In Sec. III we discuss the dependence of current on temperature difference in the non-linear regime. In Sec. IV we discuss the conditions for energy harvesting, and Sec. V is devoted to summary and conclusions.

II. FORMALISM

A. Model

The system under consideration is a DQD, illustrated in Fig. 1. Within the DQD we take into account intra-dot as well as inter-dot Coulomb interaction. The DQD is coupled to two leads characterized by different temperatures and chemical potentials. The corresponding Hamiltonian is:

\[
\mathcal{H} = \sum_{k,\sigma} \varepsilon_{kz} c_{kz\sigma}^{\dagger} c_{kz\sigma} + \mathcal{H}_{DQD} + \sum_{kz} V_{kz\sigma} c_{kz\sigma}^{\dagger} d_{\sigma} + h.c.,
\]  

(1)

where $\mathcal{H}_{DQD}$ includes the exchange interaction between the dots and the leads,\textsuperscript{35} and $V_{kz\sigma}$ is the coupling between the dot and the lead. The occupations of the lead states are given by $n_{k\sigma} = \langle c_{k\sigma}^{\dagger} c_{k\sigma} \rangle$. The current in the lead is given by:

\[
\mathcal{J}_{x\sigma} = -e \sum_k \left( n_{k\sigma} - \overline{n}_{k\sigma} \right) \langle c_{k\sigma}^{\dagger} c_{k\sigma} \rangle - n_{k\sigma} \langle c_{k\sigma} c_{k\sigma}^{\dagger} \rangle.
\]
Without loss of generality we consider different transport regimes. Our results are thus qualitatively valid as long as the inter-dot Coulomb interaction is not much larger than inter-dot one. However, our results are valid as long as the inter-dot Coulomb energy would be larger than the intra-dot one. We assume that

\[ \varepsilon_{i} \text{ is the level of the } i\text{-th dot}, \]

where \( \varepsilon_{kx} \) is the energy of an electron with momentum \( k \) in the \( x\)-th lead \((x=L,R)\), \( \hat{c}_{kx\sigma} \) creates an electron with spin \( \sigma \) and momentum \( k \) in the lead, \( \hat{d}_{i\sigma} \) annihilates an electron with spin \( \sigma \) in the \( i\)-th dot, and \( \hat{V}_{kxi\sigma} \) is the coupling between the lead and the dot. The first term is the Hamiltonian of the leads, the second describes the DQD, and the third represents the interface. The Hamiltonian of the DQD can be written as:

\[ \mathcal{H}_{DQD} = \sum_{i\sigma=A,B} \varepsilon_{i\sigma} \hat{d}_{i\sigma}^\dagger \hat{d}_{i\sigma} + U_{0} \sum_{i=A,B} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + U_{1} \hat{n}_{A} \hat{n}_{B}, \]  

where \( \varepsilon_{i\sigma} \) is the level of the \( i\)-th dot, \( U_{0}(U_{1}) \) is the inter-dot (intra-dot) Coulomb interaction strength, \( \hat{n}_{i\sigma} \equiv \hat{d}_{i\sigma}^\dagger \hat{d}_{i\sigma} \), and \( \hat{n}_{i} = \hat{n}_{i\uparrow} + \hat{n}_{i\downarrow} \). All energies are measured from the zero-energy, defined as the energy of the empty dot. For simplicity, and to reduce the number of numerical parameters, we assume that \( U_{0} = U_{1} = U \) \( \leq 34-37 \). One might expect that intra-dot Coulomb energy would be larger than inter-dot one. However, our results are valid as long as the inter-dot Coulomb interaction is not much smaller than the intra-dot interactions. Specifically (as we discuss in Sec. III), the interaction strength defines different transport regimes. Our results are thus qualitatively valid as long as \( U_{0} \) and \( U_{1} \) lie within the same regime.

The leads are modeled as free electron gases, and are characterized by their Fermi functions with temperatures \( T_{L(R)} \), and chemical potentials \( \mu_{L(R)} = \mu_{v} \pm \Delta \mu/2 \). Without loss of generality we consider different temperatures such that \( T_{R} > T_{L} \), and define the voltage as \( \Delta \mu = \mu_{L} - \mu_{R} \), which can be either positive or negative.

### B. Method

#### 1. Rate Equations

When considering the Hamiltonian of Eq. 1 one may conceptually divide it into three parts: the DQD (system), the leads (reservoirs), and the tunneling between them. There are various ways to treat such systems, each has its advantages. In this work we use rate equations. Using rate-equation formalism, one finds the population of the system (diagonal elements of the density matrix) by solving a system of linear first-order differential equations. The standard procedure is to consider the steady state solution upon which the problem reduces to an algebraic system of linear equations whose solution, i.e. the kernel of the rate matrix, is the population of the DQD. The rate-equations are valid in the weak coupling limit which requires \( \Gamma_{xij\sigma} \ll k_{B}T \) \( \leq 38,39 \), where \( \Gamma_{xij\sigma}(\omega) = \sum_{k} V_{kx\sigma} V_{kx\sigma}^* A_{kx\sigma}(\omega) \), and \( A_{kx\sigma}(\omega) \) is the spectral function. While generally \( \Gamma_{xij\sigma}(\omega) \) is a function of energy \( (\omega) \) \( \leq 39,40 \), here we employ the wide band approximation, and assume constant density of states in the reservoirs within the relevant region of the spectrum, thus \( \Gamma_{xij\sigma}(\omega) \equiv \Gamma_{xij\sigma} \).

This approach is closely related to the master equations approach, and in fact they become the same at steady state for non-degenerate systems \( \leq 41,42 \). However, rate equations can also be thought of as a consequence of time-dependent perturbation theory in first order (Fermi golden rule) \( \leq 43 \), as a consequence of solving a many body Schrödinger equation \( \leq 44 \), or as limiting case of Non-Equilibrium Green-Function approach when the width of the spectral function vanishes. The main advantage of the use of rate equations is that with relative ease one can model quite complex systems, which would require much more effort in other approaches, yet one is able to observe many of the interesting phenomena that such systems demonstrate. Additionally, they allow treatment of systems with competing energy scales with relative ease.

Within the rate-equation formalism we treat the DQD by diagonalizing \( \mathcal{H}_{DQD} \) (diagonal in position basis in our case), while the leads are treated as electronic reservoirs, and are characterized by their Fermi distributions. Electron transfer between the DQD and either lead is treated only within the rate matrix \( W \) whose elements are the rates for transition between eigenstates of the reduced (sub-)system which is the DQD. The rate equations read \( \leq 42,43,45 \):

\[ \frac{dp_{m}}{dt} = \sum_{n \neq m} W_{n \rightarrow m} p_{n} - p_{m} \sum_{n \neq m} W_{m \rightarrow n} \Leftrightarrow \dot{p} = Wp, \]  

where \( W_{n \rightarrow m} = W_{mn} \) (for off-diagonal matrix elements) is the rate of transition from the many body Fock state \( \langle n \rangle \) to \( \langle m \rangle \), and \( p_{m} \) - the probability that the system will be in the many-body state \( \langle m \rangle \) of the DQD. The rate associated with adding an electron to the system is:

\[ W_{n \rightarrow m} = \sum_{x=L,R} f(\epsilon_{m} - \epsilon_{n} - \mu_{x}) \times \]

\[ \times \sum_{i,j=A,B} \Gamma_{xij\sigma} \langle m | d_{i\sigma}^\dagger | n \rangle \langle n | d_{j\sigma} | m \rangle, \]
while for removal of an electron the associated rate is:

$$W_{n \to m} = \sum_{x=L,R} \left( 1 - f(\epsilon_n - \epsilon_m - \mu_x) \right) x \times \sum_{i,j=A,B} \Gamma_{xij} \langle m | d_{j,i} | n \rangle \langle n | d_{i,j}^{\dagger} | m \rangle,$$

where $\epsilon_m$ is the energy of the $m$-th state of the DQD, $f(\epsilon - \mu_x)$ stands for the Fermi function of the $x$-th lead, and therefore represents the probability of the lead to have an electron with the energy of the desired quasi-level and $\Gamma_{xij}$ (defined above) is the coupling of the DQD to the leads. The overlaps of the form $\langle m | d_{j,i} | n \rangle$ dictate whether the transition is allowed in the sequential tunneling regime, where allowed transitions move the system between states which differ by one electron. Eq. 3 also implies that the diagonal elements of the rate matrix read:

$$W_{mm} = -\sum_{n \neq m} W_{m \to n} = -\sum_{n \neq m} W_{nm},$$

such that the sum of elements along each column vanishes. Steady state populations are found by solving $Wp = 0$, and imposing probability conservation ($\sum_m p_m = 1$).

2. Current

By defining the total particle number in the DQD as $\hat{n} = \sum_m \hat{n}_{m\sigma}$, we may find the particle current by writing:

$$\frac{d\langle \hat{n} \rangle}{dt} = \frac{d}{dt} \left( \sum_m n_m p_m \right) = \sum_m n_m \dot{p}_m = \sum_m n_m (Wp)_m =$$

$$\sum_m \left( W_L p \right)_m + \sum_m \left( W_R p \right)_m = 0,$$

where $W = W_L + W_R$, and $W_L(R)$ is the matrix which corresponds to rates involving only the left (right) lead, and $n_m = \langle m | \hat{n} | m \rangle$ (since $[\hat{n}, \mathcal{H}_{DQD}] = 0$ the value of $n_m$ is time independent as long as the leads aren’t considered). At steady state, the latter is trivially $J_L + J_R = 0$, as the current flowing from the DQD to one lead, is exactly canceled by the current flowing into the DQD from the other. However, if we only consider the current between the DQD and one of the leads the expression does not vanish and we can express the actual electric current flowing through the system by:

$$J = J_L = \frac{e}{\hbar} \sum_m n_m (W_L p)_m.$$

The power harvested by the DQD can simply be found by:

$$P_{out} = -J \Delta \mu.$$

In this work we express all energy scales in units of temperature. For quantum dots, typical operating temperatures range from millikelvins to room temperature, and our results consider temperatures of up to 1K, as this is the range of temperatures in which relevant experimental work\(^\text{18}\) was conducted.

III. NON-MONOTONONIC CURRENT-TEMPERATURE BIAS RELATIONS

We begin the results section of the paper by describing the non-trivial dependence of the current on the temperature bias. Similar to negative differential conductance (NDC), upon considering an interacting system with distinguishable transport channels ($\Delta \epsilon > T$), one may observe negative thermal response (NTR), seen as a non-monotonic dependence of $J$ on thermal bias ($\Delta T$). However, the mechanisms responsible for the two phenomena are not the same. As discussed in\(^\text{46,47}\), in order to observe NDC one must consider unequal couplings between DQD and leads adhering to certain conditions. Even though NTR may be observed in the same regime, it may also be observed with equal couplings, as shown in recent theoretical studies\(^\text{21,48}\), and experimental work\(^\text{18}\) (which includes also a rate equations model).

Below, we discuss one of several configurations in which NTR is predicted in the presence of interactions, ranging from $U \sim \Delta \epsilon$ to strong interactions, $U \gg |\epsilon_{A,B}|, |\mu_{L,R}|, T$ (reminding that the energies and chemical potentials are measured from the empty dot, which is defined as the zero energy), and with equal couplings. Unlike previous works\(^\text{18,21,48}\), we are motivated by potential applications regarding energy harvesting, and therefore consider also finite voltage ($\Delta \mu = \mu_L - \mu_R \neq 0$). To this end, $J$ is plotted as a function of $\Delta T$ such that $T_L = T, T_R = T + \Delta T$. The thermal bias needed in order to observe the phenomena in the high temperature limit ($k_B T \gg \Gamma$), is rather large and challenging from an experimental point of view as discussed by\(^\text{18}\), and our analysis assumes the maximal thermal bias as $\max(\Delta T) = 40T$ as reported in their work, though we predict the phenomena to be evident even for smaller thermal bias.

Fig. 2(a) illustrates the discussed configuration. It shows the alignment of leads with respect to accessible transport channels pertaining to single-particle states ($\epsilon_{A,B}$ in the figure), and to two-particle states ($\epsilon_{A+B}+U$ in figure), while purple arrows indicate the direction of current flowing through each channel upon sufficient heating of the right lead. Currents flowing through each dot ($J_{A,B}$) as well as total current ($J = J_A + J_B$) are plotted vs. thermal bias for two interaction strengths in Fig. 2(b). As expected, for $\Delta T = 0$ no current flows as there is no available channel within the Fermi window. A small thermal bias ($\Delta T \sim 5T$ in the figure) induces thermal current only through single-particle channels. However, current through the lower channel (illustrated as $\epsilon_A$) prevails over current through the higher one (labeled as $\epsilon_B$).
for the following reason: since \( f_{L,R}(\epsilon_B) \leq \frac{1}{2} \leq f_{L,R}(\epsilon_A) \) the probability of the system to be in Fock states with a single particle in dot \( A \) is necessarily larger than those with a particle in dot \( B \) as insertion and extraction rates are proportional to \( f \) and \( 1 - f \). Since due to the interaction, these channels cannot support current simultaneously, current through \( \epsilon_A \) prevails. Note that this dominance is maintained even though we chose \( \mu_R \) such that it is closer to \( \epsilon_B \), i.e. \( |\mu_R - \epsilon_B| < |\mu_R - \epsilon_A| \), and thus one would expect naively the effect of \( \Delta T \) on current flowing through \( \epsilon_B \) to be greater.

For larger thermal bias, depending on the magnitude of \( U \), current may also flow through higher channels labeled as \( \epsilon_{A,B} + U \) in Fig. 2(a). Current flowing through \( \epsilon_B + U \) flows in the same direction as current through \( \epsilon_B \) (since both \( \epsilon_B, \epsilon_B + U > \mu_{L,R} \)) and the two contributions add up. This is in contrast to current through \( \epsilon_A \) and \( \epsilon_A + U \) which flow in opposite directions (since \( \epsilon_A < \mu_{L,R} < \epsilon_A + U \)), which manifests in Fig. 2(b) as a local maxima in the gray dashed curve. The suppression of \( J_A \) together with the enhanced \( J_B \) are responsible for the local maxima in \( J \) in the solid gray curve. Since the position of the maxima in \( J \) vs. \( \Delta T \) increases monotonically with \( U \), the maxima in the blue line (\( U = 300T \)) is shifted to a value of \( \Delta T \) beyond the domain depicted in the figure.

The Coulomb interaction determines the occupation of different levels. It is thus interesting to look at the correspondence between changes in current line-shape and the probability of the system to be in two-particle states. In Fig. 2(c) \( J \) is plotted vs. \( p_2 \) for various values of \( U \), where \( p_2 \) is defined as sum of probabilities for being in any of the two-particle states. The curves were obtained by finding both \( J \) and \( p_2 \) for \( \Delta T \)'s within the domain \( \Delta T \in (0, 40T) \), and the curves terminate at the maximal allowed thermal bias, i.e. \( \Delta T = 40T \) (intuitively, \( p_2 \) increases monotonically with \( \Delta T \)). As shown, the current increases as a function of \( p_2 \), up until a \( U \) dependent threshold, at which it drops sharply with the onset of

**FIG. 2:** (a) Illustration of transport channels alignments with the two chemical potentials \( \mu_{L,R} \), where purple arrows indicate the direction of current through each channel. (b) Current through each dot \((J_{A,B})\) and total current \((J = J_A + J_B)\) vs. the temperature difference \( \Delta T \) for two values of the interaction strengths: \( U = 100T, 300T \). (c) Current through the DQD \((J)\) vs. probability of the DQD to contain two particles \((p_2)\), for various values of \( U \). (d) \( J \) vs. \( \Delta T \) and \( U \). The gray and blue vertical lines indicate values of \( U \) for which cross-sections were plotted in (b), while the black curve indicates the path along which the current has a local maxima (when plotted only against \( \Delta T \)). Parameter values: \( \Gamma = T/10, \epsilon_A = 10T, \epsilon_B = 40T, \mu_L = 20T, \mu_R = 30T \).
the NTR. Quite surprisingly, Fig. 2(c) shows a huge sensitivity of the current to the occupations; a seemingly insignificant probability of 1% of the system to be in a two particle state drastically alters transport, and is enough to cause NTR.

Fig. 2(d) shows the broader picture via a contour map of $J$ vs. $\Delta T$ and $U$. The black curve shows the path along which the current has a local maxima for the specified $\Delta \epsilon, \mu_{L,R}$. The curve, which has been found numerically, illustrates approximately the minimal thermal bias required for NTR to be observed for a given value of $U$.

Generally speaking, non-monotonic current is a consequence of competition between currents flowing in opposite directions via different transport channels. One way to achieve this is by choosing a large enough interaction such that two particle states are not accessible without heating ($U \gg T$), while making it comparable with the thermal bias ($U \sim \Delta T$) so that heating may facilitate current through multi-particle channel in an opposite direction, thus diminishing overall current.

IV. ENERGY HARVESTING

Thermo-electric energy harvesting requires driving a current against a load, i.e. driving an electric current opposite to an external voltage ($\Delta \mu$) by means of a thermal bias ($\Delta T$)\textsuperscript{20}. Harvesting systems can be classified as n-type if electric current flows from the hot lead to the cold one (same direction as heat flow), or as p-type if electric current flows from the cold lead to the hot one (opposite to heat flow). The discussed DQD system may act as either type, and we discuss the required settings for each regime, limiting the discussion to equal coupling and strong interactions. We then consider also the case of weaker interaction strength ($U \sim \Delta \epsilon$).

A. Monotonic Harvesting, N-Type

Let us start with the case of very large (essentially infinite) $U$. For $\Delta \mu = \mu_{L} - \mu_{R} > 0$ energy harvesting requires $J < 0$ which occurs (approximately) when $f_{R}(\epsilon_{A}) > f_{L}(\epsilon_{A})$. This is explained by the following: Trivially, the direction of current through dot $A$ ($J_{A}$) depends solely on the above comparison. Similarly, the direction of current through dot $B$ ($J_{B}$) and $f_{L}(\epsilon_{B})$. That being said, for $\epsilon_{B} > \mu_{L}$, the current through $\epsilon_{A}$ prevails over current flowing through $\epsilon_{B}$, as, due to the strong interaction, the larger occupation of dot $A$ prohibits occupation and current flow through $\epsilon_{B}$. If, on the other hand, $\mu_{L}$ is assumed to be large enough such that occupations of the two dots are comparable, the currents through the two dots are similar too. Thus, in order to pass current against voltage (with $\Delta \mu > 0$ and strong interactions) we require: $f_{R}(\epsilon_{A(MB)}) > f_{L}(\epsilon_{M}) \Rightarrow f_{R}(\epsilon_{A}) > f_{L}(\epsilon_{A})$, since, as explained, the condition regarding $\epsilon_{B}$ is obeyed trivially if the one for $\epsilon_{A}$ does.

The last inequality yields an analytic condition for harvesting:

$$f_{R}(\epsilon_{A}) > f_{L}(\epsilon_{A}) \Rightarrow \frac{1}{e^{\frac{\Delta \epsilon - \mu_{R}}{T}} + 1} > \frac{1}{e^{\frac{\Delta \epsilon - \mu_{L}}{T}} + 1} \Rightarrow \Delta T > \Delta T_{\text{harvest}} = \frac{T(\mu_{L} - \mu_{R})}{\epsilon_{A} - \mu_{L}} = \frac{2T\Delta \mu}{2\epsilon_{A} - \mu_{av} - \Delta \mu}. \tag{10}$$

The above condition also contains another constraint, stating that for harvesting we must require $\epsilon_{A} > \mu_{L}$ as we consider $\Delta T > 0$ and $\Delta \mu = \mu_{L} - \mu_{R} > 0$. If this additional constraint is not obeyed, no amount of heating will cause current flow against the voltage (no harvesting) in the n-type regime with strong interactions (i.e. $U$ is large enough to prohibit transport via two-particle channels).

Fig. 3(a) shows a contour plot of $J$ vs. $\mu_{av}$ and $\Delta T$ for fixed $\Delta \mu$. Dashed contour lines indicate the domain in which current is negative (the DQD acts as a harvester, since $\Delta \mu > 0$), while solid contour lines indicate the domain of positive current, and the two are separated by the thick black curve indicating the curve along which current vanishes ($J = 0$). The thick red curve shows the analytic condition derived above (Eq. 10). As can be seen, the above condition approximates very well the black curve. The deviation at large $\Delta T$ stems from the fact that our derivation does not take into account the less significant current flowing through $\epsilon_{B}$.

Fig. 3(b) shows a cross section of the contour plot in Fig. 3(a) taken at $\mu_{av} = 2.5T$ as indicated by the vertical gray line in the contour map. In this regime the current is monotonic with respect to $\Delta T$, in agreement with the discussion in the previous section, as there is no competition between currents flowing in opposite directions. This is shown in the inset, which illustrates the alignment of the leads chemical potentials with the transport channels, with arrows indicating the direction of current flow through each channel upon heating of the right lead.

B. Monotonic Harvesting, P-Type

The DQD may also act as a p-type harvester and drive electric current from the cold lead to the hot one against a voltage, with the previous inequality merely changing direction, i.e. $f_{L}(\epsilon_{A}) > f_{R}(\epsilon_{A})$. Therefore the relation in Eq. 10 still determines the cross-over between harvesting and investing energy, with the only change being that now we consider $\mu_{A} < 0$, and thus the trivial condition switches to $\epsilon_{A} < \mu_{L}$.

Fig. 3(c) shows a contour plot of $J$ vs. $\mu_{av}$ and $\Delta T$, with dashed contours indicating negative current, and solid contours indicating positive current (here harvesting requires $J > 0$ as we consider $\Delta \mu < 0$). The two domains are separated by the thick black curve indicating the curve along which current vanishes ($J = 0$), while...
FIG. 3: (a) Contour plot of $J$ vs. $\mu_{av}$ and $\Delta T$ for $\Delta \mu > 0$. The black curve indicates the contour line along which $J = 0$, the red curve plots Eq. 10 and the gray vertical line ($\mu_{av} = 2.5T$) indicates the path along which the cross section in (b) is taken. (c) Contour plot of $J$ vs. $\mu_{av}$ and $\Delta T$ for $\Delta \mu < 0$. The black curve indicates the contour line along which $J = 0$, the red curve plots Eq. 10 and the gray vertical line ($\mu_{av} = 16.5T$) indicates the path along which the cross section in (d) is taken. Parameter values: $U = 1000T$, $\Gamma = T/10$, $\epsilon_A = 10T$, $\epsilon_B = 40T$, $|\Delta \mu| = 5T$.

the thick red one shows the analytic approximation derived above (Eq. 10). As before, the red curve slightly deviates from the black one for large $\Delta T$, stemming from neglecting the contribution to current through $\epsilon_B$.

Fig. 3(d) shows a cross section of the contour plot in Fig. 3(c) taken at $\mu_{av} = 16.5T$ as indicated by the vertical gray line in the contour map. The inset illustrates the alignment of the leads chemical potentials with the transport channels, and the arrows indicate the direction of current flow through each channel upon heating of the right lead. Here too, the current is monotonic with respect to $\Delta T$. Since the voltages taken into account in figures 3(b) and 3(d) are of the same magnitude, comparing current magnitudes reveals that for the discussed configurations the n-type regime allows for more power to be harvested.

C. Non-Monotonic Harvesting

1. Interaction Facilitated Harvesting

Allowing for a small enough interaction such that two-particle states may get occupied and current may flow through two-particle channels has trivial as well as surprising effects regarding harvesting. Trivially, as current flows through more channels the dominance of flow through $\epsilon_A$ is reduced, thus the analytic approximation employed before becomes less relevant. Another effect is that, if the bare levels are submerged in Fermi sea (single-particle channels are inaccessible), considering different strengths of interaction ($U$) may dictate whether the system harvests energy or not upon heating. This is easily
seen in Fig. 4(a) which shows $J$ vs. $U$ and $\Delta T$ for an n-type configuration ($\mu_L - \mu_R = 5T$). As shown, the question whether or not the system harvests energy (harvesting for $J < 0$, dashed contour lines) depends on $U$ and $\Delta T$. More notably, there are values of $U$ (for instance $U = 95T$ marked by a red vertical line in Fig. 4(a)) for which the system’s behavior changes non-monotonically with $\Delta T$ between harvesting and investing energy (Fig. 4(b)).

This is shown in Fig. 5, where the output power $P_{\text{out}}$ (Eq. 9) is plotted as a function of $\Delta T$ for opposite voltages (n-type and p-type regimes), and for two interaction strengths, $U = 45T, 300T$. The relevant configuration is the same as the one illustrated in Fig. 2(a).

As seen, in the n-type regime ($\Delta \mu > 0$) the two-particle channels assist harvesting and in fact if $U$ is too large harvesting is impossible in this configuration as the two-particle channels are blocked. On the other hand, in the p-type regime ($\Delta \mu < 0$), the interaction hinders harvesting, and therefore if the interaction is small enough to allow two-particle channels to affect transport, harvesting is possible only for a limited ($U$ dependent) range of thermal bias.

3. Maximal power and optimal thermal bias

As is implied from the previous discussion, under given conditions (negative bias, so called p-type regime, and intermediate interaction strength) the power output displays non-monotonic behavior as a function of the thermal bias. This needs to be accounted for, if one wishes to optimize the performance of the thermoelectric device as an energy harvester. To explore this situation, we have performed the following computation. A set of DQD systems, all with the same energies, average chemical potential and couplings, but with different interaction strengths ($30T < U < 150$) were considered ($\epsilon_A = 10T, \epsilon_B = 40T, \mu_{\text{avg}} = 25T, \Gamma = T/10$). The bias voltage was scanned i the range $-20T < \Delta \mu < -T$. For each system (value of $U$) and each voltage bias ($\Delta \mu$), the temperature bias $\Delta T$ for which one obtains maximal power was found. Each curve in Fig. 6 depicts that maximal power as a function of the optimal thermal bias for the different voltage biases, and a given value of $U$. The different curves correspond to different interaction strengths, according to the color coding. The first thing one notices is that the larger the interaction strength,

2. Harvesting via Single-Particle Channels

When transport is dominated by current flowing through single-particle channels, interaction may either assist or hinder harvesting depending on configuration. To understand why this is the case, one must note that in the n-type regime harvesting stems from current via channels which are aligned above the hot lead chemical potential ($\epsilon_i > \mu_R$), as illustrated in Fig. 3(b). On the other hand, in the p-type regime, harvesting stems from current via channels which are beneath the hot lead chemical potential ($\epsilon_i < \mu_R$), as illustrated in Fig. 3(d).
more power can be extracted at optimal conditions. This is easy to understand; in this case, the harvested current passes through the lower quantum dot ($\epsilon_A$). The current that flows through the quasi-level (at energies $\epsilon_A(B) + U$), in the direction of the voltage bias (suppressing harvesting), is diminished by the increasing interaction.

Second, for each value of $U$ there is an optimal thermal bias which maximizes the harvested power. This can only be understood in light of the non-monotonicity discussed throughout the paper. Indeed, in a simple device (such as a non-interacting quantum dot) such behavior will not occur. Third, one can observe a specific structure of the data; for each value of $U$ there is a well-defined "trajectory" when plotted against $\Delta T_{opt}$. The inset shows one example of such a trajectory (taken at $U = 60T$), in the $P_{\text{max}}$, $\Delta T_{opt}$ plane. The arrow indicates the direction of increasing negative voltage, from $-17T$ to $-20T$. Maximal power appears at $\Delta \mu \sim -11T$. In general, the optimal power must depend on the applied bias voltage. This is in contrast with the non-interacting case (or the strong-$U$ case, as in Fig. 4), where one expects a monotonic dependence on thermal bias. For the (intermediate) interacting case, on the other hand, the non-monotonicity in the current implies a more subtle relation between optimal power, optimal thermal bias and voltage bias, depicted in Fig. 6. To put it simply: for non-interacting systems, the larger the thermal bias is, the more energy can be harvested, while for the interacting DQD system considered here, there is an optimal finite thermal bias. This point should be considered for optimal design of an experimental apparatus.

V. SUMMARY AND CONCLUSIONS

To summarize, in this work we studied the thermoelectric current and energy harvesting in an interacting double-quantum-dot system, connected to reservoirs held at different chemical potentials and temperatures. Using a rate-equation approach, the current was evaluated for different energetic configurations of the DQD. We discuss in details the current-temperature gradient relations (the thermoelectric analog to current-voltage relations) and the conditions under which energy (from the temperature gradient) can be harvested and converted to useful electrical power.

Specifically, our main results are: (i) In the presence of interactions, the current-thermal bias relations may exhibit non-monotonic behavior, in analogy to negative differential conductance in voltage-biased interacting quantum dot junctions. (ii) there is an extreme sensitivity of the current to the populations on the dot. Specifically, a tiny change in the quantum dot occupations can alter qualitatively the current-bias voltage dependence. (iii) Energy harvesting can be enhanced or hindered by Coulomb interactions, depending on the specific conditions of the DQD. (iv) In the presence of interactions, the non-monotonic current leads to non-trivial optimal configuration of the DQD. More specifically, one can say that in the presence of interactions, it is not always the case that the larger the thermal bias the more energy can be harvested.

These results provide insight into the nature of thermoelectric transport in quantum-dot junctions. Being well within current experimental capabilities, our predictions can be tested experimentally. Going beyond the weak-coupling limit, extending the system to more than two quantum dots, and allowing for direct coupling between the quantum dots (i.e. electron tunneling between the dots) are all directions we plan to address in the future.

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