Thermoelectric phenomena in a quantum dot asymmetrically coupled to external leads

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We study thermoelectric phenomena in a system consisting of strongly correlated quantum dot coupled to external leads in the Kondo regime. We calculate linear and nonlinear electrical and thermal conductance and thermopower of the quantum dot and discuss the role of asymmetry in the couplings to external electrodes. In the linear regime electrical and thermal conductances are modified, while thermopower remains unchanged. In the nonlinear regime the Kondo resonance in differential conductance develops at non-zero source-drain voltage, which has important consequences on thermoelectric properties of the system and the thermopower starts to depend on the asymmetry. We also discuss Wiedemann-Franz relation, thermoelectric figure of merit and validity of the Mott formula for thermopower.

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I. INTRODUCTION

The Kondo effect, discovered more than seventy years ago in metals with small amounts of magnetic impurities and explained more than forty years ago [1], is now a prime example of many body phenomena in a wide class of correlated electron systems [2]. Due to recent advances in nanotechnology it became possible to fabricate artificial structures, where the Kondo effect can be studied systematically. Probably the best control over the isolated spin can be achieved in quantum dot (QD) systems. Originally, the Kondo effect in quantum dots was predicted theoretically in late eighties [3, 4, 5] and later confirmed in series of beautiful experiments [6, 7, 8, 9, 10, 11, 12, 13, 14].

The origin of the Kondo effect, both in alloys and in quantum dots comes from the formation of a singlet state between a localized spin and free electrons. This takes place at temperatures lower than the Kondo temperature $T_K$ and manifests itself in increasing conductance ($G$) of the system. The increase of $G$ is a result of the formation of a many body Kondo or Abrikosov-Suhl resonance at the Fermi energy.

The experiments have confirmed the validity of the theoretical picture but they also discovered phenomena which require new theoretical ideas. These include observation of the Kondo resonance at non-zero source-drain voltage [8, 9], absence of even-odd parity effects expected for these systems [10], observation of the singlet-triplet transition in a magnetic field [11], splitting of the Kondo resonance due to the ferromagnetism in the leads [12] or the interplay between the Kondo effect and superconductivity in carbon nanotube quantum dots coupled to superconducting leads [13].

In the present paper we shall focus our attention on the nonequilibrium Kondo effect (i.e. the Kondo effect at non-zero source drain voltage) in asymmetrically coupled quantum dots. In our previous work [15] we have found that the asymmetry in the couplings to the leads is the main reason responsible for the nonequilibrium Kondo effect. The effects of non-symmetric coupling have also been discussed in Refs. [16, 17, 18, 19, 20]. Here we present a more systematic study on the role of the asymmetry in the couplings to the leads in thermoelectric and transport properties of quantum dot system. Thermoelectric properties (thermopower and thermal conductance) in the Kondo regime have already been investigated in the quantum dots coupled to the normal [21, 22, 23] and the ferromagnetic [24, 25] leads, however the role of the asymmetry in the couplings has not been discussed so far. Thus we shall concentrate on the conductance, thermal conductance, thermopower and related quantities such as thermoelectric figure of merit which directly provides the information on the usefulness of the system for applications and Wiedemann-Franz ratio which signals breakdown of the Fermi liquid state when its normalized value differs from 1.

In order to calculate those quantities we use non-crossing approximation (NCA) [26], which is a widely accepted and reliable technique to study the equilibrium and nonequilibrium Kondo effect in quantum dots [15, 27, 28, 29]. Although this method is known to give non-Fermi liquid ground state, it remains valid down to temperatures below $T_K$ [30, 31].

The paper is organized as follows. In Sec. 11 we introduce the model and discuss some aspects of our procedure. The results of our calculations regarding the electrical and thermal transport in linear and nonlinear regime are presented and discussed in Secs. 111 and 114 respectively. Finally, conclusions are given in Sec. 115.

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II. THE MODEL AND APPROACH

The system we are studying consists of a quantum dot with a single energy level coupled to external electrodes. We describe it by the single impurity Anderson model \[ L \] with very strong on-dot Coulomb repulsion \((U \to \infty)\) and use slave boson representation \[ 33 \], in which the real on-dot electron operator \( \sigma \) is replaced by the product of boson \( b \) and fermion operators \( f_\sigma \) \((d_\sigma = b^+ f_\sigma)\) subject to the constraint \( b^+ b + \sum_\sigma f_\sigma^+ f_\sigma = 1 \). The resulting Hamiltonian reads

\[
H = \sum_{\lambda \kappa \sigma} \epsilon_{\lambda \kappa} c_{\lambda \kappa \sigma}^+ c_{\lambda \kappa \sigma} + \epsilon_d \sum_\sigma f_\sigma^+ f_\sigma + \sum_\lambda (V_{\lambda \kappa} c_{\lambda \kappa \sigma}^+ b_\sigma + H.c.),
\]

where \( \lambda = L (R) \) denotes left (right) lead, \( c_{\lambda \kappa \sigma}^+ \) (\( c_{\lambda \kappa \sigma} \)) is the creation (annihilation) operator for a conduction electron with the wave vector \( \kappa \), spin \( \sigma \) in the lead \( \lambda \) and \( V_{\lambda \kappa} \) is the hybridization parameter between localized electron on the dot with energy \( \epsilon_d \) and conduction electron of energy \( \epsilon_{\lambda \kappa} \) in the lead \( \lambda \).

In order to calculate electrical current \( J_{E \lambda} \) and energy flux \( J_{E \lambda} \) flowing from the lead \( \lambda \) to the central region we follow standard derivation \[ 34 \] and express all the currents in terms of retarded Keldysh Green functions \[ 35 \]. Moreover, we use relation \( J_{Q \lambda} = J_{E \lambda} - \mu_L J_{E \lambda} \) for thermal flux \( J_{Q \lambda} \), so the resulting expressions are

\[
J_{E \lambda} = \frac{i e}{h} \sum_\sigma \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma_\lambda(\omega) [G_\sigma^{\tau}(\omega) + 2i f_\lambda(\omega) \text{Im} G_\sigma^{\tau}(\omega)](2)
\]

\[
J_{Q \lambda} = \frac{i}{h} \sum_\sigma \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma_\lambda(\omega)(\omega - \mu_L) [G_\sigma^{\tau}(\omega) + 2i f_\lambda(\omega) \text{Im} G_\sigma^{\tau}(\omega)](3)
\]

where \( \Gamma_\lambda(\omega) = 2\pi \sum_\kappa |V_{\lambda \kappa}|^2 \delta(\omega - \epsilon_{\lambda \kappa}) \) is the coupling of the dot to the lead \( \lambda \), and \( G_\sigma^{\tau}(\omega) \) is the time Fourier transform of retarded Green function (GF) \( G_\sigma^{\tau}(t, t') = i \theta(t - t')[(b^+ (t) f_\sigma (t), f_\sigma^+ (t') b(t')]_\omega \) and \( G_\sigma^{\tau}(\omega) = i \{(f_\sigma^+ (t') b(t') + b^+ (t) f_\sigma (t))\} \) is the Fourier transform of lesser Keldysh GF \( 35 \). \( f_\lambda(\omega) = \left( \exp \left( \frac{\omega - \mu_L}{k_B T_\lambda} \right) + 1 \right)^{-1} \) is the Fermi distribution function in the lead \( \lambda \) with chemical potential \( \mu_\lambda \) and temperature \( T_\lambda \).

In order to calculate lesser GF \( G_{\lambda \sigma}^{\tau}(\omega) \) we use widely accepted Ng ansatz \[ 36 \], as in the presence of both the on-dot Coulomb interaction and tunneling between the QD and the leads, it is not possible to calculate \( G_{\lambda \sigma}^{\tau}(\omega) \) exactly. In this approach one assumes that full interacting lesser self-energy is proportional to the noninteracting one, and the resulting lesser self-energy is expressed in terms of retarded interacting and noninteracting self-energies. This approach has three advantages, (i) it is exact in nonequilibrium for \( U = 0 \), (ii) it is exact in equilibrium for any \( U \), and (iii) it satisfies the continuity equation \( J_L = -J_R \) in the steady state \[ 36 \]. In the present case it yields

\[
J_c = -\frac{2e}{h} \sum_\sigma \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \tilde{\Gamma}(\omega)[f_L(\omega) - f_R(\omega)] \text{Im} G_\sigma(\omega)(4)
\]

\[
J_Q = \frac{2}{\hbar} \sum_\sigma \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \tilde{\Gamma}(\omega)(\omega - eV)
\times [f_L(\omega) - f_R(\omega)] \text{Im} G_\sigma(\omega), (5)
\]

where \( \tilde{\Gamma} = \Gamma L \Gamma R/(\Gamma L + \Gamma R) \), and \( eV = \mu_L - \mu_R \). The on-dot retarded GF \( G_{\sigma}^{\tau}(\omega) \) is calculated within NCA in terms of boson and fermion propagators, which are expressed by the coupled integral equations \[ 15, 27, 28, 29 \].

In the numerical calculations we have used Lorentzian bands in the electrodes of width \( D = 100 eV \), and chosen \( \Gamma = \Gamma L + \Gamma R = 1 \) as an energy unit. The asymmetry in the couplings to the leads is defined as \( \delta = \Gamma_L / \Gamma_R \). Note that in our previous paper \[ 12 \], to faithfully reflect the situation in experiment \[ 9 \], we kept \( \Gamma_R = 1 \) and varied \( \Gamma_L \), so \( \Gamma_L + \Gamma_R \neq 1 \). Here we shall concentrate on scaling of various physical quantities with asymmetry parameter \( \delta \), therefore we keep \( \Gamma = 1 \), which means that both coupling parameters \( \Gamma_L \) and \( \Gamma_R \) change at the same time.

In the linear regime, i.e., for small voltage biases \((eV = \mu_L - \mu_R \to 0)\) and small temperature gradients \((\delta T = T_L - T_R \to 0)\) one defines the conductance \( G = -e^2 / T L_{11} \), thermopower \( S = -(1/T)(L_{12} / L_{11}) \) and thermal conductance \( \kappa = (1/T^2)(L_{22} - L_{12}^2 / L_{11}) \). The kinetic coefficients read

\[
L_{11} = T \sum_\sigma \int d\omega \tilde{\Gamma}(\omega) \text{Im} G_\sigma(\omega) \left( \frac{\partial f(\omega)}{\partial \omega} \right)_T \quad (6)
\]

\[
L_{12} = \frac{T^2}{h} \sum_\sigma \int d\omega \tilde{\Gamma}(\omega) \text{Im} G_\sigma(\omega) \left( \frac{\partial f(\omega)}{\partial T} \right)_\mu \quad (7)
\]

\[
L_{22} = \frac{T^2}{h} \sum_\sigma \int d\omega \tilde{\Gamma}(\omega)(\omega - eV) \text{Im} G_\sigma(\omega) \left( \frac{\partial f(\omega)}{\partial T} \right)_\mu \quad (8)
\]

with equilibrium Fermi distribution function \( f_L(\omega) = f_R(\omega) = f(\omega) \).

In the nonlinear regime, i.e., for any voltages and temperature differences, it is not possible to use kinetic coefficients, as they have been derived for small deviations from equilibrium \[ 27 \]. Therefore, in general case, the thermoelectric quantities have to be calculated from proper general definitions, like the nonlinear differential conductance \( G(eV) = dJ_c / d(eV) \), thermopower \( S = \Delta V / \Delta T |_{J_c = 0} \) or thermal conductance \( \kappa = -J_Q / \Delta T |_{J_c = 0} \).

III. LINEAR REGIME

In the linear regime the transport coefficients depend on the asymmetry parameter via \( \tilde{\Gamma} \) only. This stems from
the fact that the equilibrium density of states (DOS) entering Eqs. (3-8) is independent of asymmetry as long as \( \Gamma_L + \Gamma_R = 1 \). As a result \( L_{ij}(\delta) \) can be easily obtained from its behavior in the case of symmetric couplings (\( \delta = 1 \)) as

\[
L_{ij}(\delta) = 4L_{ij}^0 \frac{\delta}{(1 + \delta)^2}, \tag{9}
\]

where by \( L_{ij}^0 \) we denoted the corresponding kinetic coefficient for symmetrically coupled QD, i.e. \( L_{ij}^0 = L_{ij}(\delta = 1) \).

Knowing \( \delta \) dependence of the \( L_{ij} \), one can deduce what will be the modifications of the transport properties due to the asymmetry in the couplings. For example, the linear conductance is directly related to the kinetic coefficient \( L_{11} \) via the previously mentioned relation \( G = -(e^2/T)L_{11} \). Thus \( G(\delta) \) shows the same scaling with \( \delta \) as \( L_{ij} \) does. It is linear in \( \delta \) in the limit \( \delta << 1 \) and goes as \( 1/\delta \) for \( \delta >> 1 \).

We shall not present the numerical results for conductance and thermal conductance as for \( \delta = 1 \) as they are well known from previous studies \[21, 22, 24] \. The same is true for thermopower. However in Fig. 1 we show the temperature dependence of the (\( \delta \) independent) thermopower calculated from kinetic coefficients \( L_{ij} \) (solid line) and compare it to that obtained from so called Mott formula \[38 \] (dashed line).

![FIG. 1: The linear thermopower \( S \) (solid line) and the Mott thermopower \( S_M \) (dashed line) as a function of temperature. Both quantities are not sensitive to the asymmetry \( \delta \).](image)

Thermopower is very useful measure of the Kondo correlations \[21, 22, 24, 39] \. Below the Kondo temperature \( T_K \) it is negative, indicating particle like transport and changes sign at higher temperatures, where the transport is hole-like. At \( T \approx T_K \) it shows a broad maximum associated with single particle excitations. The sign change can also be understood from the fact that \( S \) is sensitive to the slope of the density of states at the Fermi energy. With decreasing temperature, the Kondo correlations lead to development of a narrow peak in the DOS slightly above the Fermi energy, and thus to a slope change at the \( E_F \).

At this point we would like to comment on the validity of so-called Mott formula for thermopower \[38 \], which is widely used in the literature \[40, 41, 42] \, while explaining experimental results. This formula relates thermopower and the linear conductance, i.e.

\[
S_M = -\frac{\pi^2 k_B^2 T}{3e} \frac{\partial G}{\partial E_F}, \tag{10}
\]

and is expected to work well in noninteracting systems at low temperatures \[43 \]. As we can see in Fig. 1 the Mott formula works surprisingly well in the interacting systems in the whole temperature range. The qualitative behavior of \( S_M \) is approximately the same as of linear thermopower \( S \) (solid line), giving slightly different temperature at which \( S_M \) changes sign. It also does not depend on the asymmetry \( \delta \). Thus one can conclude that the interactions do not necessarily lead to the violation of the Mott formula. Similar conclusions have been recently obtained for interacting quantum wires \[44 \].

The low temperature thermopower has been measured in the Kondo regime in quantum dot system by Scheibner et al. \[40 \]. These authors found departures from the Mott formula and attributed them to development of the narrow Kondo resonance. On the other hand, the measurements of thermopower in carbon nanotubes \[41 \] and in Zn nanowires \[42 \] qualitatively agree with the Mott formula, Eq. (10).

Figure 2 shows temperature dependence of the Wiedemann-Franz (WF) law which relates thermal and electrical transport via relation

\[
\kappa = \frac{\pi^2 k_B^2}{3e^2} T G \tag{11}
\]

This law describes transport in Fermi liquid bulk metals and in general is not obeyed in QD systems where the transport takes place through a small confined region \[21, 22, 24 \]. However, at very low temperatures, where the Kondo effect develops and the ground state of the system
has Fermi liquid nature, the WF law is recovered. At high
temperatures, the WF law is violated as the transport is
due to sequential processes leading to the suppression of
the thermal transport [24]. The WF relation does
not depend on the asymmetry δ, as both electrical and
thermal conductances show similar scaling with δ.

At this point we would like to comment on the valid-
ity of NCA, as it is well known, that this approach gives
non-Fermi liquid ground state [30, 31]. However, around
the Kondo temperature and slightly below \( T_K \) it provides
a proper description. In our case \( T_K = 5.3 \cdot 10^{-2} \Gamma \), thus
NCA is believed to give reliable results in the tempera-
ture range studied here. However, at lower temperatures
we expect a violation of WF law.

Thermoelectric figure of merit \( Z = S^2G/\kappa \) is a direct
measure of the usefulness of the system for applications.
For simple systems it is inversely proportional to opera-
tional temperature. Figure 3 shows which numerical value is an indicator of the system per-
ture. At temperature at which thermopower
fluctuations around \( T = \Gamma \) and the other one associated
with the Kondo correlations below the Kondo tempera-
ture. At temperature at which thermopower \( S \) changes
sign it vanishes. However, as one can see, its value never
exceeds 1, which indicates limited practical applicability
of the system for cryogenic purposes. Similarly as the
Wiedemann-Franz relation, it also does not depend on
the asymmetry in the couplings.

![FIG. 3: Temperature dependence of the thermoelectric figure of merit \( ZT = S^2GT/\kappa \). This quantity does not depend on the asymmetry \( \delta \).](image-url)

IV. NONLINEAR TRANSPORT

In nonlinear regime we do not expect simple scaling of
thermoelectric quantities with the asymmetry \( \delta \), as the \( \delta \)
will modify not only the effective coupling \( \tilde{\Gamma} \) in Eqs. 41
and 42 but also the QD density of states. In general,
under nonequilibrium conditions the Kondo resonance in
the DOS will be split by the bias voltage \( eV = \mu_L - \mu_R \). To be precise, there will be two resonances, one
located at \( \omega = \mu_L \) and the other one located at \( \omega = \mu_R \).
If we introduce asymmetry in the couplings \( \delta \), one of
the resonances will be broader and higher and the other
one will be narrower and lower, depending which of the
couplings \( \Gamma_L \) or \( \Gamma_R \) is stronger [15]. As a results, all
the thermoelectric quantities will be modified in more
complicated way.

The most pronounced example is the differential con-
ductance \( G_{\text{diff}} = dJ_e/d(eV) \). In symmetrically coupled
QD there is a resonance at zero source-drain bias. The
introduction of asymmetry in the couplings \( \delta \neq 1 \) shifts
the resonance to non-zero voltage [15]. Figure 4 shows
differential conductance \( G_{\text{diff}} \) for different values of the
asymmetry parameter \( \delta \). The spectra are calculated for
temperature \( T = 10^{-2} \Gamma \), below the Kondo tempera-
ture \( T_K \), which is equal to \( 5.3 \cdot 10^{-2} \Gamma \) in this case. One ob-
serves the evolution of the position of the maximum to-
wards negative voltages with increasing of the \( \delta \). This is
the effect of different heights and widths of two Kondo
resonances in the density of states, one at \( \mu_L \) and the
other one at \( \mu_R \) [15]. It is also clearly seen that the asym-
metry suppresses the conductance for all bias voltages.
This is due to suppression of the effective coupling \( \tilde{\Gamma} \) in
Eq. 4.8 by the asymmetry parameter \( \delta \). At lower tem-
peratures the maximum of the conductance is narrower
due to temperature effects and similar suppression of the
conductance with increasing \( \delta \) is observed. In Fig. 4
the position of the conductance maximum \( eV_0 \) is plotted
as a function of the asymmetry parameter \( \delta \) for various
temperatures. The reason for larger shifts of the max-
ima at higher temperatures stems from two facts. First
one is the asymmetry in the couplings, which leads to
a broader and higher (narrower and lower) Kondo res-
onance associated with tunneling between QD and left
(right) lead. The second reason, more important, is the
temperature effect. At very low temperature, the energy

![FIG. 4: Differential conductance \( G_{\text{diff}} \) as a function of the source-drain voltage \( eV \) for different coupling asymmetries \( \delta = 1, 2, 5 \) and 10 (from top to bottom), calculated at tempera-
ture \( T = 10^{-2} \Gamma \). Note evolution of the position of the resonance with \( \delta \).](image-url)
window accessible to the transport is bounded by the positions of the chemical potentials $\mu_L$ and $\mu_R$. So in fact, only half of each Kondo resonance contributes to transport. At higher temperatures, this energy window becomes wider, thus it increases effect of the asymmetry, leading to larger shifts of the maxima in $G_{\text{diff}}$. All curves in Fig. 5 show $f(\delta) = a + b/\delta$ behavior with temperature dependent coefficients $a$ and $b$.

Similarly, the maximal value of the differential conductance (associated with the Kondo effect), i.e. $G_{\text{diff}}(\epsilon V_0)$, also depends on the asymmetry parameter $\delta$, as shown in Fig. 6.

Another quantity affected by the asymmetry in the couplings is the nonlinear thermopower, shown in Fig. 7 as a function of the left lead temperature $T_L$. The right lead is kept at fixed temperature equal to $10^{-3}$T. Similarly as in the linear regime, the thermopower is negative in the low temperature (Kondo) regime and positive at high temperatures. The asymmetry $\delta$ leads to shallower minimum of $S$ in the Kondo regime, and to higher values of $S$ at temperature around $\Gamma$, associated with single particle excitations. The change of the minimum of $S$ at low temperatures reflects a fact that asymmetry changes the slope of the QD density of states at the Fermi energy due to splitting of the Kondo resonance under nonequilibrium. There is no simple scaling behavior of nonlinear thermopower with asymmetry $\delta$.

Another interesting fact is that $S$ changes sign at different temperatures for different asymmetries $\delta$, leading to different Kondo temperatures. At first sight this is in contradiction with the conclusions reached from the linear behavior of the thermopower. However one has to keep in mind that for the system out of equilibrium ($\mu_L \neq \mu_R$ and $T_L \neq T_R$) one of the Kondo peaks may disappear. For the parameters studied we always have the Kondo effect associated with the right lead but not with the left one. The density of states has a Kondo resonance at the energy $\omega = \mu_R$, while the Kondo resonance at $\mu_L$ vanishes with increasing $T_L$. If the asymmetry in the couplings is introduced, the Kondo resonance at $\mu_R$ contributes less and less to electrical and thermal transport. As a result, for large asymmetry $\delta$ one observes lower minimum at low $T_L$ and larger values of $S$ at higher temperatures.

The nonlinear thermal conductance is not sensitive to the Kondo correlations due to small contribution of low energy excitations to it. As a result the nonlinear thermal conductance $\kappa$ shows similar scaling behavior with $\delta$ as the linear one. In other words, the nonlinear $\kappa$ can be calculated, with good accuracy, from its linear values.

It is the nonlinear differential conductance which changes with $\delta$ in qualitative way. In symmetrically coupled QD ($\delta = 1$) it shows a maximum at zero bias, while the asymmetry tends to move the maximum to non-zero
vVoltages. All the other quantities change with \( \delta \) only quantitatively.

V. CONCLUSIONS

We have studied electrical and thermal transport in a quantum dot asymmetrically coupled to external leads. Those studies revealed that the most suitable tool to study the asymmetry in the couplings \( \delta \) is nonlinear differential conductance. The presence of the maximum at non-zero bias voltage directly signals that the system is asymmetrically coupled to the electrodes. In the linear regime electrical and thermal conductances show simple scaling behavior with \( \delta \) while thermopower does not depend on it. Finally, we also checked the Mott formula of thermopower and found that this quantity is a good approximation even in the presence of strong Coulomb interactions, provided the changes in the spectrum are confined to small region around chemical potential.

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