Thermoelectric effects in Kondo correlated quantum dots

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Abstract. – In this Letter we study thermoelectric effects in ultra small quantum dots. We study the behaviour of the thermopower, Peltier coefficient and thermal conductance both in the sequential tunneling regime and in the regime where Kondo correlations develop. Both cases of linear response and non-equilibrium induced by strong temperature gradients are considered. The thermopower is a very sensitive tool to detect Kondo correlations. It changes sign both as a function of temperature and temperature gradient. We also discuss violations of the Wiedemann-Franz law.

Introduction. The Kondo effect, studied since several decades in metals, has been shown to significantly affect the transport properties through quantum dots (QDs). Anticipated theoretically in Refs. it was demonstrated experimentally in Ref. Current research in this area aims at exploring features previously unaccessible in bulk systems. Examples are the study of non-equilibrium effects, decoherence, the phase sensitive transport, or more exotic models like the singlet-triplet transition. A yet unexplored problem is the study of thermoelectric effects in QDs in the presence of Kondo correlations, which allow to probe for the slope of the Kondo resonance.

Transport in the presence of electrical and thermal gradients is a well studied phenomenon in bulk systems. More recently it became possible to explore those effects in nanostructured systems, like e.g. quantum point contacts and QDs, however, in comparison with electrical transport thermoelectric effects are still much less studied. Yet they can provide additional information on the kinetics of carriers not contained in the measurement of current-voltage characteristics. Moreover they may have interesting technological applications using fabricated devices as micro-refrigerators. In single electron tunneling devices research has focused primarily on the linear thermopower. More recently the case of small

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level spacing was investigated [17,18]. Non-equilibrium effects induced by large temperature gradients were not studied so far.

In this Letter we probe the Kondo resonance by studying the thermoelectric power, the Peltier effect and the thermal conductance in a QD connected by tunnel barriers to two leads. We consider non-equilibrium situations either due to an applied bias voltage or to a temperature gradient. We make clear predictions which can be verified in experiment. The thermopower $S(T)$ changes sign at low temperatures by entering in the Kondo regime. If the Kondo resonance is splitted by a magnetic field $S(T)$ shows an additional re-entrance. Similar effects are shown when the dot is driven out of equilibrium by a strong temperature difference between the electrodes. We finally discuss the Wiedemann-Franz law and experimental realizations.

Model. In ultra-small QDs the level spacing is larger than other energy scales like the level broadening or temperature. Moreover interaction and interference lead to a repulsion of other levels from the “transport-active” one [19]. Transport can be discussed using the Hamiltonian for a single spin-degenerate level, tunnel coupled to two leads $H = H_{\text{res}} + H_{\text{dot}} + H_T$. The reservoir part $H_{\text{res}} = \sum_{k\sigma} \epsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma}$, the dot part including the interaction $H_{\text{dot}} = \sum_{k\sigma} \epsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + U n_{\uparrow} n_{\downarrow}$ and the tunneling term $H_T = \sum_{k\sigma} \langle t_{k\sigma}^r \rangle (c_{k\sigma}^\dagger + c_{k\sigma}) + \text{h.c.}$. In this work we take the charging energy $U \to \infty$, i.e. away from the symmetric regime. The bias voltage shall be applied symmetrically, i.e. $\mu_{L/R} = \mp eV/2 = \mp eV/2$. The temperature gradient is realized by heating the left reservoir by $\Delta T$, therefore $T_L = T_R + \Delta T = T + \Delta T$. We introduce the coupling $\Gamma = \Gamma^L + \Gamma^R$ through $\Gamma^r(\omega) = 2\pi \sum_k |t_{k\sigma}^r|^2 \delta(\omega - \epsilon_{k\sigma})$, which we assume to be independent of energy and spin. Furthermore we take $\Gamma^l = \Gamma/2$. The electrical current and the heat current at time $\tau$ are defined respectively as

$$I^r(\tau) = i e \sum_{k\sigma} \langle [a_{k\sigma}^\dagger (a_{k\sigma})](\tau) \rangle - \text{h.c.}, \quad (1)$$

$$I_Q^r(\tau) = i \sum_{k\sigma} \langle (\epsilon_{k\sigma} - \mu_r) [a_{k\sigma}^\dagger (a_{k\sigma})](\tau) \rangle - \text{h.c.}. \quad (2)$$

Note that due to charge conservation the electrical currents in the left and right reservoirs are the same (up to a sign) while the heat current due to the factor $\mu_r$ differs by $eV\Gamma^r$. This difference is the rate of entropy production across the QD for an out of equilibrium situation. In the following we concentrate on $I_Q^L$.

In the linear response regime the electrical current and the heat current are related to the bias voltage $V$ and the temperature difference $\Delta T$ by the constitutive equations

$$\begin{pmatrix} I \\ I_Q \end{pmatrix} = \begin{pmatrix} G(0) & L(0) \\ M(0) & K(0) \end{pmatrix} \begin{pmatrix} V \\ \Delta T \end{pmatrix}. \quad (3)$$

Experimentally however, the current $I$ is an independent variable rather than the bias voltage and correspondingly

$$\begin{pmatrix} V \\ I_Q \end{pmatrix} = \begin{pmatrix} R(0) & S(0) \\ \Pi(0) & -K(0) \end{pmatrix} \begin{pmatrix} I \\ \Delta T \end{pmatrix}. \quad (4)$$

The Onsager relations tell us that in equilibrium $S(0) = \Pi(0)/T$ and $L(0) = -M(0)/T$.

Perturbation theory. For temperatures $T > \Gamma$ an expansion in the coupling $\Gamma$ is sufficient. We obtain the currents for $\Delta \epsilon = 0$ ($\Delta \epsilon$ being the level splitting)

$$I(V, \Delta T) = 2e \Gamma^L \Gamma^R \sum_{\tau} \Gamma^r(\tau) (f_{\tau}(\epsilon) + 1) \quad (5)$$

$$I_Q^L(V, \Delta T) = \frac{\epsilon - \mu_r}{-e} I(V, \Delta T), \quad (6)$$
where \( f_r(\epsilon) \) is the Fermi function in reservoir \( r \). Having defined \( \gamma = 2\Gamma^L\Gamma^R/\left[(2\cosh(\beta\epsilon/2))^2 \sum_r \Gamma^r (f_r(\epsilon) + 1)\right] \) we calculate the thermo-electric coefficients in linear response to

\[
\begin{align*}
L^{(0)} &= e\gamma\beta\epsilon, \\
S^{(0)} &= -\beta\epsilon/e, \\
K^{(0)} &= -\gamma\beta^2\epsilon^2, \\
M^{(0)} &= -e\gamma\beta\epsilon, \\
\Pi^{(0)} &= -\epsilon/e, \\
\kappa^{(0)} &= 0.
\end{align*}
\]

(7)

The Wiedemann-Franz law is violated due to the strong energy dependence of the tunneling rates. The thermal conductance \( \kappa^{(0)} \) vanishes also out of equilibrium (up to exponentially small corrections). The reason is that in first order any thermal transport is associated with charge transport. For a measurement of \( \kappa^{(0)} \) however, the electrical current is held at zero, and hence \( \kappa^{(0)} \) has to vanish. This is not true when higher orders are included, and therefore \( \kappa \) can be used as a primary measurement for coherent contributions which manifest themselves in a non-zero value. A similar observation holds for the Peltier coefficient \( \Pi^{(0)} \) which is independent of temperature in first order. Out of equilibrium the expressions become more complicated, however, the following analytical relations can be obtained:

\[
\begin{align*}
K(V = 0, \Delta T) &= -\frac{\epsilon}{e} L(V = 0, \Delta T) \\
M(V, \Delta T = 0) &= -\left(\frac{\epsilon}{e} + V\right) G(V, \Delta T = 0).
\end{align*}
\]

(8) (9)

In particular this predicts a sign change of \( M(V, \Delta T = 0) \) at an applied bias voltage of \( V = -\epsilon/e \), when the transport switches from hole to particle like.

**Kondo regime.** For \( T < \Gamma \) perturbation theory is no longer sufficient. For levels below the Fermi edge spin-fluctuations become increasingly important and lead for \( T < T_K \) to a new strongly correlated ground state, in which the dot’s spin is completely screened. The transition to the Kondo regime is accompanied by the development of a sharp peak at the Fermi edge in the spectral density. As the thermo-electric coefficients are highly sensitive to the structure (value and slope) of the spectral density near the Fermi edge, they also display a strong temperature dependence.

In order to describe this regime (\( T_K < T < \Gamma \)) we employ the non-perturbative resonant tunneling approximation \([5]\) without vertex corrections, which is equivalent to the equation of motion method used in Ref. \([4]\), and gives qualitatively correct results for temperatures above the Kondo temperature \( T_K \). The high-energy cut-off required is chosen to be of Lorentzian-shape around 50\( \Gamma \) throughout this work. Low \( T \) calculations for the transport coefficients of the Anderson model in equilibrium have been performed using the numerical renormalization group \([20]\).

The linear thermopower as a function of temperature is shown in Fig. \( \[\] \) for three different level positions and hence different Kondo temperatures. For \( T \approx T_K \) the thermopower approaches a local minimum (not resolvable in our approximation) from which a logarithmic increase leads to a broad maximum associated to the single particle excitation before it reaches the perturbative \( 1/T \) regime. The important point is a change of the sign of \( S^{(0)} \) due to the suppression of the Kondo peak in the spectral density, which should be easy to measure. The change of sign expresses the change from particle like transport at low temperatures to hole like transport away from the Kondo regime. It can also be understood from the fact that the thermopower is sensitive to the slope of the spectral density at \( E_F \). For a level below (above) \( E_F \) the slope is negative (positive). The Kondo correlations lead to the development of a narrow peak slightly above \( E_F \), which therefore transforms the negative slope into a positive one, hence the change of sign. Note that the thermopower does not show universal scaling behavior, i.e. is not a function of \( T/T_K \). In the Kondo regime it is determined by the potential
Fig. 1 – Linear thermopower $S^{(0)}(T)$ for different level positions $\epsilon = -1.5\Gamma$ (dashed), $-2\Gamma$ (solid), and $-2.5\Gamma$ (dotted). 

scattering term, which breaks the particle hole symmetry making $S^{(0)}$ non zero and which does not scale.

The introduction of a Zeeman splitting can lead to additional structure as shown in Fig. 2. At temperatures larger than the level splitting $\Delta \epsilon$, $S^{(0)}(T)$ remains unchanged, around $T \approx \Delta \epsilon$ a local minimum is reached resulting in a change of sign at even lower $T$. This can be understood from the density of states where for $T > \Delta \epsilon$ the Kondo peaks starts to develop, leading to a negative $S^{(0)}$ at low $T$. Around $T \approx \Delta \epsilon$ the peak splits into two, changing the sign of $S^{(0)}$ and eventually suppressing it when the local minimum is developed at the Fermi edge.

The Seebeck effect is related to the voltage drop which is established through the device due to a temperature gradient. The thermopower, given by the ratio $V/\Delta T$ is a measure of this effect. Upon increasing $\Delta T$ non-linear effects may appear. We investigate these non-

Fig. 2 – Linear thermopower $S^{(0)}(T)$ for different level splittings $\Delta \epsilon = 0$ (solid), $0.01\Gamma$ (dotted), $0.02\Gamma$ (dashed) and $0.1\Gamma$ (dot-dashed) around $\epsilon = -1.5\Gamma$. 

linearities by evaluating the voltage $V_t$ at which no current is flowing at a given value of $\Delta T$. For convenience we consider the differential thermopower $S(\Delta T) = \partial V_t/\partial \Delta T|_{I=0}$ and show the results in Fig. 3. Also $S(\Delta T)$ displays a change of sign. After departing from the linear regime, characterized by $\Delta T < \max\{T_K, T\}$, the previously unaffected Kondo peak is destroyed, resulting in a logarithmic dependence. Eventually at an energy scale set by the level position $|\epsilon|$ the single particle peak is reached, where the physics is dominated by strong charge fluctuations.

The Peltier effect is related to the heat current which flows through the device in consequence of an electrical current. In linear response the Onsager relations relate $\Pi(0)$ to the thermopower. Out of equilibrium however, we study the differential Peltier coefficient defined as $\Pi(I) = \partial I_Q/\partial I|_{\Delta T=0}$. In Fig. 4 we compare the pairs of coefficients $(S, \Pi/T)$ and $(L, -M/T)$ which split upon leaving the linear regime, break the Onsager symmetries, and in addition can change the sign.

The thermal conductance $\kappa$ is not suitable to measure the Kondo correlations. As coherent processes measured by $\kappa$ are quadratically weighted by the energy, contributions close to the Fermi edge are negligible. However, we observe a peak related to the single particle excitation at higher temperatures, as shown in Fig. 5. The deviations from first order perturbation theory, even at higher temperatures, are evident and emphasize the importance of higher order processes.

The Wiedemann-Franz law relates thermal and electrical transport in metals by the relation $\kappa(0) = L_0 T G(0)$, with the Lorentz number $L_0 = \pi^2/3e^2$. In QDs transport occurs differently and consequently the Wiedemann-Franz law does not hold in general. At high temperatures transport is dominated by sequential tunneling events which leads to a suppression of $\kappa(0)$ relative to $G(0)$. At low temperatures however, the Kondo correlations create an effective Fermi liquid state for which the Wiedemann-Franz law holds again. The crossover shows the logarithmic temperature dependence characteristic for the onset of Kondo correlations modified by the peak seen in Fig. 5.

Finally we remark on the experimental check of our results. The definition of the Seebeck and Peltier coefficients imply current control, which due to non-perfect measurement devices and finite impedances of the external circuit might not be perfectly achieved. Although
Summary. We have studied electrical and heat transport through voltage and temperature biased QDs. Using perturbation theory we found analytic expressions for all coefficients in first order. We found that the measurement of the thermal conductance $\kappa$ allows to directly probe higher-order coherent contributions, and that the Wiedemann-Franz law is strongly violated at higher temperatures. We have shown that the development of Kondo correlations leads to logarithmic dependencies in the thermoelectric coefficients, temperature dependent changes of sign, and made clear predictions for experimental observation.

Fig. 4 – Differential Seebeck and Peltier coefficient vs. their driving force. $T = 0.005\Gamma$, and $\epsilon = -1.5\Gamma$. Inset: Differential $L$ and $-M/T$ in units of $e/h$.

Fig. 5 – Linear thermal conductance $\kappa^{(0)}$ for different level positions $\epsilon = -1.5\Gamma$ (dashed), $-2\Gamma$ (solid), and $-2.5\Gamma$ (dotted).
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