Universal out-of-equilibrium transport in Kondo-correlated quantum dots: a renormalized superperturbation theory on the Keldysh contour

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(Dated: November 18, 2011)

The non-linear conductance of semiconductor heterostructures and single molecule devices exhibiting Kondo physics has recently attracted attention. We address the observed sample-dependence across various systems by considering additional electronic contributions present in the effective low-energy model underlying these experiments. To this end we develop a novel version of the superperturbation theory in terms of dual fermions on the Keldysh contour. We analyze the role of particle hole asymmetry on the transport coefficients. Our approach systematically extends the work of Yamada and Yosida and others to the particle-hole asymmetric Anderson model. It correctly describes the strong coupling physics and is free of internal inconsistencies that would lead to a breakdown of current conservation.

PACS numbers: 72.10.Bg, 72.15.Qm, 73.21.La, 75.30Mb, 73.23.Hk, 71.27.+a, 73.63.Kv, 73.63.Rt

The influence of electron-electron correlation and electron-lattice interaction on the flow of charge and energy in matter has been a central topic of both explorative and applied materials science research. While the calculation of transport coefficients on the basis of the fluctuation-dissipation theorem is fairly well developed, no generally valid methods exist to go beyond the linear-response regime, as e.g. the Boltzmann equation based approach relies on well-defined quasi-particles and relaxation-time-like approximations. Of particular current interest is therefore the effect of strong electron-electron correlation on electrical and thermal conductivities beyond the linear response regime. Kondo-correlated quantum dots have served as ideal model systems to address this interplay between out-of-equilibrium dynamics and strong correlations both experimentally and theoretically. In equilibrium, the Kondo effect leads to an enhancement of the linear conductance \( G = dI/dV |_{V=0} \) to close to twice the quantum of conductance at sufficiently low temperatures and symmetric coupling (\( I \) is the current through the quantum dot and \( V \) the applied bias voltage) independent of details of e.g. the density of states of the leads. The fate of this universality away from equilibrium has been a subject of intense research. Recently, the universal aspects of steady-state charge transport in the Kondo regime beyond linear response through semiconductor heterostructures and various single molecule devices have been addressed experimentally and theoretically. It was found that the prefactors \( \alpha \) and \( \gamma \) of the non-linear conductance, defined via (\( k_B = 1 \))

\[
(G_0 - G(T, V))/(c_T G_0) = \left( \frac{T}{T_K} \right)^2 + \alpha \left( \frac{eV}{T_K} \right)^2 - \gamma c_T \left( \frac{eV T}{T_K} \right)^2
\]

(1)

differ significantly across different classes of devices. Here, \( T \) is temperature, \( G_0 = G(T \to 0, 0) \) and \( T_K \) is a dynamically generated low energy scale, the Kondo temperature.

Our primary motivation is to address the discrepancy between the results reported in (\( \alpha_G = 0.1, \gamma_G = 0.5 \)) and (\( \alpha_S = 0.05, \gamma_S = 0.1 \)) within the single-level Anderson impurity model (SIAM) as the effective low-energy model for these devices. In the strong coupling regime, this model is equivalent to the Kondo model plus a potential scattering term generated away from particle-hole (p-h) symmetry. As realistic devices are generally not p-h symmetric it is important to understand the effect of p-h asymmetry on transport properties.

Although, Kondo physics has been at the center and forefront of condensed matter physics not much is known about departures from the linear response behavior, as coded to lowest order in the coefficients \( \alpha \) and \( \gamma \). A full solution of the SIAM out of equilibrium is not available and the calculation of these transport coefficients is challenging. Results for \( \alpha \) obtained from exactly solvable cases are not directly applicable. Standard approaches, e.g. the numerical renormalization group (NRG) yield only linear response transport coefficients. Selfconsistent methods can in principle be extended to the non-linear response regime. They are conserving by construction but either fail to capture the correct ground state as in the case of the non crossing approximation or the extension onto the Keldysh contour is too involved. Bare perturbation theory is in principle applicable but its non-equilibrium extension suffers from an internal inconsistency that leads to non-conservation of the charge current away from p-h symmetry. Theoretical treatment of the non-linear transport within such frameworks therefore assumes p-h symmetry to avoid issues with current conservation. In the strong-coupling limit and at p-h symmetry...
\( \alpha \approx 0.15 \) has been obtained independently of the amount of asymmetry in the lead-dot coupling between the two leads \( \textbf{14, 23, 26} \).

As the potential scattering term is a marginally irrelevant perturbation it is expected to modify the transport coefficients but its effect should be perturbatively accessible. To address its effect in a current conserving approximation we develop a novel 'superperturbation theory' on the Keldysh contour for the SIAM. The term superperturbation theory was first used by Hafermann et al. in their treatment of a quantum impurity \( \textbf{25, 28} \): a small number of bath states defines a reference system to set up a superperturbative expansion around the numerical solution in terms of dual fermions. This numerical approach also has been extended to the Keldysh contour \( \textbf{29} \). Our approach is rather different: We organize a superperturbation theory in terms of the p-h asymmetry around the SIAM. This systematically extends the work of Yamada and Yosida and others to the asymmetric SIAM \( \textbf{21, 24, 33} \). The reference system is solved in terms of the renormalized perturbation theory of Hewson for the strong-coupling limit of the p-h symmetric SIAM extended to the Keldysh contour \( \textbf{10, 21, 34} \). This allows us to calculate the transport coefficients analytically and give a full characterization of the devices in \( \textbf{14, 15} \) in terms of the SIAM.

The SIAM Hamiltonian is \( \hat{H} = \hat{H}_c + \hat{H}_d + \hat{H}_{d-c} \), where we define

\[
\hat{H}_c = \sum_{\lambda=L,R} \sum_{k,\sigma} \epsilon_{k\lambda} \hat{c}_{k\lambda\sigma}^\dagger \hat{c}_{k\lambda\sigma},
\]

\[
\hat{H}_d = \sum_{\sigma} E_d \hat{d}_\sigma^\dagger \hat{d}_\sigma + U \left( \hat{d}_\uparrow^\dagger \hat{d}_\uparrow - \frac{1}{2} \right) \left( \hat{d}_\downarrow^\dagger \hat{d}_\downarrow - \frac{1}{2} \right) - \frac{U}{4},
\]

\[
\hat{H}_{d-c} = \sum_{\lambda=L,R} \sum_{k,\sigma} \left( V_{k\lambda} \hat{d}_\sigma^\dagger \hat{c}_{k\lambda\sigma} + V_{k\lambda}^* \hat{c}_{k\lambda\sigma}^\dagger \hat{d}_\sigma \right).
\]

Here, \( \hat{H}_c \) is the Hamiltonian for electrons in a single conduction band at the metallic leads. \( \hat{H}_d \) is the Hamiltonian for localized states in the dot, including the Coulomb interaction, and \( \hat{H}_{d-c} \) is the coupling term between the dot and the leads. We have defined \( E_d = \epsilon_d + U/2 \). For the p-h symmetric case \( \epsilon_d = -U/2 \) and hence \( E_d = 0 \). The generating functional on the Keldysh contour is given by

\[
Z = \int \mathcal{D}[\hat{\psi}^\dagger, \hat{\psi}^\dagger, \hat{\psi}, \hat{\psi}] e^{iS[\hat{\psi}^\dagger, \hat{\psi}, \hat{\Phi}^\dagger, \hat{\Phi}],}
\]

where the action on the Keldysh contour is expressed in terms of a functional integral over time-dependent Grassmann fields. \( \hat{\psi}_{k\lambda\sigma}(t) = (c_{k\lambda\sigma}^\dagger(t), c_{k\lambda\sigma}(t))^\dagger \) and \( \hat{\Phi}(t) = (d_{\sigma}^\dagger(t), d_\sigma(t))^\dagger \). Here, the indexes \( \pm \) refer to the time-ordered (–) and anti-time-ordered (+) path along the closed Keldysh contour \( \textbf{33} \). The lead electrons are non-interacting and the resulting Gaussian integrals in Eq. \( \textbf{44} \) can be carried out. This results in

\[
Z = \int \mathcal{D}[\hat{\Phi}_{\sigma\omega}, \hat{\Phi}_{\sigma\omega}] e^{iS[\hat{\Phi}_\sigma, \hat{\Phi}_{\sigma\omega}]},
\]

The effective action \( S \) is defined as

\[
S[\hat{\Phi}_\sigma, \hat{\Phi}_{\sigma\omega}] = S_U[\hat{\Phi}_\omega, \hat{\Phi}_{\sigma\omega}] = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \sum_{\sigma} \hat{\Phi}_\omega^\dagger E_d \hat{\Phi}_{\sigma\omega},
\]

where

\[
S_U[\hat{\Phi}_\sigma, \hat{\Phi}_{\sigma\omega}] = S_U^\text{int}[\hat{\Phi}_\omega, \hat{\Phi}_{\sigma\omega}] + \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \sum_{\sigma} \hat{\Phi}_\omega^\dagger (\omega + (G_L + G_R)\sigma_3) \hat{\Phi}_{\sigma\omega},
\]

is the effective action for a p-h symmetric \( (E_d = 0) \) and interacting \( (U \neq 0) \) system, and

\[
\Gamma_\lambda = -\sum_{k,\sigma} \frac{|V_{k\lambda}|^2}{\omega - \epsilon_{k\lambda} + i\eta^+} \quad \text{for} \quad \lambda = L, R.
\]

For simplicity, we assume that the density of states of left and right lead, \( \rho_\lambda(\omega) = \sum_\delta \delta(\omega - \epsilon_{k\lambda}) \), are identical and p-h symmetric. P-h asymmetry in \( \rho_\lambda(\omega) \) will have a similar effect as the local p-h asymmetry generated by a finite \( E_d \). Each lead \( (L/R) \) is in equilibrium and is characterized by temperature \( (T) \) and its chemical potential \( (\mu_L/\mu_R) \).

We use the superperturbation scheme \( \textbf{27} \) to treat the term proportional to \( E_d \) in the effective action, Eq. \( \textbf{15} \), by using as a reference system the p-h symmetric and interacting system described by the effective action \( S_U[\hat{\Phi}_\sigma, \hat{\Phi}_{\sigma\omega}] \) in Eq. \( \textbf{45} \). This yields

\[
G_{\sigma\omega} = -E_d^{-1} \hat{\delta}_3 + (g_{\sigma\omega} \Delta E_d \hat{\delta}_3)^{-1} G_{\sigma\omega}^f (E_d \hat{\delta}_3 g_{\sigma\omega})^{-1},
\]

where \( g_{\sigma\omega} \) is the Green’s function matrix for the interacting \( (U \neq 0) \) asymmetric \( (E_d \neq 0) \) SIAM, \( \hat{\delta}_3 \) is the third Pauli matrix, and \( g_{\sigma\omega} \) is the Green’s function for the interacting \( (U \neq 0) \) and symmetric \( (E_d = 0) \) SIAM. Finally, \( G_{\sigma\omega}^f \) is the dual fermion matrix Green’s function, obtained from the solution of the matrix Dyson equation

\[
G_{\sigma\omega}^f = G_{\sigma\omega}^{f(0)} + G_{\sigma\omega}^{f(0)} \Sigma_{\sigma\omega}^f G_{\sigma\omega}^{f(0)},
\]

and the bare dual fermion Green’s function is defined by the expression

\[
G_{\sigma\omega}^{f(0)} = -g_{\sigma\omega} (g_{\sigma\omega} - E_d^{-1} \hat{\delta}_3)^{-1} g_{\sigma\omega}.
\]

By construction, the expansion works for small and large \( E_d \) \( \textbf{27} \). The explicit expression for the retarded self-energy obtained from our superperturbation scheme, up to \( O(T^2V^2) \), is

\[
\Sigma_{E_d} = \left( 1 - \chi_{++} \right) \omega + E_d - \chi_{++}^{-1} E_d \left( \frac{U}{\pi \Delta} \right) \left\{ 1 - \frac{\chi_{++}^2}{3} \right\} \times \left[ \left( \frac{\pi T}{\Delta} \right)^2 + \frac{\zeta(eV/\Delta)^2}{3} \right] + 2 \frac{\zeta^4}{3} \chi_{++} \left( \frac{\pi T eV}{\Delta^2} \right)^2 \right\} -i \frac{\Delta}{2} \left( \frac{U}{\pi \Delta} \right)^2 \left( \frac{\chi_{++}^2}{3} \right) + \chi_{++} \left( \frac{\pi T}{\Delta} \right)^2 + \frac{\zeta(eV/\Delta)^2}{3} \right\}.
\]

\( \chi_{++} \) is the real part of the zero-temperature self-energy for symmetric leads, which is a function of the temperature, chemical potential, and coupling constant. The explicit expression for the retarded self-energy obtained from our superperturbation scheme, up to \( O(T^2V^2) \), is

\[
\Sigma_{E_d} = \left( 1 - \chi_{++} \right) \omega + E_d - \chi_{++}^{-1} E_d \left( \frac{U}{\pi \Delta} \right) \left\{ 1 - \frac{\chi_{++}^2}{3} \right\} \times \left[ \left( \frac{\pi T}{\Delta} \right)^2 + \frac{\zeta(eV/\Delta)^2}{3} \right] + 2 \frac{\zeta^4}{3} \chi_{++} \left( \frac{\pi T eV}{\Delta^2} \right)^2 \right\} -i \frac{\Delta}{2} \left( \frac{U}{\pi \Delta} \right)^2 \left( \frac{\chi_{++}^2}{3} \right) + \chi_{++} \left( \frac{\pi T}{\Delta} \right)^2 + \frac{\zeta(eV/\Delta)^2}{3} \right\}.
\]
with $\tilde{x}_{++} = 1 + (3 - \pi^2/4)(U/\pi\Delta)^2 + O(U^4)$ and $\zeta = 3\beta/(1 + \beta)^2$ where $\beta = \Gamma_L/\Gamma_R$ measures the asymmetry in the lead-to-dot couplings. It is easy to see that our results reproduce the resonant level case for $(U = 0)$ and also the special case $E_d = U/2$ at $V = 0$ considered in [21] thereby establishing the power of this approach.

We now turn to the evaluation of the transport coefficients. The steady-state current through the dot follows from the continuity equation [30]

$$I = \left(\frac{e}{h}\right) \int_{-\infty}^{+\infty} \frac{d\omega}{\Gamma_R + \Gamma_L} [f_L(\omega) - f_R(\omega)] A(\omega, V), \quad (12)$$

where $A(\omega, V)$ is the local spectral function (in the presence of the dot-lead coupling); $f_L/f_R$ is Fermi function in the left/right lead, respectively and $V = (\mu_L - \mu_R)/e$.

We now demonstrate that this scheme gives the correct conductance even away from p-h symmetry. Current conservation holds if $G^<(\omega)\Sigma^>(\omega) - G^>(\omega)\Sigma^<(\omega) = 0$ [22]. This in turn is ensured if $F_U(\omega) = F_U(\omega)$, where the distribution functions are defined by $G^<(\omega) \equiv -F_U(\omega)(G^>(\omega) - G^0(\omega))$ and $\Sigma^<(\omega) \equiv -F_U(\omega)(\Sigma^>(\omega) - \Sigma^0(\omega))$. It is easy to see that any self-consistent theory will fulfill this condition [14]. To demonstrate that our scheme is indeed current conserving up to the order of approximation $O(U^2, E_d^2)$, we note that the retarded component of the self-energy is $\Sigma_{\alpha}^{R} = -\Sigma_{\alpha}^{++} + \Sigma_{\alpha}^{+} + \Sigma_{\alpha}^{ hotted}$ so that

$$F_U(\omega)(\Sigma_{\alpha}^{R} - \Sigma_{\alpha}^{0}) = f_{eff}(\omega)(\Sigma_{\alpha}^{R} - \Sigma_{\alpha}^{0}) + O(U^4)$$

$$= -i\Delta \left(\frac{U}{2\Delta}\right)^2 \left[\left(\frac{eU}{\Delta}\right)^2 + \left(\frac{eV}{\Delta}\right)^2 + \zeta \frac{(eV)^2}{\Delta^2}\right] f_{eff}(\omega)$$

$$= \Sigma_{\alpha}^{++}, \quad (13)$$

which implies that this is indeed a current-conserving approximation. In Eq. [13] we have chosen the chemical potentials as to satisfy $\Gamma_L|\mu_L| = \Gamma_R|\mu_R|$ for convenience. For the special case of identical leads, the distribution function of the local Green function is given by $f_{eff}(\omega) = (f_L(\omega)\Gamma_L + f_R(\omega)\Gamma_R)/(\Gamma_L + \Gamma_R)$ [37]. To the same order of approximation as the self-energies are calculated, the effective distribution function for the interacting system is $F_U(\omega) \sim f_{eff}(\omega) - (\zeta/12)(U/\pi\Delta)^2(\pi eV/\Delta)^2 f_0^0(\omega)$, with $f_0^0$ the Fermi-Dirac distribution at $\mu = 0$. This again demonstrates the current-conserving nature of our scheme. The non-linear conductance follows from Eq. [12] and the approximation for $A(\omega, V)$, obtained from the retarded self-energy Eq. [11];

$$G(T, V = 0) = G_0 \left[1 - c_T \left(\frac{k_B T}{\Delta}\right)^2\right], \quad (15)$$

where

$$c_T = \frac{\pi^2}{3} \frac{1}{2}\left(-2e_2^2 + e_2^2(3 - 8\upsilon^2)\right)$$

The renormalized parameters are defined as $\tilde{\epsilon}_d = E_d/\Delta, \tilde{\Delta} = \tilde{x}_{++}^{-1}, \tilde{\upsilon} = \tilde{x}_{++}^{-1}(U/\pi\Delta)$, and $G_0 = (2e^2/\hbar)^2 \frac{\epsilon_2}{2}(1 + (1 - \upsilon^2)\epsilon_2^2)^{-1}$ which reproduces the exact result from Friedel’s sum rule up to order $O(\epsilon_2^2)$ as $[\sin(\xi/\beta) - \tan^{-1}(\epsilon_d)]^2 \sim 1 - \epsilon_d^2$. The transport coefficients $c_{TV}$, $c_{TV}$ are to $O(\epsilon_2^2)$ given by

$$c_{TV} = \frac{1}{2}(\epsilon_2^2 + (1 - 2\xi)(5\epsilon_2^2 - 1) + (5 - 8\xi))) \upsilon^2 + (1 + 2\xi)(2\xi^2 + \upsilon^2)$$

$$c_{TV} = \frac{1}{2}(\epsilon_2^2 + (1 - 2\xi)(5\epsilon_2^2 - 1) + (5 - 8\xi))) - \xi^2(1 - \upsilon^2)^2$$

and $c_{TV}$ and $c_{TV}$ are proportional to the product of lead-dot asymmetry $\beta$ and p-h asymmetry $\tilde{\epsilon}_d$. This product and hence $c_{TV}$ and $c_{TV}$ are expected to be small in the most experimental realizations. Interestingly, the ratio of $c_{TV}$ and $c_{TV}$ is only a function of $\upsilon$.

In Fig. [1] we show our results for $\alpha$ and $\gamma$ for various cuts through parameter space $(\tilde{\upsilon}, \tilde{\epsilon}_d, \beta)$. Note, that in the strong coupling limit $(\tilde{\upsilon} \rightarrow 1)$ the dependence on $\tilde{\epsilon}_d$ vanishes reflecting the fact that this limit is p-h symmetric (see Fig. [1](a) and (b)). $\gamma$ retains its dependence on $\beta$ in this limit while $\alpha$ becomes independent of $\beta$ for $\tilde{\upsilon} \rightarrow 1$. Fig. [1](c) and (d) show the ratio $\gamma/\alpha$. Eq. [17] shows that $c_{TV}$ and $c_{TV}$ are proportional to the product of lead-dot asymmetry $\beta$ and p-h asymmetry $\tilde{\epsilon}_d$. This product and hence $c_{TV}$ and $c_{TV}$ are expected to be small in most experimental realizations. Interestingly, the ratio of $c_{TV}$ and $c_{TV}$ is only a function of $\upsilon$. For the p-h symmetric case our expressions reduce to the results obtained by Oguri and others [10, 29].

We are now in a position to address the experimental results for $\alpha = c_{TV}/c_T$ and $\gamma = c_{TV}/c_T^2$ mentioned in the introduction [14, 15]. A major experimental challenge is to reliably extract the dynamically generated low-energy scale $\Delta = T_K$. The phenomenological formula

$$G(T, 0) = \frac{G_0}{(1 + (2s - 1)(T/T_K)^2)^s} \quad (18)$$

is commonly employed to extract $T_K$ [3]. The parameter $s$ apparently fixes $c_T$ from Eqs. [15] and [18] ($s = 0.21$ as in [14] leads to $c_T \approx 5.5$ and $s = 0.22$ [15] results in $c_T \approx 4.9$). It is clear from Eq. [19] that $c_T$ is not only
Our approach is current conserving. This scheme may hence be used to characterize nano-structured devices in terms of the underlying model beyond empirical formulas. It can also be applied to obtain magneto- and thermal transport properties. Finally, it is possible to generalize the results for the single-level Anderson model to more complex systems.

We thank A. Antipov, D. Natelson and in particular T. Costi for many useful discussions. E.M. and S.K. acknowledge support by the Comisión Nacional de Investigación Científica y Tecnológica (CONICYT), grant No. 11100064 and the German Academic Exchange Service (DAAD) under grant No. 52636698.

Note added After completion of this work we became aware of Ref. [38], which addresses the effect of p-h asymmetry on $\alpha$ within a perturbation theory around the p-h asymmetric case. A problem with this approach is that it fails to recover p-h symmetry at $\tilde{u} = 1$ and in addition it gives an unphysical linear in T term in the spectral density away from half filling $n = 1$.

![FIG. 1: (Color online) (a) $\alpha$ and (b) $\gamma$ versus $\tilde{u}$ for various $\tilde{\varepsilon}_d$ and $\beta$. (c) ratio $\gamma/\alpha$ versus $\tilde{\varepsilon}_d$ and (d) the asymmetry in the lead-dot couplings $\beta$ for selected $\tilde{u}$. In (a)-(d), we used the full expressions in $\tilde{\varepsilon}_d$ whereas Eqs. (16) and (17) include terms only up to order $O(\tilde{\varepsilon}_d^2)$.](image)

a function of $\tilde{u}$ but also depends on the p-h asymmetry through $\tilde{\varepsilon}_d$. This complicates the experimental extraction of $T_K$. On the other hand, $\alpha$ and $\gamma$ are ratios of transport coefficients and insensitive to the precise definition of $T_K$. Fig. 1(c) shows the ratio $\gamma/\alpha$ versus p-h asymmetry. From the reported values [14, 15] it then becomes clear that charge fluctuations (i.e., no p-h symmetry) are present in both experiments. According to [14, 12] the coefficients $c_{\nu E_d}$ and $c_{\nu V E_d}$ are indeed vanishingly small. Yet, they may have been detected in [15]. The experimental values reported in [15] are compatible with e.g. $\tilde{u} = 0.45, \tilde{\varepsilon}_d = 0.1, \beta = 1$ yielding $\alpha = 0.1$ and $\gamma = 0.47$.

While we can reproduce $\alpha S$ or $\gamma S$ of [15] independently, it is not possible to reproduce both consistently within the SIAM. The underlying low-energy model of the experiment [15] cannot simply be the SIAM. One possible generalization is that more than one level participates. The renormalized low-energy properties. Then, already $G_0$ is no longer given solely in terms of the occupation $n_d$ and the lead-to-dot couplings will enter explicitly [38]. A more likely alternative is that local phonon modes renormalize the transport coefficients $\alpha$ and $\gamma$ differently.

Our scheme thus constitutes a convenient analytic way of characterizing an experimental device in terms of the renormalized parameters $\tilde{u}, \tilde{\varepsilon}_d$ and $\beta$ and the low-energy scale $\Delta$. Inverting the definitions for the renormalized parameters will give access to the bare ones [34].

In summary, we have developed a novel analytic scheme based on dual fermions to obtain non-linear transport coefficients for the strong coupling lines of the Anderson model. This approach is reliable and controlled as demonstrated. It is rooted in the marginal irrelevance of the potential scattering term generated by p-h asymmetry and on the approaches of [10, 32, 34] as a reference system as well as on the dual fermion approach of [27].
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