Universal Out-of-Equilibrium Transport in Kondo-Correlated Quantum Dots: Renormalized Dual Fermions on the Keldysh Contour

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The nonlinear conductance of semiconductor heterostructures and single molecule devices exhibiting Kondo physics has recently attracted attention. We address the observed sample dependence of the measured steady state transport coefficients by considering additional electronic contributions in the effective low-energy model underlying these experiments that are absent in particle-hole symmetric setups. A novel version of the superperturbation theory of Hafermann *et al.* in terms of dual fermions is developed, which correctly captures the low-temperature behavior. We compare our results with the measured transport coefficients.

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Quantum matter out of equilibrium is currently investigated in a wide range of settings ranging from cold atom setups and light-matter systems to various condensed matter systems. Depending on the context, the focus ranges from thermalization of quantum matter to the description of relaxation processes to the microscopic characterization of nonthermal steady states. In condensed matter systems, with couplings to well-defined heat and particle reservoirs, current-carrying steady states are of particular interest [1,2].

In this Letter, we are concerned with the nonlinear conductance of a model system of strong electron-electron interaction. Traditionally, the calculation of transport properties on the basis of the fluctuation-dissipation theorem is fairly well developed. Yet, no generally valid method exists to go beyond the linear-response regime, as e.g., a Boltzmann equation based approach relies on well-defined quasiparticles 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 (I is the current through the quantum dot and V the applied bias voltage) independent of any details of e.g., the density of states of the leads. The fate of this universality away from equilibrium has been the subject of intense research [3–7]. 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 [8–10]. It was found that the prefactors α and γ of the nonlinear conductance, defined via ($k_B = 1$)

$$[G_0 - G(T,V)]/(c_T G_0) = \left(\frac{T}{T_K}\right)^2 + \alpha\left(\frac{eV}{T_K}\right)^2 - \gamma c_T\left(\frac{eVT}{T_K^2}\right)^2 \quad (1)$$

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

Our primary motivation is to address the systematic difference between the results reported in Ref. [8] ($\alpha_G = 0.1$, $\gamma_G = 0.5$) and [9] ($\alpha_S = 0.05$, $\gamma_S = 0.1$) 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. Particle-hole symmetry can easily be broken either locally on the device itself [see below Eq. (2)] or in the leads connected to the device (see below Eq. (7) [11]). Consequently, realistic devices are generically not p-h symmetric and it is important to understand the effect of p-h asymmetry on transport properties. An immediate consequence of p-h asymmetry is that the number of electrons localized on the device is no longer fixed to be $1/2$ (per spin component).

Theoretically, not much is known about α and γ. A full solution of the SIAM out of equilibrium is not available and the calculation of these transport coefficients is challenging. Results for α obtained from exactly solvable cases are not directly applicable [3,12]. Standard approaches, e.g., the numerical renormalization group, yield only linear response transport coefficients [13]. Selfconsistent methods can in principle be extended to the nonlinear response regime. They are conserving by construction [14] but

either fail to capture the correct ground state as, e.g., the noncrossing approximation or the extension onto the Keldysh contour is too involved [15]. As the potential scatterer is a marginally irrelevant perturbation it is expected to modify the transport coefficients but its effect should be pertubatively accessible. 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 [5,16–19].

The SIAM Hamiltonian is $\hat{H} = \hat{H}_c + \hat{H}_d + \hat{H}_{d-c}$, where

$$\hat{H}_c = \sum_{\lambda=L,R} \sum_{k,\sigma} \epsilon_{k\lambda} \hat{c}^\dagger_{k\lambda\sigma} \hat{c}_{k\lambda\sigma},$$
$$\hat{H}_d = \sum_\sigma E_d \hat{d}^\dagger_\sigma \hat{d}_\sigma + U\left(\hat{d}^\dagger_\uparrow \hat{d}_\uparrow - \frac{1}{2}\right)\left(\hat{d}^\dagger_\downarrow \hat{d}_\downarrow - \frac{1}{2}\right) - \frac{U}{4},$$
$$\hat{H}_{d-c} = \sum_{\lambda=L,R} \sum_{k,\sigma} (V_{k\lambda} \hat{d}^\dagger_\sigma \hat{c}_{k\lambda\sigma} + V^*_{k\lambda} \hat{c}^\dagger_{k\lambda\sigma} \hat{d}_\sigma). \quad (2)$$

Here, $\hat{H}_c$ is the Hamiltonian for electrons in the metallic leads labeled by $\lambda = L$ and $\lambda = R$. $\hat{H}_d$ describes the 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 $E_d = 0$.

Beyond setting up a systematic expansion for α and γ in terms of E_d and up to $O(V^2)$, we also address the issue of current conservation beyond $O(V^2)$. Away from p-h symmetry, a proper treatment of the (renormalized) interaction vertex is necessary to reproduce, e.g., the correct local occupation already in equilibrium. Since, by continuity, particle flow is connected to the rate of change of the local occupation, any sensible approximation has to respect the corresponding symmetries of the interaction vertex in order to be current-conserving [14]. As discussed by Hershfield *et al.* [21], for perturbation theory in U, steady state current conservation holds only in the p-h symmetric SIAM. We therefore develop an approach to transport in the p-h asymmetric SIAM based on dual fermions [22] that is based on perturbation theory in U for the p-h symmetric SIAM [5,20,23,24]. As demonstrated explicitly, our results are rigorous up to $O(V^2)$ and are current conserving [beyond $O(V^2)$]. As the p-h symmetric SIAM includes Coulomb interactions, the expansion around it is delicate. We use the dual fermion method [25,26] which yields a formal expansion built around the four-point vertex of the reference system with $E_d = 0$. This systematically extends the work of Yamada and Yosida and Zlatić and Horvatić to the asymmetric SIAM [20,24,27–29] and results in a controlled expansion for the transport coefficients up to, and including, $O(U^2 E_d^2)$. A generalization to higher orders is possible [30].

The generating functional on the Keldysh contour is given by

$$Z = \int \mathcal{D}[\hat{\psi}^\dagger, \hat{\psi}] \mathcal{D}[\hat{\Phi}^\dagger, \hat{\Phi}] e^{iS[\hat{\psi}^\dagger, \hat{\psi}, \hat{\Phi}^\dagger, \hat{\Phi}]}, \quad (3)$$

where the action on the Keldysh contour is expressed in terms of a functional integral over time-dependent Grassmann fields, $\hat{\psi}^\dagger_{k\lambda\sigma}(t) = (c^-_{k\lambda\sigma}(t), c^+_{k\lambda\sigma}(t))^\dagger$ and $\hat{\Phi}^\dagger(t) = (d^-_\sigma(t), d^+_\sigma(t))^\dagger$. Here, the indices $\pm$ refer to the time-ordered $(-)$ and anti-time-ordered $(+)$ path along the closed Keldysh contour. Each lead (L/R) is taken to be in equilibrium and characterized by its temperature $(T_L = T_R = T)$ and its chemical potential (μ_L/μ_R).

The lead electrons are noninteracting and the resulting Gaussian integrals can be carried out, resulting in

$$Z = \int \mathcal{D}[\hat{\Phi}^\dagger_{\sigma\omega}, \hat{\Phi}_{\sigma\omega}] e^{iS[\hat{\Phi}^\dagger_{\sigma\omega}, \hat{\Phi}_{\sigma\omega}]}, \quad (4)$$

where the effective action S is given by

$$S[\hat{\Phi}^\dagger_{\sigma\omega}, \hat{\Phi}_{\sigma\omega}] = S_U[\hat{\Phi}^\dagger_{\sigma\omega}, \hat{\Phi}_{\sigma\omega}] - \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \sum_\sigma \hat{\Phi}^\dagger_{\sigma\omega} E_d \hat{\sigma}_3 \hat{\Phi}_{\sigma\omega}, \quad (5)$$

and

$$S_U[\hat{\Phi}^\dagger_{\sigma\omega}, \hat{\Phi}_{\sigma\omega}] = S^{\text{int}}_U[\hat{\Phi}^\dagger_{\sigma\omega}, \hat{\Phi}_{\sigma\omega}] + \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \times \sum_\sigma \hat{\Phi}^\dagger_{\sigma\omega}[\omega + (\Gamma_L + \Gamma_R)] \hat{\sigma}_3 \hat{\Phi}_{\sigma\omega} \quad (6)$$

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

$$\Gamma_\lambda = -\sum_{k,\sigma} \frac{|V_{k\lambda}|^2}{\omega - \epsilon_{k\lambda} + i\eta^+} \qquad \text{for } \lambda = L, R. \quad (7)$$

For simplicity, we assume that the density of states of the left and right lead, $\rho_\lambda(\omega) = \sum_k \delta(\omega - \epsilon_{k\lambda})$, are identical and p-h symmetric, $\rho_\lambda(\omega) = \rho_\lambda(-\omega)$ [11]. In the wide band limit, we set $i\Delta = \Gamma_L + \Gamma_R$. To generate an expansion in terms of E_d, we decouple the 2nd term on the right-hand side of Eq. (5) into $\phi^\dagger_{\sigma\omega} \mathbf{g}^{-1}_{\sigma\omega} \hat{\Phi}_{\sigma\omega}$ via a fermionic Hubbard-Stratonovich transformation, where $\mathbf{g}_{\sigma,\omega}$ is the Green's function for the interacting $(U \neq 0)$ and symmetric $(E_d = 0)$ SIAM [22]. One can show [22,30]

$$\mathbf{G}_{\sigma,\omega} = -E_d^{-1} \hat{\sigma}_3 + (\mathbf{g}_{\sigma,\omega} E_d \hat{\sigma}_3)^{-1} \mathbf{G}^f_{\sigma,\omega} (E_d \hat{\sigma}_3 \mathbf{g}_{\sigma,\omega})^{-1}, \quad (8)$$

where $\mathbf{G}_{\sigma,\omega}$ is the Green's function matrix for the interacting $(U \neq 0)$ asymmetric $(E_d \neq 0)$ SIAM, $\hat{\sigma}_3$ is the third Pauli matrix, and $\mathbf{G}^f_{\sigma,\omega}$ is the dual fermion matrix Green's function, obtained from the solution of the matrix Dyson equation

$$\mathbf{G}^f_{\sigma,\omega} = \mathbf{G}^{f(0)}_{\sigma,\omega} + \mathbf{G}^{f(0)}_{\sigma,\omega} \mathbf{\Sigma}^f_{\sigma,\omega} \mathbf{G}^f_{\sigma,\omega}, \quad (9)$$

where the bare dual fermion Green's function is defined by $\mathbf{G}^{f(0)}_{\sigma,\omega} = -\mathbf{g}_{\sigma,\omega}(\mathbf{g}_{\sigma,\omega} - E_d^{-1}\hat{\sigma}_3)^{-1}\mathbf{g}_{\sigma,\omega}$. The dual fermion

self-energy $\mathbf{\Sigma}^{f}_{\sigma,\omega}$ is given in terms of $\mathbf{g}_{\sigma,\omega}$ and the four-point vertex of the interacting ($U \neq 0$) and symmetric ($E_d = 0$) SIAM [30]. So far, no approximation has been made and this expansion is expected to work for small as well as large E_d [25]. We proceed by solving the reference system ($E_d = 0$) within the renormalized perturbation theory around the strong coupling fixed point [5,23,28]. For a systematic expansion in E_d up to $O(E_d^2)$, we keep only the first two terms in the Dyson series for $\mathbf{G}^{f(0)}_{\sigma,\omega}$. As a result, the explicit expression for the retarded self-energy at finite bias voltage $\mu_L - \mu_R = \mathrm{eV}$ obtained from our scheme up to, including, $O(T^2V^2)$, is [30,31]

$$\begin{aligned}\Sigma^r_{E_d} =& (1-\tilde{\chi}_{++})\omega + E_d - \tilde{\chi}^{-1}_{++}E_d\left(\frac{U}{\pi\Delta}\right)\\ &\times\left\{1-\frac{\tilde{\chi}^2_{++}}{3}\left[\left(\frac{\pi T}{\Delta}\right)^2+\zeta\left(\frac{eV}{\Delta}\right)^2\right]+7\frac{\zeta}{9}\tilde{\chi}^4_{++}\left(\frac{\pi TeV}{\Delta^2}\right)^2\right\}\\ &-i\frac{\Delta}{2}\left(\frac{U}{\pi\Delta}\right)^2\left[\left(\frac{\omega}{\Delta}\right)^2+\left(\frac{\pi T}{\Delta}\right)^2+\zeta\left(\frac{eV}{\Delta}\right)^2\right.\\ &\left.-\frac{\zeta}{3}\left(\frac{\pi TeV}{\Delta^2}\right)^2\tilde{\chi}^2_{++}\right],\end{aligned} \tag{10}$$

with $\tilde{\chi}_{++} = 1 + (3 - \pi^2/4)(U/\pi\Delta)^2 + O(U^4)$ [20,23] and $\zeta = 3\kappa/(1+\kappa)^2$ where $\kappa = \Gamma_L/\Gamma_R$ measures the asymmetry in the lead-to-dot couplings. Notice that there are no terms of $O(E_d^2U)$ nor $O(E_d^2U^2)$ in Eq. (10). The next leading correction to the retarded self-energy is $O(E_d^3U, E_dU^3)$ [30]. For $U = 0$, Eq. (10) reduces to the corresponding result of the resonant level model.

We now turn to a discussion of the current. The steady-state current through the dot [21,32],

$$I = \left(\frac{e}{\hbar}\right)\int_{-\infty}^{+\infty} d\omega \frac{4\Gamma_R\Gamma_L}{\Gamma_R+\Gamma_L}[f_L(\omega) - f_R(\omega)]A(\omega,T,V), \tag{11}$$

follows from the continuity equation and relies on current conservation $I_L + I_R = 0$ in the steady state to recast I entirely in terms of the spectral density. As a result, Eq. (11) poses a strong constraint on admissible local distribution functions $F(\omega, T, V)$, where F is defined through $G^{-+} = F(\omega,T,V)(G^a - G^r)$ [30]. Here, $I_{L/R}$ is the current from the left or right lead to the dot, $A(\omega, T, V)$ is the local spectral density (in the presence of the dot-lead coupling) and f_L/f_R is the Fermi function in the left or right lead, respectively. A second local distribution function $\tilde{F}(\omega, T, V)$ can be introduced via $\Sigma^{-+} = \tilde{F}(\omega,T,V)(\Sigma^a - \Sigma^r)$. For the SIAM considered here one can show that $F(\omega, T, V) = \tilde{F}(\omega, T, V)$ in the steady state limit. This in turn implies $G^{-+}\Sigma^{+-} = G^{+-}\Sigma^{-+}$ which ensures current conservation [21,30]. Note, that in general one cannot conclude $F = \tilde{F}$ away from equilibrium.

Current conservation of our approach beyond $O(V^2)$ follows from the general relations $\Sigma^{++}_{E_d} + \Sigma^{--}_{E_d} - \Sigma^{+-}_{E_d} - \Sigma^{-+}_{E_d} = 0$, $\Sigma^{++}_{E_d} = -(\Sigma^{--}_{E_d})^*$, $\Sigma^r_{E_d} = \Sigma^{--}_{E_d} - \Sigma^{-+}_{E_d}$, and Eq. (10) which imply

$$\begin{aligned}&F(\omega,T,V)(\Sigma^a_{E_d} - \Sigma^r_{E_d})\\ &= i\Delta\left(\frac{U}{\pi\Delta}\right)^2\left[\left(\frac{\omega}{\Delta}\right)^2+\left(\frac{\pi T}{\Delta}\right)^2+\zeta\left(\frac{eV}{\Delta}\right)^2\right.\\ &\left.-\frac{\zeta}{3}\left(\frac{\pi TeV}{\Delta^2}\right)^2\tilde{\chi}^2_{++}\right]f_{\mathrm{eff}}(\omega,T,V) = \Sigma^{-+}_{E_d},\end{aligned} \tag{12}$$

where we introduced $f_{\mathrm{eff}}(\omega, T, V) = (\kappa f_L + f_R)/(1+\kappa)$. Equation (12) shows that within our scheme $F(\omega, T, V) = \tilde{F}(\omega, T, V)$. The local distribution function F turns out to be [30]

$$F(\omega,T,V) = \frac{\Gamma_L f_L + \Gamma_R f_R - f_{\mathrm{eff}}(\omega,T,V)\mathrm{Im}\Sigma^r}{1 - \mathrm{Im}\Sigma^r}. \tag{13}$$

The nonlinear conductance follows from Eq. (11) and the approximation for $A(\omega, T, V) = -\pi^{-1}\mathrm{Im}G^r$, where $G^r = (\omega + i\Delta - \Sigma^r_{E_d})^{-1}$ is the retarded Green function. We are primarily interested in the transport coefficients in the vicinity of the strong coupling fixed point, where our expansion is in terms of renormalized parameters [23]. The renormalized parameters are defined as $\tilde{\epsilon}_d = E_d/\Delta$, $\tilde{\Delta} = \tilde{\chi}^{-1}_{++}\Delta$, $\tilde{u} = \tilde{\chi}^{-1}_{++}(U/\pi\Delta)$. In terms of these, one finds

$$\begin{aligned}\frac{G(T,0) - G(T,V)}{G_0} =& c_V\left(\frac{eV}{\tilde{\Delta}}\right)^2 - c_{TV}\left(\frac{eV}{\tilde{\Delta}}\right)^2\left(\frac{k_BT}{\tilde{\Delta}}\right)^2\\ &- c_{VE_d}\left(\frac{eV}{\tilde{\Delta}}\right) + c_{TVE_d}\left(\frac{eV}{\tilde{\Delta}}\right)\left(\frac{k_BT}{\tilde{\Delta}}\right)^2,\end{aligned} \tag{14}$$

where

$$\begin{aligned}G(T,V=0) &= G_0\left[1 - c_T\left(\frac{k_BT}{\tilde{\Delta}}\right)^2\right],\\ c_T &= \frac{\pi^2}{3}\frac{1+2\tilde{u}^2+\tilde{\epsilon}_d^2[(8-5\tilde{u})\tilde{u}-3]}{[1+(1-\tilde{u})^2\tilde{\epsilon}_d^2]^2}.\end{aligned} \tag{15}$$

The zero-temperature linear conductance $G_0 = (2e^2/h)\times\frac{4\zeta}{3}(1+(1-\tilde{u})^2\tilde{\epsilon}_d^2)^{-1}$ reproduces the exact result from Friedel's sum rule up to $O(\tilde{u}^2\tilde{\epsilon}_d^2)$ as $\sin^2(\pi n_d) \sim 1-(1-\tilde{u})^2\tilde{\epsilon}_d^2$, for n_d the local occupation per spin component. For the transport coefficients in Eq. (14), we find

$$c_V = 1 + \frac{\tilde{u}^2}{2} - \zeta(1-\tilde{u}^2) - \tilde{\epsilon}_d^2(1-\tilde{u})H_V(\tilde{u},\zeta) + O(\tilde{\epsilon}_d^4), \tag{16}$$

$$\begin{aligned}c_{TV} =& \pi^2\left[2(1-\zeta)+\frac{\tilde{u}^2}{2}(9-5\zeta)\right]\\ &- \tilde{\epsilon}_d^2(1-\tilde{u})H_{TV}(\tilde{u},\zeta) + O(\tilde{\epsilon}_d^4),\end{aligned} \tag{17}$$

$$c_{VE_d} = 2\left(\frac{1-\kappa}{1+\kappa}\right)(1-\tilde{u})\tilde{\epsilon}_d + O(\tilde{\epsilon}_d^3), \tag{18}$$

$$c_{TVE_d} = -2\pi^2\left(\frac{1-\kappa}{1+\kappa}\right)(2+3\tilde{u}^2)(1-\tilde{u})\tilde{\epsilon}_d + O(\tilde{\epsilon}_d^3), \tag{19}$$

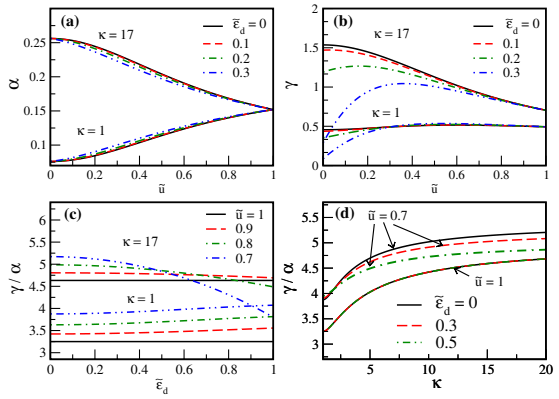

FIG. 1 (color online). Coefficients α (upper set) and γ (lower set) versus the different degrees of lead-to-dot asymmetry coupling: $\kappa = 20$ (left) and $\kappa = 2$ (right) are compared, for different values of particle-hole asymmetry $\tilde{\epsilon}_d$.

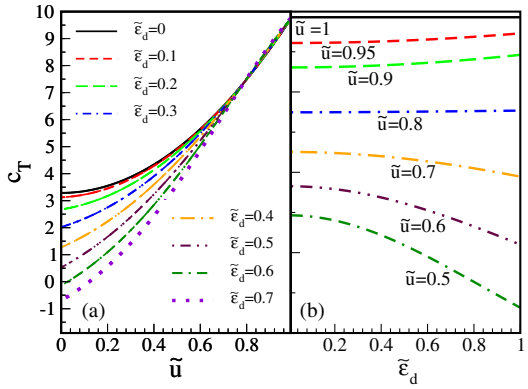

FIG. 2 (color online). Prefactor of $k_B^2 T^2/\tilde{\Delta}^2$ of the linear conductance (a) vs renormalized coupling strength (b) vs p-h asymmetry.

where we have defined the functions $H_V(\tilde{u}, \zeta) = 5 - 5\tilde{u} + \tilde{u}^2 - \zeta(5 - 3\tilde{u} - 2\tilde{u}^2)$ and $H_{TV}(\tilde{u}, \zeta) = \pi^2[28 - 16\tilde{u} + \frac{81}{2}\tilde{u}^2 - \zeta(28 - \frac{22}{3}\tilde{u} + \frac{76}{3}\tilde{u}^2)]$.

In Fig. 1, we show our results for α and γ for various cuts through parameter space ($\tilde{u}$, $\tilde{\epsilon}_d$, κ). Note, that in the strong coupling limit ($\tilde{u} \to 1$) the dependence on $\tilde{\epsilon}_d$ vanishes reflecting the fact that this limit is p-h symmetric [see Figs. 1(a) and 1(b)]. γ retains its dependence on κ in this limit while α becomes independent of κ for $\tilde{u} \to 1$. Figures 1(c) and 1(d) show the ratio γ/α. According to Eqs. (18) and (19) c_{VE_d} and c_{TVE_d} are proportional to the product of lead-dot asymmetry κ and p-h asymmetry $\tilde{\epsilon}_d$ and hence may be small in most experimental realizations. For the p-h symmetric case our expressions reduce to the results of Oguri and others [5,16,17].

We are now in a position to address the experimental results for $\alpha = c_V/c_T$ and $\gamma = c_{TV}/c_T^2$ [8,9]. A major experimental challenge is to reliably extract the dynamically generated low-energy scale $\tilde{\Delta} \sim T_K$ ($\tilde{\Delta} = 4k_B T_K/\pi$ at $\tilde{u} = 1$). The phenomenological formula $G(T, 0) = G_0/[1 + (2^{1/s} - 1)(T/T_K)^2]^s$ is commonly employed to extract T_K [33]. Evidently, the parameter s fixes c_T ($s = 0.21$ as in Ref. [8] leads to $c_T \approx 5.5$ and $s = 0.22$ [9] results in $c_T \approx 4.9$). Equation (16) shows that c_T is not only a function of $\tilde{u}$ but also depends on the p-h asymmetry through $\tilde{\epsilon}_d$, see Fig. 2. This complicates the experimental extraction of T_K. In theory, T_K is not unique away from p-h symmetry but will depend on the physical quantity used for its definition.

The reported values [8,9] suggest that charge fluctuations are present in both experiments and the coefficients c_{VE_d} and that c_{TVE_d} are indeed vanishingly small. Yet, they may have been detected in Ref. [9]. The experimental values reported in Ref. [8] are compatible with, e.g., $\tilde{u} = 0.45$, $\tilde{\epsilon}_d = 0.1$, $\kappa = 1$ yielding $\alpha = 0.1$ and $\gamma = 0.51$. While we can reproduce γ_S of Ref. [9], it is not possible to reproduce both consistently within the SIAM. The value $\alpha_S \sim 0.05$ is too small to be explained within the SIAM, as the minimum value for α within the SIAM is $\alpha_{\min} = 3/(4\pi^2) \approx 0.076$ (corresponding to $\tilde{\epsilon}_d = 0$, $\tilde{u} = 0$, $\kappa = 1$). The underlying low-energy model of the experiment [9] can therefore not simply be the SIAM. One possible generalization is that more than one level participates in the 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 [34]. A more likely alternative is that local phonon modes renormalize the transport coefficients α and γ differently.

In summary, we have developed a novel analytic scheme based on dual fermions to obtain nonlinear transport coefficients for the Anderson model. This approach gives a controlled expansion around the weak and strong coupling fixed points even away from particle-hole symmetry and allows for a consistent calculation of charge and energy currents. A generalization to nonlinear magneto- and thermal-transport properties is possible. Our scheme thus constitutes a convenient analytic way of characterizing nanostructured devices in terms of renormalized parameters $\tilde{u}$, $\tilde{\epsilon}_d$, and κ, and the low-energy scale $\tilde{\Delta}$ of an underlying model. With the current interest in strongly correlated systems away from equilibrium our approach should prove useful as it provides controlled results against which more general schemes [35] might be tested.

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Note added.—Recently, we became aware of Ref. [36], which addresses the effect of p-h asymmetry on α 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 gives a linear in T term in the spectral density away from half filling $n = 1$ in contradiction to certain Ward identities [5].

[1] *Physical Properties of Nanosystems*, edited by J. Bonca and S. Kruchinin (Springer, New York, 2010).

[2] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, England, 1993).

[3] A. Schiller and S. Hershfield, Phys. Rev. B **51**, 12896 (1995).

[4] A. Kaminski, Y. V. Nazarov, and L. I. Glazman, Phys. Rev. B **62**, 8154 (2000).

[5] A. Oguri, Phys. Rev. B **64**, 153305 (2001).

[6] R. M. Konik, H. Saleur, and A. Ludwig, Phys. Rev. B **66**, 125304 (2002).

[7] A. C. Hewson, J. Bauer, and A. Oguri, J. Phys. Condens. Matter **17**, 5413 (2005).

[8] M. Grobis, I. G. Rau, R. M. Potok, H. Shtrikman, and D. Goldhaber-Gordon, Phys. Rev. Lett. **100**, 246601 (2008).

[9] G. D. Scott, Z. K. Keane, J. W. Ciszek, J. M. Tour, and D. Natelson, Phys. Rev. B **79**, 165413 (2009).

[10] A. V. Kretinin, H. Shtrikman, D. Goldhaber-Gordon, M. Hanl, A. Weichselbaum, J. von Delft, T. Costi, and D. Mahalu, Phys. Rev. B **84**, 245316 (2011).

[11] Particle-hole asymmetry in $\rho_\lambda(\omega)$ will have a similar effect as the local p-h asymmetry generated by a finite E_d.

[12] K. Majumdar, A. Schiller, and S. Hershfield, Phys. Rev. B **57**, 2991 (1998).

[13] T. A. Costi, A. C. Hewson, and V. Zlatić, J. Phys. Condens. Matter **6**, 2519 (1994).

[14] G. Baym and L. P. Kadanoff, Phys. Rev. **124**, 287 (1961).

[15] S. Kirchner and J. Kroha, J. Low Temp. Phys. **126**, 1233 (2002).

[16] A. Oguri, J. Phys. Soc. Jpn. **74**, 110 (2005).

[17] J. Rincón, A. A. Aligia, and K. Hallberg, Phys. Rev. B **79**, 121301 (2009).

[18] E. Sela and J. Malecki, Phys. Rev. B **80**, 233103 (2009).

[19] Note that the use of Ward identities in Ref. [5] would result in a current conserving approximation up to order V^3 even away from p-h symmetry but the derivative of the (renormalized) susceptibilities with respect to (the renormalized) E_d is not known [20].

[20] K. Yamada, Prog. Theor. Phys. **62**, 354 (1979).

[21] S. Hershfield, J. H. Davies, and J. W. Wilkins, Phys. Rev. B **46**, 7046 (1992).

[22] A. N. Rubtsov, M. I. Katsnelson, and A. I. Lichtenstein, Phys. Rev. B **77**, 033101 (2008).

[23] A. C. Hewson, Phys. Rev. Lett. **70**, 4007 (1993).

[24] V. Zlatić and B. Horvatić, Phys. Rev. B **28**, 6904 (1983).

[25] H. Hafermann, C. Jung, S. Brenner, M. I. Katnelson, A. N. Rubtsov, and A. I. Lichtenstein, Europhys. Lett. **85**, 27 007 (2009).

[26] C. Jung, A. Lieder, S. Brener, H. Hafermann, B. Baxevanis, A. Chudnovskiy, A. N. Rubtsov, M. I. Katsnelson, and A. I. Lichtenstein, Ann. Phys. (Amsterdam) **524**, 49 (2012).

[27] K. Yosida and K. Yamada, Prog. Theor. Phys. Suppl. **46**, 244 (1970).

[28] K. Yamada, Prog. Theor. Phys. **54**, 316 (1975).

[29] B. Horvatić, D. Šokčević, and V. Zlatić, Phys. Rev. B **36**, 675 (1987).

[30] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.110.016601 for technical details on the dual fermion perturbation theory on the Keldysh contour, including explicit calculations leading to the self energy Eq. (10), and to the nonequilibrium distribution function Eq. (13).

[31] We have chosen the chemical potentials to satisfy $\Gamma_L|\mu_L| = \Gamma_R|\mu_R|$ for convenience. Results for the general case are available [30].

[32] Y. Meir and N. S. Wingreen, Phys. Rev. Lett. **68**, 2512 (1992).

[33] D. Goldhaber-Gordon, J. Göres, M. A. Kastner, H. Shtrikman, D. Mahalu, and U. Meirav, Phys. Rev. Lett. **81**, 5225 (1998).

[34] J. Kroha, S. Kirchner, G. Sellier, P. Wölfle, D. Ehm, F. Reinert, S. Hüfner, and C. Geibel, Physica (Amsterdam) **18**, 69 (2003).

[35] M. Pletyukhov and H. Schoeller, Phys. Rev. Lett. **108**, 260601 (2012).

[36] A. A. Aligia, J. Phys. Condens. Matter **24**, 015306 (2012).