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Transport calculations based on density functional theory, Friedel's sum rule, and the Kondo effect

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Friedel's sum rule provides an explicit expression for a conductance functional $\mathcal{G}[n]$, valid for the single-impurity Anderson model at zero temperature. The functional is special because it does not depend on the interaction strength U. As a consequence, the Landauer conductance for the Kohn-Sham (KS) particles of density functional theory (DFT) coincides with the true conductance of the interacting system. The argument breaks down at temperatures above the Kondo scale, near integer filling, $n_{\mathrm{d}\sigma} \approx 1/2$ for spins $\sigma = \uparrow\downarrow$. Here, the true conductance is strongly suppressed by the Coulomb blockade, while the KS conductance still indicates resonant transport. Conclusions of our analysis are corroborated by DFT studies with numerically exact exchange-correlation functionals reconstructed from calculations employing the density matrix renormalization group.

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I. INTRODUCTION

The ground-state density functional theory (DFT) owes its success to the fact that it proves enormously useful in the prediction of electronic properties of molecules, solids, and surfaces.[1,2] Therefore, applications toward the electronic transport properties of single molecules and self-assembled monolayers came together quite naturally with the corresponding experimental successes in the field.[3,4] Besides being useful for quantitative calculations, more fundamental properties of DFTs and the corresponding exact functionals have been an issue of intensive research. As an example we mention the question of what exactly is the nature of the approximations when using ground-state DFT in combination with the Landauer formalism for transport calculations.[5,6]

The Landauer approach, formulated in terms of nonequilibrium Green's functions[7,8] and its validity for Kohn-Sham (KS) particles, is also our topic in this work. It relates a single-particle Hamiltonian H^{KS} to the conductance via the transmission function, $G = T^{\mathrm{KS}}(\varepsilon_{\mathrm{F}})e^2/h$. The definition is

$$T^{\mathrm{KS}}(E) = \mathrm{Tr}\, \Gamma^{\mathrm{KS}}_{\mathcal{R}}(E) G(E) \Gamma^{\mathrm{KS}}_{\mathcal{L}}(E) G^{\dagger}(E), \tag{1}$$

where $\Gamma^{\mathrm{KS}}_{\alpha} = \mathrm{i}(\Sigma_\alpha - \Sigma^{\dagger}_{\alpha})$, $G = (E - H^{\mathrm{KS}} - \Sigma_{\mathcal{R}} - \Sigma_{\mathcal{L}})^{-1}$ and the trace is over the Hilbert space associated with H^{KS}. The self-energies Σ_α describe the coupling of the KS system to external reservoirs, $\alpha = \mathcal{L},\mathcal{R}$. They are given by a golden rule expression, $\Sigma_\alpha(E) = |V|^2 g^{\mathrm{KS}}_{\alpha}(E)$, where V is the coupling matrix element and g^{KS}_{α} is a Green's function of the leads.[9]

Often quantum transport is dominated by a single orbital of the molecule or the quantum dot (QD) only. Therefore, in studies of correlated electron transport interacting level models are standard, e.g., the single-impurity Anderson model [SIAM, see also (4)]:

$$\hat{H}_{\mathrm{QD}} = \varepsilon_{\mathrm{d}} \hat{N}_{\mathrm{d}} + U \left(\hat{n}_{\mathrm{d}\uparrow} - \tfrac{1}{2}\right)\left(\hat{n}_{\mathrm{d}\downarrow} - \tfrac{1}{2}\right), \tag{2}$$

where $\hat{N}_{\mathrm{d}} = \hat{n}_{\mathrm{d},\uparrow} + \hat{n}_{\mathrm{d},\downarrow}$ with $\hat{n}_{\mathrm{d},\sigma} = \hat{d}^{\dagger}_{\sigma}\hat{d}_{\sigma}$ and spin $\sigma = \uparrow, \downarrow$. In such models an analog version of (1) is valid, featuring retarded and advanced Green's functions of the interacting system ($\Gamma_{\alpha,\sigma\sigma'} = \Gamma_\alpha \delta_{\sigma\sigma'}$),

$$T(E) = \frac{\Gamma_{\mathcal{L}}\Gamma_{\mathcal{R}}}{\Gamma_{\mathcal{L}} + \Gamma_{\mathcal{R}}} \mathcal{A}_{\mathrm{d}}(E), \tag{3}$$

where we have introduced the spectral function of the interacting QD, $\mathcal{A}_{\mathrm{d}}(E) = \mathrm{i}\mathrm{Tr}_\sigma\, [G^{\mathrm{r}}(E) - G^{\mathrm{a}}(E)]$.[10]

By comparing Eqs. (3) and (1), one might suspect that in order to accurately reproduce the true value for the transmission, $T(\varepsilon_{\mathrm{F}}) = T^{\mathrm{KS}}(\varepsilon_{\mathrm{F}})$, it is necessary for the KS theory to also reproduce the true spectral function $\mathcal{A}_{\mathrm{d}}(E)$. It is easy to see that the latter is not possible, however, unless $U = 0$. To this end we recall that $\mathcal{A}_{\mathrm{d}}(E)$ carries the two Hubbard peaks at energies $\varepsilon_{\mathrm{d}} \pm U/2$.[11] These peaks are not seen by the KS system because the model (2) does not exhibit magnetism, so both spin channels ($\uparrow$, $\downarrow$) are equivalent. Therefore, H^{KS} is diagonal, $H^{\mathrm{KS}}_{\mathrm{d}} = \varepsilon^{\mathrm{KS}}_{\mathrm{d}}\delta_{\sigma\sigma'}$, and the KS spectral function, $\mathcal{A}^{\mathrm{KS}} = \mathrm{i}\mathrm{Tr}_\sigma(G - G^{\dagger})$, supports a single peak centered at about $\varepsilon^{\mathrm{KS}}_{\mathrm{d}}$, only. Despite the absence of the Hubbard peaks in $\mathcal{A}^{\mathrm{KS}}$, we argue that in addition to the ground-state density also the KS conductance coincides with the true value of the interacting system, $T(\varepsilon_{\mathrm{F}}) = T^{\mathrm{KS}}(\varepsilon_{\mathrm{F}})$, at least in the case of symmetric coupling, $\Gamma_{\mathcal{L}} = \Gamma_{\mathcal{R}}$.

Moreover, we maintain that this statement is correct, even though H^{KS} is not unique in the sense that the exchange-correlation (XC) on-site potential, $v^{\mathrm{KS}}_{\mathrm{d}} = \varepsilon^{\mathrm{KS}}_{\mathrm{d}} - \varepsilon_{\mathrm{d}}$, can be complemented by an XC contribution to the couplings, $V \to V^{\mathrm{KS}}$, as well. Different combinations (V^{KS},v^{KS}_{x}) produce an effective single-particle Hamiltonian with the correct ground-state density.[12] In fact, as will be demonstrated below, the on-site exchange-correlation potential and the coupling to the leads can be drastically different. Correspondingly, the resonance position $\varepsilon^{\mathrm{KS}}_{\mathrm{d}}$ and broadenings $\Gamma_{\mathcal{L},\mathcal{R}}$ that determine $\mathcal{A}^{\mathrm{KS}}$ will be strongly XC-functional dependent. We will see that despite this ambiguity in H^{KS}, the KS conductance is an observable that takes a unique value coinciding with the true conductance.

II. CONDUCTANCE FUNCTIONALS AND GROUND-STATE DFT

We start with a general recollection regarding features of ground-state DFT that follow directly from the first Hohenberg-Kohn theorem.[13] According to this theorem, we can reconstruct the external potential $v_{\rm ex}(\mathbf{r})$ that an interacting gas of N electrons is exposed to, if the ground-state density $n(\mathbf{r})$ together with the Hamiltonian $\hat{H}_0$ in the absence of any $v_{\rm ex}(\mathbf{r})$ is known. Thus we reproduce the full Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}_{\rm ex}$ from our knowledge of $n(\mathbf{r})$ (up to a constant shift in energy).

Knowing $\hat{H}$ we can calculate, in principle, all equilibrium response functions of the N-particle system, provided that the ground state is unique, once $n(\mathbf{r})$ has been specified. Hence, we can consider such correlators to be a functional of the ground-state density.

A correlation function of special interest to us is the current-current correlation function. It determines the conductance in transport experiments that operate with electrodes for which the Fermi-liquid description holds true. Hence, in such geometries also the conductance is a functional of the ground-state density $\mathcal{G}[n]$. Hence, this conductance can be calculated using the ground-state DFT if the proper functional $\mathcal{G}[n]$ is being used.

The previous statement is as correct as it is useless for practical purposes unless a good approximation for $\mathcal{G}[n]$ can be given. Of course, even if such an approximation would be known, in practice calculations would still suffer from inaccuracies in approximate XC functionals used to obtain $n(\mathbf{r})$.

A. Friedel's sum rule for the SIAM

The complete definition of the SIAM Hamilonian reads[14]

$$\hat{H} = \hat{H}_{\rm QD} + \sum_{\alpha=\mathcal{L},\mathcal{R}} \hat{H}_\alpha + \hat{H}_{\rm T}, \tag{4}$$

$$\hat{H}_\alpha = -t \sum_{x=1,\sigma}^{M-1} (\hat{c}^\dagger_{x+1,\sigma,\alpha}\hat{c}_{x,\sigma,\alpha} + \text{H.c.}), \tag{5}$$

$$\hat{H}_{\rm T} = -V \sum_{\sigma,\alpha} (\hat{c}^\dagger_{1,\sigma,\alpha}\hat{d}_\sigma + \hat{d}^\dagger_\sigma \hat{c}_{1,\sigma,\alpha}). \tag{6}$$

The $\hat{c}^{(\dagger)}_{x,\sigma,\alpha}$ denote fermionic annihilation (creation) operators at site x, lead $\alpha = \mathcal{R},\mathcal{L}$. The model is of interest to us because it affords the Friedel sum rule,[14,15] which relates the extra scattering phase shift $\delta(\varepsilon_{\rm F})$ induced by decreasing $\varepsilon_{\rm d}$ (down from ∞) to the extra spectral weight $\Delta\mathcal{A}(E)$ thus generated:

$$\frac{\delta(\varepsilon_{\rm F})}{\pi} = \int_{-\infty}^{\varepsilon_{\rm F}} dE\, \Delta\mathcal{A}_\sigma(E) = \Delta N_\sigma. \tag{7}$$

The right-hand side denotes the total change in the particle number per spin, $\Delta N_\sigma = \sum_x \Delta N_{x\sigma}$, associated with the occupation of this extra weight. The scattering phase shift in turn determines the conductance,

$$\mathcal{G}[n] = \frac{2e^2}{h}\frac{\Gamma_{\mathcal{L}}\Gamma_{\mathcal{R}}}{(\Gamma/2)^2}\sin^2[\delta(\varepsilon_{\rm F})], \tag{8}$$

where $\Gamma = \Gamma_{\mathcal{L}} + \Gamma_{\mathcal{R}}$. The identity constitutes an exact analytical expression for a conductance functional $\mathcal{G}[n]$. While it has not been considered as such, $\mathcal{G}[n]$ was frequently employed before in treatments of correlated electron systems, e.g., see 14.

It is remarkable that in the case of symmetric coupling, $\Gamma_{\mathcal{L}} = \Gamma_{\mathcal{R}}$, Eq. (8) relates the conductance associated with $\hat{H}$ to single-system characteristics only, which is the number of particles (per spin) displaced by the Anderson impurity ΔN_σ. This implies that any change in the parameters of $\hat{H}$ leaves the conductance invariant, provided that ΔN_σ is unchanged and that the conditions of applicability of (8) are still valid. One requirement for this is that both leads are (effectively) noninteracting and free of backscattering, so that the Fermi-liquid picture holds at zero temperature.[16]

B. Implications of Friedel's sum rule for KS transport

We now investigate the specific consequences of (8) for KS transport calculations of ground-state DFT. The importance of Friedel's sum rule for KS-based transport calculations was discussed also in previous work.[17,18] In contrast to these papers, we present rigorous statements available for the model system, SIAM, in the Kondo regime. Similar conclusions to ours were concomitantly reached by the authors of Refs. 19 and 20, however see Ref. 21. In the following we have a particular focus on the fate of the Abrikosov-Suhl resonance in KS theory and how despite a completely unphysical KS spectral function, KS transport can still be exact (at zero temperature).

The KS Hamiltonian of ground-state DFT reads

$$\hat{H}^{\rm KS} = \varepsilon^{\rm KS}_{\rm d}\hat{N}_{\rm d} + \sum_\alpha \left[\hat{H}_\alpha + \sum_{x=1,\sigma}^{M} v^{\rm KS}_{x,\alpha}\hat{N}_{x,\alpha}\right] + \hat{H}_{\rm T}, \tag{9}$$

where the on-site XC potentials, $\varepsilon^{\rm KS}_d[N] = \varepsilon_{\rm d} + v^{\rm KS}_{\rm d}[N]$ and $v^{\rm KS}_{x,\alpha}[N]$, are functionals of the local particle density N_x.

One can use the associated KS orbitals to construct scattering states and a KS Landauer conductance, Eq. (1). The scattering phase $\delta^{\rm KS}(\varepsilon_{\rm F})$ that appears in this construction also obeys an equation analog to (7): $\delta^{\rm KS}(\varepsilon_{\rm F})/\pi = \Delta N^{\rm KS}_\sigma$. In particular, parametric details of the Hamiltonian, such as V,U, etc., or the on-site potentials do not enter explicitly. Since the displaced charge of the KS system that appears here is by definition identical to the true displaced charge, $\Delta N_\sigma = \Delta N^{\rm KS}_\sigma$, we conclude that the true quasiparticles and the KS particles experience the same phase shift: $\delta(\varepsilon_{\rm F}) = \delta^{\rm KS}(\varepsilon_{\rm F}) = \Delta N_\sigma$.

Functional (8) is valid at any value of the interaction strength U and at temperatures below the Kondo scale, $T_{\rm K} = c\sqrt{\Gamma U/2}e^{-\pi U/8\Gamma}$, $c \approx 0.41\ldots$ near integer filling $N_{\rm d} = n_{{\rm d}\uparrow} + n_{{\rm d}\downarrow} \approx 1$.[14] In this context the Abrikosov-Suhl resonance underlying the Kondo effect plays a crucial role. At temperatures above $T_{\rm K}$ it is not developed and the conductance is strongly suppressed due to the Coulomb blockade; relation (8) is strongly violated. The XC functional of DFT must be very sensitive to Kondo physics. This is obvious for the following reason: we have seen that KS transport reproduces the exact transmission. To reproduce the resonant transport (Kondo) scenario in the regime $N_{\rm d} \approx 1$, the KS level of the quantum dot must be half filled for each spin, implying $|\varepsilon_{\rm F} - \varepsilon^{\rm KS}_{\rm d}| \lesssim \Gamma$ in the Kondo regime, even if the bare position of the level $-\varepsilon_{\rm d} \gg \Gamma$.

III. EXACT GROUND-STATE DFT VIA THE DENSITY MATRIX RENORMALIZATION GROUP METHOD

To illustrate and extend our analysis we have calculated the ground-state density and corresponding exact XC potentials employing the density matrix renormalization group (DMRG) method[22] and "backward" DFT. The approach has proven useful before in the context of the interacting resonant level model (IRLM) and resonant chains.[23] We have adapted our technology here to treat the SIAM.[24]

In our calculations we consider a coupling to a single lead only in order to reduce the computational effort. As far as the ground state is concerned, the case with two leads and symmetric couplings, $\Gamma_{\mathcal{L}} = \Gamma_{\mathcal{R}}$, has an exact mapping into the single-lead case, essentially because the odd combination of tunneling operators, $c^{(\dagger)}_{\text{odd},1} = (c^{(\dagger)}_{\mathcal{L},1} - c^{(\dagger)}_{\mathcal{R},1})/\sqrt{2}$, decouples from the QD.

A. Ground-state density

In Fig. 1 we display the evolution of the density in a QD with ε_{d} slightly above ε_{F}. Without interactions, the QD is empty. Upon increasing U, the dot fills because in the spirit of a Jellium model we have defined the interaction in (2) with respect to density fluctuations against a background $n^{\text{bg}}_{\text{d}\sigma} = 1/2$. The density in the leads, $x \geqslant 1$, exhibits the typical Friedel oscillations with their $2k_{\text{F}}$ periodicity. Their amplitude is controlled by the boundary condition which is set by the QD. Since its occupation changes from $N_{\text{d}} \approx 0$ to $N_{\text{d}} \approx 1$, the phase shift associated with backscattering increases by $2\pi n_{\text{d},\sigma}$. This is why at large U the Friedel oscillations are antiphase with the case $U = 0$ (inset Fig. 1). We witness a signature of Kondo physics (cf. Ref. 25).

B. KS potential

Figure 2 shows the KS potential on the QD, $\varepsilon^{\text{KS}}_{\text{d}}$, and in the lead, v^{KS}_x, corresponding to the evolution of the density (Fig. 1). We thus provide a constructive proof of v representability: An effective XC potential exists for the SIAM, such that the associated effective single-particle (KS) problem exhibits a density that coincides everywhere with the exact density of the true many-body ground state. With respect to the Kondo effect, we conclude that by construction, all of its signatures that can be read off the ground-state density alone, like the size of the Kondo cloud, will also be reflected by the KS system.

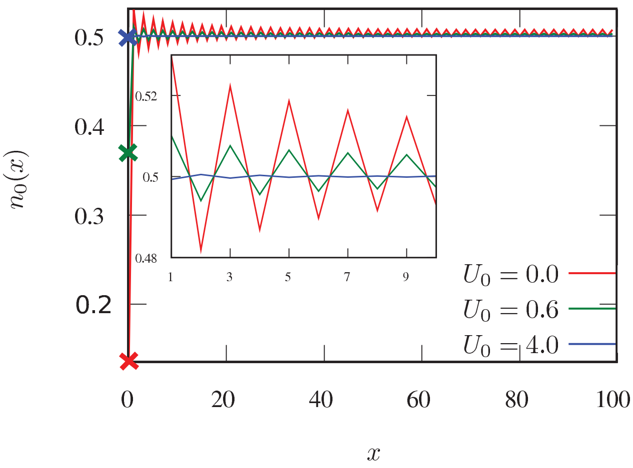

FIG. 1. (Color online) Ground-state density per spin of a QD (at $x = 0$) coupled to a noninteracting reservoir with $M = 100$ sites for growing on-QD interaction $U = 0.0, 0.6, 4.0$. Parameters: $\varepsilon_{\text{d}} = 0.2, V = 0.3$, bandwidth of conduction electrons: $2t = 2$.

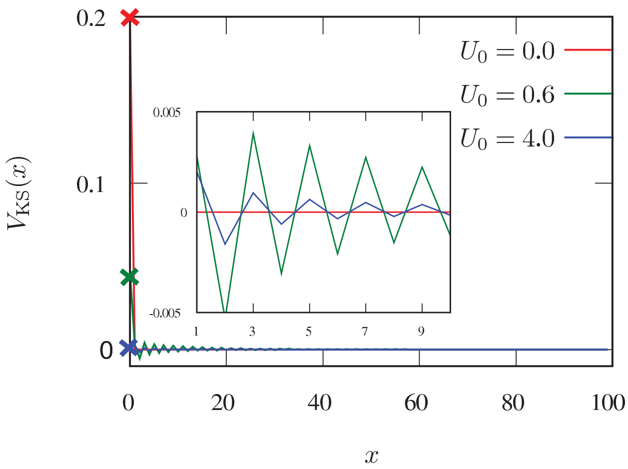

FIG. 2. (Color online) XC correlation potential corresponding to the evolution of the density shown in Fig. 1. The on-dot potential is denoted $v^{\text{KS}}_0 = \varepsilon^{\text{KS}}_{\text{d}}$.

In the absence of an analytical expression for the exact XC potential, our data in Fig. 2 can illustrate how v^{KS}_x modulates the noninteracting KS density into the proper "interacting" shape. Moreover, the data may also serve as a reference for benchmarking later approximative schemes for XC potentials. On the dot v^{KS}_x shows the expected behavior from $\varepsilon^{\text{KS}}_{\text{d}} = \varepsilon_{\text{d}}$ at $U = 0$ to $\varepsilon^{\text{KS}}_{\text{d}} \approx 0$ at large interaction, here $U = 4$. In the leads the potential oscillations follow the Friedel oscillations of the density and introduce the interaction corrections. The oscillation amplitude depends in a nonmonotonic way on U, increasing from the noninteracting fixpoint and decreasing again when approaching the strong-coupling, Kondo fixpoint (see inset).

In a KS theory based on a local KS potential, the hybridization V coincides with the one from the original coupling $\hat{H}_{\text{T}}$. This suggests that the on-dot spectral function $\mathcal{A}^{\text{KS}}(E)$ of the KS system has a width close to the noninteracting one, $\Gamma \approx \Gamma^{\text{KS}}$. Figure 3 fully supports this point of view. In addition, it also shows that indeed there is only a single maximum (Hubbard peaks do not exist in KS theory) which will be getting closer to ε_{F} with increasing interaction strength U.

C. KS transmission and spectral function

Figure 4 compares transmission curves obtained via the Friedel sum rule[26] with a Landauer-type expression ($\Gamma_{\mathcal{L}} = \Gamma_{\mathcal{R}}$),

$$T(\varepsilon_{\text{F}}) = \frac{[\Gamma/2]^2}{\left[\varepsilon^{\text{KS}}_{\text{d}}(V_{\text{g}}, V^{\text{KS}}) - \varepsilon_{\text{F}}\right]^2 + [\Gamma/2]^2}, \qquad (10)$$

that is valid in the wide band limit. The good agreement between the results obtained with both methods illustrates the point emphasized above: Even though the KS spectral function

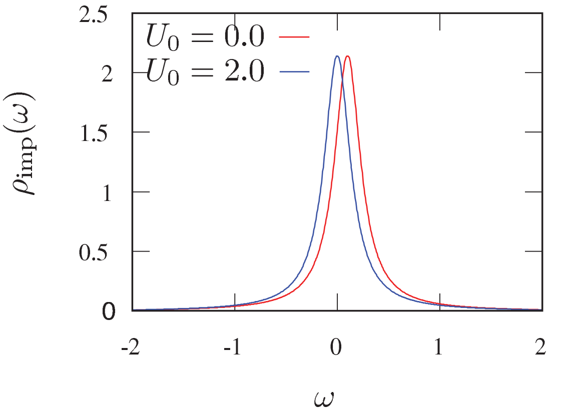

FIG. 3. (Color online) Local spectral function of the KS system, $\mathcal{A}_d^{\rm KS} = 2\pi\rho_{\rm imp}$, on the QD for interactions $U = 0,2$. Parameters: $\varepsilon_{\rm d} = 0.1$, $V = 0.15\ldots$, $\varepsilon_{\rm F} = 0$.

is not physical, the associated Landauer transmission can be exact.

We briefly mention that this point can be highlighted further by constructing a version of KS theory in which not only the diagonal elements of the density matrix, i.e., $n_{x\sigma}$ but also off-diagonal elements are faithfully reproduced. This can be achieved by adding to the on-site potential also a modification of hopping matrix elements, i.e., $V \to V^{\rm KS\prime}$, such that the expectation value of the kinetic energy of the impurity coupled to the first lead site within the KS description matches the one obtained from DMRG[24]; for the technical details see Ref. 24. Responding to this change, the width of the KS spectral function of the modified theory is no longer close to the original one, $\Gamma^{\rm KS} \to \Gamma^{\rm KS\prime}$. Since the charge in the QD must remain unchanged, we expect a compensating shift in the on-site energy, $\varepsilon_{\rm d}^{\rm KS} \to \varepsilon_{\rm d}^{\rm KS\prime}$. As can be seen from Fig. 4, despite a substantial change, e.g., $V = 0.3$ is replaced by $V = 0.16$ at $U = 1.8$, the transmission when evaluated via (10) is not changed. In view of Eq. (10) this finding is easily understood: $T(\varepsilon_{\rm F})$ is determined by the same ratio, $\Gamma^{\rm KS}/(\varepsilon_{\rm d}^{\rm KS} - \varepsilon_{\rm F})$, that also fixes the density. Since by construction in all KS models the density is the same, we have $\Gamma^{\rm KS}/(\varepsilon_{\rm d}^{\rm KS} - \varepsilon_{\rm F}) = \Gamma^{\rm KS\prime}/(\varepsilon_{\rm d}^{\rm KS\prime} - \varepsilon_{\rm F})$, so the transmission remains the same, also.

IV. FURTHER REMARKS AND CONCLUSIONS

We briefly discuss three generalizations of the preceding analysis. First, our analysis heavily relies on the Kondo effect restoring full transmission in the case of single occupation of the dot. Suppose that the Kondo temperature is very low and that the measurement is done at slightly higher temperatures. Then transport is dominated by the Coulomb blockade and the conductance is strongly suppressed, $T(\varepsilon_{\rm F}) \sim (\Gamma/U)^2 \lll 1$. On the other hand, the particle density is essentially still the ground-state one $n(\mathbf{r})$, i.e., it is largely insensitive to this change and in particular, $n_{\rm d\sigma} \approx 1/2$. We conclude that in this case the Friedel sum rule (8) does not hold and that KS theory (without breaking spin rotational invariance) does not reflect this change, i.e., we still have $T^{\rm KS}(\varepsilon_{\rm F}) \approx 1$.

The preceding observation can be reformulated in more general terms. The connection from the exact ground-state density to transport coefficients, that we rely upon, is mediated by Friedel's sum rule which is valid in the form we use it, Eq. (7), only for systems near the Fermi-liquid (FL) ground state. Therefore, we rely upon the FL ground-state property of the Kondo effect. At temperatures $T > T_{\rm K}$ the system is far away from the FL fixed point and our arguments need to be modified. The same is true also for multichannel Kondo systems.

Second, our paper and the claims therein focus on ground-state properties only. We expect that the discussion can be generalized to equilibrium situations at finite temperatures, also, but for this situation one needs to introduce a notion of thermal XC functionals.[28] Also, for this situation a conductance functional should exist that translates thermal equilibrium densities into conductances at finite temperatures. One would expect that such functionals correctly describe the breakdown of the Kondo FL at temperatures exceeding the Kondo scale.[29]

Third, the Friedel sum rule (7) is strictly valid for a single-level system only, while a real QD, e.g., a molecule, generally exhibits several levels. One expects that the main conclusion remains correct as long as all levels contribute independently

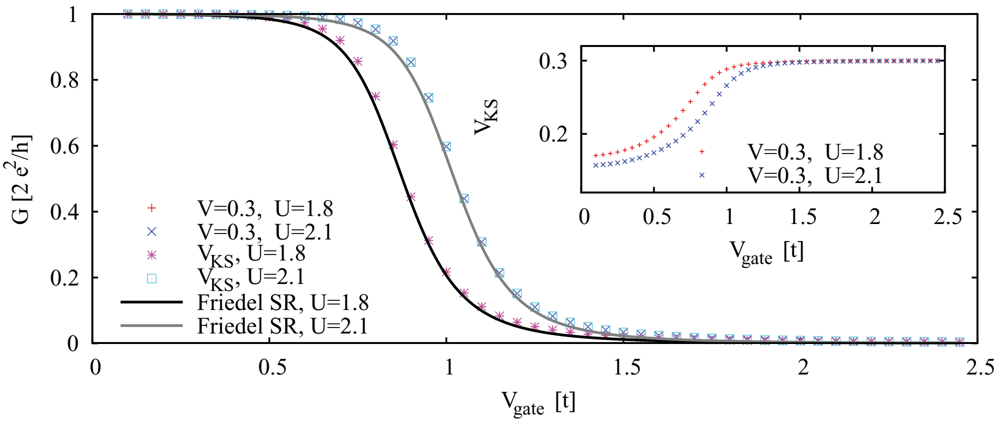

FIG. 4. (Color online) Comparison of conductances calculated via the Friedel sum rule (FSR) and directly from the Landauer formula for KS particles (10). Two parameter combinations, $V = 0.3, U = 1.8, 2.1$ are shown. Inset: Detuned KS couplings $V^{\rm KS}$ that together with the belonging XC potential reproduce the evolution of the dot occupation and hence the conductance seen in main plot.

to the transport current. In particular, the sum rule (8) should remain a useful approximation for the true density functional. Of course, transport is not always described by independent quantum channels. A good testing ground for investigations of the effect of channel cross talk is provided by two-level quantum dots.[16] They can exhibit phases different from the normal FL, with the associated phase transitions. The related conductance functional does not vary smoothly with the density and therefore the description along Eqs. (7) and (8) needs to be modified. We leave this challenging issue for future research.

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[12] We here employ the familiar fact that the XC potential is uniquely defined only after the mean-field contribution to the interaction energy has been specified together with the kinetic energy term. The conventional choice for the latter is $V = V^{\mathrm{KS}}$, i.e., the noninteracting one. This choice, however, is not mandatory. One can also construct an XC potential such that a Hamiltonian with a detuned kinetic term $V \neq V^{\mathrm{KS}}$ reproduces the exact ground-state density. The degree of freedom that emerges here can readily be used in order to generalize conventional DFT into a density matrix functional theory.

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[21] The authors of Ref. 20 emphasize the importance of the discontinuity in the XC potential to describe Coulomb-blockade and Kondo physics. However, as we investigate in detail in a previous work,[27] the XC potential of the SIAM does not exhibit such a discontinuity, strictly speaking. Specifically, the zero-temperature discontinuity of the XC potential of any isolated, finite system is smeared out, generically, by lifetime effects (level broadening Γ) that tend to appear when particles are exchanged with an external reservoir. Hence, a "discontinuity" (at $T = 0$) in the sense of Ref. 20 should be understood with a grain of salt: it indicates a large slope in the density susceptibility, $dn/d\mu \sim U/\Gamma$ when $U \gg \Gamma$, but not a real mathematical singularity. The discontinuity proper is reached in the isolated limit, $\Gamma = 0$.

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[26] We have made a replacement in this calculation $\Delta N_\sigma \longrightarrow N_{\mathrm{d}}$. This approximation becomes exact in the wide band limit, but our experience with the IRLM[22] shows that it works very well also in the more general case.

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[29] A breakdown of the Kondo effect can also be driven by applying magnetic fields beyond a critical field strength B_{K}. At $B > B_{\mathrm{K}}$ transport is no longer resonant and we return to Coulomb-blockade physics (now in finite B fields). This case has been studied by two of us in a generalized spin density functional theory. (Tröster and Schmitteckert, unpublished). We have ascertained, that spin DFT faithfully monitors the particle density and magnetization in all B-field regimes, including the destruction of the Kondo FL fixed point.