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Thermoelectric transport through a SU(N) Kondo impurity

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We investigate thermoelectric transport through a SU(N) quantum impurity in the Kondo regime. The strong-coupling fixed-point theory is described by the local Fermi-liquid paradigm. Using Keldysh technique we analyze the electric current through the quantum impurity at both finite bias voltage and finite temperature drop across it. The theory of a steady state at zero current provides a complete description of the Seebeck effect. We find pronounced nonlinear effects in temperature drop at low temperatures. We illustrate the significance of the nonlinearities for enhancement of thermopower by two examples of SU(4) symmetric regimes characterized by a filling factor m: (i) particle-hole symmetric at $m = 2$ and (ii) particle-hole nonsymmetric at $m = 1$. We analyze the effects of potential scattering and coupling asymmetry on the transport coefficients. We discuss connections between the theory and transport experiments with coupled quantum dots and carbon nanotubes.

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Introduction. Recent progress in the understanding of thermoelectric phenomena on the nanoscale stimulated experiments [1–3] and the development of theoretical approaches to this problem (see, e.g., [4] for review). One of the fundamental properties of the quantum transport through nanosized objects [quantum dots (QDs), carbon nanotubes (CNTs), quantum point contacts (QPCs), etc.] is associated with the charge quantization [5]. It offers a very efficient tool for the quantum manipulation of the single-electron devices being building blocks for quantum information processing. The universality of the heat flows in the quantum regime, scales of the quantum interference effects, and limits of the tunability are the central questions of the emergent field of the quantum heat transport [1–3,6–8]. Besides, the effects of strong electron correlations and resonance scattering become very pronounced at low temperatures and can be measured with high controllability (e.g., external electric and magnetic fields, geometry, temperature, etc.) of the semiconductor nanodevices. Therefore, investigation of the quantum effects and influence of strong correlations and resonance scattering on the heat transport (both experimentally and theoretically) is one of the cornerstones of quantum electronics.

As follows from the Fermi-liquid (FL) theory, the thermoelectric power (Seebeck effect) of bulk metals is directly proportional to the temperature T and inversely proportional to the Fermi energy ε_F [9]. The resonance scattering on a quantum impurity, however, dramatically enhances this effect due to the emergence of quasiparticle resonances at the Fermi level described by the Kondo effect [10–12]. The contribution to the Seebeck effect proportional to the concentration of impurities at low T, as a result, scales as T/T_K [9,12] where T_K is a characteristic energy defining the width of the Kondo resonance, the Kondo temperature (Fig. 1). The Kondo effect in nanodevices is key for enhancing the thermoelectric transport coefficients [8]. The tunable thermotransport through nanodevices controlling the heat flow is needed for efficient operation of quantum circuits elements: single-electron transistors, quantum diodes, etc., to perform controllable heat guiding.

The Kondo effect has been observed in the experiments on the semiconductor quantum dots and the single wall carbon nanotubes [13–16]. The effect manifests itself by complete screening of spin of the quantum impurity and, as a result, the FL behavior in the strong-coupling (low temperatures) regime [10,11,17]. Here we use the local FL paradigm [18–25] which is a powerful tool for the description of thermodynamic and transport properties of quantum impurity in the strong-coupling regime. It has also been applied recently for explanation of "0.7 anomaly" in QPCs [26,27]. The $s = 1/2$ SU(2) Kondo impurity physics arises at the half-filled particle-hole (PH) symmetric regime. We refer to "electrons" as quasiparticles above ε_F and "holes" as the excitations below ε_F. The PH symmetry, being responsible for the enhancement of the electric conductance, suppresses however the thermoelectric transport: the thermocurrent carried by electrons is completely compensated by heat current carried by holes challenging us however to investigate Kondo models in the regime away from PH symmetry. To achieve appreciable thermopower, the occupation factor of the quantum impurity should be integer (Coulomb blockade valleys [5]), while the particle-hole symmetry should be lifted. Such properties are generic for the SU(N) Kondo models with the filling factors different from 1/2.

The SU(N) Kondo physics with $N = 4$ is experimentally realized in CNTs [28–31] and double QDs [32]. The SU(N) Kondo model has also been proposed to investigate in ultracold gases experiments [33,34]. There are several theoretical suggestions for realization of SU(N) Kondo physics with $N = 3$ [34,35], $N = 6$ [36], and $N = 12$ reported in [37]. While the electron transport through SU(N) Kondo impurity is well understood theoretically [21,38–40], the thermoelectric transport in the Kondo regime remains challenging [41–45].

In this Rapid Communication we present a full-fledged theory for the Seebeck effect of the SU(N) Kondo model for the strong-coupling regime $T \ll T_K$. Our approach is based on real time out-of-equilibrium Keldysh calculations. We used the local Fermi-liquid paradigm for constructing a perturbative expansion for the electric current around the strong-coupling fixed point of the model. We illustrate the

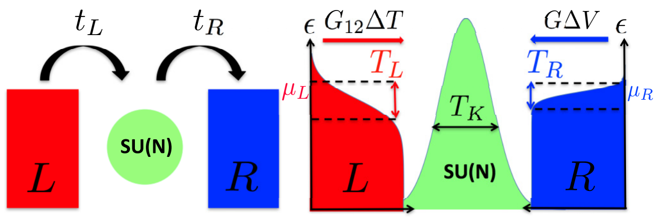

FIG. 1. Left panel: Cartoon for the tunneling $t_{L/R}$ through the SU(N) quantum impurity (see the main text). Right panel: Fermi distribution functions of the left (hot) and right (cold) leads at temperatures $T_{L/R}$ and chemical potential $\mu_{L/R}$. The thermovoltage $\Delta V = |\mu_R - \mu_L|/e$ is applied to achieve the steady state with zero net current across the impurity. Left (red)/right (blue) arrows show directions of thermo- and electric currents. Resonance Kondo peak of width T_K in density of states is shown by the green color.

thermoelectric properties of the SU(N) Kondo model on two particular examples, namely, $N = 4$ with the filling factors 1/4 and 1/2. We compute the thermoelectric power for arbitrary temperature drop between the electron reservoirs and discuss the significance of nonlinear effects in temperature drop.

Setup. We consider an SU(N) quantum impurity (such as a CNT or coupled QDs) sandwiched between two leads (Fig. 1). The model geometry resembles the experimental setup [8]. The temperature of the drain electrode (R) is taken as the reference temperature of the system. The temperature of the source electrode (L) is controlled by the Joule heat released due to the finite current flowing along the lead [8]. Thus, the temperature drop ΔT is fixed for all measurements. The bias voltage ΔV is applied between the source and the drain in order to stop the thermocurrent (Fig. 1, right panel):

$$I = 0 = G(T)\Delta V + G_{12}(T)\Delta T. \qquad (1)$$

The differential thermoelectric power is defined at the total current across the impurity tuned to zero:

$$S(T) = -\lim_{\Delta T\to 0} \frac{\Delta V}{\Delta T}\bigg|_{I=0} = \frac{G_{12}(T)}{G(T)}, \qquad (2)$$

$G = \partial I/\partial\Delta V|_{\Delta T=0}$ is the electric conductance, and $G_{12} = \partial I/\partial \Delta T|_{\Delta V=0}$ is the thermoelectric coefficient.

Model. The tunneling of electrons through the SU(N) quantum impurity (Fig. 1, left panel) is described by the Anderson model [21]:

$$H = \sum_{k\alpha r} \varepsilon_k c^\dagger_{\alpha kr} c_{\alpha kr} + \sum_r \epsilon_0 d^\dagger_r d_r + \sum_{r<r'} U d^\dagger_r d_r d^\dagger_{r'} d_{r'} + \sum_{k\alpha r} t_\alpha d^\dagger_r c_{\alpha kr} + \text{H.c.} \qquad (3)$$

Here d_r annihilates an electron in the dot level ϵ_0 with orbitals $r = 1,2,\dots N$, $c_{\alpha kr}$ annihilates a conduction electron with the momentum k and orbital r in the leads $\alpha = L,R$ and U is the Coulomb repulsion (charging) energy in the dot, t_α is lead-dot tunneling, and $\varepsilon_k = \epsilon_k - \varepsilon_F$ is the linearized conductance electron's dispersion. We assume that the charging energy U is the largest energy scale of the model and therefore take into account only the "last" occupied state. We project out the charge states by applying the Schrieffer-Wolff transformation [46]. As a result we obtain the effective SU(N) Kondo model describing the physics at the weak-coupling $T \gg T_K$ limit:

$$\mathcal{H}_K = J_K^{\alpha\beta}(\mathbf{c}^\dagger_\alpha \lambda^\mu \mathbf{c}_\beta)(\mathbf{d}^\dagger \Lambda^\mu \mathbf{d}), \qquad (4)$$

where $\mathbf{c}^\dagger = (c_1^\dagger,\dots,c_N^\dagger)$ is a row vector of the electron states in the leads and $\mathbf{d}^\dagger = (d_1^\dagger,\dots,d_N^\dagger)$ represents the local states in the dot. The SU(N) generators λ^μ and Λ^μ for $\mu = 1,2,\dots N^2-1$ are traceless $N\times N$ Hermitian matrices of the fundamental representation, satisfying the commutation relations $[\lambda^\mu,\lambda^\nu] = if^{\mu\nu\rho}\lambda^\rho$ where $f^{\mu\nu\rho}$ is the set of fully antisymmetric structure factors. As a last step we diagonalize the matrix $J_K^{\alpha\beta} \sim |t_\alpha t_\beta|/U$ in the subspace of two leads $\alpha,\beta = L,R$ performing the Glazman-Raikh rotation [47–49]. Similarly to the SU(2) Kondo model, the antisymmetric combination of the electron states in the leads $a^\dagger = (c_L^\dagger - c_R^\dagger)/\sqrt{2}$ is fully decoupled from the Hamiltonian while the symmetric combinations $b^\dagger = (c_L^\dagger + c_R^\dagger)/\sqrt{2}$ remain coupled to the quantum impurity [50] (without loss of generality we present here the results for symmetric $t_L = t_R$ dot-lead coupling; general equations for arbitrary coupling are presented in the Supplemental Material [51]).

The FL Hamiltonian describing the strong-coupling $T \ll T_K$ regime is obtained by applying the standard point-splitting procedure to $:(\mathbf{b}^\dagger\lambda^\mu\mathbf{b})\cdot(\mathbf{b}^\dagger\lambda^\mu\mathbf{b}):$ (see [17] for the details), $H_{FL} = H_0 + H_\alpha + H_\phi$ [52]:

$$H_0 = \nu \sum_r \int_\varepsilon \varepsilon[a^\dagger_{\varepsilon r} a_{\varepsilon r} + b^\dagger_{\varepsilon r} b_{\varepsilon r}],$$

$$H_\alpha = -\sum_r \int_{\varepsilon_{1-2}} \left[\frac{\alpha_1}{2\pi}(\varepsilon_1+\varepsilon_2) + \frac{\alpha_2}{4\pi}(\varepsilon_1+\varepsilon_2)^2\right] b^\dagger_{\varepsilon_1 r} b_{\varepsilon_2 r},$$

$$H_\phi = \sum_{r<r'} \int_{\varepsilon_{1-4}} \left[\frac{\phi_1}{\pi\nu} + \frac{\phi_2}{4\pi\nu}\left(\sum_{i=1}^4 \varepsilon_i\right)\right] : b^\dagger_{\varepsilon_1 r} b_{\varepsilon_2 r} b^\dagger_{\varepsilon_3 r'} b_{\varepsilon_4 r'} :. \qquad (5)$$

The *Kondo floating* paradigm [10,11,17–25] leads to the following FL identities: $\alpha_1 = (N-1)\phi_1$ and $\alpha_2 = (N-1)\phi_2/4$, ν is the density of states at ε_F. The connection between α_1 and α_2 is given by the Bethe ansatz [21]. We use $\alpha_1 = 1/T_K$ as the definition of the Kondo temperature [49].

Charge current. The current operator at position x is expressed in terms of first-quantized operators ψ attributed to the linear combinations of the Fermi operators in both leads,

$$\hat{I}(x) = \frac{\hbar e}{2mi} \sum_r [\psi^\dagger_r(x)\partial_x\psi_r(x) - \partial_x\psi^\dagger_r(x)\psi_r(x)]. \qquad (6)$$

For the expansion of Eq. (6), we choose the basis of scattering states that includes completely elastic and Hartree terms [21] to get it in compact form:

$$\hat{I} = \frac{e}{2\nu h} \sum_r [a^\dagger_r(x) b_r(x) - a^\dagger_r(-x) S b_r(-x) + \text{H.c.}], \qquad (7)$$

where $a_r(x) = \sum_k a_{kr}e^{ikx}$, $b_r(x) = \sum_k b_{kr}e^{ikx}$, $Sb_r(x) = \sum_k S_k b_{kr} e^{ikx}$, and the $N\times N$ S matrix is expressed in terms of a phase shift $\delta(\varepsilon_k)$ as $S_k = e^{2i\delta(\varepsilon_k)}$.

Elastic current. Calculation of the expectation value of (7) in the absence of interactions is equivalent to using the

Landauer-Büttiker formalism [5]:

$$I_{el} = \frac{Ne}{h}\int_{-\infty}^{\infty} d\varepsilon \mathcal{T}(\varepsilon)\Delta f(\varepsilon), \qquad (8)$$

where $\Delta f(\varepsilon) = f_L(\varepsilon) - f_R(\varepsilon)$, $f_{L/R}$ are Fermi distribution functions of L/R leads; $\mu_L - \mu_R = e\Delta V \ll T_K$ are the chemical potentials, $T_R = T$ is the reference temperature, and $T_L = T + \Delta T$ (Fig. 1 right panel). The energy dependent transmission $\mathcal{T}(\varepsilon) = \sin^2[\delta(\varepsilon)]$. Following Ref. [21], we Taylor-expand the phase shift for all flavors r in the presence of voltage bias $e\Delta V$ and temperature drop ΔT as $\delta_r(\varepsilon) = \delta_0 + \alpha_1\varepsilon + \alpha_2(\varepsilon^2 - \mathcal{A})$, where $\mathcal{A} = [\frac{(e\Delta V)^2}{4} + \frac{(\pi T)^2}{3} + \frac{\pi^2 T\Delta T}{3}]$ and $\delta_0 = \pi m/N$ for the quantum impurity's occupation $m = 1,\ldots,N-1$. The zero energy transmission is given by $\mathcal{T}_0 = \sin^2\delta_0$. Using the above equation for the phase shift δ_r we expand the current up to the second order in $T/T_K \ll 1$ to get the elastic contribution. The linear response result is

$$\frac{I_{el}}{Ne/h} = \left[\sin^2\delta_0 + \frac{\alpha_1^2}{3}(\pi T)^2\cos 2\delta_0\right]e\Delta V - \left[\frac{\alpha_1}{3T}(\pi T)^2\sin 2\delta_0\right]\Delta T. \qquad (9)$$

Inelastic current. The inelastic contribution to the current is computed using the nonequilibrium Keldysh formalism [53]:

$$\delta I_{\rm in} = \langle T_c \hat{I}(t)e^{-i\int dt' H_\phi(t')}\rangle, \qquad (10)$$

where C denotes the double side $\eta = \pm$ Keldysh contour [53]. Here T_C is the time ordering operator on a contour and the average is performed with the Hamiltonian H_0 whereas the contribution from H_α is already accounted in I_{el}. As discussed in detail in Ref. [22], the second-order interaction correction to the current is expressed in terms of the self-energies,

$$\delta I_{\rm in} = \mathcal{S}\int_{-\infty}^{\infty}\frac{d\varepsilon}{2\pi}[\Sigma^{-+}(\varepsilon) - \Sigma^{+-}(\varepsilon)]i\pi\nu\Delta f(\varepsilon); \qquad (11)$$

we used the notation $\mathcal{S} = \frac{N(N-1)e\pi}{h}\cos 2\delta_0$. The self-energies in Eq. (11) are defined in terms of the Green's functions as $\Sigma^{\eta_1,\eta_2}(t) = (\frac{\phi_1}{\pi\nu^2})^2\sum_{k_1,k_2,k_3} G_{bb}^{\eta_1,\eta_2}(k_1,t)G_{bb}^{\eta_2,\eta_1}(k_2,-t)G_{bb}^{\eta_1,\eta_2}(k_3,t)$. The *local* Green's functions of aa/bb fermions and the mixed ab fermions in real time are given by

$$\mathcal{G}_\pm(t) = -\frac{\pi\nu}{2}\left[\frac{T_L e^{-i\mu_L t}}{\sinh(\pi T_L t)} \pm \frac{T_R e^{-i\mu_R t}}{\sinh(\pi T_R t)}\right], \qquad (12)$$

with $\mathcal{G}_+(t) = G_{aa/bb}^{+-}(t)$ and $\mathcal{G}_-(t) = G_{ba/ab}(t) = i\pi\nu\Delta f(t)$. Computing the integrals in the presence of the finite bias voltage and finite temperature drop one gets

$$\Sigma^{-+}(\varepsilon) - \Sigma^{+-}(\varepsilon) = \frac{\phi_1^2}{i\pi\nu}\left[\frac{3}{4}(e\Delta V)^2 + \frac{\Delta T}{T}(\pi T)^2 + \varepsilon^2 + (\pi T)^2\right]. \qquad (13)$$

In the limit $\Delta T \to 0$, $\Delta f(\varepsilon) = -(\Delta T\varepsilon/T)\partial f/\partial\varepsilon$ and the FL self-energies, being even functions of ε, do not contribute to the thermocurrent at $\Delta V = 0$. Therefore, the thermoelectric coefficient G_{12} at $\Delta T \to 0$ is fully determined by elastic processes [54]. The linear-response inelastic contribution to the current at $\Delta T = 0$ reads

$$\frac{\delta I_{\rm in}}{Ne/h} = \left[\frac{2}{3}(\pi T)^2(N-1)\phi_1^2\cos 2\delta_0\right]e\Delta V. \qquad (14)$$

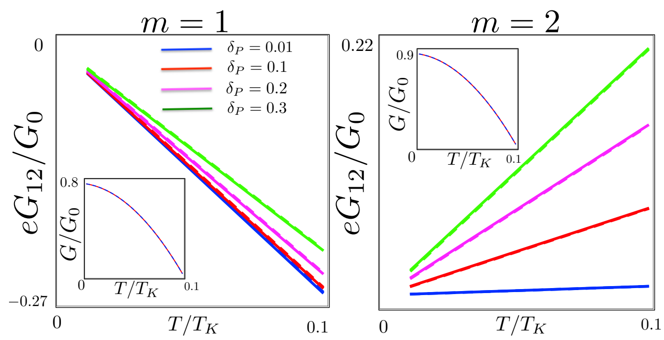

FIG. 2. Main frame: Thermoelectric coefficient G_{12} as a function of the reference temperature $T = T_R$ at different values of potential scattering δ_P. Inset: differential conductance G as a function of T for $\delta_P = 0.3$. The legend is shown in the left panel. Solid lines: numerical solution obtained from (1) with (S6)-(S7), (S11)-(S12) [51]. Dashed lines: analytical solution given by (15) and (16). Solid and dashed lines are almost indistinguishable.

The equation for the total current beyond the linear response is cumbersome and given in the Supplemental Material [51]. Finally, the differential conductance G and differential thermoelectric coefficient G_{12} are given by

$$G(T) = G_0\left[\sin^2\delta_0 + \frac{\alpha_1^2}{3}\frac{N+1}{N-1}(\pi T)^2\cos 2\delta_0\right], \qquad (15)$$

$$G_{12}(T) = -G_0\left[\frac{\alpha_1}{3e}\pi^2 T\sin 2\delta_0\right], \qquad (16)$$

where $G_0 = Ne^2/h$ is the unitary conductance.

Potential scattering. As is seen from (16), the G_{12} at given reference temperature T in linear response is proportional to $\sin 2\delta_0$. For the particle-hole symmetric (PHS) case in the absence of potential scattering $m = N/2$ (we assume N even), $\delta_0 = \pi/2$ and both G_{12} and the thermoelectric power are zero—the particle thermocurrent exactly compensates the hole thermocurrent (Fig. 2 right panel, blue curve). The potential scattering explicitly breaks the PH symmetry. It can be accounted for by replacement of δ_0 by $\tilde{\delta}_0 = \delta_0 + \delta_P$, $\delta_P \ll \delta_0$. As a result, the finite G_{12} and thermopower arises. The results of calculations obtained from the zero-current conditions for SU(4) quantum impurity are illustrated in Fig. 2. First, for the case of singly occupied quantum impurity $m = 1$ (Fig. 2, left panel), $\delta_0 = \pi/4$ and the PH symmetry is explicitly broken. At zero potential scattering inelastic effects associated with the finite bias voltage vanish and the thermoelectric power is completely defined by elastic processes. The effect of potential scattering is twofold: (i) it detunes G_{12} from its maximal value and (ii) it results in finite-temperature inelastic corrections to the conductance (see Fig. 2, inset). For the PHS case of double occupation $m = 2$ (Fig. 2, right panel) $\delta_0 = \pi/2$. Therefore the finite potential scattering results in finite G_{12} and differential thermopower S is proportional to $\sin 2\delta_P$. Crossover between $m = 2$ SU(4) and $m = 1$ SU(2) has been studied recently experimentally [55]. Note that the current across the dot symmetrically coupled to the leads contains only odd powers of the voltage both for the PHS $m = 2$ and the PH-nonsymmetric (PHN) $m = 1$ cases [56].

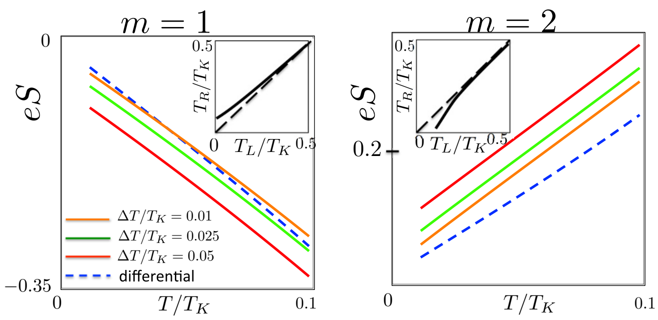

FIG. 3. Main frame: Thermoelectric power S as a function of the reference temperature $T = T_R$; blue dashed curve: differential S given by (2) with (15) and (16); solid curves correspond to S defined under zero-current condition by (1) with (S6)-(S7), (S11)-(S12) [51] at different values of ΔT (see the legend); $\delta_P = 0.3$. Inset: evolution of the zero current steady state as a function of the temperatures of L-R leads at finite voltage $e\Delta V/T_K = 0.02$.

Seebeck effect. In the limit $T \to 0$ the differential thermopower is given by [57]

$$S(T) = -2eL_0 T \cot\tilde{\delta}_0 \left.\frac{\partial\tilde{\delta}(\varepsilon)}{\partial\varepsilon}\right|_{\varepsilon=0} = -\frac{\pi\gamma T}{e}\cot\tilde{\delta}_0, \qquad (17)$$

where $L_0 = \pi^2/(3e^2)$ is the Lorentz number and $\gamma = 2\pi\alpha_1/3 \sim 1/T_K$ in accordance with the FL theory [12].

The thermopower *measurements* [8] refer however *not* to the differential Seebeck effects (see Fig. 1). Since there were no independent measurements of T_L and T_R, the temperature drop was estimated from the Joule heat. It appeared that the ΔT was finite and *not* fulfilling the condition $|\Delta T| \ll T_R$. To demonstrate the significance of nonlinear effects associated with finite temperature drop we show in Fig. 3 the thermopower of the SU(4) model computed by two different methods: (i) the dashed blue line stands for the differential thermopower $S(T) = G_{12}/G$ where G_{12} is obtained at zero voltage drop while G is calculated at equal temperatures of the leads; (ii) the solid lines correspond to $S = -\Delta V/\Delta T$ resembling the experimental situation in [8]: the temperature drop is fixed at $\Delta T/T_K = 0.05$ (red), 0.025 (green), 0.01 (orange), and the thermovoltage is obtained from the zero current condition. As one can see, at small reference temperatures "finite ΔT" thermopower always *overshoots* the differential S. The effect is more pronounced in the PHS regime [58]. This observation can explain the thermovoltage offset observed in the experiment [8] in the Kondo limit of SU(2) quantum impurity (PHS regime). According to our calculations this offset is associated with a nonlinear ΔT dependence of the current at low reference temperatures (see Fig. 3, insets). We suggest to check this statement experimentally by performing Seebeck effect measurements varying the temperature in the "hot" lead.

Coupling asymmetry. The effect of coupling asymmetry $t_L \neq t_R$ in (3) manifests itself in the following way: for the broken PH-symmetry case it results in an asymmetric $I-V$ curve due to a contribution to the current quadratic in voltage which, in turn, depends linearly on the coupling asymmetry. For both PHS and PHN cases the coupling asymmetry results in (i) renormalization of the elastic contribution to the charge current [see [51], Eq. (S5)]; (ii) renormalization of the Kondo temperature due to tunneling rates asymmetry [51]; and (iii) renormalization of the coefficient in front of the term cubic in voltage. The magnitude of current is suppressed by the coupling asymmetry. Besides, it also affects the thermocurrent. However, this effect is proportional to $\Delta V \cdot \Delta T$ and therefore beyond the linear-response theory (see the Supplemental Material [51] for details).

Peltier effect. In order to compute other thermoelectric coefficients, e.g., the Peltier effect, one needs to define and compute the heat current. To proceed with full-fledged Keldysh calculations one can, e.g., deal with the Luttinger "gravitational potential" approach [59–61]. Such a theory would access the effects nonlinear in ΔT [62]. In the linear-response theory the Peltier coefficient $\Pi(T)$ can be calculated using the transport integrals method (see, e.g., [63] for details) based on calculation of different momenta of the single-particle lifetime $\tau(\epsilon,T)$ (see the Supplemental Material [51]) and is related to the thermopower by the Onsager's relation $\Pi(T) = S(T)T$ [64].

Summary. The full-fledged theory based on Keldysh out-of-equilibrium calculations of the electric current is constructed for the SU(N) Kondo quantum impurity subject to a finite bias voltage and a finite temperature drop. The transport coefficients, conductance G, thermoelectric coefficient G_{12}, and thermopower S, are computed under the condition of zero-current state for the strong-coupling regime of the quantum impurity. It is shown that pronounced nonlinear effects in temperature drop influence the transport coefficients at the low-temperature limit. These effects are likely sufficient to resolve the experimental puzzle of the thermotransport through the Kondo impurity at the strong coupling.

Note added. While completing this work, a paper [65] appeared where the significance of nonlinear effects in temperature drop on the temperature-driven current through SU(2) quantum impurity has been reported.

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