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Orbital caloritronic transport in strongly interacting quantum dots

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Abstract

We discuss population imbalances between different orbital states due to applied thermal gradients. This purely thermoelectric effect appears quite generically in nanostructures with a pseudospin (orbital) degree of freedom. We define an orbital Seebeck coefficient that characterizes the induced orbital bias generated across a quantum conductor in response to a temperature difference applied to the attached reservoirs. We analyze a two-terminal strongly interacting quantum dot with two orbital states and find that the orbital thermopower acts as an excellent tool to describe the crossover between $SU(4)$ and $SU(2)$ Kondo states. Our conclusions are reinforced with a detailed comparison to the charge thermopower using exact numerical renormalization group calculations.

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1. Introduction

The discovery of the spin Seebeck effect [1] has ignited research in spin caloritronics, a field where the focus is put on the generation of spin-polarized electric currents by applying thermal gradients [2]. Nonequilibrium spin accumulations can thus be generated in response to a temperature difference across a junction even when the charge current vanishes [3].

In addition to electronic spin, many nanostructures offer the possibility of an extra degree of freedom—the orbital quantum number. This property naturally arises in carbon nanotubes as a result of the two ways (clockwise and anticlockwise) that electrons possess to move around the tube axis [4] or can be artificially realized in double quantum dot structures since each individual dot state can be viewed as the possible outcome of two-level pseudospin (orbital) measurements [5].

We here put forward the idea of generating different orbital populations using temperature gradients (orbital caloritronics). We thus define the orbital or pseudospin Seebeck coefficient which measures the orbital bias voltage generated across a mesoscopic conductor under the conditions of vanishing charge and orbital currents. Remarkably, we find that the orbital thermopower acts as an efficient probe to characterize pseudospin-driven quantum crossovers in quantum dots and carbon nanotubes.

The orbital degree of freedom plays an essential role in the formation of highly symmetric Kondo states. While the conventional $SU(2)$ Kondo resonance arises, at low temperature T, from the many-body exchange interaction between a localized spin $\frac{1}{2}$ (the quantum impurity) and conduction band electrons (the Fermi sea), the $SU(4)$ Kondo physics occurs because the entangled spin and orbital degrees of freedom form a hyperspin with higher dimensionality that undergoes simultaneous flip processes both in the spin and the orbital sectors [9]. Hence, the crossover between $SU(2)$ and $SU(4)$ Kondo effects involves two strongly correlated states with rather different temperature scales (the Kondo temperatures $T_{\rm K}^{SU(2)}$ and $T_{\rm K}^{SU(4)}$ that typically fulfill $T_{\rm K}^{SU(4)} \gg T_{\rm K}^{SU(2)}$). Such crossover has been investigated both experimentally [6–8] and theoretically [9–16]. In particular, an applied magnetic field in nanotubes couples differently to the spin and orbital quantum numbers [4], lifting the degeneracy and allowing for a tunable conversion from $SU(4)$ Kondo physics to a purely spin or orbital Kondo effect [6]. On the other hand, pseudospin resolved transport has been achieved very recently in double quantum dots [17].

Thermoelectric properties of $SU(2)$ Kondo impurities show not only clear changes depending on the ratio $T/T_{\rm K}$ [18] but also deviations from the semiclassical Mott formula [19]. These are important features for strongly interacting quantum dots that might potentially work as nanoscale thermoelectric coolers or heat-to-electricity converters. When the pseudospin degree of freedom is created by charged states in negative charging energy quantum dots, the Seebeck coefficient can be substantially enlarged [20]. Moreover, pure spin currents can be thermally generated from an artificial Kondo impurity coupled to ferromagnetic leads [21] or in the presence of magnetic fields [22]. Recently, the Seebeck coefficient has been proposed as a sensitive probe of the crossover between $SU(2)$ and $SU(4)$ Kondo states [23]. It is thus natural to ask whether the generalization to orbital thermopower can provide additional insight on that crossover. Below, we demonstrate that the orbital Seebeck coefficient shows a characteristic minimum that signals the crossover from one Kondo state to another. Therefore, investigation of orbital thermoelectric effects is interesting from both viewpoints—the practical motivation that leads to the generation of orbital polarizations and the fundamental study of orbital driven crossovers.

2. Orbital and charge Seebeck coefficients

We consider a generic mesoscopic conductor with interacting electrons and coupled to left (L) and right (R) leads. Let $\nu = \pm$ be the orbital index that labels the two orbital states present both in the sample and in the leads. For completeness, we also take into account the spin index $\sigma = \uparrow / \downarrow$, although in what follows we will assume spin degeneracy in order to focus on orbital effects only. The exact formula for the current at channel ν reads [24, 25]

$$I_\nu = \frac{e}{h} \sum_\sigma \int \mathrm{d}\varepsilon \; (f_{\mathrm{L}\nu}(\varepsilon) - f_{\mathrm{R}\nu}(\varepsilon))\, \mathcal{T}_{\nu\sigma}(\varepsilon), \tag{1}$$

where the generalized transmission function is

$$\mathcal{T}_{\nu\sigma}(\varepsilon) = \frac{4\pi \Gamma_\mathrm{L} \Gamma_\mathrm{R}}{\Gamma_\mathrm{L} + \Gamma_\mathrm{R}} A_{\nu\sigma}(\varepsilon) \tag{2}$$

in terms of level broadenings $\Gamma_\alpha = \pi \sum_k |t_\alpha|^2 \delta(\varepsilon - \varepsilon_{\alpha k})$ with t_α the tunnel amplitude from lead $\alpha = \mathrm{L}, \mathrm{R}$. The total linewidth is then $\Gamma = \sum_\alpha \Gamma_\alpha$. The dot spectral weight $A_{\nu\sigma}(\varepsilon)$ in an orbital ν with spin σ is obtained from the retarded dot Green's function $\mathcal{G}^r_{\nu\sigma,\nu\sigma}(\varepsilon)$ by $A_{\nu\sigma}(\varepsilon) = -\mathrm{Im}\, \mathcal{G}^r_{\nu\sigma,\nu\sigma}(\varepsilon)/\pi$.

In equation (1), the leads are Fermi reservoirs with distribution function $f_{\alpha\nu}(\varepsilon) = 1/[\exp((\varepsilon - \mu_{\alpha\nu})/k_\mathrm{B} T_\alpha) + 1]$, where $\mu_{\alpha\nu} = E_\mathrm{F} + eV_{\alpha\nu}$ (E_F is the Fermi energy) and $T_\alpha = T + \theta_\alpha$ (T is the background temperature). It is worthy to note that the electrochemical potential $\mu_{\alpha\nu}$ depends on the orbital index ν that labels the bias $V_{\alpha\nu}$. This model is valid for, e.g. a long carbon nanotube with a depleted region acting as a quasi-localized level (the quantum dot). It has been experimentally confirmed that the orbital index is conserved during tunneling across a highly symmetric carbon-nanotube quantum dot [6]. Thus, possible orbital polarizations are determined from the imbalance $\mu_{\alpha+} \neq \mu_{\alpha-}$ [26][5]. Finally, θ_α is the temperature shift applied to lead α.

We define the *orbital* current as $I_\mathrm{o} = I_+ - I_-$ while the electric (charge) current is accordingly given by $I_\mathrm{c} = I_+ + I_-$. The applied thermal difference is denoted with $\Delta T = \theta_\mathrm{L} - \theta_\mathrm{R}$. With the electrochemical potential parametrization $\mu_\alpha = (\mu_{\alpha+} + \mu_{\alpha-})/2$, the electric voltage bias ΔV and the orbital bias ΔV_o become

$$e\Delta V = \mu_\mathrm{L} - \mu_\mathrm{R}, \tag{3}$$

$$e\Delta V_\mathrm{o} = (\mu_{\mathrm{L}+} - \mu_{\mathrm{L}-}) - (\mu_{\mathrm{R}+} - \mu_{\mathrm{R}-}). \tag{4}$$

Notice that Rejec *et al* [22] propose analogous expressions for the pure spin case.

We define the orbital thermopower

$$S_\mathrm{o} = -\left.\frac{e\Delta V_\mathrm{o}}{\Delta T}\right|_{I_\mathrm{c}=0, I_\mathrm{o}=0} \tag{5}$$

[5] In the same way one can define spin polarizations or even Kramers polarizations between time-reversal pair states in the presence of spin–orbit coupling see [26].

as the ratio between the induced orbital voltage $\Delta V_{\rm o}$ and the applied temperature difference ΔT, in close analogy with the charge Seebeck coefficient,

$$S_{\rm c} = -\frac{e\Delta V}{\Delta T}\bigg|_{I_{\rm c}=0, I_{\rm o}=0}. \tag{6}$$

We emphasize that the two coefficients are calculated under the condition that *both* orbital and charge currents simultaneously vanish.

In linear response, the differences ΔT, $e\Delta V$, and $e\Delta V_{\rm o}$ are small and we expand $I_{\rm o}$ and $I_{\rm c}$ to first order:

$$I_{\rm o} = \frac{e}{h}\left[(\mathcal{I}_{1+}-\mathcal{I}_{1-})\frac{\Delta T}{T} + (\mathcal{I}_{0+}-\mathcal{I}_{0-})e\Delta V + \frac{1}{2}(\mathcal{I}_{0+}+\mathcal{I}_{0-})e\Delta V_{\rm o}\right], \tag{7}$$

$$I_{\rm c} = \frac{e}{h}\left[(\mathcal{I}_{1+}+\mathcal{I}_{1-})\frac{\Delta T}{T} + (\mathcal{I}_{0+}+\mathcal{I}_{0-})e\Delta V + \frac{1}{2}(\mathcal{I}_{0+}-\mathcal{I}_{0-})e\Delta V_{\rm o}\right]. \tag{8}$$

Here, $\mathcal{I}_{n\nu}$ is the transport integral defined by

$$\mathcal{I}_{n\nu} = \sum_{\sigma}\int {\rm d}\varepsilon\, \varepsilon^n\, (-\partial_\varepsilon f_0(\varepsilon))\, \mathcal{T}_{\nu\sigma}(\varepsilon) \tag{9}$$

with $f_0(\varepsilon) = 1/({\rm e}^{\varepsilon/T}+1)$ the equilibrium distribution function (we set $E_{\rm F}=0$ and $k_{\rm B}=1$).

To have purely orbital currents, the charge current must vanish. This is accomplished by the application of the electric bias

$$e\Delta V = -\frac{1}{2}\left(\frac{\mathcal{I}_{1+}}{\mathcal{I}_{0+}} + \frac{\mathcal{I}_{1-}}{\mathcal{I}_{0-}}\right)\frac{\Delta T}{T}. \tag{10}$$

Therefore, the orbital Seebeck coefficient becomes

$$S_{\rm o} = \frac{1}{T}\left(\frac{\mathcal{I}_{1+}}{\mathcal{I}_{0+}} - \frac{\mathcal{I}_{1-}}{\mathcal{I}_{0-}}\right). \tag{11}$$

This is a general result. We expect the formation of an orbital bias $\Delta V_{\rm o} = -eS_0\Delta T$ in the leads when the transmission depends on the orbital index, similarly to the temperature driven generation of spin biases in junctions showing spin-dependent scattering [3].

At low temperatures, it is useful to consider the Sommerfeld expansion [27]. Then,

$$S_{\rm o} \underset{T\to 0}{\longrightarrow} \frac{\pi^2 T}{3}\left(\frac{\sum_\sigma \partial_\varepsilon A_{+\sigma}(E_{\rm F})}{\sum_\sigma A_{+\sigma}(E_{\rm F})} - \frac{\sum_\sigma \partial_\varepsilon A_{-\sigma}(E_{\rm F})}{\sum_\sigma A_{-\sigma}(E_{\rm F})}\right) \tag{12}$$

to leading order in T. This expression is a generalization of the Mott formula [28] valid for orbital bias driven quantum systems.

For comparison, we also give the expression of the charge Seebeck coefficient

$$S_{\rm c} = \frac{1}{2T}\left(\frac{\mathcal{I}_{1+}}{\mathcal{I}_{0+}} + \frac{\mathcal{I}_{1-}}{\mathcal{I}_{0-}}\right), \tag{13}$$

which in the limit of $T \to 0$ becomes

$$S_c \xrightarrow[T\to 0]{} \frac{\pi^2 T}{6}\left(\frac{\sum_\sigma \partial_\varepsilon A_{+\sigma}(E_F)}{\sum_\sigma A_{+\sigma}(E_F)} + \frac{\sum_\sigma \partial_\varepsilon A_{-\sigma}(E_F)}{\sum_\sigma A_{-\sigma}(E_F)}\right). \tag{14}$$

Equations (11) and (13) are valid for generic nanostructures with two orbital states. As an illustration, we now consider a quantum dot with energy levels

$$\varepsilon_\nu = \varepsilon_d + \nu\frac{\delta}{2}, \tag{15}$$

where δ is the orbital splitting induced by any symmetry breaking mechanism such as a magnetic field along a nanotube axis [12] and ε_d is the mean energy level measured with respect to E_F. Formally, the problem is equivalent to a spin-split quantum dot with a single energy level. However, the difference is that spin and orbital states couple differently to an external magnetic field since their associated magnetic moments generally differ; e.g. for a carbon-nanotube quantum dot, orbital splittings of the order of δ are 10–20 times larger than spin splittings at a fixed magnetic field [4]. We shall first consider noninteracting electrons and then discuss in detail the strongly correlated case where the orbital degree of freedom plays a crucial role.

3. Noninteracting limit

For noninteracting electrons, the exact expression for the dot spectral weight is

$$A_{\nu\sigma}(\varepsilon) = \frac{1}{\pi}\frac{\Gamma}{(\varepsilon - \varepsilon_\nu)^2 + \Gamma^2}. \tag{16}$$

Using this equation in equations (12) and (14) we find the low temperature behavior of the Seebeck coefficients

$$S_o \xrightarrow[T\to 0]{} \frac{2\pi^2 T}{3}\left(\frac{\varepsilon_d + \delta/2}{(\varepsilon_d + \delta/2)^2 + \Gamma^2} - \frac{\varepsilon_d - \delta/2}{(\varepsilon_d - \delta/2)^2 + \Gamma^2}\right), \tag{17a}$$

$$S_c \xrightarrow[T\to 0]{} \frac{\pi^2 T}{3}\left(\frac{\varepsilon_d + \delta/2}{(\varepsilon_d + \delta/2)^2 + \Gamma^2} + \frac{\varepsilon_d - \delta/2}{(\varepsilon_d - \delta/2)^2 + \Gamma^2}\right), \tag{17b}$$

which are plotted in figure 1. We observe in figure 1(b) that when $\delta = 0$ the charge thermopower S_c changes sign when the dot level ε_d lies above or below E_F. This is an expected behavior due to the ability of S_c to indicate electron- or hole-like transport [29]. As δ increases, S_c remains roughly constant until the split level crosses E_F and S_c then changes sign. Importantly, the charge thermopower vanishes at the particle symmetry point ($\varepsilon_d = 0$) regardless of the δ value.

More interestingly, the orbital thermopower S_o shows distinct features, see figure 1(a). It vanishes in both limits, $\delta \to 0$ and $\delta \gg \Gamma$. This is expected since no orbital bias can be induced if the two orbitals are degenerate or they lie far apart. Furthermore, the orbital thermopower is quite generally nonzero when particle–hole symmetry takes place at $\varepsilon_d = 0$, unlike S_c. The two Seebeck coefficients also differ when transport is electron- or hole-like. While S_c changes its sign when ε_d is reversed with respect to E_F, the orbital Seebeck coefficient is insensitive to

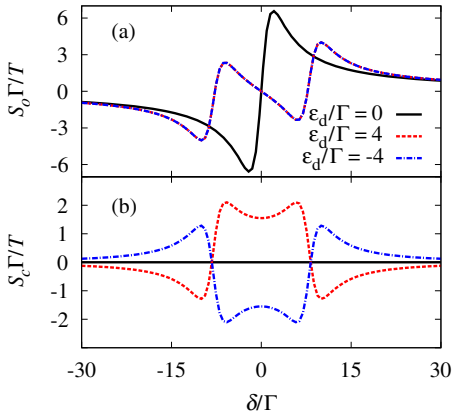

Figure 1. (a) Orbital (S_o) and (b) charge (S_c) Seebeck coefficients as a function of the level splitting δ in the noninteracting limit and temperature $T \to 0$ for various values of the level position ε_d.

whether transport is dominated by electron or hole excitations since both curves for $\varepsilon_d = 4\Gamma$ and $\varepsilon_d = -4\Gamma$ in figure 1(a) are identical ($S_o(\varepsilon_d) = S_o(-\varepsilon_d)$ in equation (17*a*)). In addition, for $\varepsilon_d = 0$ S_0 reaches an optimal value when the splitting δ is of the order of Γ because charge fluctuations are maximal precisely at that level position. The optimal value shifts with $\varepsilon_d \neq 0$ and new peaks arise due to the passage of the split level ε_ν across $\sim \pm\Gamma$ above and below the Fermi energy. This demonstrates a full tunability of the generated orbital population with the aid of an external gate voltage.

4. Strong coupling regime

Consider now electron–electron interactions described by $\sum_{\nu\sigma\neq\nu'\sigma'} U n_{\nu\sigma} n_{\nu'\sigma'}$, where $n_{\nu\sigma}$ is the occupation of the dot spin–orbital state (ν, σ) and U is the onsite charging energy. Using the Friedel–Langreth sum rule [30, 31], the spectral weight $A_{\nu\sigma}(\varepsilon)$ at $\varepsilon = E_F$ can be expressed in terms of $n_{\nu\sigma}$

$$A_{\nu\sigma}(E_F) = \frac{\sin^2(n_{\nu\sigma}\pi)}{\pi\Gamma}. \tag{18}$$

It follows that its energy derivative takes the form [23]

$$\partial_\varepsilon A_{\nu\sigma}(E_F) = \frac{1}{\pi\Gamma\widetilde{\Gamma}_{\nu\sigma}} \sin(2n_{\nu\sigma}\pi)\sin^2(n_{\nu\sigma}\pi). \tag{19}$$

Here, the tunnel broadening $\widetilde{\Gamma}_{\nu\sigma} = z_{\nu\sigma}\Gamma$ becomes renormalized by the quasi-particle weight factor $z_{\nu\sigma} = 1/[1 - \partial_\varepsilon Re\Sigma^r_{\nu\sigma}(E_F)]$, where $\Sigma^r_{\nu\sigma}$ is the retarded self-energy contribution due to interaction effects [31].

Combining equations (18) and (19), we find the thermopowers

$$S_o = \frac{\pi^2 T}{3}(\mathcal{S}_+ - \mathcal{S}_-), \tag{20a}$$

$$S_c = \frac{\pi^2 T}{6}(\mathcal{S}_+ + \mathcal{S}_-), \tag{20b}$$

where

$$\mathcal{S}_\nu \equiv \frac{\mathcal{I}_{1\nu}}{\mathcal{I}_{0\nu}} = \frac{\sum_\sigma \left[\sin(2n_{\nu\sigma}\pi)\sin^2(n_{\nu\sigma}\pi)/\widetilde{\Gamma}_{\nu\sigma}\right]}{\sum_\sigma \sin^2(n_{\nu\sigma}\pi)}. \tag{21}$$

Since our system is spin rotationally invariant, we have $n_{\nu\sigma} = n_\nu/2$ and $\widetilde{\Gamma}_{\nu\sigma} = \widetilde{\Gamma}_\nu$. Thus, equation (21) can be further simplified

$$\mathcal{S}_\nu = \frac{1}{\widetilde{\Gamma}_\nu}\sin(n_\nu\pi), \tag{22}$$

where $n_\nu = \sum_\sigma n_{\nu\sigma}$.

Equations (20) and (22) are formally exact in the strong coupling regime, i.e. when temperature is much lower than the characteristic Kondo temperature of the system. In the crossover from $SU(4)$ to $SU(2)$ Kondo physics, the $SU(4)$ Kondo temperature $T_{\rm K}^{SU(4)}$ is larger than the $SU(2)$ Kondo temperature $T_{\rm K}^{SU(2)}$. Thus, in what follows we perform calculations in the limit $T \to 0$ and our results will be qualitatively valid even for $T < T_{\rm K}^{SU(2)}$. Our goal is to find the orbital occupation n_ν, which fully determines both the orbital and charge Seebeck coefficients. One possibility is to employ a slave-boson mean-field theory [23]. However, this approach neglects the orbital index in the renormalized hybridization function, $\widetilde{\Gamma}_\nu \simeq \widetilde{\Gamma}$. This is qualitatively correct in the limit $\delta \to 0$ but it breaks down as δ increases because $\widetilde{\Gamma}_\nu$ will be renormalized differently for $\nu = \pm$, similarly to the spin Kondo effect in the presence of ferromagnetism [33–35]. Since our main goal in the remainder of the paper is to discuss a qualitative picture of the orbital themoelectric effect in a strongly correlated system, we prefer not to delve into complicated details and consider instead the *scaled* thermopowers

$$\tilde{S}_{\rm o} = \frac{\pi^2}{3}(\mathcal{S}_+\widetilde{\Gamma}_+ - \mathcal{S}_-\widetilde{\Gamma}_-) = \frac{\pi^2}{3}(\sin(n_+\pi) - \sin(n_-\pi)), \tag{23a}$$

$$\tilde{S}_{\rm c} = \frac{\pi^2}{6}(\mathcal{S}_+\widetilde{\Gamma}_+ + \mathcal{S}_-\widetilde{\Gamma}_-) = \frac{\pi^2}{6}(\sin(n_+\pi) + \sin(n_-\pi)). \tag{23b}$$

Next, we follow two different routes for assessing n_ν. First, we consider a variational approach that yields analytical results for the Kondo temperature and the dot orbital occupation. Then, we perform a numerical renormalization group (NRG) analysis which fully takes into account Kondo fluctuations in the orbital states.

5. Variational approach

We consider the limit $U \to \infty$. Since the Kondo ground state is a many-body singlet, we take the trial wave function [32]

$$|\psi_0\rangle = \left(\alpha + \sum_k^{k_{\rm F}} \sum_{\nu,\sigma} \beta_{k\nu} d^\dagger_{\nu\sigma} c_{k\nu\sigma}\right)|F\rangle, \tag{24}$$

where $c^\dagger_{k\nu\sigma}(c_{k\nu\sigma})$ $(d^\dagger_{\nu\sigma}(d_{\nu\sigma}))$ annihilates (creates) a conduction (dot) electron with momentum k and spin σ in a channel ν and $|F\rangle$ represents the filled Fermi sea ground state when the dot states are empty.

To calculate the variational energy of the trial wave function, we use the energy functional

$$E_0[|\psi_0\rangle] = \frac{\langle\psi_0|\mathcal{H}|\psi_0\rangle}{\langle\psi_0|\psi_0\rangle}, \tag{25}$$

where the system Hamiltonian reads

$$\mathcal{H} = \sum_{\alpha,k,\nu,\sigma} \varepsilon_{\alpha k} c^{\dagger}_{\alpha k\nu\sigma} c_{\alpha k\nu\sigma} + \sum_{\nu,\sigma} \varepsilon_{\nu} d^{\dagger}_{\nu\sigma} d_{\nu\sigma} + \sum_{\alpha,k,\nu,\sigma} \left(t_{\alpha} c^{\dagger}_{\alpha k\nu\sigma} d_{\nu\sigma} + \text{h.c.} \right) \tag{26}$$

with the constraint that the dot occupation is always 1 due to the infinite charging energy limit.

On minimizing equation (25) with respect to α and $\beta_{k\nu}$ we find

$$E_0 = \sum_{k,\sigma} \frac{t^2}{\varepsilon_k - T_{\text{K}}} + \sum_{k,\sigma} \frac{t^2}{\varepsilon_k - T_{\text{K}} - \delta}, \tag{27}$$

where $t = \sqrt{t_{\text{L}}^2 + t_{\text{R}}^2}$. The Kondo temperature is defined as $T_{\text{K}} = \varepsilon_- - E_0$, i.e. the energy difference between the lowest orbital level (we take $\delta > 0$) and the ground state energy. We transform in equation (27) the sums over k into integrals. Hence [23, 36],

$$T_{\text{K}}(\delta) = \left\{ D(D+\delta) \exp\left[\frac{\pi\varepsilon_-}{2\Gamma} \right] + \frac{\delta^2}{4} \right\}^{1/2} - \frac{\delta}{2}, \tag{28}$$

where D is the lead bandwidth. For $\delta = 0$, we have a strongly correlated four-fold degenerate state and the resulting Kondo state possesses $SU(4)$ symmetry with a Kondo temperature $T_{\text{K}}^{SU(4)} = T_{\text{K}}(0) = D\exp[\pi\varepsilon_-/4\Gamma]$. As δ increases orbital flip transitions become energetically costly and in the limit $\delta \to \infty$ we recover purely spin Kondo physics characterized with a Kondo temperature $T_{\text{K}}^{SU(2)} = T_{\text{K}}(\infty) = D\exp[\pi\varepsilon_-/2\Gamma]$. Due to a different numerical factor inside the exponential, one has $T_{\text{K}}^{SU(4)} \gg T_{\text{K}}^{SU(2)}$, as expected [9].

The average dot occupation is given by

$$n_{\nu} = \frac{\left\langle\psi_0\right| \sum_{\sigma} d^{\dagger}_{\nu\sigma} d_{\nu\sigma} \left|\psi_0\right\rangle}{\langle\psi_0|\psi_0\rangle} = \frac{\sum_{k,\sigma} \beta^2_{k\nu}}{\alpha^2 + \sum_{k,\nu,\sigma} \beta^2_{k\nu}}. \tag{29}$$

The minimization procedure and the integration over the k-space yield

$$n_{\nu}(\delta) = \frac{2\Gamma T_{\text{K}} \left(T_{\text{K}} + \delta\right) / \left(T_{\text{K}} + \Lambda\right)}{\pi T_{\text{K}} \left(T_{\text{K}} + \delta\right) + 2\Gamma \left(2T_{\text{K}} + \delta\right)}, \tag{30}$$

where $\Lambda = \delta(0)$ if $\nu = +(-)$. We recall that T_{K} is a function of δ, cf equation (28). When $\delta = 0$, the occupation is the same for both orbital levels

$$n_{\nu} \xrightarrow[\delta=0]{} \frac{2\Gamma}{\pi T_{\text{K}}^{SU(4)} + 4\Gamma}. \tag{31}$$

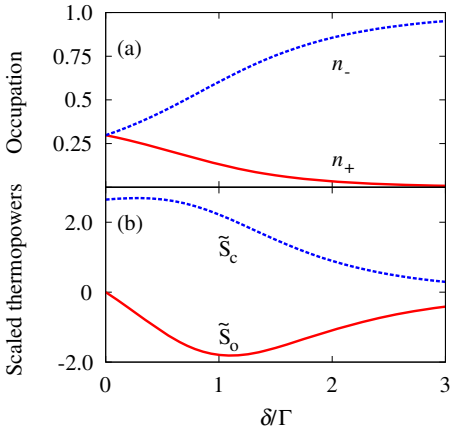

Figure 2. (a) Dot occupation n_ν for the orbital quantum number $\nu = \pm$ as a function of the level splitting δ obtained from a variational approach. (b) Scaled thermopowers (charge $\bar{S}_c$ and orbital $\bar{S}_o$) as a function of δ. Parameters: $\varepsilon_d/\Gamma = -4$, $D/\Gamma = 20$, $U \to \infty$ and $T \to 0$.

As δ increases, the orbital $\nu = +$ becomes less populated due to the level splitting, as depicted in figure 2(a) with solid lines. In the $SU(2)$ Kondo limit ($\delta \to \infty$), equation (30) gives

$$n_\nu \underset{\delta\to\infty}{\longrightarrow} \begin{cases} \pi T_K^{SU(2)} & \text{for } \nu = +, \\ 2\Gamma/\left(\pi T_K^{SU(2)} + 2\Gamma\right) & \text{for } \nu = -. \end{cases} \tag{32}$$

In general, the $SU(2)$ Kondo temperature is much smaller than the hybridization width, $T_K^{SU(2)} \ll \Gamma$. Therefore, $n_- = 1$ and $n_+ = 0$ to a good extent (see figure 2(a)) and we recover the $1/2$ value of the population per spin obtained at very low temperatures [31].

Clearly, the orbital level occupations differ depending on the Kondo state symmetry. As a consequence, the thermopowers (orbital and charge) will be significantly altered as a function of the level splitting δ. Furthermore, for a system with $SU(2)$ symmetry the Kondo resonance develops at the Fermi level E_F, see figure 3(a). We below discuss the numerical method that generates figure 3. Therefore, the charge thermopower S_c will attain an exceedingly small value at low temperatures since the derivative of the spectral weights $\partial_\varepsilon A_{\nu\varepsilon}(\varepsilon)$ vanishes at E_F. The dashed line in figure 2(b) at $\delta \gg \Gamma$ precisely reflects this property. On the other hand, for a system with $SU(4)$ symmetry the Kondo resonance develops at $\varepsilon \approx T_K^{SU(4)}$, as shown in figure 3(b). This is a crucial difference with the $SU(2)$ case since $\partial_\varepsilon A_{\nu\varepsilon}(E_F) \neq 0$ and $\tilde{S}_c$ then reaches a finite value at $\delta = 0$.

More interestingly, the orbital Seebeck coefficient $\tilde{S}_o$ reaches a maximum (in absolute value) at intermediate values of the level splitting, see figure 2(b). At $\delta = 0$, $\tilde{S}_o$ vanishes because $n_+ = n_-$. For $\delta \gg \Gamma$, $\tilde{S}_o$ tends to zero for the same reason that the charge thermopower decreases—the Kondo resonance remains pinned at E_F. Then, an extremum must arise for a nonzero value of δ. We find that a maximal orbital bias is generated when the splitting is of the order of $T_K^{SU(4)}$. Since this energy scale is precisely of the order of the level broadening, our results can be understood in terms of a resonance which behaves effectively as a noninteracting system with renormalized parameters. This picture is valid in the low temperature regime where Kondo correlations simultaneously quench spin and charge fluctuations [31].

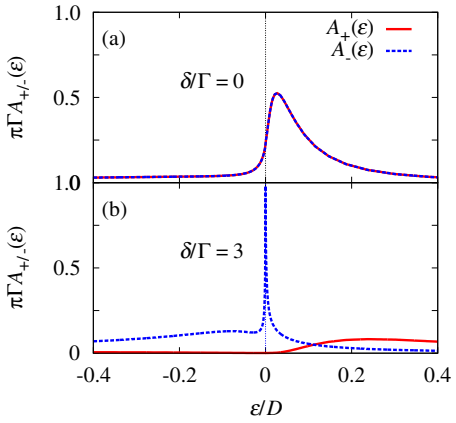

Figure 3. NRG calculation of the dot spectral weight A_ν as a function of energy ε for the two orbital states $\nu = \pm$. (a) $SU(4)$ Kondo resonance clearly develops for level splitting $\delta = 0$ (the spectral weights for both orbitals coincide). (b) $SU(2)$ Kondo resonance forms when δ is tuned beyond the crossover point between the two Kondo states in which case the contribution from the $\nu = +$ channel to the Kondo resonance is negligible. Parameters: $\varepsilon_d/\Gamma = -4$, $D/\Gamma = 20$, $U/\Gamma = 200$ and $T \to 0$. The vertical dotted line is a guide to the eye.

6. Numerical results

Our previous results were restricted to $U \to \infty$ case. We now consider large (but finite) charging energies using a NRG formalism.

In the Lehmann representation, the dot spectral weight takes the form

$$A_{\nu\sigma}(\varepsilon) = \frac{1}{\mathcal{Z} f_0(\varepsilon)} \sum_{p,q} \mathrm{e}^{-E_p/T} |\langle p|d^\dagger_{\nu\sigma}|q\rangle|^2 \delta\left(\varepsilon - (E_p - E_q)\right), \tag{33}$$

where $\mathcal{Z} = \sum_p \mathrm{e}^{-E_p/T}$ is the partition function and E_p, E_q are many-body eigenenergies calculated within NRG [37]. We use equation (33) to calculate the dot local densities of states shown in figure 3.

The orbital occupation is readily obtained from equation (33) as

$$n_\nu = \sum_\sigma \int \mathrm{d}\varepsilon\, A_{\nu\sigma}(\varepsilon) f_0(\varepsilon). \tag{34}$$

In figure 4(a), we depict n_ν for $U = 200\,\Gamma$ as a function of δ. For vanishingly small level splittings, the occupations are equal, $n_+ = n_-$, as expected. Importantly, their exact values are smaller than $1/2$. This can be understood with the aid of equation (31). Unlike the exponentially small $SU(2)$ Kondo temperature $T_K(\infty)$, the higher $SU(4)$ Kondo temperature is $T_K^{SU(4)} \simeq 0.864\Gamma$ for the parameters used in figure 4. Therefore, its contribution cannot be neglected in the denominator of equation (31). This is a crucial difference with the $SU(2)$ case. In addition, when δ increases n_+ tends to vanish since the level ε_+ is pushed up and its occupation is energetically hindered. At the same time, n_- shows the opposite behavior.

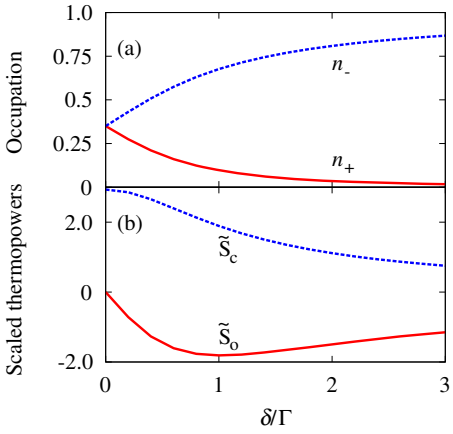

Figure 4. (a) NRG dot occupation n_ν for the orbital quantum number $\nu = \pm$ as a function of the level splitting δ. (b) NRG scaled thermopowers (charge $\tilde{S}_c$ and orbital $\tilde{S}_o$) as a function of δ. Parameters: $\varepsilon_d/\Gamma = -4$, $D/\Gamma = 20$, $U/\Gamma = 200$ and $T \to 0$.

Figure 4(b) shows the scaled thermopowers obtained from our NRG calculations. Our results strongly resemble those obtained with the variational approach, cf figure 2(b). This confirms our previously discussed picture of the orbital thermopower minimum signaling the crossover from $SU(4)$ to $SU(2)$ Kondo physics as the level splitting is increased. Notice that here we have analyzed scaled Seebeck coefficients since they are easier to understand (they depend on the occupation only, see equations (23)). We do not expect qualitative changes if the exact S were calculated using, e.g. the methods discussed in [22, 38, 39].

7. Conclusions

We have investigated the formation of orbital accumulations in systems with spin and pseudospin degrees of freedom under the influence of externally applied temperature differences. We have defined the orbital Seebeck coefficient from an open-circuit pure orbital bias. We have found that orbital thermopower is really sensitive to changes in level splitting fields possibly present in the system. Thus, we propose to use the occurrence of orbital thermopower peaks as the 'smoking gun' of the crossover between Kondo states with distinct symmetry types.

The presence of orbital polarizations could be experimentally detected using the different coupling of circularly polarized light to the unequal population of electronic orbital states [40]. An alternative scheme might measure the magnetization response using ultrasmall magnetometers [41]. Further work is thus needed to test the effects discussed in this paper.

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