Adiabatic pumping through a quantum dot in the Kondo regime: Exact results at the Toulouse limit

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Transport properties of ultrasmall quantum dots with a single unpaired electron are commonly modeled by the nonequilibrium Kondo model, describing the exchange interaction of a spin-$\frac{1}{2}$ local moment with two leads of noninteracting electrons. Remarkably, the model possesses an exact solution when tuned to a special manifold in its parameter space known as the Toulouse limit. We use the Toulouse limit to exactly calculate the adiabatically pumped spin current in the Kondo regime. In the absence of both potential scattering and a voltage bias, the instantaneous charge current is strictly zero for a generic Kondo model. However, a nonzero spin current can be pumped through the system in the presence of a finite magnetic field, provided the spin couples asymmetrically to the two leads. Tunneling through a Kondo impurity thus offers a natural mechanism for generating a pure spin current. We show, in particular, that one can devise pumping cycles along which the average spin pumped per cycle is closely equal to $\hbar$. By analogy with Brouwer's formula for noninteracting systems with two driven parameters, the pumped spin current is expressed as a geometrical property of a scattering matrix. However, the relevant scattering matrix that enters the formulation pertains to the Majorana fermions that appear at the Toulouse limit rather than the physical electrons that carry the current. These results are obtained by combining the nonequilibrium Keldysh Green function technique with a systematic gradient expansion, explicitly exposing the small parameter controlling the adiabatic limit.

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I. INTRODUCTION

The act of pumping is well known from everyday life. By repeatedly operating a periodic sequence of steps, one can transfer a certain amount of fluid or gas between reservoirs held at equal potential. The same principle applies to electrical charge. By periodically modulating spatially confined potentials, it is possible to generate a nonzero dc current between leads that are kept at equal temperature and electrochemical potential. When operated sufficiently slow such that the typical scattering time for electrons is much faster than the time over which the scattering potentials vary, this process is known as adiabatic quantum pumping. Recently, there has been considerable theoretical[1–19] and experimental[20–23] interest in adiabatic quantum pumping in confined nanostructures. Besides the fundamental and technological importance of understanding time-dependent phenomena in nanodevices such as semiconductor and carbon nanotube quantum dots, adiabatic quantum pumping offers new possibilities that otherwise are difficult to realize in conventional dc transport measurements with a finite voltage bias. Most notably, the ability to pump a quantized amount of charge per cycle,[20,22] which is of potential metrological importance. In this paper, we address another such example, the generation of pure, possibly quantized spin current without any charge current.[13–19,23]

In the absence of interactions, adiabatic pumping is by now well understood. In particular, building on the scattering approach of Büttiker *et al.*,[24] Brouwer has elegantly shown[2] that the adiabatically pumped current can be expressed in terms of the instantaneous (equilibrium) scattering matrix. In the case of two driven parameters, the pumped charge per cycle reduces to a geometrical property of the equilibrium scattering matrix, pertaining to the area enclosed in parameter space by the pumping cycle. All other details of the pumping cycle, i.e., the explicit time dependences of the scattering potentials, are irrelevant as long as pumping is adiabatic.

Far less understood are the effects of interactions, where efforts have focused thus far on zero-dimensional[3,11,13] and one-dimensional[12,14] systems. The difficulty with incorporating interactions lies in the need to treat retardation effects beyond the static limit. Indeed, recent attempts to generalize Brouwer's formula so as to include interactions[25–27] have required either the introduction of complicated vertex corrections[25,26] or the application of a gradient expansion to interaction-induced self-energies.[27] Both formulations can only be implemented approximately at this stage, urging the need for benchmark results against which approximate treatments can be tested. In this paper, we provide such an exact result for the pumped currents through a Kondo impurity.

Kondo-assisted tunneling has been observed by now in an abundance of nanostructures, ranging from semiconductor[28] and nanotube[29] quantum dots to single-atom[30] and single-molecule[31] transistors. In the Kondo regime, these systems are described by the well-known Kondo model: a spin-$\frac{1}{2}$ local moment undergoing antiferromagnetic spin exchange with the conduction electrons in the leads. The nonequilibrium Kondo model, either with a static or a time-dependent voltage bias, is a difficult problem. Remarkably, it possesses an exact solution when tuned to a special manifold in its parameter space, known as the Toulouse limit.[32,33] At the Toulouse limit, one can apply a suitable canonical transformation to recast the interacting problem in free, quadratic form. This requires the introduction of new fermionic degrees of freedom having no simple relation to the physical

electrons in the leads. The resulting solution, which generalizes previous exact results for the equilibrium Kondo problem,[34,35] does not correspond to realistic parameters. It requires large values of certain exchange couplings (see below), rendering it incapable of describing weak-coupling physics. However, the Toulouse limit is expected to correctly describe the strong-coupling regime of the nonequilibrium Kondo effect, as different microscopic models are governed by the same strong-coupling fixed point. Indeed, previous applications of the model to dc,[32,33] ac,[36] and pulsed-bias potentials[37] have shown all the qualitative features of Kondo-assisted tunneling: a zero-bias anomaly that splits in an applied magnetic field, Fermi-liquid characteristics in the low-T and low-V differential conductance, side peaks in the differential conductance at $eV=\pm n\hbar\omega$ for an ac drive of frequency ω, and a hierarchy of time scales for the rise, saturation, and falloff of the current in response to a pulsed-bias potential. The Toulouse limit was also recently applied to compute the full counting statistics for tunneling through a Kondo impurity.[38,39]

In this paper, we take the solution one step further by exactly computing the adiabatically pumped currents on the Toulouse manifold. Contrary to previous applications of the Toulouse limit to Kondo-assisted tunneling, we set the voltage bias to zero but consider a general periodic modulation of the transverse exchange couplings and the local magnetic field (the free parameters on the Toulouse manifold). In the limit of slow time variations, we obtain an exact analytic expression for the adiabatically pumped spin current. In particular, we show that a nonzero spin current can be pumped through the system for a time-varying magnetic field, provided the couplings to the two leads are made asymmetric. Such a condition is easily met in practical devices. Unlike the spin current, the instantaneous charge current is strictly zero in the absence of both potential scattering and a voltage bias, as follows from general symmetry considerations. This feature is generic to the Kondo model, independent of the adiabatic and Toulouse limits. Hence, tunneling through a Kondo impurity offers a natural mechanism for the realization of a spin battery, i.e., a source of pure spin current without any charge current. This statement, valid both in the adiabatic limit and beyond, is in qualitative agreement with earlier slave-boson mean-field studies of adiabatic pumping through an Anderson impurity,[13,25] indicating that no fine tuning of model parameters is required as long as one operates in the Kondo regime. Finally, we show that one can devise suitable pumping cycles that operate as a quantized spin pump. Namely, a spin closely quantized to $\hbar$ is pumped per cycle without an accompanying charge.

As indicated above, the solution at the Toulouse limit relies on a nonlocal transformation that converts the original spin-exchange Hamiltonian to free-fermion form.[35,33] In contrast to conventional quadratic Hamiltonians, though, the number of fermions (not to be confused with the physical electrons in the system) is not conserved, excluding the application of Brouwer's formula in its existing form. To generalize Brouwer's result to this somewhat unconventional case, we follow a path similar to the one taken by Vavilov *et al.*[7] in studying the photovoltaic effect in open chaotic cavities. Starting from the nonequilibrium Keldysh Green function technique, we show how the adiabatic limit is obtained from a systematic gradient expansion. In this manner, we are able to express the instantaneous spin current in terms of an energy-shift matrix,[6] leading to a Brouwer-type formula for the adiabatically pumped spin current.

The formalism outlined above has three notable advantages over the scattering approach[24] originally used by Brouwer to derive his result: (i) It conveniently accommodates the case where particles are not conserved. (ii) All orders of perturbation theory are summed up in the Keldysh technique, thus exceeding linear response. (iii) Based on a systematic gradient expansion, one can easily read off the small parameter controlling the adiabatic limit. We emphasize, however, that the resulting Brouwer-type formula for the electronic spin current is formally expressed in terms of the scattering matrix for the Majorana fermions that appear in the transformed Hamiltonian. While technically useful, these degrees of freedom have neither a simple representation nor a simple interpretation in terms of the physical electrons in the leads, thus obscuring a clear physical picture. It remains to be seen whether a similar expression can be written down for the spin current directly in terms of the scattering properties of the lead electrons which carry the current.

The remainder of the paper is organized as follows. In Sec. II, we briefly review the Toulouse limit, introducing the different Green functions that will be used later in the course of the calculation. In Sec. III, we present general symmetry considerations and apply them to the problem at hand. In particular, we show that the instantaneous charge current is strictly zero in the absence of potential scattering, whereas the spin current is zero unless the dot couples asymmetrically to the two leads. Proceeding with quantitative calculations, we combine the Keldysh technique with a gradient expansion in Sec. IV to derive a Brouwer-type formula for the adiabatically pumped spin current in the Toulouse limit. Using this formula, a specific class of pumping cycles is analyzed in detail in Sec. V. In particular, we demonstrate a pumping cycle for which the total spin pumped per cycle is closely equal to $\hbar$, thus operating as a quantized spin pump. Finally, we present our conclusions in Sec. VI.

II. PHYSICAL MODEL AND TOULOUSE LIMIT

We begin with a brief review of the Toulouse limit and with introducing the different Green functions that will be used later in calculating the pumped spin current. The physical system under consideration is shown schematically in Fig. 1. A spin-$\frac{1}{2}$ local moment τ is embedded between two leads of noninteracting spin-$\frac{1}{2}$ electrons, undergoing a spin-exchange interaction with the local conduction-electron degrees of freedom on either side of the junction. As emphasized in Sec. I, the impurity moment τ can represent either an ultrasmall quantum dot with a single unpaired electron[28] or an actual magnetic impurity as in single-atom[30] and single-molecule[31] transistors.

Since scattering off the impurity is restricted to the s-wave channel, one can reduce the conduction-electron degrees of freedom that couple to the impurity to one-dimensional fields $\psi_{\alpha\sigma}(x)$, where $\alpha=R,L$ labels the lead

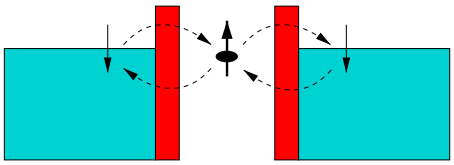

FIG. 1. (Color online) Schematic description of the physical system. A spin-$\frac{1}{2}$ local moment τ is placed in between two leads of noninteracting spin-$\frac{1}{2}$ electrons. The local moment τ experiences a spin-exchange interaction with the local conduction-electron degrees of freedom near the junction, as described by the Hamiltonian of Eq. (1). Tunneling between the leads is facilitated by spin-exchange terms that scatter an electron across the junction.

(right or left) and $\sigma=\uparrow,\downarrow$ specifies the spin orientations. In terms of the one-dimensional fields, coupling to the impurity takes place via the local spin densities at the origin: $\vec{s}_{\alpha\beta}=\frac{1}{2}\Sigma_{\sigma,\sigma'}\psi^{\dagger}_{\alpha\sigma}(0)\vec{\sigma}_{\sigma,\sigma'}\psi_{\beta\sigma'}(0)$. The most general form of a spin-exchange Hamiltonian is therefore

$$\mathcal{H}=iv_F\sum_{\alpha=L,R}\sum_{\sigma=\uparrow,\downarrow}\int_{-\infty}^{\infty}\psi^{\dagger}_{\alpha\sigma}(x)\partial_x\psi_{\alpha\sigma}(x)dx + \sum_{\alpha,\beta=L,R}\sum_{\lambda=x,y,z}J^{\alpha\beta}_{\lambda}(t)\tau^{\lambda}s^{\lambda}_{\alpha\beta}-\mu_B g_i H(t)\tau^z, \quad (1)$$

where we have allowed for different exchange couplings $J^{\alpha\beta}_{\lambda}=J^{\beta\alpha}_{\lambda}$ and for a local magnetic field H acting on the impurity spin. Here, μ_B and g_i are the Bohr magneton and impurity Landé g factor, respectively. Throughout the paper, we use units for which $\hbar=k_B=1$, while the electronic charge is taken to be $-e$. Proper units will be reinstated in some of the final expressions presented below.

The Hamiltonian of Eq. (1) is written for general time-dependent exchange couplings $J^{\alpha\beta}_{\lambda}(t)$ and local magnetic field $H(t)$. Our interest, however, will be in slow periodic modulations of the transverse couplings $J^{\alpha\beta}_{x}(t)=J^{\alpha\beta}_{y}(t)$ and the local magnetic field. The longitudinal couplings $J^{\alpha\beta}_{z}$ will be taken to be constant in time and equal to particular values as detailed below. It is the fine tuning of $J^{\alpha\beta}_{z}$ that defines the Toulouse manifold and enables our exact solution.

A. Toulouse limit

The spin-exchange Hamiltonian of Eq. (1) is conventionally derived from the more basic Anderson impurity model via the Schrieffer-Wolff transformation.[40] The couplings $J^{\alpha\beta}_{\lambda}$ generated in this case are weak, isotropic (i.e., independent of λ), and satisfy $J^{LL}J^{RR}=(J^{LR})^2$. The Toulouse limit corresponds to a different sector in the parameter space of the Kondo Hamiltonian where $J^{LL}_z=J^{RR}_z=2\pi v_F$ and $J^{LR}_z=0$. The transverse couplings $J^{\alpha\beta}_{x}(t)=J^{\alpha\beta}_{y}(t)\equiv J^{\alpha\beta}_{\perp}(t)$ and the local magnetic field $H(t)$ are allowed to be arbitrary and will be subsequently taken to be periodically modulated in time. Physically, this choice of parameters implies that tunneling is always accompanied by a spin flip. Although quite remote from the situation encountered in real quantum dots, this model is expected to correctly describe the strong-coupling regime of the Kondo effect, as argued in Sec. I and elaborated on in Refs. 32, 33, and 37. In particular, it has been shown[33] that the strong-coupling physics of the Anderson impurity model is best described both in and out of equilibrium by couplings that satisfy

$$J^{LL}_{\perp}J^{RR}_{\perp}=(J^{LR}_{\perp})^2. \quad (2)$$

As described in detail in Ref. 33, the Hamiltonian of Eq. (1) can be mapped under the conditions listed above onto a free-fermion form. The mapping involves a sequence of steps, comprised of (i) bosonizing the fermion fields, (ii) a nonlocal canonical transformation involving the conduction-electron spin degrees of freedom, and (iii) refermionization of the boson fields to form four new fermion fields: $\psi_{\nu}(x)$ with $\nu=c,s,f,sf$. Here c, s, f, and sf stand for charge, spin, flavor (left minus right), and spin-flavor fields. In addition, the impurity spin τ, which has been mixed by the canonical transformation with the conduction-electron spin degrees of freedom, is represented in terms of two real Majorana fermions: $\hat{a}=-\sqrt{2}\tau^y$ and $\hat{b}=-\sqrt{2}\tau^x$. At the conclusion of these steps, one arrives at a quadratic Hamiltonian conveniently written in the form

$$H'=\sum_{\nu=c,s,f,sf}\sum_{k}\epsilon_k\psi^{\dagger}_{\nu,k}\psi_{\nu,k}+i\mu_B g_i H(t)\hat{b}\hat{a}+iJ^{+}_{sf}(t)\hat{\chi}^{+}_{sf}\hat{b} + iJ^{-}_{sf}(t)\hat{\chi}^{-}_{sf}\hat{a}+iJ^{-}_{f}(t)\hat{\chi}^{-}_{f}\hat{a}, \quad (3)$$

where we have introduced the three couplings

$$J^{+}_{sf}(t)=\frac{J^{LL}_{\perp}(t)+J^{RR}_{\perp}(t)}{2\sqrt{2\pi a}}, \quad (4)$$

$$J^{-}_{sf}(t)=\frac{J^{LL}_{\perp}(t)-J^{RR}_{\perp}(t)}{2\sqrt{2\pi a}}, \quad (5)$$

$$J^{-}_{f}(t)=\frac{J^{LR}_{\perp}(t)}{\sqrt{2\pi a}}. \quad (6)$$

Here, the energies ϵ_k are equal to $-v_F k$, a is an ultraviolet momentum cutoff corresponding to a lattice spacing, and L is the effective size of the leads (i.e., k is discretized in units of $2\pi/L$). The fields $\hat{\chi}^{\pm}_{\nu}$ $(\nu=f,sf)$ are local Majorana fermions, defined as

$$\hat{\chi}^{+}_{\nu}=\frac{1}{\sqrt{2L}}\sum_{k}(\psi^{\dagger}_{\nu,k}+\psi_{\nu,k}), \quad (7)$$

$$\hat{\chi}^{-}_{\nu}=\frac{1}{i\sqrt{2L}}\sum_{k}(\psi^{\dagger}_{\nu,k}-\psi_{\nu,k}). \quad (8)$$

Relaxation of each of the conditions $J^{LR}_z=0$, $J^{LL}_z+J^{RR}_z=4\pi v_F$, and $J^{LL}_z-J^{RR}_z=0$ introduces a different interaction term into the Hamiltonian of Eq. (3), as discussed in Ref. 41 and detailed below.

Although noninteracting, the Hamiltonian of Eq. (3) is unconventional in the sense that it does not conserve the number of ψ fermions (not to be confused with the physical electrons in the system). Indeed, the fermion fields $\psi_{\nu}(x)$ with $\nu=c,s,f,sf$ have neither a simple representation nor a

simple interpretation in terms of the original electronic degrees of freedom. Consequently, not all observables can be computed based on the mapping of Eq. (3). Only observables that have a simple representation in terms of the ψ fields are accessible. Fortunately, both the charge and spin currents fall in this category.

To derive the transformed forms of the electronic charge and spin currents, it is necessary to go back to their original representation in terms of the physical electrons in the leads. Denoting the total number operator for electrons with spin projection σ in lead α by $\hat{N}_{\alpha\sigma}$, the charge current flowing from right to left is given by

$$\hat{I}_c = -ie[\mathcal{H}, \hat{N}_{L\uparrow} + \hat{N}_{L\downarrow}] = ie[\mathcal{H}, \hat{N}_{R\uparrow} + \hat{N}_{R\downarrow}]. \tag{9}$$

Here, $\mathcal{H}$ is the Kondo Hamiltonian of Eq. (1). Since charge fluctuations are excluded on the dot, the instantaneous charge current outgoing from the left lead (left commutator) is identical to the instantaneous charge current flowing into the right lead (right commutator). This is no longer the case with the spin current, defined as half the difference in particle currents between the spin-up and spin-down electrons. (The factor of one-half comes from the electronic spin projection in the z direction.) Indeed, the spin currents associated with the left and right leads differ by a term proportional to $d\tau^z/dt$, which stems from conservation of the total spin projection S^z_{total} of the entire system. Fortunately, this difference in currents has no significance for our purposes, since $d\tau^z/dt$ averages to zero over a single pumping cycle. This grants us the freedom to work with our operator of choice. In the following, we shall concentrate on the symmetrized spin current, i.e., the average of the spin currents to the left and to the right of the impurity, which turns out to be the most convenient current combination to work with. With this convention, the (symmetrized) spin current flowing from left to right is written as

$$\hat{I}_s = \frac{i}{4}[\mathcal{H}, \hat{N}_{R\uparrow} - \hat{N}_{R\downarrow} - \hat{N}_{L\uparrow} + \hat{N}_{L\downarrow}]. \tag{10}$$

Equations (9) and (10) specify the electronic charge and spin currents in terms of the physical electrons. The transformed operators, $\hat{I}'_c$ and $\hat{I}'_s$, are obtained by repeating the same sequence of steps as applied to the Hamiltonian, namely, bosonization, a nonlocal canonical transformation, and refermionization. Skipping the details of the algebra,[33] we quote here only the end result:

$$\hat{I}'_c = ieJ^-_f(t)\hat{\chi}^+_f\hat{a} \tag{11}$$

and

$$\hat{I}'_s = \frac{i}{2}[J^-_{sf}(t)\hat{\chi}^+_{sf}\hat{a} - J^+_{sf}(t)\hat{\chi}^-_{sf}\hat{b}]. \tag{12}$$

Note that although these expressions are written in terms of Majorana fermions, they describe the actual electronic charge and spin currents flowing in the system. The unconventional forms of the currents stem from the nonlocal transformation that has been applied.

B. Keldysh Green functions

To compute the spin current, we shall make use of the nonequilibrium Keldysh Green function technique. The basic ingredients of the theory are the greater, lesser, retarded, and advanced Majorana Green functions, defined as[42]

$$G^>_{\alpha\beta}(t,t') = \langle \hat{\alpha}(t)\hat{\beta}(t')\rangle, \tag{13}$$

$$G^<_{\alpha\beta}(t,t') = \langle \hat{\beta}(t')\hat{\alpha}(t)\rangle, \tag{14}$$

$$G^{r,a}_{\alpha\beta}(t,t') = \mp i\theta(\pm t \mp t')\langle\{\hat{\alpha}(t), \hat{\beta}(t')\}\rangle. \tag{15}$$

Here, $\alpha, \beta \in \{a, b\}$, while the upper and lower signs in Eq. (15) correspond to the retarded (r) and advanced (a) Green functions, respectively. The curly brackets in Eq. (15) denote the anticommutator.

In thermal equilibrium, the Majorana Green functions are easily found by summing all orders of the perturbation theory in the time-independent couplings $J^{\pm}_{sf}$, J^-_f, and H. Specifically, switching over to the energy domain and assuming the wide-band limit, one obtains

$$G^{r,a}(\epsilon) = \frac{1}{(\epsilon \pm i\Gamma_a)(\epsilon \pm i\Gamma_b) - (\mu_B g_i H)^2} \times \begin{bmatrix} \epsilon \pm i\Gamma_b & -i\mu_B g_i H \\ i\mu_B g_i H & \epsilon \pm i\Gamma_a \end{bmatrix}. \tag{16}$$

Here, we have adopted a 2×2 matrix notation, with the indices 1 and 2 corresponding to a and b, respectively.

Equation (16) features two new energy scales,

$$\Gamma_a = \pi\rho_0[(J^-_f)^2 + (J^-_{sf})^2] \tag{17}$$

and

$$\Gamma_b = \pi\rho_0(J^+_{sf})^2, \tag{18}$$

where $\rho_0 = 1/2\pi v_F$ is the density of states per unit length in the leads. These two scales determine the widths of the various Majorana spectral functions, and thus play the role of Kondo temperatures at the Toulouse limit. The conventional single-channel Kondo effect is best described by the case where $\Gamma_a = \Gamma_b \equiv T_K$, corresponding to the condition specified in Eq. (2). The equilibrium greater and lesser Green functions are related in turn to $G^{r,a}(\epsilon)$ through standard identities:

$$G^>_{\alpha\beta}(\epsilon) = i[1 - f(\epsilon)][G^r_{\alpha\beta}(\epsilon) - G^a_{\alpha\beta}(\epsilon)], \tag{19}$$

$$G^<_{\alpha\beta}(\epsilon) = if(\epsilon)[G^r_{\alpha\beta}(\epsilon) - G^a_{\alpha\beta}(\epsilon)], \tag{20}$$

where $f(\epsilon)$ is the Fermi-Dirac distribution function.

As emphasized above, Eqs. (16)–(20) are restricted to thermal equilibrium. They do not apply when any of the couplings $J^{\pm}_{sf}$, J^-_f, and H is time dependent, which is the case of interest here. Indeed, time-dependent couplings are generally difficult to treat analytically even for noninteracting systems. Below, we shall first derive the instantaneous spin current for a general time-dependent setting but will eventually be interested in slow periodic modulations of the four coupling constants listed above. In terms of the original spin-

exchange Hamiltonian of Eq. (1), we allow for general time variation of the couplings $J_{\perp}^{\alpha\beta}$ and field H but demand that the longitudinal exchange couplings $J_z^{\alpha\beta}$ be held fixed at their Toulouse-limit values. We exclude variations in the phase of $J_{\perp}^{LR}=(J_{\perp}^{RL})^*$, as this corresponds to biasing the system. Accordingly, we take $J_{\perp}^{LR}=J_{\perp}^{RL}$ to be real throughout the paper.

C. Deviations from the Toulouse limit

We conclude this section by briefly describing the modifications that are introduced into the Hamiltonian and the current operators upon departure from the Toulouse manifold. As discussed in Ref. 41, the Hamiltonian of Eq. (3) is supplemented by three new interaction terms away from the Toulouse limit:

$$\mathcal{H}' \to \mathcal{H}' + \mathcal{H}_{\text{int}}, \tag{21}$$

with

$$\mathcal{H}_{\text{int}} = -J_z^{LR}\hat{b}\hat{a}\hat{\chi}_f^-\hat{\chi}_{sf}^+ - i\frac{J_z^+}{L}\hat{b}\hat{a}\sum_{k,k'} :\psi_{s,k}^\dagger\psi_{s,k'}:$$

$$-i\frac{J_z^-}{L}\hat{b}\hat{a}\sum_{k,k'} :\psi_{sf,k}^\dagger\psi_{sf,k'}:. \tag{22}$$

Here, $\hat{\chi}_\nu^\pm$ are the local Majorana fields of Eqs. (7) and (8), while: $\psi_{\nu,k}^\dagger\psi_{\nu,k'}$: stands for normal ordering with respect to the unperturbed Fermi sea of the ψ fermions. The three couplings $J_z^-=(J_z^{LL}-J_z^{RR})/2$, $J_z^+=(J_z^{LL}+J_z^{RR})/2-2\pi\hbar v_F$, and J_z^{LR} measure the deviations from the Toulouse manifold in each of the three possible directions in parameter space. The new tunneling term J_z^{LR}, also modifies the current operators $\hat{I}_c'$ and $\hat{I}_s'$, which take the general forms

$$\hat{I}_c' = ieJ_f^-(t)\hat{\chi}_f^+\hat{a} - eJ_z^{LR}(t)\hat{\chi}_f^+\hat{\chi}_{sf}^+\hat{b}\hat{a} \tag{23}$$

and

$$\hat{I}_s' = \frac{i}{2}[J_{sf}^-(t)\hat{\chi}_{sf}^+\hat{a} - J_{sf}^+(t)\hat{\chi}_{sf}^-\hat{b}] + \frac{J_z^{LR}(t)}{2}\hat{\chi}_f^-\hat{\chi}_{sf}^-\hat{b}\hat{a}. \tag{24}$$

Here, we have explicitly allowed for time variation of the new coupling constant J_z^{LR}.

III. SYMMETRY CONSIDERATIONS

Before proceeding to detailed calculations, in this section we first present general symmetry considerations applicable to any two-lead system. By analyzing their implications for the Kondo Hamiltonian of Eq. (1), we identify necessary conditions for finite charge and spin currents to be pumped through the system.

A. Particle-hole symmetry acting separately on each lead

Consider a general two-lead system where each lead is represented by a single spinful channel. The charge current flowing into lead α ($\alpha=L,R$) is given by

$$\hat{I}_{c,\alpha} = -ie[\mathcal{H},\hat{N}_{\alpha\uparrow}+\hat{N}_{\alpha\downarrow}], \tag{25}$$

while the symmetrized spin current $\hat{I}_s$ flowing from left to right is specified in Eq. (10). Here, $\hat{N}_{\alpha\sigma}$ denotes the total number operator for electrons with spin projection σ on lead α. Let us consider the situation where the time-dependent Hamiltonian $\mathcal{H}$ is invariant under a particle-hole transformation that converts particles on each lead to opposite-spin holes on the *same* lead (i.e., $c_{\alpha,k,\sigma}^\dagger \to e^{i\varphi_{\alpha\sigma}}c_{\alpha,-k,\bar{\sigma}}$, where $\bar{\sigma}$ is the spin index opposite to σ; the phases $\varphi_{\alpha\sigma}$ are arbitrary). The total number operator for electrons on lead α, $\hat{N}_\alpha \equiv \hat{N}_{\alpha\uparrow}+\hat{N}_{\alpha\downarrow}$, is converted under such a transformation to $n_\alpha - \hat{N}_\alpha$, where n_α marks the total number of electronic states in lead α. Consequently, $\hat{I}_{c,\alpha}$ transforms according to

$$\hat{I}_{c,\alpha} = -ie[\mathcal{H},\hat{N}_\alpha] \to -ie[\mathcal{H},n_\alpha-\hat{N}_\alpha] = -\hat{I}_{c,\alpha}. \tag{26}$$

If the system begins its evolution from equilibrium, i.e., the statistical averaging at time t depends solely on the Hamiltonian at previous times, then the instantaneous charge current $I_{c,\alpha}(t)\equiv\langle\hat{I}_{c,\alpha}(t)\rangle=-\langle\hat{I}_{c,\alpha}(t)\rangle$ must necessarily be zero.

The above argumentation is quite general, making no reference to the microscopic details of $\mathcal{H}$ nor to the temperature T. Its usefulness lies in revealing the necessary (but not sufficient) condition for a finite instantaneous charge current to flow: Either the Hamiltonian is not permanently invariant under the particle-hole transformation indicated above or the statistical averaging is not determined by the Hamiltonian alone (as in the case of a finite voltage bias). Note that this symmetry bears no information on the spin current, as the latter is invariant under the particle-hole transformation specified above.

B. Particle-hole symmetry that interchanges the two leads

An equivalent statement can be made about the symmetrized spin current $I_s(t)=\langle\hat{I}_s(t)\rangle$ in the case of a particle-hole symmetry that simultaneously interchanges the two leads. Indeed, let us now assume that $\mathcal{H}$ is invariant under a transformation where particles on each lead are converted to opposite-spin holes on the *opposite* lead (i.e., $c_{\alpha,k,\sigma}^\dagger \to e^{i\varphi_{\alpha\sigma}}c_{\bar{\alpha},-k,\bar{\sigma}}$, where $\bar{\alpha}$ is the lead index opposite to α). Under such a transformation, $\hat{I}_{c,\alpha}$ is converted to $-\hat{I}_{c,\bar{\alpha}}$, while $\hat{I}_s$ is transformed to $-\hat{I}_s$. Hence, the instantaneous spin current $I_s(t)$ must necessarily be zero whenever evolution begins from thermal equilibrium. By contrast, no general statement can be made about the charge current in this case, apart from the obvious identity $I_{c,\alpha}(t)=-I_{c,\bar{\alpha}}(t)$.

C. Application to the Kondo Hamiltonian

Our discussion thus far was quite general. We now apply the symmetry arguments presented above to the Kondo Hamiltonian of Eq. (1). It is easy to verify that Eq. (1) is invariant under the particle-hole transformation

$$\psi_{\alpha\uparrow}^\dagger(x) \to \psi_{\alpha\downarrow}(x), \quad \psi_{\alpha\downarrow}^\dagger(x) \to -\psi_{\alpha\uparrow}(x) \tag{27}$$

(corresponding to $\psi_{\alpha,k,\sigma}^\dagger \to \pm\psi_{\alpha,-k,\bar{\sigma}}$), regardless of the local field H and the Kondo couplings $J_\lambda^{\alpha\beta}=J_\lambda^{\beta\alpha}$. Hence, the instan-

taneous charge current for tunneling through a Kondo impurity is strictly zero in the absence of a voltage bias, as follows from the general discussion of Sec. III A. In particular, no charge can be pumped through the system unless a finite amplitude for potential scattering is introduced into the Hamiltonian. Although the description of real quantum dots typically requires the inclusion of a potential-scattering term, the latter can be made negligibly small by operating the device deep in the Kondo regime. In this manner, charge transport can be excluded.

Similarly, it is straightforward to confirm that the Hamiltonian of Eq. (1) is invariant under the combined transformation

$$\psi^{\dagger}_{\alpha\uparrow}(x) \to \psi_{\bar{\alpha}\downarrow}(x), \quad \psi^{\dagger}_{\alpha\downarrow}(x) \to -\psi_{\bar{\alpha}\uparrow}(x) \tag{28}$$

(corresponding to $\psi^{\dagger}_{\alpha,k,\sigma} \to \pm \psi_{\bar{\alpha},-k,\bar{\sigma}}$), provided the intralead exchange couplings obey $J^{LL}_{\lambda}=J^{RR}_{\lambda}$. Thus, the instantaneous spin current is strictly zero if the spin couples equally to the two leads, as follows from the general discussion of Sec. III B. Spin pumping therefore requires asymmetric coupling to the two leads at least in some stretches of time.

It is instructive to rederive these results based on the symmetries of the transformed Hamiltonian $\mathcal{H}'+\mathcal{H}_{\rm int}$, which serves primarily as a check for the correctness of Eqs. (3) and (22). Other than the free kinetic-energy term, the flavor field ψ_f enters both $\mathcal{H}'$ and $\mathcal{H}_{\rm int}$ only in the form of $\hat{\chi}^{-}_{f}$, which is invariant under the particle-hole transformation

$$\psi^{\dagger}_{f,k} \to -\psi_{f,-k}. \tag{29}$$

Note that the latter transformation is restricted to the flavor sector. Consequently, $\mathcal{H}'+\mathcal{H}_{\rm int}$ is invariant under the transformation of Eq. (29), while the charge-current operator, being proportional to $\hat{\chi}^{+}_{f}$, transforms according to $\hat{I}'_c \to -\hat{I}'_c$ [see Eq. (23)]. This in turn demands that $I_c(t)$ be zero in the absence of a voltage bias, in agreement with the general symmetry considerations of Eq. (27).

Similarly, when $J^{LL}_{\lambda}=J^{RR}_{\lambda}$, the couplings J^{-}_{sf} and J^{-}_{z} drop from the transformed Hamiltonian $\mathcal{H}'+\mathcal{H}_{\rm int}$, which now depends on the field ψ_{sf} either through the free kinetic-energy term or in the form of $\hat{\chi}^{+}_{sf}$. As a result, the transformed Hamiltonian is invariant under the spin-flavor particle-hole transformation

$$\psi^{\dagger}_{sf,k} \to \psi_{sf,-k}, \tag{30}$$

while the spin-current operator, being proportional to $\hat{\chi}^{-}_{sf}$, acquires an extra minus sign: $\hat{I}'_s \to -\hat{I}'_s$ [see Eq. (24)]. This in turn implies that the instantaneous spin current $I_s(t)$ is strictly zero if the leads couple equally to the spin, in agreement with the symmetry considerations of Eq. (28). Interestingly, $I_s(t)$ remains zero for symmetric coupling also in the presence a finite bias, as the latter couples solely to the flavor field. This result, originally derived in Ref. 33 for nonequilibrium steady state, also extends to time-dependent couplings and time-dependent bias.

IV. PUMPED SPIN CURRENT

Having established that the instantaneous charge current vanishes for a generic Kondo model in the absence of a voltage bias, we focus our attention hereafter on the spin current. To this end, we evaluate $I_s(t)=\langle \hat{I}_s(t)\rangle$ exactly on the Toulouse manifold by summing all orders of the perturbation theory in the couplings $J^{-}_{f}(t), J^{\pm}_{sf}(t)$, and $H(t)$. We show that a finite spin current can indeed be pumped through the system by applying a nonzero magnetic field, provided the spin couples asymmetrically to the two leads. The calculation proceeds in three steps. Using the Keldysh technique, we first derive a formal expression for the instantaneous spin current in terms of the Majorana Green functions of Eqs. (13)–(15). This portion of the derivation makes no assumption on the time-dependent couplings, apart from the restriction to the Toulouse manifold and the exclusion of an applied voltage bias. The resulting expression is recast in turn in Sec. IV B in terms of a time-dependent scattering matrix for the Majorana fermions, defined in Eq. (46) below. The latter scattering matrix reduces in equilibrium to the Fourier transform (with respect to energy) of the conventional single-particle scattering matrix. Finally, a systematic gradient expansion is carried out in Sec. IV C for the case of slowly varying potentials, resulting in a Brouwer-type formula for the adiabatically pumped spin current.

A. General formulation

We begin by formally deriving the instantaneous spin current $I_s(t)$ using the Keldysh technique for general time-dependent couplings on the Toulouse manifold. As is always the case with the Keldysh approach, we assume that the perturbations $J^{\pm}_{sf}$, J^{-}_{f}, and H have been switched on at some distant time in the past, $t_0 \to -\infty$, prior to which the system was in thermal equilibrium.

To set the stage for the Keldysh formalism, the spin current $I_s(t)$ is first written as

$$I_s(t) = \frac{i}{2}[J^{-}_{sf}(t)G^{<}_{a,sf+}(t,t) - J^{+}_{sf}(t)G^{<}_{b,sf-}(t,t)], \tag{31}$$

where

$$G^{<}_{a,sf+}(t,t') = \langle \hat{\chi}^{+}_{sf}(t')\hat{a}(t)\rangle, \tag{32}$$

$$G^{<}_{b,sf-}(t,t') = \langle \hat{\chi}^{-}_{sf}(t')\hat{b}(t)\rangle \tag{33}$$

[see Eq. (12)]. Using standard diagrammatics, each of the latter correlators is expressed in an exact manner as

$$G^{<}_{a,sf+}(t,t') = -i\int_{-\infty}^{\infty} J^{+}_{sf}(\tau)[G^{<}_{ab}(t,\tau)g^{a}_{sf,+,+}(\tau,t') + G^{r}_{ab}(t,\tau)g^{<}_{sf,+,+}(\tau,t')]d\tau, \tag{34}$$

$$G^{<}_{b,sf-}(t,t') = -i\int_{-\infty}^{\infty} J^{-}_{sf}(\tau)[G^{<}_{ba}(t,\tau)g^{a}_{sf,-,-}(\tau,t') + G^{r}_{ba}(t,\tau)g^{<}_{sf,-,-}(\tau,t')]d\tau, \tag{35}$$

where

$$g^{<}_{\nu,p,p'}(t,t') = \langle \hat{\chi}^{p'}_{\nu}(t')\hat{\chi}^{p}_{\nu}(t)\rangle_0 \tag{36}$$

and

$$g^a_{\nu,p,p'}(t,t') = i\theta(t'-t)\langle\{\hat{\chi}^p_\nu(t),\hat{\chi}^{p'}_\nu(t')\}\rangle_0 \tag{37}$$

are the unperturbed Green functions for the local Majorana fields. Here, $\nu=f,sf$ and $p,p'=\pm 1$. The zero subscripts in Eqs. (36) and (37) come to indicate that both the time evolution and statistical averaging are taken with respect to the unperturbed Hamiltonian, i.e., the free kinetic-energy part of Eq. (3).

In writing Eqs. (34) and (35), we have used the fact that $g^<_{f,-,+}$ and $g^a_{f,-,+}$ identically vanish as long as no voltage bias is applied.[43] Indeed, in the wide-band limit, Eqs. (36) and (37) take the explicit forms

$$g^<_{\nu,p,p'}(t,t') = 2\pi\rho_0\delta_{pp'}\mathcal{F}(t-t') \tag{38}$$

and

$$g^a_{\nu,p,p'}(t,t') = i\pi\rho_0\delta_{pp'}\delta(t-t'), \tag{39}$$

where $\mathcal{F}(t)$ is the Fourier transform of the Fermi function $f(\epsilon)$:

$$\mathcal{F}(t) = \lim_{\eta\to 0^+}\int_{-\infty}^{\infty}\frac{d\epsilon}{2\pi}e^{-i\epsilon t}e^{-|\epsilon|\eta}f(\epsilon). \tag{40}$$

The limiting procedure used in Eq. (40) corresponds to regularizing the conduction-electron density of states per unit length according to $\rho(\epsilon)=\rho_0 e^{-|\epsilon|\eta}$ and taking the wide-band limit $D=1/\eta\to\infty$. Equation (39) is slightly modified for a finite bandwidth D (Ref. 44) but remains proportional to $\delta_{pp'}$. Inserting Eqs. (38) and (39) into Eqs. (34) and (35) and plugging the resulting expressions into Eq. (31), one obtains

$$\begin{aligned} I_s(t) = {} & i\frac{\pi\rho_0}{2}J^+_{sf}(t)J^-_{sf}(t)[G^<_{ab}(t,t)-G^<_{ba}(t,t)] \\ & + \pi\rho_0\int_{-\infty}^{\infty}[J^-_{sf}(t)G^r_{ab}(t,\tau)J^+_{sf}(\tau) \\ & - J^+_{sf}(t)G^r_{ba}(t,\tau)J^-_{sf}(\tau)]\mathcal{F}(\tau-t)d\tau. \end{aligned} \tag{41}$$

It is easy to see at this point that the instantaneous spin current vanishes in the absence of an applied magnetic field, as it physically should. Indeed, setting $H=0$ in Eq. (3), the two Majorana fermions $\hat{a}$ and $\hat{b}$ decouple within the Hamiltonian $\mathcal{H}'$. As a result, the Green functions G_{ab} and G_{ba} identically vanish, as does I_s. It is also apparent that I_s is strictly zero unless the impurity couples asymmetrically to the two leads, in accordance with the general symmetry arguments of Sec. III C. In fact, $I_s(t)$ vanishes not only when $J^{LL}_\perp=J^{RR}_\perp$ but also for $J^{LL}_\perp=-J^{RR}_\perp$, which stems from yet another symmetry of the Toulouse-limit Hamiltonian. Specifically, Eq. (3) is invariant for $J^+_{sf}=0$ under the particle-hole transformation $\psi_{sf,k}\to-\psi^\dagger_{sf,-k}$, while $\hat{I}_s$ transforms according to $\hat{I}_s\to-\hat{I}_s$. Consequently, $I_s(t)=-I_s(t)$ must necessarily vanish when $J^{LL}_\perp=-J^{RR}_\perp$.

B. Time-dependent scattering matrix

Although formally exact, Eq. (41) requires knowledge of the time-dependent Green functions G_{ab} and G_{ba}, which are difficult to compute for a general time-dependent setting. In order to implement the adiabatic limit, it is useful to first recast Eq. (41) in terms of a time-dependent scattering matrix to be defined below. This goal requires a sequence of steps, starting with expressing the lesser Green functions $G^<_{ab}$ and $G^<_{ba}$ in terms of the retarded and advanced Green functions. Since the Hamiltonian of Eq. (3) is quadratic, one has the identities

$$\begin{aligned} G^<_{ab}(t,t) = {} & 2\pi\rho_0\int_{-\infty}^{\infty}d\tau\int_{-\infty}^{\infty}d\tau'[G^r_{aa}(t,\tau)\{J^-_{sf}(\tau)J^-_{sf}(\tau') \\ & + J^-_f(\tau)J^-_f(\tau')\}G^a_{ab}(\tau',t) \\ & + G^r_{ab}(t,\tau)J^+_{sf}(\tau)J^+_{sf}(\tau')G^a_{bb}(\tau',t)]\mathcal{F}(\tau-\tau'), \end{aligned} \tag{42}$$

$$\begin{aligned} G^<_{ba}(t,t) = {} & 2\pi\rho_0\int_{-\infty}^{\infty}d\tau\int_{-\infty}^{\infty}d\tau'[G^r_{ba}(t,\tau)\{J^-_{sf}(\tau)J^-_{sf}(\tau') \\ & + J^-_f(\tau)J^-_f(\tau')\}G^a_{aa}(\tau',t) \\ & + G^r_{bb}(t,\tau)J^+_{sf}(\tau)J^+_{sf}(\tau')G^a_{ba}(\tau',t)]\mathcal{F}(\tau-\tau'). \end{aligned} \tag{43}$$

Substituting Eqs. (42) and (43) into Eq. (41), it is convenient to introduce the scattering T matrix associated with the Majorana fields $\hat{\chi}^\pm_{sf}$ and $\hat{\chi}^-_f$,

$$\mathbf{T}^{r,a}(t,t') = 2\pi\rho_0\begin{bmatrix} J^+_{sf}(t)G^{r,a}_{bb}(t,t')J^+_{sf}(t') & J^+_{sf}(t)G^{r,a}_{ba}(t,t')J^-_{sf}(t') & J^+_{sf}(t)G^{r,a}_{ba}(t,t')J^-_f(t') \\ J^-_{sf}(t)G^{r,a}_{ab}(t,t')J^+_{sf}(t') & J^-_{sf}(t)G^{r,a}_{aa}(t,t')J^-_{sf}(t') & J^-_{sf}(t)G^{r,a}_{aa}(t,t')J^-_f(t') \\ J^-_f(t)G^{r,a}_{ab}(t,t')J^+_{sf}(t') & J^-_f(t)G^{r,a}_{aa}(t,t')J^-_{sf}(t') & J^-_f(t)G^{r,a}_{aa}(t,t')J^-_f(t') \end{bmatrix}. \tag{44}$$

Here, the row and column indices $i=1,2,3$ are identified with $(sf,+)$, $(sf,-)$, and $(f,-)$, respectively. In terms of the T matrix specified above, the spin current is written as

$$I_s(t) = \frac{1}{4}\Bigg[\int_{-\infty}^{\infty} d\tau\{\mathbf{T}^r(t,\tau)\mathcal{F}(\tau-t) - \mathcal{F}(t-\tau)\mathbf{T}^a(\tau,t)\} + i\int_{-\infty}^{\infty} d\tau\int_{-\infty}^{\infty} d\tau'\mathbf{T}^r(t,\tau)\mathcal{F}(\tau-\tau')\mathbf{T}^a(\tau',t)\Bigg]_{(sf-,sf+)} - \frac{1}{4}\Bigg[\int_{-\infty}^{\infty} d\tau\{\mathbf{T}^r(t,\tau)\mathcal{F}(\tau-t) - \mathcal{F}(t-\tau)\mathbf{T}^a(\tau,t)\} + i\int_{-\infty}^{\infty} d\tau\int_{-\infty}^{\infty} d\tau'\mathbf{T}^r(t,\tau)\mathcal{F}(\tau-\tau')\mathbf{T}^a(\tau',t)\Bigg]_{(sf+,sf-)}. \tag{45}$$

Finally, the time-dependent scattering matrix for the Majorana fields $\hat{\chi}_\nu^{\pm}$ is defined as

$$\widetilde{\mathbf{S}}(t,t') = \delta(t-t')\mathbf{1} - i\mathbf{T}^r(t,t'), \tag{46}$$

$$\widetilde{\mathbf{S}}^\dagger(t,t') = \delta(t-t')\mathbf{1} + i\mathbf{T}^a(t,t'), \tag{47}$$

allowing us to compactly rewrite Eq. (45) in the form

$$I_s(t) = \frac{1}{2}\,\mathrm{Im}\int_{-\infty}^{\infty} d\tau\int_{-\infty}^{\infty} d\tau'[\widetilde{\mathbf{S}}(t,\tau)\mathcal{F}(\tau-\tau')\widetilde{\mathbf{S}}^\dagger(\tau',t)]_{(sf+,sf-)}. \tag{48}$$

A word is in order at this point about the time-dependent scattering matrix of Eq. (46). Physically, $\widetilde{\mathbf{S}}(t,t')$ describes the scattering of an incoming Majorana fermion at time t' to an outgoing Majorana fermion at time t. It reduces in equilibrium to the Fourier transform (with respect to energy) of the conventional single-particle scattering matrix and remains an exclusive function of the time difference $\Delta t = t - t'$ under general steady-state conditions. Although this ceases to be the case in the presence of time-varying fields, $\widetilde{\mathbf{S}}(t,t')$ continues to satisfy the generalized unitarity relation

$$\int_{-\infty}^{\infty} d\tau\widetilde{\mathbf{S}}(t,\tau)\widetilde{\mathbf{S}}^\dagger(\tau,t') = \delta(t-t')\mathbf{1}, \tag{49}$$

to be utilized below.

C. Gradient expansion and Brouwer-type formula

The main achievement of Eq. (48) is the expression of the instantaneous spin current in terms of the time-dependent scattering matrix $\widetilde{\mathbf{S}}(t,t')$. For a general periodic modulation of the couplings $J_{sf}^{\pm}$, J_f^-, and H, the instantaneous spin current at time t depends on the specifics of the pumping cycle, for example, the history and rates at which parameters are varied. As we show below, this is not the case in the adiabatic limit, where the only information needed to predict the pumped spin per cycle is (i) the shape of the pumping trajectory in parameter space and (ii) the equilibrium S matrix along the trajectory. Similar to adiabatic quantum pumping in noninteracting systems, the adiabatic limit is approached when the characteristic modulation frequency Ω obeys $\Omega \ll \Gamma_a, \Gamma_b$ at each point along the pumping trajectory. Here, Γ_a and Γ_b are the energy scales defined in Eqs. (17) and (18), respectively.

To substantiate these claims and devise a Brouwer-type formula for adiabatic quantum spin pumping in the Kondo regime, we resort to a systematic gradient expansion of Eq. (48). To this end, we first introduce the Wigner transform of the time-dependent scattering matrix,

$$\mathbf{S}(\epsilon,T) = \int_{-\infty}^{\infty} d\tau e^{i\epsilon\tau}\widetilde{\mathbf{S}}\left(T+\frac{\tau}{2}, T-\frac{\tau}{2}\right). \tag{50}$$

Next, we apply the well-developed machinery of the gradient expansion.[45] For example, the Wigner transform of the convolution of two functions,

$$[A\star B](\epsilon,T) = \int_{-\infty}^{\infty} d\tau e^{i\epsilon\tau}\int_{-\infty}^{\infty} d\tau' A\left(T+\frac{\tau}{2},\tau'\right)B\left(\tau', T-\frac{\tau}{2}\right), \tag{51}$$

has the formally exact representation[45]

$$[A\star B](\epsilon,T) = \exp\left[\frac{1}{2i}\{\partial_T^A\partial_\epsilon^B - \partial_T^B\partial_\epsilon^A\}\right]A(\epsilon,T)B(\epsilon,T) = A(\epsilon,T)B(\epsilon,T) + \frac{1}{2i}(\partial_T A\partial_\epsilon B - \partial_\epsilon A\partial_T B) + \cdots. \tag{52}$$

Here, ∂^A and ∂^B stand for differential operators that act on $A(\epsilon,T)$ and $B(\epsilon,T)$, respectively. The usefulness of Eq. (52) comes into play when the expansion on the right-hand side is controlled by a small parameter. This is indeed the case in the present context, where the double convolution of Eq. (48) possesses an analogous expansion in gradients of $S(\epsilon,T)$. Each combined derivative $\partial_T\partial_\epsilon$ is parametrically reduced for Eq. (48) by a factor of $\Omega/\bar{\Gamma}$, where $\bar{\Gamma}$ is some characteristic value of either Γ_a or Γ_b in the relevant time interval. The scale $\bar{\Gamma}$ is bounded from below by the minimum of Γ_a and Γ_b along the pumping cycle, a quantity denoted hereafter by Γ. Hence, for $\Omega \ll \Gamma$, one can settle with linear order in $\partial_T\partial_\epsilon$ to obtain

$$I_s(t) = \mathrm{Im}\int_{-\infty}^{\infty}\frac{d\epsilon}{4\pi}\Bigg[\Bigg\{\mathbf{S}\mathbf{S}^\dagger + \frac{1}{2i}[(\partial_t\mathbf{S})(\partial_\epsilon\mathbf{S}^\dagger) - (\partial_\epsilon\mathbf{S})(\partial_t\mathbf{S}^\dagger)]\Bigg\}f(\epsilon) + \frac{1}{2i}\{\mathbf{S}(\partial_t\mathbf{S}^\dagger) - (\partial_t\mathbf{S})\mathbf{S}^\dagger\}\times\left(-\frac{\partial f(\epsilon)}{\partial\epsilon}\right)\Bigg]_{(sf+,sf-)}. \tag{53}$$

Here and in the following, T coincides with t. All terms omitted in Eq. (53) are of order $(\Omega/\Gamma)^2$ or higher, and thus can be safely neglected.[46]

The term proportional to the Fermi function $f(\epsilon)$ in Eq. (53) is purely diagonal to order $\mathcal{O}(\Omega/\Gamma)$, as can be seen by expanding the unitarity relation of Eq. (49) to first order in time gradients:

$$\mathbf{S}\mathbf{S}^\dagger + \frac{1}{2i}[(\partial_t \mathbf{S})(\partial_\epsilon \mathbf{S}^\dagger) - (\partial_\epsilon \mathbf{S})(\partial_t \mathbf{S}^\dagger)] + \mathcal{O}[(\Omega/\Gamma)^2] = \mathbf{1}. \tag{54}$$

Since Eq. (53) requires an off-diagonal matrix element of the expression in the square brackets, the instantaneous spin current reduces in the adiabatic limit to

$$I_s(t) = \mathrm{Re}\int_{-\infty}^{\infty} \frac{d\epsilon}{8\pi} f'(\epsilon)[\mathbf{S}(\partial_t \mathbf{S}^\dagger) - (\partial_t \mathbf{S})\mathbf{S}^\dagger]_{(sf+,sf-)}. \tag{55}$$

This expression can be further simplified by noting that $S(\epsilon,t)$ is equal to leading order in Ω/Γ to the instantaneous scattering matrix, i.e., the equilibrium scattering matrix with all system parameters $J_{sf}^{\pm}$, J_f^-, and H frozen at their instantaneous values at time t:

$$\mathbf{S}(\epsilon,t) = \mathbf{S}_{\mathrm{eq}}(\epsilon; J_{sf}^{\pm}(t), J_f^-(t), H(t)) + \mathcal{O}(\Omega/\Gamma). \tag{56}$$

Consequently, one can substitute $\mathbf{S}_{\mathrm{eq}}$ in for $\mathbf{S}$ in Eq. (55). Lastly, one can exploit the unitarity of the equilibrium S matrix, $\mathbf{S}_{\mathrm{eq}}\mathbf{S}_{\mathrm{eq}}^\dagger = \mathbf{1}$, to replace $(\partial_t \mathbf{S}_{\mathrm{eq}})\mathbf{S}_{\mathrm{eq}}^\dagger$ with $-\mathbf{S}_{\mathrm{eq}}(\partial_t \mathbf{S}_{\mathrm{eq}}^\dagger)$ in Eq. (55). This yields the final expression for the spin current,

$$I_s(t) = \mathrm{Re}\left\{\int_{-\infty}^{\infty} \frac{d\epsilon}{4\pi} f'(\epsilon)[\mathbf{S}(\partial_t \mathbf{S}^\dagger)]_{(sf+,sf-)}\right\}. \tag{57}$$

Here and in the remainder of the paper, the symbol $\mathbf{S}$ is used as a shorthand for the instantaneous scattering matrix $\mathbf{S}_{\mathrm{eq}}(\epsilon; J_{sf}^{\pm}(t), J_f^-(t), H(t))$.

Equation (57) is exact in the adiabatic limit, $\Omega \to 0$. Its derivation was based on a systematic truncation of higher order terms in Ω, controlled by the expansion parameter Ω/Γ. It therefore encompasses all pumping trajectories and all coupling regimes, whether weak or strong. This should be contrasted with the commonly used linear-response theory, which is restricted, strictly speaking, to weak coupling only.

In the following, we shall consider examples of pumping cycles where two system parameters, generically termed X_1 and X_2, are varied slowly and periodically in time along a certain closed trajectory $\mathcal{C}$ in parameter space. The quantity of interest in this case is the total magnetization in the z direction (or spin) transferred from left to right in a single pumping cycle. The latter quantity is defined as

$$\langle S \rangle = \oint_{\mathcal{C}} I_s(t)dt, \tag{58}$$

where $I_s(t)$ is the instantaneous spin current. Using Eq. (57), one can express $\langle S \rangle$ as a line integral along the contour $\mathcal{C}$,

$$\langle S \rangle = \mathrm{Re}\left\{\int_{-\infty}^{\infty} \frac{d\epsilon}{4\pi} f'(\epsilon) \oint_{\mathcal{C}} [\mathbf{S}\underline{\nabla}\mathbf{S}^\dagger]_{(sf+,sf-)} \cdot d\underline{X}\right\}. \tag{59}$$

This expression applies to the variation of any number of system parameters $X_1, \ldots, X_N$. In the particular case where $N=2$, one can make use of Green's theorem to express the spin pumped per cycle as a geometric property of the Majorana-fermion scattering matrix, analogous to Brouwer's formula for noninteracting systems. Explicitly, $\langle S \rangle$ assumes the form

$$\langle S \rangle = \mathrm{Re}\int_{-\infty}^{\infty} \frac{d\epsilon}{4\pi} f'(\epsilon) \int_{\mathcal{A}} dX_1 dX_2 \times [\partial_{X_1}\mathbf{S}\partial_{X_2}\mathbf{S}^\dagger - \partial_{X_2}\mathbf{S}\partial_{X_1}\mathbf{S}^\dagger]_{(sf+,sf-)}, \tag{60}$$

where $\mathcal{A}$ is the (oriented) area in parameter space enclosed by the contour $\mathcal{C}$.

Equation (60) is the central result of our study. We devote the remainder of the paper to analyzing its implications for a particular class of pumping trajectories defined below.

V. APPLICATIONS

We conclude our analysis by applying the formula derived above to study a particularly simple class of pumping cycles where one parameter, X_1, controls the transverse Kondo couplings, and the other parameter, X_2, controls the applied magnetic field. Based on our Toulouse-limit calculations, we will show that such a cycle can be used to realize a pure quantized spin pump, namely, *quantized* spin pumping without any charge transport.

To make contact with realistic systems such as quantum dots, we impose hereafter the condition $J_\perp^{LL} J_\perp^{RR} = (J_\perp^{LR})^2$, corresponding to $(J_{sf}^-)^2 + (J_f^-)^2 = (J_{sf}^+)^2$. As mentioned above, this condition best describes the strong-coupling physics of the Anderson impurity model, where a single Kondo scale $\Gamma_a = \Gamma_b \equiv T_K$ emerges. Keeping J_{sf}^+, and thus T_K, fixed, we parametrize J_{sf}^-, J_f^-, and $\mu_B g_i H$ according to

$$J_{sf}^- = X_1, \tag{61}$$

$$J_f^- = \sqrt{(J_{sf}^+)^2 - X_1^2}, \tag{62}$$

and

$$\mu_B g_i H = X_2. \tag{63}$$

In terms of the original Kondo couplings to the two leads, Eqs. (61) and (62) translate to

$$J_\perp^{LL/RR} = \sqrt{2\pi a}(J_{sf}^+ \pm X_1), \tag{64}$$

$$J_\perp^{LR} = \sqrt{2\pi a}\sqrt{(J_{sf}^+)^2 - X_1^2}. \tag{65}$$

The pumping cycle under consideration is depicted schematically in Fig. 2. It consists of four segments, two in which X_1 is tuned from $\pm J_{sf}^+$ to $\mp J_{sf}^+$ while X_2 is kept fixed [lines (a) and (c)] and two in which X_2 is tuned from $\pm h$ to $\mp h$ while X_1 is held fixed [lines (b) and (d)]. The cycle $\mathcal{C}$ thus consists of periodic opening and closing of the transverse couplings to the left and right leads, followed by inversion of the applied magnetic field at points where spin-flip scattering is restricted to one lead only. The analogous cycle for real quantum dots comprises of periodic opening and closing of the tunneling rates to the left and right leads, followed by inversion of the applied magnetic field at points where tunneling is restricted to one lead only.

Combining Eq. (60) for $\langle S \rangle$ with Eqs. (46), (47), (44), and (16) for the instantaneous S matrix, one obtains the following result after some straightforward but tedious algebra:

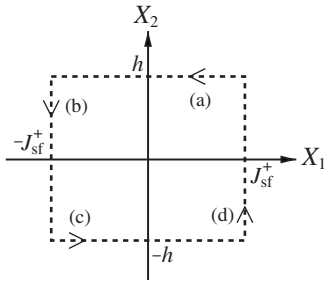

FIG. 2. The pumping cycle under consideration in Sec. V. The first pumping parameter, X_1, controls the Kondo couplings J_{sf}^{-} and J_f^{-}, which vary according to $J_{sf}^{-}=X_1$ and $J_f^{-}=\sqrt{(J_{sf}^{+})^2-X_1^2}$. The third Kondo coupling, J_{sf}^{+}, is held fixed throughout the cycle, along with T_K. The second pumping parameter, X_2, controls the Zeeman splitting $\mu_B g_i H$, which varies according to $\mu_B g_i H=X_2$.

$$\langle S\rangle = -\frac{2\hbar}{\pi}\int_{-\infty}^{\infty} d\epsilon f'(\epsilon)\left[\mathrm{Re}\left\{\frac{hT_K}{h^2+(T_K-i\epsilon)^2}\right\} + \arctan\left(\frac{h+\epsilon}{T_K}\right)\right]. \tag{66}$$

Here, we have restored $\hbar$ for proper units of $\langle S\rangle$. Representative plots of $\langle S\rangle$ as a function of both h and T are shown in Fig. 3. As expected of the Kondo regime, $\langle S\rangle$ is an exclusive function of the rescaled parameters h/T_K and T/T_K. In particular, at $T=0$, one finds

$$\langle S\rangle = \frac{2\hbar}{\pi}\left[\left(\frac{h}{T_K}+\frac{T_K}{h}\right)^{-1}+\arctan\left(\frac{h}{T_K}\right)\right], \tag{67}$$

which has the formal expansion $\langle S\rangle/\hbar=1-\mathcal{O}[(T_K/h)^3]$. Hence, the pumped spin per cycle is closely quantized to $\hbar$

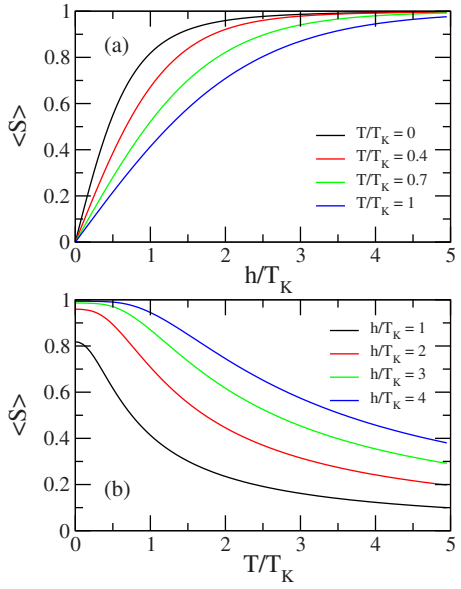

FIG. 3. (Color online) The spin pumped per cycle, $\langle S\rangle$, in units of $\hbar$ (a) plotted as a function of h/T_K for different T and (b) plotted as a function of T/T_K for different h. For $T<T_K$, the spin pumped per cycle rapidly approaches $\hbar$ with increasing h. Explicitly, $\langle S\rangle$ exceeds $0.9\hbar$ for all $T\leqslant 0.45T_K$ when $h=2T_K$.

when the magnetic field H performs a large enough excursion along the pumping cycle. The effect of a temperature is to reduce the spin pumped per cycle. However, $\langle S\rangle$ remains closely quantized to $\hbar$ when $h\gg T_K,T$. Importantly, when $T\ll T_K$, it suffices that h will only moderately exceed T_K in order for $\langle S\rangle$ to closely approach $\hbar$. For example, at $T=0$, the pumped spin per cycle is equal to $0.82\hbar$ $(0.96\hbar)$ by the time $h=T_K$ $(h=2T_K)$.

The above results were derived at the Toulouse limit, which does not correspond to any realistic parameters. It is therefore pertinent to question the relevance of these results to actual quantum dots. Since any exact solution can be used to extract universal low-energy properties of the Kondo effect, we expect Eq. (66) to be *quantitatively* correct when $T,h\ll T_K$. Equation (66) should remain qualitatively correct as one of the parameters T or h becomes comparable to T_K, though quantitative deviations are expected. Still, since $\langle S\rangle$ approaches $\hbar$ quite rapidly with increasing h (essentially by $h\sim T_K$) and since the departure from strong coupling is only logarithmically slow in h, we expect $\langle S\rangle$ to remain nearly quantized in real quantum dots provided $T\ll T_K$. This picture is further supported by a naive application of Brouwer's formula using the exact $T=0$ single-particle scattering matrix[47] and by slave-boson mean-field theory of the corresponding Anderson model.[13] The Toulouse limit fails, however, to describe the weak-coupling regime, as certain bare couplings are required to be large. In particular, Eq. (66) should neither be quantitatively nor qualitatively correct when $T_K\ll T$.

A simple interpretation of Eq. (66) follows from the observation that the ground state of the Kondo model is that of a local Fermi liquid. Only resonant elastic scattering takes place at the Fermi level when $T=0$, as reflected in the Abrikosov-Suhl resonance. The latter resonance is pinned to the Fermi energy when $H=0$ and is split by an applied magnetic field. This basic phenomenology can be mimicked by a simple noninteracting resonant-level model,

$$\mathcal{H}_{\mathrm{RLM}} = \sum_{\alpha=L,R}\sum_{k,\sigma}\epsilon_k\psi_{k\alpha\sigma}^{\dagger}\psi_{k\alpha\sigma} - \mu_B g_i H(d_{\uparrow}^{\dagger}d_{\uparrow}-d_{\downarrow}^{\dagger}d_{\downarrow}) + \sum_{k,\alpha,\sigma} V_{\alpha}\{\psi_{k\alpha\sigma}^{\dagger}d_{\sigma}+\mathrm{H.c.}\}, \tag{68}$$

which is studied below. Here, $\psi_{k\alpha\sigma}^{\dagger}$ creates an electron with wave number k and spin projection σ on lead α $(\alpha=L,R)$, while $d_{\sigma}^{\dagger}$ creates a localized electron on the level.

Allowing for slow periodic modulation of H and V_{α} in Eq. (68), we extract the adiabatically pumped spin and charge along a closed pumping cycle analogous to the one shown in Fig. 2. For a generic trajectory $\mathcal{C}$ in the parameter space (X_1,X_2) defined below, the adiabatically pumped spin and charge are given for $\mathcal{H}_{\mathrm{RLM}}$ by the standard Brouwer formula

$$\langle S\rangle = \hbar\sum_{\sigma}\sigma\int_{-\infty}^{\infty}\frac{d\epsilon}{4\pi i}f'(\epsilon)\int_{\mathcal{A}}dX_1dX_2[\partial_{X_1}\mathbf{S}_{\sigma}\partial_{X_2}\mathbf{S}_{\sigma}^{\dagger} - \partial_{X_2}\mathbf{S}_{\sigma}\partial_{X_1}\mathbf{S}_{\sigma}^{\dagger}]_{LL} \tag{69}$$

and

$$\langle Q\rangle = -e\sum_{\sigma}\int_{-\infty}^{\infty}\frac{d\epsilon}{2\pi i}f'(\epsilon)\int_{\mathcal{A}}dX_1dX_2[\partial_{X_1}\mathbf{S}_{\sigma}\partial_{X_2}\mathbf{S}_{\sigma}^{\dagger} - \partial_{X_2}\mathbf{S}_{\sigma}\partial_{X_1}\mathbf{S}_{\sigma}^{\dagger}]_{LL}. \quad (70)$$

Here, $\sigma=\uparrow,\downarrow$ and $\sigma=\pm 1$ are used interchangeably to label the spin projection. The domain of integration, $\mathcal{A}$, is the (oriented) area in parameter space enclosed by the contour $\mathcal{C}$. The instantaneous S matrix pertaining to $\mathcal{H}_{\mathrm{RLM}}$ is written in the $L-R$ basis as

$$\mathbf{S}_{\sigma}(\epsilon) = \begin{bmatrix} 1-2i\Gamma_L G_{\sigma}^{r}(\epsilon) & -2i\sqrt{\Gamma_L\Gamma_R}G_{\sigma}^{r}(\epsilon) \\ -2i\sqrt{\Gamma_L\Gamma_R}G_{\sigma}^{r}(\epsilon) & 1-2i\Gamma_R G_{\sigma}^{r}(\epsilon) \end{bmatrix}, \quad (71)$$

where

$$G_{\sigma}^{r}(\epsilon) = \frac{1}{\epsilon-\sigma\mu_B g_i H + i\Gamma_+} \quad (72)$$

is the associated dot Green function. Here, $\Gamma_+=\Gamma_L+\Gamma_R$ with $\Gamma_{\alpha}=\pi\rho_0 V_{\alpha}^2$ is the resonance width, which plays the role of the Kondo temperature in the Kondo model.

By analogy with the cycle of Fig. 2, we vary the two pumping parameters $X_1=\Gamma_L-\Gamma_R$ and $X_2=\mu_B g_i H$ while Γ_+ is held fixed. As before, the cycle is composed of four segments, two in which X_1 is tuned from $\pm\Gamma_+$ to $\mp\Gamma_+$ while X_2 is kept fixed and two in which X_2 is tuned from $\pm h$ to $\mp h$ while X_1 is held fixed. Using Eqs. (69)–(72) for this cycle, one obtains $\langle Q\rangle=0$ and

$$\langle S\rangle = -\frac{2\hbar}{\pi}\int_{-\infty}^{\infty}d\epsilon f'(\epsilon)\left[\mathrm{Re}\left\{\frac{h\Gamma_+}{h^2+(\Gamma_+-i\epsilon)^2}\right\} + \arctan\left(\frac{h+\epsilon}{\Gamma_+}\right)\right]. \quad (73)$$

Both results are identical to those obtain at the Toulouse limit, provided Γ_+ is identified with T_K. Thus, the physical picture underlying Eq. (66) is consistent with that of simple resonant elastic scattering, where a single resonance is symmetrically split about the Fermi energy by an applied magnetic field.

From a theoretical standpoint, it is clear that one can realize a quantized spin pump using either a quantum dot in the Kondo regime or a Zeeman-split single-particle resonance that is tuned to the Fermi energy. However, practical considerations make the Kondo-dot scenario a more promising candidate for the realization of such a device. Indeed, modulation of the couplings to the two leads is typically accompanied in real devices by a capacitive shift of the dot level. In the case of a simple resonance, the induced modulation of the dot level will generally produce a finite charge current and is likely to spoil the quantization of the pumped spin. The Kondo-dot scenario is immune to such fluctuations, as these produce only a tiny shift of the Abrikosov-Suhl resonance. Indeed, as discussed in Sec. III, charge transport is strictly forbidden as long as the Coulomb-blockaded dot can be described in terms of a pure Kondo Hamiltonian having no potential scattering. Although a realistic description of quantum dots generally requires the inclusion of potential scattering, the latter term can be made negligibly small by operating the device deep in the Kondo regime. In this manner, charge transport can be excluded.

VI. CONCLUSIONS

In this paper, we have presented an exact analysis of adiabatic quantum pumping through a quantum dot in the Kondo regime. It follows from general symmetry arguments that the instantaneous charge current is strictly zero in the absence of potential scattering and for zero voltage bias. A similar statement applies to the symmetrized spin current either in the absence of an applied magnetic field or for symmetric coupling to the leads. Pumping of a spin current therefore requires both a finite magnetic field and for left-right symmetry to be simultaneously broken. Both conditions are readily met in practical devices, making ultrasmall quantum dots a natural candidate for the realization of a spin battery.

To quantify this statement, we have computed the pumped spin current exactly at the Toulouse limit. Exploiting the mapping onto a quadratic Hamiltonian and performing a controlled expansion in the small parameter Ω/T_K (Ω being the characteristic modulation frequency, T_K is the Kondo temperature), we have expressed the pumped spin per cycle as a geometric property of the scattering matrix associated with three flavors of Majorana fermions, which are the effective degrees of freedom at the Toulouse limit. In particular, employing the coupling to the leads as one pumping parameter and the applied magnetic field as another, we have shown that one can devise pumping cycles that realize a pure quantized spin pump, namely, a device for which the average spin pumped per cycle is closely equal to $\hbar$ but where no accompanying charge current is produced. We expect the pumped spin per cycle to remain nearly quantized in real quantum dots provided that one operates at $T\ll T_K$.

There have been by now a number of different proposals in the literature for the realization of spin pumps, employing diverse setups such as chaoticity in quantum dots,[15] ferromagnetic leads,[17] spin-orbit interactions,[16,19] classical turnstile cycles,[18] one-dimensional Luttinger-liquid physics,[14] and, finally, the Kondo effect in quantum dots.[13] While all these proposals reported schemes to realize a pure adiabatic spin pump along specific cycles, the quantization of the spin pumped per cycle has been shown to be the case only in the classical turnstile setup[18] and for a Luttinger liquid.[14] In contrast to Ref. 18, the pumping scheme investigated in this paper offers an interesting possibility to realize a *coherent* quantized spin pump, in which the absence of charge current is essentially warranted along all possible cycles (including beyond the adiabatic limit).

The quantization of the pumped spin per cycle reported in this paper is subject to small deviations as the temperature T becomes of order T_K or as the magnetic-field excursion is altered. Moreover, it applies only to the *average* spin pumped per cycle. In order to better characterize such a quantum pump, a detailed study of its noise properties (and full counting statistics) is desirable. A study of the statistical properties of the Kondo pump is a challenge left for future work.

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