Perturbative expansion of the magnetization in the out-of-equilibrium Kondo model

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This paper is concerned with the out-of-equilibrium two-lead Kondo model, considered as a model of a quantum dot in the Kondo regime. We revisit the perturbative expansion of the dot's magnetization, and conclude that, even at order 0 in the Kondo interactions, the magnetization is not given by the usual equilibrium result. We use the Schwinger-Keldysh method to derive a Dyson equation describing the steady state induced by the voltage between the two leads, and thus present the correct procedure for calculating perturbative expansions of steady-state properties of the system.

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I. INTRODUCTION

In recent years, much experimental and theoretical work has been devoted to exploring the properties of so-called ''quantum dots.''[1] These are mesoscopic devices in which a ''dot'' containing a small number of electrons is isolated from two macroscopic leads (denoted ''left'' and ''right'') by potential barriers, through which tunneling processes take place. Experimentally, these systems are small devices fabricated using a two-dimensional electron gas[2–5] or carbon nanotubes.[6] The number of electrons on the dot, N, is controlled by a gate voltage V_g. From the experimental point of view, one is primarily interested in the current flowing through the dot as a function of V_g and of the potential difference V between the two leads. When tunneling is weak enough, the Coulomb blockade phenomenon appears:[1] the conductance through the dot is essentially zero except in the vicinity of certain special values of V_g, where the energy difference between the ground states of the dot with N and $N+1$ electrons vanishes. At these points, conductance peaks are observed.

A simple model for such a system is the Anderson model, where the localized level represents the dot and the hopping term describes its hybridization with the leads. In this paper, we shall restrict ourselves to the regime where the occupation of the dot is not fluctuating, and where naïve application of Coulomb blockade ideas would predict a strongly suppressed conductance. However, when N is odd there is exactly one unpaired spin, which is coupled to the leads via a Kondo interaction. In this case, in the linear response regime, spin physics opens up a new transmission channel via the Kondo effect. Although the occupancy of the dot remains fixed, spin-flip interactions permit the formation of strong dot-lead hybridization for temperatures $T<T_K$, where T_K is the Kondo temperature. As the temperature approaches zero, this leads to unitary limit conductance ($G=G_0\equiv 2e^2/h$) via a sharp resonance at the Fermi surface—the Abrikosov-Suhl resonance. This effect was predicted in the context of quantum dots fourteen years ago,[7,8] and was recently observed in a series of experiments.[2–6]

By contrast, in the large-voltage regime, full nonequilibrium calculations are required, and much less is known than close to equilibrium. The out-of-equilibrium Anderson and Kondo models have been studied by various methods in the last decade. Much of this work[9–11] has concentrated on the noncrossing approximation approach, which adopts a slave-boson description of the problem, and then renders it tractable by neglecting certain vertex corrections. This yields a picture in which the Kondo resonance in the density of states is both split and broadened as the voltage V between the leads is increased. Recent work[12] has explored a new approach, where one attempts to use the Bethe ansatz results for the Anderson model to construct a Landauer-type picture of transport through the dot. This approach also involves approximation, when one comes to construct the ''in'' and ''out'' scattering states from the dressed excitations of the model. Another thread[13,14] has involved studying the Anderson model via perturbation theory in the on-site Coulomb repulsion U. While these works provide approximate information on the behavior of the current-voltage characteristic, they shed little light on the nature of the many-body state of the system when $V>T_K$.

In particular, a basic question recently debated[11,15] is whether the Kondo problem has a strong coupling regime at low temperature and high voltage. In discussing this point, a previous paper[15] used a second-order perturbative expression for the magnetic susceptibility in the out-of-equilibrium steady state induced by V. This putative result was however incorrect, even at order 0 in J, the strength of the Kondo couplings; in this paper, we correct this result at order 0, discuss the related physics and present a systematic method for calculating higher-order corrections in J. Our main conclusion is that, even at order 0 in J, the Keldysh function of the spin deviates from its equilibrium value and therefore the steady-state magnetization of the dot is not given by $M_{\rm eq} \equiv \frac{1}{2}\tanh(B/2T)$. Rather, it must be computed by solving a transport equation. We emphasize that this issue is not directly related to the so called ''decoherence time''[11] but is a basic point about perturbation theory to be addressed before discussing out-of-equilibrium renormalization group equations and the existence of a strong coupling regime at large voltage. In particular, it has ramifications for other physical quantities, e.g., the current and the current-current correlation function in a magnetic field (see Sec. III C 2).

The paper is organized as follows. In Sec. II, we present the model, our main result [the magnetization at order 0 given by Eq. (4)], and the associated physical discussion. In Sec. III, we present a detailed pedagogical derivation of Eq. (4) using the Keldysh method. Finally, in Appendix D, we present more details about the computation presented in Ref. 15, and explain why it was incorrect.

II. RESULTS AND DISCUSSION

Our starting point is the Hamiltonian of the two-lead Kondo model. For a discussion of the modeling of the quantum dot, and for a derivation of this model from the Anderson model via an out-of-equilibrium Schrieffer-Wolff transformation, we refer the reader to the literature, in particular Ref. 16 and references therein. The Hamiltonian is

$$H=\sum_{\alpha\mathbf{k}\sigma}(\varepsilon_{\mathbf{k}}+\mu_\alpha)c^\dagger_{\alpha\mathbf{k}\sigma}c_{\alpha\mathbf{k}\sigma}+H_{\rm refl}+H_{\rm trans}$$
$$-B_sS_z-B_c\sum_{\alpha\mathbf{k}\sigma}\sigma c^\dagger_{\alpha\mathbf{k}\sigma}c_{\alpha\mathbf{k}\sigma},$$
$$H_{\rm refl}=\left(J_R\sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma'}(c^\dagger_{R\mathbf{k}\sigma}\vec{\sigma}_{\sigma\sigma'}c_{R\mathbf{k}'\sigma'})\vec{S}\right)+(R\leftrightarrow L),$$
$$H_{\rm trans}=\left(J_{RL}\sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma'}(c^\dagger_{R\mathbf{k}\sigma}\vec{\sigma}_{\sigma\sigma'}c_{L\mathbf{k}'\sigma'})\vec{S}\right)+(R\leftrightarrow L), \tag{1}$$

where $c^\dagger_{\alpha\mathbf{k}\sigma}$ creates an electron in lead $\alpha\in\{L,R\}$ with momentum $\mathbf{k}$ and spin σ, and J_L, J_R and $J_{LR}=(J_{RL})^*$ are Kondo coupling constants between the electrons and the spin of the dot $\vec{S}$. The first term in H describes the electrons in the leads, with $\varepsilon_{\mathbf{k}}$ being the bare energy of an electron of momentum $\mathbf{k}$ at zero voltage (the same for each lead) and μ_α is the potential in the lead α. Each lead consists of a free electron gas with a density of states $\rho(\epsilon)$ of bandwidth D: ultimately, we will be interested in the result in the large bandwidth ($D\to\infty$) limit, but the computations are first performed for finite D. We will make the physical assumption that the leads are in thermal equilibrium at a temperature T. The voltage is applied by taking the chemical potentials of the two leads to be different, $\mu_L-\mu_R=V$. $H_{\rm refl}$ describes regular Kondo processes, where an electron from a given lead is spin-flip scattered back into the *same* lead; $H_{\rm trans}$ describes ''spin-flip cotunneling,'' where an electron from one lead is spin-flip scattered into the *other* lead. If the model (1) is derived from the Anderson model, one finds that $J_RJ_L=|J_{RL}|^2$. In this paper, however, we relax this relation between the coupling constants and treat them as independent parameters. Finally, the last terms represent the coupling to the magnetic field. We allow two different magnetic fields, B_s for the spin and B_c for the lead electrons; this permits the calculation of the total and local spin susceptibilities within the same computation.

In this paper, we shall be interested in the values taken by the following quantities in the nonequilibrium steady state induced by the voltage V: the dot magnetization $M_{\rm dot}$, the magnetization of the leads $M_{\rm leads}$, the total magnetization $M_{\rm tot}$, and the total and local susceptibilities. These are given by

$$M_{\rm dot}(B_s,B_c)=\langle S_z\rangle, \tag{2a}$$
$$M_{\rm leads}(B_s,B_c)=\left\langle\sum_{\alpha\mathbf{k}\sigma}\sigma c^\dagger_{\alpha\mathbf{k}\sigma}c_{\alpha\mathbf{k}\sigma}\right\rangle, \tag{2b}$$
$$M_{\rm tot}(B)=M_{\rm dot}(B,B)+M_{\rm leads}(B,B)-M_{\rm Pauli}, \tag{2c}$$
$$\chi_{\rm tot}=\left.\frac{\partial M_{\rm tot}(B)}{\partial B}\right|_{B=0}, \tag{2d}$$
$$\chi_{\rm loc}=\left.\frac{\partial M_{\rm dot}(B_s,B_c=0)}{\partial B_s}\right|_{B_s=0}, \tag{2e}$$

where angular-brackets $\langle\cdots\rangle$ denote an expectation value taken in the steady (i.e., long-time) state of the system. $M_{\rm Pauli}$ is simply the Pauli paramagnetic contribution from the lead electrons which would be present even in the absence of the impurity, and which we therefore exclude from $M_{\rm tot}$. We consider the perturbative expansions of these steady state quantities; more precisely, we define

$$J_R\equiv\theta_RJ,\quad J_L\equiv\theta_LJ,\quad J_{RL}\equiv\theta_{RL}J \tag{3}$$

and we let J go to zero while keeping the coefficients θ_R, θ_L, and θ_{RL} fixed. In the following, the expression ''order n'' refers to the order n of this expansion in J.

Our main result is the order 0 term of the perturbative expansion of the magnetization

$$M_{\rm tot}(B)=M_{\rm dot}(B,B_c)+O(J)=\frac{1}{2}\tanh\left(\frac{B}{2T}\right)\frac{\varphi\left(\frac{B}{T}\right)\left(1+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}\right)}{\frac{1}{2}\left[\varphi\left(\frac{B+V}{T}\right)+\varphi\left(\frac{B-V}{T}\right)\right]+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}\varphi\left(\frac{B}{T}\right)}+O(J), \tag{4}$$

where φ is defined by

$$\varphi(x) \equiv \frac{x}{\tanh\left(\frac{x}{2}\right)}. \tag{5}$$

[At this order, $M_{\rm dot}(B,B_c)$ does not depend on B_c.] As a result, the magnetic susceptibility at order 0 is

$$\chi(T,V) = \frac{1}{4T} \frac{1+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}}{\frac{1}{2}\varphi\left(\frac{V}{T}\right)+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}} + O(J), \tag{6}$$

and in particular for $|V|\to\infty$,

$$\chi(T,V) \sim \frac{1+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}}{2|V|}. \tag{7}$$

A striking feature of Eq. (4) is that the magnetization at order 0 in J is not given by the equilibrium expression $M_{\rm eq} = \frac{1}{2}\tanh(B/2T)$. This may seem surprising: since the couplings to the leads are relaxed to zero, why should we not find $M_{\rm eq}$, the magnetization of a free spin? Physically, the answer is that $M_{\rm eq}$ is not the "magnetization of a free spin," but rather the magnetization of a spin weakly coupled to an *equilibrium* bath. On the other hand, Eq. (4) gives the magnetization of a spin weakly coupled, via the Hamiltonian (1), to two leads with different chemical potentials. It is simply an out-of-equilibrium extension of the Curie law. In particular, we emphasize that the finite susceptibility at $T=0$ cannot be interpreted as a renormalization (or a screening) of the spin due to the voltage.

At long times, the state of the spin is completely determined by the properties of these leads, and hence so is its distribution function, which describes the population of its two states as a function of temperature, voltage, and magnetic field. Since the whole system is not in equilibrium, this steady state is not described by the Gibbs distribution; in particular, the fluctuation-dissipation theorem (FDT) need not hold. Hence the magnetization need not be (and is not) $M_{\rm eq}$ at order 0 in J. Rather it should be computed by solving a transport equation in the steady-state regime, i.e., a quantum Boltzmann equation. (At dominant order this procedure is equivalent to using a semi-classical master equation; see Appendix A.)

The crucial point is that J_R, J_L, J_{RL} are relaxed to zero, *assuming that they are still bigger than the coupling of the spin to any other thermal bath*. If we were to take into account such a coupling (denoted by J_0) then the result would cross over to $M_{\rm eq}$ when $J_R, J_L, J_{RL} \ll J_0$ (all the couplings going to zero while maintaining fixed ratios). In fact, Eq. (4) implies that the equilibrium value is only an upper bound: $0 \leqslant M_{\rm tot}(J=0)/M_{\rm eq} \leqslant 1$, which follows from the convexity of φ. This bound is saturated only in equilibrium, i.e., for $\theta_{RL}\to 0$ or $V\to 0$.

Moreover, as expected on physical grounds, the nonequilibrium result is much less universal than the equilibrium one. In equilibrium, the magnetization of a spin weakly coupled to a bath depends neither on the properties of the bath except the temperature T nor on the form of the couplings between the spin and the bath. Neither of these statements holds true for the nonequilibrium Kondo model: $M_{\rm dot}$ (at order 0) depends not only on T but also on the voltage V and on a ratio involving the parameters θ_R, θ_L, and θ_{RL}. Thus $M_{\rm dot}$ is perturbative in J, but not in the three variables J_R, J_L, J_{RL}, and in particular it is not analytic in J_R, J_L, J_{RL} around $(0,0,0)$. One can find such a dependence on the ratio of couplings even in a simple free model (a single level coupled to two leads), as illustrated in Appendix B. Furthermore, it should be remembered that we computed $M_{\rm dot}$ with free leads: introducing interactions in the leads would change the function φ (the important quantity being the electron-electron bubble in the leads). For example, we expect a different result to hold for a spin coupled to Luttinger liquids, even at order 0.

It should be noted that the result (4) gives a nontrivial expression for the magnetization even at zero temperature. Since $\phi(x)\to|x|$ as $|x|\to\infty$, we find that

$$M_{\rm tot}(B,V)\big|_{T=0} = \frac{B}{2}\left(\frac{1+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}}{\frac{1}{2}(|B+V|+|B-V|)+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}|B|}\right)+O(J). \tag{8}$$

We then have two cases (we can take $B,V>0$): for $B>V>0$, we obtain the equilibrium result $M_{\rm tot}(B,V)=\mathrm{sgn}(B)/2+O(J)$, but for $0<B<V$, the magnetization is a still a function of B/V,

$$M_{\rm tot}\big|_{T=0} \approx \frac{B}{2}\left(\frac{1+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}}{|V|+\frac{\theta_R^2+\theta_L^2}{2\theta_{RL}^2}|B|}\right), \tag{9}$$

in agreement with Eq. (7) in the limit $|V|\gg|B|$. Physically, there are two sources of energy available to flip the spin: the thermal fluctuations of both baths (represented by T) and the fact that an electron can go from L to R and give to the spin an energy of order V. If we decrease B from high values ($B\gg T,V$), the spin is locked until B reaches the largest of these energy scales. Thus the magnetization at zero temperature is expected to saturate only for $B>V$. Similarly, the susceptibility is in general expected to behave as $\chi\sim 1/E$, where E is the largest energy available to flip the spin. The fact that our result is still nontrivial at zero temperature implies that it could be seen in numerical computations, such as the density matrix renormalization group (DMRG) approach of Cazalilla and Marston.[17]

III. DERIVATION OF Eq. (4)

The purpose of this section is to present the derivation of Eq. (4) and more generally the procedure for obtaining the perturbative expansion of physical quantities in the steady state. It is organized as follows: in Sec. III A, we first give an overview of the derivation; full details are given in the following sections (Secs. III B and III C) and in the appendixes, including a presentation of the Keldysh method.

A. Overview

When doing perturbation theory in the steady state, there are two important small couplings: J, the strength of the Kondo couplings, and s, the small regulator that appears in Green's functions such as the ''spin'' retarded function (defined in Sec. III B 3),

$$R_B^0(\omega)=\frac{1}{\omega-B+is}. \tag{10}$$

The scale s should be thought of as being due to the coupling to an auxiliary thermal bath, which in the physical system would be the substrate. If the impurity is coupled to such a bath whose density of states is ρ_2 by a coupling g, one finds $s\sim g^2\rho_2$ (in the large bandwidth limit). In the physical quantum dot system, $J\gg s$, meaning that the correct order of limits to take is $s\to 0$ followed by $J\to 0$, as pointed out in Sec. II.

However, ''straightforward'' perturbation theory in J takes the limit in the opposite order: one first expands in J while keeping s finite, and only then takes $s\to 0$ term by term in the perturbation series. In equilibrium, these two limits commute, but out of equilibrium they do not. This is explicitly shown in Sec. III C 1; the out-of-equilibrium Keldysh Green's function is not analytic around $(s,J)=(0,0)$. This nonanalytic behavior is quite generic in situations where an impurity is coupled to several leads, and may be seen even in a simple free model (see Appendix B). The signature of that noncommutativity is that ''straightforward'' perturbation theory fails: its terms [starting at $O(J^2)$] exhibit divergences of a $1/s$ form. These are similar to the infrared divergences in equilibrium perturbation theory that signal an incorrect choice of reference state. As shown below, $1/s$ divergences at order J^2 signal an incorrect choice for the Keldysh Green's function at order 0, or equivalently of the distribution function which describes the nonthermal population of the two levels of the spin. Consequently, contrary to the claims of Ref. 15 there is no possibility of regulating these $1/s$ divergences order by order in J. We discuss the putative regulation procedure of Ref. 15 in Appendix D, and explain why it is incorrect.

The solution is to begin with the Dyson equation in the steady state, considered as a functional equation for the full Green's function G, using the skeleton self-energy diagrams (see Appendix E for further details). After taking the $s\to 0$ limit in this equation, a perturbative expansion for G may be inserted into it, and a solution obtained order by order in J. Solving the Keldysh component of this Dyson equation is equivalent to solving the quantum Boltzmann equation; as shown below, the solution is nonthermal even at order 0 in J. Another procedure for solving the problem of $1/s$ divergences was proposed in Ref. 18 in the context of the $U=\infty$ out-of-equilibrium Anderson model: its method was to *choose* the zeroth-order spin Keldysh function (see Sec. III B 3) in order precisely to cancel the divergences at order J^2. This is completely equivalent to our approach, as explained in Sec. III C 2.

Finally, we note that one may give a simple semiclassical derivation of the out-of-equilibrium result (4) based on a master equation. This derivation is due to Glazman and Kaminski,[19] and is presented in Appendix A. The success of such a semiclassical approach (at this lowest order) is related to the fact that one can compute the equilibrium magnetization of a free quantum spin using a classical Ising model. In fact, at this order, the semiclassical master equation is strictly equivalent to the Keldysh component of the steady-state Dyson equation, so the two apparently disparate derivations yield the same result.

B. Technical preliminaries: the Keldysh method

Let us now turn to the technical details. In the following, for simplicity, we will write some equations for a generic fermionic field ψ, which will be specialized afterwards to the fields representing the electrons and the spin.

1. Generalities

The basic idea of the nonequilibrium Keldysh method[20–22] consists in taking the system at an initial time $t=0$ in an initial state described by a density matrix ρ_0 and letting the system relax, using the Hamiltonian evolution given by H, to a long-time regime. In order to ensure that the system relaxes, it may be necessary to add some additional coupling terms to the Hamiltonian, in particular to break conservation laws: see the discussion in Appendix E. Depending on the system, the long-time regime can be an equilibrium state, a nonequilibrium steady state, a non-time-translation-invariant steady state, or even an aging regime (in glassy systems). In the quantum dot problem, we assume that the system reaches at finite voltage V a nonequilibrium steady state, in which we want to compute physical quantities.

A ''Keldysh'' average of any quantity A is defined by

$$\langle A(t)\rangle_{\mathcal{K}}\equiv\langle e^{iHt}Ae^{-iHt}\rangle_0\equiv\mathrm{Tr}(\rho_0 e^{iHt}Ae^{-iHt}), \tag{11}$$

where $\langle\cdots\rangle_0$ is the average taken using the *initial* density matrix of the system. The steady-state average is given by

$$\langle A\rangle=\lim_{t\to\infty}\langle A(t)\rangle_{\mathcal{K}}.$$

Using the usual representation of the evolution operator e^{-iHt} as a T-ordered exponential in the interaction picture (and the anti-T-ordered one for e^{iHt}), one can obtain an expansion in the coupling constant J. Following the usual conventions, it is convenient to keep track of the two exponentials using a closed time contour, running from 0 to $+\infty$ and back to 0 (Ref. 21): we denote by $+$ the upper contour (from

0 to ∞), which arises from expanding e^{-iHt}, and by $-$ the lower contour. In accordance with this notation, we define the four Green's functions,

$$G_{\psi}^{++}(t,t') \equiv -i\langle T\psi(t)\psi^{\dagger}(t')\rangle_{\mathcal{K}},$$

$$G_{\psi}^{-+}(t,t') \equiv -i\langle \psi(t)\psi^{\dagger}(t')\rangle_{\mathcal{K}},$$

$$G_{\psi}^{--}(t,t') \equiv -i\langle \tilde{T}\psi(t)\psi^{\dagger}(t')\rangle_{\mathcal{K}},$$

$$G_{\psi}^{+-}(t,t') \equiv i\langle \psi^{\dagger}(t')\psi(t)\rangle_{\mathcal{K}}. \tag{12}$$

Here T is the time-ordering operator, $\tilde{T}$ the anti-time-ordering operator, and ψ is any *fermionic* field. The two indices of the matrix G will be called "indices in Keldysh space." In the following, Eqs. (12) will be summarized with the notation

$$G_{\psi}(t,t') = -i\langle\langle \psi(t)\psi^{\dagger}(t')\rangle\rangle_{\mathcal{K}} \tag{13}$$

and G will always denote a 2×2 Keldysh matrix.

The Green's functions (12) are not independent but can be expressed as functions of the retarded, advanced, and Keldysh Green's functions defined, respectively, by[29]

$$R_{\psi}(t,t') \equiv -i\,\theta(t-t')\langle\{\psi(t),\psi^{\dagger}(t')\}\rangle_{\mathcal{K}},$$

$$A_{\psi}(t,t') \equiv i\,\theta(t'-t)\langle\{\psi(t),\psi^{\dagger}(t')\}\rangle_{\mathcal{K}},$$

$$K_{\psi}(t,t') \equiv -i\langle[\psi(t),\psi^{\dagger}(t')]\rangle_{\mathcal{K}}, \tag{14}$$

as is shown from the transformation[22]

$$G_{\psi} \equiv \begin{pmatrix} G_{\psi}^{++} & G_{\psi}^{+-} \\ G_{\psi}^{-+} & G_{\psi}^{--} \end{pmatrix}, \quad \tilde{G}_{\psi} \equiv \begin{pmatrix} R_{\psi} & K_{\psi} \\ 0 & A_{\psi} \end{pmatrix},$$

$$\tilde{G}_{\psi} = \frac{1}{2}\begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix} G_{\psi} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix}. \tag{15}$$

We will denote the first set of Green's functions (12) "the $\pm$ basis," and the second (14) the "Larkin-Ovchinnikov (LO) basis."

There are further relations between these Green's functions. In general, $R_{\psi}(t,t') = [A_{\psi}(t',t)]^{*}$. Moreover, in equilibrium FDT reads:

$$K_{\psi}(\omega) = h_{\mathrm{eq}}(\omega)[A_{\psi}(\omega) - R_{\psi}(\omega)], \quad h_{\mathrm{eq}}(\omega) \equiv -\tanh\left(\frac{\omega}{2T}\right), \tag{16}$$

thus the retarded Green's function is the only remaining independent Green's function, and so contains all the information about the state of the system. Out of equilibrium, however, $K_{\psi}(\omega)$ and $R_{\psi}(\omega)$ should be independently determined.

In order to write the diagrammatic expansion, we also define the "bare" counterparts of the Green's functions in Eqs. (12) and (14) by

$$G_{\psi 0}(t,t') \equiv -i\langle\langle \psi_{\mathrm{int}}(t)\psi_{\mathrm{int}}^{\dagger}(t')\rangle\rangle_{0},$$

where the field ψ_{int} is in the interaction picture. From the expansions of the evolution operators, we obtain the diagrammatic expansion, provided that $\langle\cdots\rangle_0$ satisfies Wick's theorem. For the problem at hand, we take as the initial condition the density matrix of the model with $J_R = J_L = J_{RL} = 0$ and for the spin a finite s (arising from the coupling of the spin to a thermal bath; see Sec. III A). Going to the long time limit, every function becomes a function of the difference of the times, and transforming to Fourier space we obtain standard Feynman rules.

2. Lead electrons

Let us begin with the lead electrons. We care here only about the local Green's functions, so we will drop the spatial indices. We denote by $(\mathbf{G_c})_{\alpha\sigma,\beta\sigma'}(t,t')$ the Green's function describing the creation in lead β of an electron with spin σ' at time t', and a corresponding annihilation in lead α of an electron with spin σ at time t,

$$(\mathbf{G_c})_{\alpha\sigma,\beta\sigma'}(t,t') = -i\langle\langle c_{\alpha\sigma}(t)c_{\beta\sigma'}^{\dagger}(t')\rangle\rangle_{\mathcal{K}}. \tag{17}$$

(Here the indices $\alpha,\beta \in \{L,R\}$, while $\sigma,\sigma' = +,-$, and the local electron operators are defined by $c_{\alpha\sigma} \propto \Sigma_{\mathbf{k}} c_{\alpha\mathbf{k}\sigma}$.) The Green's function $\mathbf{G_c}$ is thus *a priori* a 4×4 matrix (in lead-spin space), whose entries are themselves 2×2 Keldysh matrices. In the following, a bold notation with subscript c always designates such a 4×4 matrix. The bare Green's function $\mathbf{G_{c0}}$ is, however, diagonal. The bare (diagonal) density of states is

$$(\boldsymbol{\rho}_{\mathbf{c}\,\mathbf{0}})_{\alpha\sigma,\beta\sigma'} = \rho(\omega + \sigma B_c - \mu_{\alpha})\,\delta_{\alpha\beta}\delta_{\sigma\sigma'}$$

(the energy levels are the same in both leads, but shifted by the Zeeman energy and the voltage). We assume that the baths, being much bigger than the impurity, are permanently in thermal equilibrium so that the bare electrons' functions read

$$\mathbf{R_{c0}}(\omega) = \int d\epsilon \frac{\boldsymbol{\rho}_{\mathbf{c0}}(\epsilon)}{\omega - \epsilon + i0^{+}}, \tag{18a}$$

$$\mathbf{K_{c0}}(\omega) = 2i\pi \mathbf{h_{c0}}(\omega)\boldsymbol{\rho}_{\mathbf{c0}}(\omega), \tag{18b}$$

$$(\mathbf{h_{c0}})_{\alpha\sigma,\beta\sigma'} \equiv h_{\mathrm{eq}}(\omega - \mu_{\alpha})\,\delta_{\alpha\beta}\delta_{\sigma\sigma'}, \tag{18c}$$

where the first two equations are matricial, and μ_{α} is the potential of the lead $\alpha = R,L$. The voltage difference between the leads is given by $V = \mu_L - \mu_R$.

3. Spin

Since the spin operator is not appropriate for diagrammatic computations (it does not satisfy Wick's theorem), we represent the spin 1/2 by three Majorana fermions $\eta^a, a \in \{x,y,z\}$ which satisfy Wick's theorem and the relations

$$S^a = -\frac{i}{2}\epsilon^{abc}\eta^b\eta^c, \tag{19a}$$

$$(\eta^a)^{\dagger} = \eta^a, \tag{19b}$$

$$\{\eta^a, \eta^b\} = \delta^{ab}. \tag{19c}$$

Using Eq. (19a), one can easily show that $\vec{S}$ satisfies the correct commutation relations and that $\vec{S}^2 = 3/4$. Note that this last constraint is *automatically* satisfied, unlike in the case of a Dirac fermion representation, where a Lagrange multiplier would have been required to fix the magnitude of the spin: the Majorana representation therefore makes the computation simpler. In this paper, we consider only spin 1/2, but our computations could be extended to higher spin, provided that one used another representation for $\vec{S}$.

Let us now discuss the propagators G_{ab} $(a,b \in \{x,y,z\})$ of the η, in the presence of a magnetic field B_s along the z direction. The general form is

$$\mathbf{G} = \begin{pmatrix} G_{xx} & G_{xy} & 0 \\ G_{yx} & G_{yy} & 0 \\ 0 & 0 & G_{zz} \end{pmatrix},$$

$$G_{ab}(t,t') \equiv -i\langle\langle \eta^a(t)\, \eta^b(t')\rangle\rangle_{\mathcal{K}}, \tag{20}$$

where the elements are 2×2 Keldysh matrices. Indeed $G_{xz} = G_{yz} = 0$ by symmetry. To prove this, note that the Hamiltonian is invariant under a π rotation around the z axis, which is implemented by $(\eta^x, \eta^y, \eta^z) \to (-\eta^x, -\eta^y, \eta^z)$ (and the corresponding rotation for the c electrons). Furthermore, making a $\pi/2$ rotation around the z axis, implemented by $(\eta^x, \eta^y, \eta^z) \to (\eta^y, -\eta^x, \eta^z)$, we obtain the relations

$$G_{xy} = -G_{yx}, \quad G_{xx} = G_{yy}. \tag{21}$$

The Hamiltonian is also invariant under a π rotation around the x axis $[(\eta^x, \eta^y, \eta^z) \to (-\eta^x, \eta^y, \eta^z)]$ together with a change of sign of the magnetic fields B_s and B_c, and hence G_{xy} and G_{xx} are respectively odd and even in the magnetic field. In particular, for $B_s = B_c = 0$, $G_{xy} = 0$. The foregoing arguments apply to the full propagator and to the free propagator G^0 (computed with only the magnetic field B_s).

A different basis is also useful: defining the Dirac fermion f and its Green's function G_B by

$$f \equiv \frac{\eta^x - i\,\eta^y}{\sqrt{2}}, \tag{22}$$

$$G_B(t) = -i\langle\langle f(t) f^\dagger(0)\rangle\rangle_{\mathcal{K}}, \tag{23}$$

we have the relations

$$G_{xx}(\omega) = G_{yy}(\omega) = \frac{1}{2}[G_B(\omega) + G_{-B}(\omega)], \tag{24a}$$

$$G_{xy}(\omega) = -G_{yx}(\omega) = \frac{i}{2}[G_B(\omega) - G_{-B}(\omega)]. \tag{24b}$$

In the (f, η^z) basis, the propagator is diagonal, so it is more convenient, for example to write the Dyson equation, whereas the original basis (η^x, η^y, η^z) is more convenient for the diagrammatics. The bare propagators in the (f, η^z) basis are given by

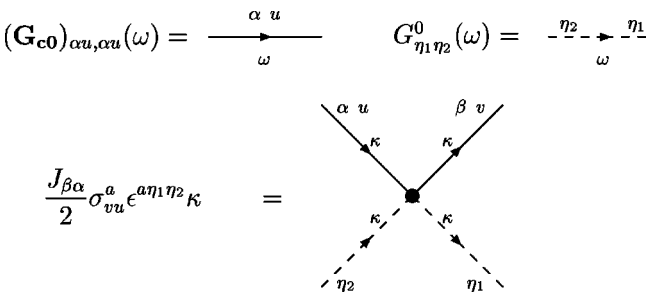

FIG. 1. Feynman rules: we represent the Majorana fermions with dashed lines and the electrons with solid lines. α, β are lead indices; u, v are spin indices for the electrons; κ is a Keldysh index $\kappa = \pm 1$; $\eta_i, a \in \{x,y,z\}$ are flavor indices for the Majorana fermion; and σ are the Pauli matrices. The vertex is given in the $\pm$ basis.

$$R^0_z(\omega) = \frac{1}{\omega + is}, \tag{25a}$$

$$K^0_z(\omega) = 2ih_{\rm eq}(\omega)\frac{s}{\omega^2 + s^2}, \tag{25b}$$

$$R^0_B(\omega) = \frac{1}{\omega - B + is}, \tag{25c}$$

$$K^0_B(\omega) = 2ih_{\rm eq}(\omega)\frac{s}{(\omega - B)^2 + s^2}, \tag{25d}$$

where s is a small regulator which, as discussed in Sec. III A, should be thought of as the width due to coupling to an auxiliary thermal bath.

4. Vertex factors and Dyson equations

The vertex factors can be extracted simply from the Hamiltonian, and the Feynman rules are summarized in Fig. 1 (in the $\pm$ basis). Note that we have oriented the Majorana fermion lines, despite the operator property that $(\eta^\dagger) = \eta$; a proof that the lines are orientable is given in Appendix C.

We now derive the expression for the Dyson equation describing the η fields. In general, a Dyson equation reads

$$\mathbf{G}^{-1}(\omega) = \mathbf{G_0}^{-1}(\omega) - \mathbf{\Sigma}(\omega), \tag{26}$$

where the inversion has to be taken in the tensor product of the x,y,z space and the Keldysh space (thus with 6×6 matrices). The free propagator is given by Eq. (25a). In the x,y,z basis, the self energy can be written as

$$\mathbf{\Sigma}(\omega) = \begin{pmatrix} \Sigma_d(\omega) & i\Sigma_a(\omega) & 0 \\ -i\Sigma_a(\omega) & \Sigma_d(\omega) & 0 \\ 0 & 0 & \Sigma_z(\omega) \end{pmatrix},$$

$$\Sigma_\mu(\omega) = \begin{pmatrix} \Sigma^R_\mu(\omega) & \Sigma^K_\mu(\omega) \\ 0 & \Sigma^A_\mu(\omega) \end{pmatrix}, \quad \mu \in \{a,d,z\}. \tag{27}$$

Using the definitions (24a), we find

$$G_{\pm B}=\begin{pmatrix} \dfrac{1}{\omega\mp B-\Sigma^R_{\pm B}(\omega)+is} & \dfrac{\Sigma^K_{\pm B}(\omega)+2ish_{\rm eq}(\omega)}{|\omega\mp B-\Sigma^R_{\pm B}(\omega)+is|^2} \\ 0 & \dfrac{1}{\omega\mp B-\Sigma^A_{\pm B}(\omega)-is} \end{pmatrix},$$

$$\Sigma_{\pm B}(\omega)\equiv\Sigma_d(\omega)\pm\Sigma_a(\omega). \tag{28}$$

C. Perturbation expansion in the steady state

After these preliminaries, let us now come back to the perturbative expansion in the steady state and develop on the points introduced in Sec. III A. In Sec. III C 1 we show that the limits $J\to 0$ and $s\to 0$ do not commute; in Sec. III C 2 we solve that problem and present the derivation of Eq. (4) together with an algorithm for computing higher-order terms in the perturbative expansion.

1. Noncommutativity of limits

We are interested in the steady-state values of the magnetizations (2a) and (2b), which can be expressed in the following way:

$$M_{\rm dot}=\frac{i}{4}\int\frac{d\omega}{2\pi}[K_B(\omega)-K_{-B}(\omega)], \tag{29a}$$

$$M_{\rm leads}=\frac{i}{2}\int\frac{d\omega}{2\pi}\sum_{\substack{\alpha=R,L\\ u=+,-}}(-1)^u(\mathbf{K_c})_{\alpha u,\alpha u}. \tag{29b}$$

Indeed, the dot magnetization is given by

$$M_{\rm dot}=\langle S^z\rangle=-\frac{i}{2}\langle[\eta^x,\eta^y]\rangle=\frac{1}{2}K_{xy}(t=0).$$

$M_{\rm leads}$ is derived analogously.

We begin with the expression for the dot magnetization derived from Eq. (29a) using the Dyson equation (28):

$$M_{\rm dot}=\frac{1}{4\pi}\int d\omega\,{\rm Im}\left(\frac{h_{\rm eq}(\omega)}{\omega-B-\Sigma^R_B(\omega)+is}\right)$$
$$-\int\frac{d\omega}{8i\pi}\frac{\Sigma^K_B(\omega)-h_{\rm eq}(\omega)[\Sigma^A_B(\omega)-\Sigma^R_B(\omega)]}{|\omega-B-\Sigma^R_B(\omega)+is|^2}$$
$$-(B\to -B), \tag{30}$$

where the last term denotes an antisymmetrization in B. The first term in Eq. (30) can be transformed into a sum over Matsubara frequencies and thus we see that the limits $J\to 0$ and $s\to 0$ commute in that term. However, the second term is more interesting. First, it vanishes in equilibrium since the numerator cancels, as required by the FDT. Moreover, using (for $\epsilon\to 0$)

$$\frac{1}{|\omega-B+i\epsilon|^2}\sim\frac{1}{\epsilon}\delta(\omega-B), \tag{31}$$

we see that in that second term these limits do not commute.

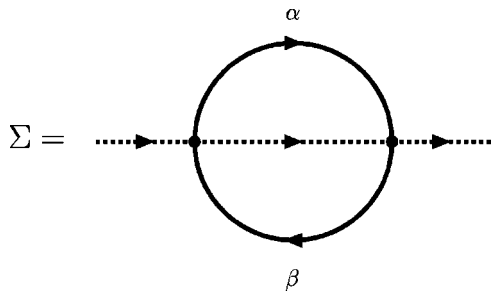

FIG. 2. Diagram of the skeleton self-energy at second order.

$J\to 0$ and then $s\to 0$. We first take $\Sigma^R_B=0$ in the denominator. We find that this second term in Eq. (30) gives a diverging term at second order in J proportional to J^2_{RL}/s. More precisely, using the expression for the self-energy at second order derived in Appendix F, and Eq. (31) with $\epsilon=s$, we find

$$M_{\rm dot}\sim\frac{J^2_{RL}}{s}\tanh\left(\frac{B}{2T}\right)\left\{\varphi\left(\frac{B}{T}\right)-\frac{1}{2}\left[\varphi\left(\frac{B+V}{T}\right)+\varphi\left(\frac{B-V}{T}\right)\right]\right\}$$

up to a finite (i.e., not diverging as $s\to 0$) term of order J^2 and to $O(J^3)$ terms [φ is defined in Eq. (5)].

$s\to 0$ and then $J\to 0$. Using Eq. (31) with $\epsilon={\rm Im}\,\Sigma^R_B(B)$ (we work at dominant order in J), we find

$$M_{\rm dot}=-\frac{1}{4}\left(\frac{\Sigma^K_B(B)}{\Sigma^A_B(B)-\Sigma^R_B(B)}\right)-(B\to -B)+O(J), \tag{32}$$

where the self-energy has to be expanded at order 2 in J [the first term in Eq. (30) cancels a part of the second term].

The second limit is the physical one (as explained in Sec. II) and it gives a formula (32) for the magnetization at order 0. However, to make use of this formula one needs to know the Keldysh $[\Sigma^K_B(B)]$ and spectral $[\Sigma^A_B(B)-\Sigma^R_B(B)]$ parts of the self-energy. The leading terms of these self-energies are of order 2 in J, but because the ratio of them is taken, they determine the magnetization at order 0. The crucial point is that Eq. (32) is in fact an implicit equation for $h_B(B)$, which appears on the left-hand side because $M_{\rm dot}$ is defined in terms of it by Eq. (29), and on the right-hand side because the self-energies at $O(J^2)$ depend on the Keldysh Green's function at $O(1)$, and hence on h_B. The most elegant way to capture this ''feedback'' effect is to reformulate the problem in terms of the perturbative expansion of the steady-state Green's function in powers of J, starting from the Dyson equation in which the $s\to 0$ limit has already been taken. We now describe this method in more detail, and specify the procedure for calculating the steady-state Green's functions perturbatively in J to arbitrary order.

2. Perturbative expansion of steady-state quantities

Let us now give a general method for computing the perturbative expansion of the Green's functions, and use it to derive Eq. (4). First, we reformulate slightly the diagrammatic expansion in terms of the full Green's function G_ψ and

of skeleton diagrams, and we explicitly derive the steady-state Dyson equation as a functional equation for G_ψ (again, for simplicity, we write some equations for a generic field ψ). The ''generic'' Dyson equation reads

$$G_\psi(J,s,\omega)=\{G_{\psi 0}^{-1}(\omega)-\Sigma_{\rm skel}[G_\psi](J,s,\omega)\}^{-1}, \quad (33)$$

where $\Sigma_{\rm skel}$ is a functional of G_ψ defined by the skeleton expansion of the self-energy (see Appendix E for a derivation). We first take the $s\to 0$ limit in Eq. (33) and then solve it order by order in J. It is, in principle, sufficient to solve Eq. (33) for G_ψ since the magnetization can be directly extracted from a Green's function, and other physical quantities (e.g., currents) are given by their skeleton expansions.

We first derive the explicit form of the Dyson equation for all fields. Defining

$$\sigma_\psi^K \equiv \frac{\Sigma_\psi^K}{2i\pi}, \quad \sigma_\psi \equiv \frac{\Sigma_\psi^A-\Sigma_\psi^R}{2i\pi}, \quad (34)$$

the full set of Dyson equations for the electrons and the Majorana fields can be rewritten explicitly as

$$\mathbf{R_c}(\omega)=\frac{1}{\mathbf{R_{c0}^{-1}}-\mathbf{\Sigma}_R(\omega)}, \quad (35)$$

$$\mathbf{K_c}(\omega)=(1-\mathbf{R_{c0}\Sigma}_R)^{-1}\mathbf{K_{c0}}(1-\mathbf{\Sigma_A A_{c0}})^{-1} + (\mathbf{R_{c0}^{-1}}-\mathbf{\Sigma_R})^{-1}\mathbf{\Sigma_K}(\mathbf{A_{c0}^{-1}}-\mathbf{\Sigma_A})^{-1}, \quad (36)$$

$$R_B(\omega)=\frac{1}{\omega-B-\Sigma_B^R(\omega)+i0^+}, \quad (37)$$

$$K_B(\omega)=h_B(\omega)[A_B(\omega)-R_B(\omega)], \quad (38)$$

$$h_B(\omega)\equiv\frac{\sigma_B^K(\omega)}{\sigma_B(\omega)}, \quad (39)$$

$$R_z(\omega)=\frac{1}{\omega-\Sigma_z^R(\omega)+i0^+}, \quad (40)$$

$$K_z(\omega)=h_z(\omega)[A_z(\omega)-R_z(\omega)], \quad (41)$$

$$h_z(\omega)\equiv\frac{\sigma_z^K(\omega)}{\sigma_z(\omega)}, \quad (42)$$

where the bold symbols are 4×4 matrices (in lead-spin space). For completeness, we have also written the definitions of the h functions. We have three blocks of equations, for the electrons, the f field, and the η^z field, respectively. Within these blocks, we have an equation for the retarded function (35),(37),(40), an equation for the Keldysh function (36),(38),(41), and for the Majorana fermions the definition of the h function (39),(42). We define h_B with Eq. (39) rather than with Eq. (38) since the spectral density is a delta peak at order 0, whereas the self-energy is a smooth function.

The spin and the lead electrons appear on a different footing: the order 0 part of the electronic Keldysh function is given by $\mathbf{K_{c0}}$ whereas the order 0 parts of h_B and h_z must be computed using Eqs. (39) and (42). Note that to obtain the order n part of h_B, one needs to compute the self-energies at order $n+2$. The method of obtaining the perturbative expansion in J order by order is as follows (denoting by $f^{(n)}$ the order n part of any function f).

(1) Assume that we have the expansion of all functions to order $n-1$.

(2) Since $\Sigma_{\rm skel}$ is at least of order 1, using Eqs. (35), (37), and (40), we compute $R_B^{(n)}$, $R_z^{(n)}$, $\mathbf{R_c}^{(n)}$, and $\mathbf{K_c}^{(n)}$.

(3) Since $\Sigma_{\rm skel}^K$ and $\Sigma_{\rm skel}''$ begin at order 2, we compute the σ^K and σ to order $n+2$, as functions of the unknowns $h_B^{(n)}$ and $h_z^{(n)}$.

(4) We then obtain closed equations for $h_B^{(n)}$ and $h_z^{(n)}$ from Eqs. (39) and (42) (expanded to order n).

Thus the order 0 part of the impurity magnetization is given by [from Eq. (29a)]

$$M_{\rm dot}=-\frac{1}{2}h_B^{(0)}(B). \quad (43)$$

The order 0 parts of R_B and K_B are

$$R_B^{(0)}=\frac{1}{\omega-B+i0^+}, \quad K_B^{(0)}=2i\pi h_B^{(0)}(B)\,\delta(\omega-B), \quad (44)$$

and the bare Green's functions of the electrons are given by Eq. (18). We compute $h_B(\omega=B)$ at order 0 by expanding the self-energies at second order (given by the diagram of Fig. 2), and then solving (39) for $h_B(B)$. Finally we find Eq. (4) in the large bandwidth limit ($D\to\infty$). The computation is presented in detail in Appendix F. This completes the computation of the Green's function to order 0.

Note that $\mathbf{K_c}$ is thermal at order 0, which expresses the fact that the leads are in thermal equilibrium. Therefore the leads' contribution to the total magnetization at order 0 is given by the Pauli term, which was explicitly excluded from $M_{\rm tot}$; hence $M_{\rm tot}=M_{\rm dot}$ at this order, as claimed in Eq. (4). Moreover, we have not written explicitly the full forms of the functions $h_B(\omega)$ and $h_z(\omega)$ to this order, since they are not required in the calculation of the zeroth-order Keldysh functions: $h_B(B)$ and $h_z(0)=0$ are sufficient, since the spectral density (at this order) is a delta peak. The full functions would, however, be needed to compute at second order; the function $h_B(\omega)$ can easily be extracted from Appendix F, and the calculation of $h_z(\omega)$ proceeds along similar lines.

Our earlier interpretation of the $1/s$ divergences in ''straightforward'' perturbation theory is borne out by this result. As stated above, the $1/s$ divergences result from an incorrect choice of zeroth-order distribution function h. If we insert the $O(1)$ part of Eq. (39) into Eq. (30) in place of $h_{\rm eq}$, we see that the divergences are cancelled at order J^2, since to this order we are now using the correct long-time distribution function. This shows that our method and that of Ref. 18 are equivalent.

It is important to note that these corrections to the zeroth-order terms in perturbation series are in no way restricted to quantities such as the magnetization. On the contrary, since what we have really calculated is the correction to the zeroth-

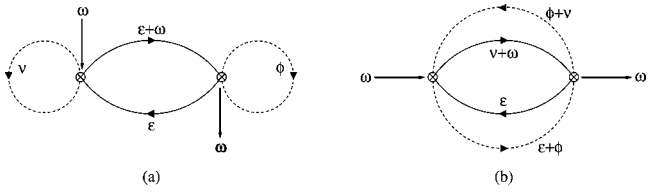

FIG. 3. The two skeleton diagrams contributing to the current-current correlator at leading order. In each diagram, one of the electrons (represented by the solid lines) is from the left lead, and the other is from the right. The vertices represent I, the current operator.

order Keldysh Green's function, they manifest themselves in many quantities. As an example, we may consider the current-current correlator: the leading terms in this quantity are of order J^2, and are calculated by inserting the zeroth-order Green's functions into the skeleton diagrams shown schematically in Fig. 3. The result is that

$$\langle I(-\omega)I(\omega)\rangle = \frac{\pi(\rho_0 J_{LR})^2}{2\hbar^2}\{[h_B^{(0)}(B)]^2 p(\omega,V) - h_B^{(0)}(B)[p(\omega+B,V)-p(\omega-B,V)] + [p(\omega,V)+p(\omega-B,V)+p(\omega+B,V)]\}, \tag{45}$$

where the function $p(x,y)$ is defined by

$$p(x,y) = \frac{x-y}{e^{\beta(x-y)}-1} + \frac{x+y}{e^{\beta(x+y)}-1}. \tag{46}$$

The result (45) is a function of $h_B^{(0)}(B)$, and is therefore clearly sensitive to the corrections made to the zeroth-order Green's function, as expected on the basis of the discussion above.

IV. CONCLUSION

The purpose of this paper has been to present the expression for the order 0 magnetization in the Kondo model out of equilibrium [Eq. (4)], and a systematic procedure for obtaining higher-order corrections to this result. The result we obtain may seem surprising, in the sense that even at order 0 in J it does not coincide with the equilibrium expression $\frac{1}{2}\tanh(B/2T)$. Indeed, the out-of-equilibrium distribution function which describes the population of the two levels of a weakly coupled spin is in general not thermal but must be computed by solving a transport equation: it is determined by the steady state into which the voltage difference forces the system. Moreover, this distribution function also enters the computation of other physical quantities (e.g., currents) and their perturbative expansions therefore exhibit similar phenomena.

Finally, we stress that the issue discussed in this paper is not directly related to the so-called ''decoherence time'' issue. Answering the question of strong coupling at finite voltage requires computations at higher orders in J. It is clear from the above, however, that the behavior of the out-of-equilibrium perturbative expansion will be markedly different from that of its equilibrium counterpart. For example, the $O(J)$ contribution to the Keldysh Green's function of the Majorana fermions depends on the $O(J^3)$ contribution to the self-energies. These, however, contain terms that diverge like $\ln D$ in the $D\to\infty$ limit, and so such logarithmic divergences may be expected to appear at $O(J)$ in some of the Green's functions, and therefore in physical properties such as the magnetization. Indeed, this phenomenon has been reported recently;[23] the full interpretation of this striking departure from equilibrium behavior merits further work.

ACKNOWLEDGMENTS

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APPENDIX A: A SEMICLASSICAL DERIVATION OF EQ. (4)

The magnetization at order 0 in J (4) can also be derived using a semiclassical master equation approach.[19] By symmetry, the reduced density matrix of the spin is diagonal, so we consider the spin as classical and characterize its state by the probability of its being up, $P_\uparrow$, or down, $P_\downarrow$. The spin dynamics is governed by a master equation,

$$\frac{dP_\uparrow(t)}{dt} = \Gamma_{\downarrow\uparrow}P_\downarrow(t) - \Gamma_{\uparrow\downarrow}P_\uparrow(t), \tag{A1a}$$

$$\frac{dP_\downarrow(t)}{dt} = \Gamma_{\uparrow\downarrow}P_\uparrow(t) - \Gamma_{\downarrow\uparrow}P_\downarrow(t), \tag{A1b}$$

where Γ_{AB} is the rate of the spin-flip process $A\to B$ induced by the Kondo terms. At second order in perturbation theory, these rates are given by

$$\Gamma_{\downarrow\uparrow} \approx \sum_{\alpha,\beta\in\{R,L\}} J_{\alpha\beta}^2 \int d\epsilon_1 d\epsilon_2 \rho(\epsilon_1 - B_c - \mu_\alpha) \times \rho(\epsilon_2 + B_c - \mu_\beta) n_F(\epsilon_1 - \mu_\alpha)(1 - n_F(\epsilon_2 - \mu_\beta)) \times \delta(\epsilon_1 - \epsilon_2 + B_s), \tag{A2a}$$

$$f(\omega, B, V_1, \sigma_1, V_2, \sigma_2) =$$

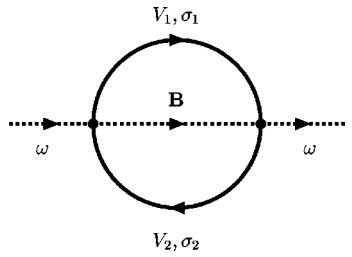


FIG. 4. Diagram of the "generic" self-energy at second order.

$$\Gamma_{\uparrow\downarrow} \approx \sum_{\alpha,\beta\in\{R,L\}} J_{\alpha\beta}^2 \int d\epsilon_1 d\epsilon_2 \rho(\epsilon_1 + B_c - \mu_\alpha) \times \rho(\epsilon_2 - B_c - \mu_\beta) n_F(\epsilon_1 - \mu_\alpha)(1 - n_F(\epsilon_2 - \mu_\beta)) \times \delta(\epsilon_1 - \epsilon_2 - B_s). \quad \text{(A2b)}$$

Taking the large bandwidth ($D \to \infty$) limit and integrating, the rates become

$$\Gamma_{\uparrow\downarrow} = (J_R^2 + J_L^2)\frac{B_s}{e^{B_s/T} - 1} + J_{RL}^2\left(\frac{B_s - V}{e^{(B_s - V)/T} - 1} + \frac{B_s + V}{e^{(B_s + V)/T} - 1}\right), \quad \text{(A3a)}$$

$$\Gamma_{\downarrow\uparrow} = (J_R^2 + J_L^2)\frac{-B_s}{e^{-B_s/T} - 1} + J_{RL}^2\left(\frac{-B_s - V}{e^{-(B_s + V)/T} - 1} + \frac{-B_s + V}{e^{-(B_s - V)/T} - 1}\right). \quad \text{(A3b)}$$

The steady-state value of the probability is given by equating the left-hand sides of Eq. (A1a) to 0, and the magnetization is given by $M_{\text{dot}} = (P_\uparrow - P_\downarrow)/2$, thus

$$M_{\text{dot}} = \frac{1}{2}\left(\frac{\Gamma_{\downarrow\uparrow} - \Gamma_{\uparrow\downarrow}}{\Gamma_{\downarrow\uparrow} + \Gamma_{\uparrow\downarrow}}\right), \quad \text{(A4)}$$

which leads to Eq. (4).

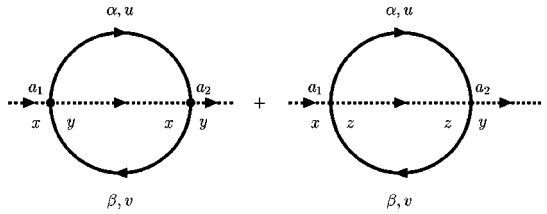
One can see that the Dyson equation in steady state, at second order in J, maps exactly to the master equation, although they appear to have different transients. So at order 0, this computation is just a reformulation of the one presented above.

APPENDIX B: A FREE MODEL

In this appendix, we recall[14,25] the solution of a simple free model in an out-of-equilibrium steady-state regime, which displays a result very similar to Eq. (4). We consider a free level coupled to some reservoirs of free electrons via hopping terms. The Hamiltonian is given by

$$H = \sum_{\alpha\mathbf{k}} (\varepsilon_{\mathbf{k}} + \mu_\alpha) c_{\alpha\mathbf{k}}^\dagger c_{\alpha\mathbf{k}} + \sum_\alpha g_\alpha (c_\alpha^\dagger d + d^\dagger c_\alpha) + \epsilon_d d^\dagger d, \quad \text{(B1)}$$

$\Sigma_{yx} =$



$\Sigma_{xx} =$

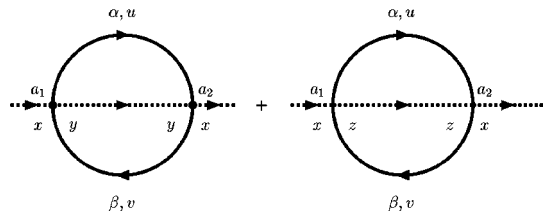


FIG. 5. Diagrams of Σ_{xx} and Σ_{xy}.

where the local reservoir-electron operator c_α is defined by $c_\alpha = c_\alpha(\mathbf{x} = \mathbf{0}) \equiv \Omega^{-1/2}\Sigma_{\mathbf{k}} c_{\alpha\mathbf{k}}$. ($\Omega$ is the usual normalization factor related to the volume of the reservoirs.) As before, we assume that the reservoirs are thermalized with the same temperature as each other, but with different chemical potentials μ_α. In particular, we are interested in a model with three reservoirs: 1 and 2 are the leads (at different potentials) and 3 an additional thermal bath (with $\mu_3 = 0$) to which the level is coupled. Since it is a Gaussian model, we can simply solve the Dyson equation,

$$G^{-1} = G_0^{-1} - \Sigma. \quad \text{(B2)}$$

We use the (d, c_1, c_2, c_3) basis. The inverse bare Green's function is given by

$$G_0^{-1}(\omega) = \begin{pmatrix} \omega - \epsilon_d & 0 & 0 & 0 \\ 0 & G_1^{-1} & 0 & 0 \\ 0 & 0 & G_2^{-1} & 0 \\ 0 & 0 & 0 & G_3^{-1} \end{pmatrix}, \quad \text{(B3)}$$

where G_α is the bare Green's function of the reservoir α, given in the large bandwidth limit by (in the LO basis)

$$G_\alpha^{-1}(\omega) = \frac{1}{\pi\rho_0}\begin{pmatrix} i & -2ih_{\text{eq}}(\omega - \mu_\alpha) \\ 0 & -i \end{pmatrix}, \quad \text{(B4)}$$

where ρ_0 is the density of states, which we take to be the same for each reservoir. The self-energy is given by

$\Sigma_{+-} =$

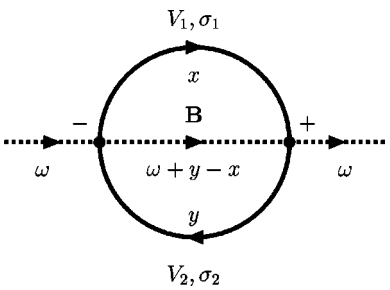


FIG. 6. Computation of the Keldysh structure of the generic diagram.

$$\Sigma(\omega)=\begin{pmatrix} 0 & g_1 & g_2 & g_3 \\ g_1 & 0 & 0 & 0 \\ g_2 & 0 & 0 & 0 \\ g_3 & 0 & 0 & 0 \end{pmatrix}. \tag{B5}$$

Solving Eq. (B2), we find the occupation of the dot in the steady state

$$n(\epsilon_d)\equiv\langle d^\dagger d\rangle. \tag{B6}$$

In the limit where g_1, g_2, g_3 go to zero in fixed ratios, we obtain

$$n(\epsilon_d)=\frac{g_1^2 n_F(\epsilon_d-\mu_1)+g_2^2 n_F(\epsilon_d-\mu_2)+g_3^2 n_F(\epsilon_d)}{g_1^2+g_2^2+g_3^2}, \tag{B7}$$

where n_F is the Fermi function. The properties of this result are similar to those of Eq. (4): the occupation of the dot, in the limit of zero couplings, depends on the ratios of these couplings and is not in general given by the Fermi function. Moreover, if $g_3 \gg g_1, g_2$, we recover the equilibrium result since the physics is dominated by the thermal bath 3. If, on the other hand, we take $g_3 \ll g_1, g_2$, we find a nonequilibrium result since the occupation at order 0 in perturbation theory in the g's is determined by the leads 1 and 2.

APPENDIX C: ORIENTABILITY OF MAJORANA FERMION LINES

In this appendix, we demonstrate that we can treat the Majorana lines as oriented in the diagrams. To show this, it is simplest to take the lines to represent the bare zero-field Majorana functions, and to treat both the magnetic field and the Kondo interaction as vertices. We have three species of Majorana fermion, $\{\eta^x, \eta^y, \eta^z\}$; formally, we may represent each of these as the sum of the creation and annihilation operators of a Dirac fermion,

$$\eta^a=\frac{1}{\sqrt{2}}(f_a+f_a^\dagger); \tag{C1}$$

the dual operator $f_a-f_a^\dagger$ decouples from the problem and may be ignored. In the absence of a magnetic field and interactions, all Majorana Green's functions are diagonal, so the only Green's functions that occur are

$$\begin{aligned} G_a &= -i\langle\langle \eta^a(t)\,\eta^a(0)\rangle\rangle_{\mathcal{K}} \\ &= -\frac{i}{2}\langle\langle f_a(t)f_a^\dagger(0)+f_a^\dagger(t)f_a(0)\rangle\rangle_{\mathcal{K}} \\ &= \left(-\frac{i}{2}\langle\langle f_a(t)f_a^\dagger(0)\rangle\rangle_{\mathcal{K}}\right)+p\text{-}h, \end{aligned} \tag{C2}$$

where the notation "p-h" stands for "particle-hole," i.e., $f\leftrightarrow f^\dagger$. We thus see that the bare Majorana Green's function may be written simply as the sum of two bare Dirac fermion Green's functions of opposite orientations.

It is easy to see that the vertex factors at the magnetic field and Kondo vertices do not depend on whether the Dirac fermion lines are incoming or outgoing. In the case of the magnetic field, for example, the interaction term is

$$H_{\rm mag}=-BS^z=iB\,\eta^x\eta^y=\frac{iB}{2}(f_x+f_x^\dagger)(f_y+f_y^\dagger), \tag{C3}$$

so we see that the vertex factor is the same irrespective of the orientation of the two f lines; a parallel argument may be given for the Kondo interaction vertex. Hence, each diagram consists of a sum of 2^N diagrams that differ only in the orientations of their N Dirac fermion lines.

But these orientational differences do not alter the value of the diagram, since the bare Green's function of the f fermion is particle-hole symmetric, and hence Eq. (C2) corresponds to the Green's function for a single orientation of the Dirac fermion line, with the prefactor 1/2 removed. Hence we may represent the Majorana Green's functions in all diagrams using oriented lines.

APPENDIX D: ERRATUM TO REF. 15

In a previous paper,[15] a formula [Eq. (2)] was proposed for the (second order) perturbative expansion of the magnetic susceptibility in the out-of-equilibrium steady state which was of the form

$$\chi(T,V)=\frac{1}{4T}+O(J). \tag{D1}$$

In this appendix, we briefly rediscuss its derivation and explain why it is incorrect. The method used was straightforward Keldysh perturbation theory to second order with s finite in which the bare Keldysh function of the Majorana spin was taken to be thermal. The $1/s$ divergences were regulated using a "point splitting" procedure: since they occur due to the coincidence of two poles in the integrals, one splits these poles on the real axis to a distance δ and sends δ to 0 at the end of the computation, term by term. The justification given in footnote 20 of Ref. 15 is, however, incorrect: endowing the Majorana fermions on the dot with a fictitious dispersion does not lead to this prescription, since divergences reappear when the bandwidth is sent to zero. The "point splitting" regularization prescription can, however, be described physically as follows: let the applied magnetic fields B_s and B_c oscillate slowly at a frequency ω_0; compute the total magnetization at frequency ω_0 up to second order in J; then take the limit $\omega_0\to 0$ in the coefficient of each power of J.

However, this regulation procedure is based on an interchange of the order of the limits $\omega_0\to 0$ and $J\to 0$. We want to calculate the static magnetic response, and consequently wish to take $\omega_0\to 0$ before $J\to 0$; but in fact the technique used in Ref. 15 does the opposite: it expands in J $(J\to 0)$ *before* taking the $\omega_0\to 0$ limit. It is simple to show that these limits do not commute; this can be seen explicitly from the form of the second term of Eq. (30),

$$\mathrm{Re}\int d\omega \frac{g(\omega)}{(\omega+\omega_0+i\gamma)(\omega-\omega_0-i\gamma)}, \tag{D2}$$

where g is some function and $\gamma\sim\Sigma''(B)$ (the limit $s\to 0$ having already been taken). If we take the limits $J\to 0$ (i.e., $\gamma\to 0$) and then $\omega_0\to 0$ we obtain the result of the point splitting prescription of[15]

$$\int d\omega\left(\frac{g'(\omega)}{\omega}\right), \tag{D3}$$

whereas if we take them in the opposite order, $\omega_0\to 0$ and then $J\to 0$, we recover Eq. (4).

APPENDIX E: THE STEADY-STATE DYSON EQUATION

In this appendix, we present a formal derivation of the steady-state Dyson equation (33). First of all, let us emphasise that our calculation is carried out *in the steady-state regime*, that is, in the long-time limit after the switching on of the interactions. We assume that, in this limit, the system evolves to a time-independent steady state under the time evolution described by its Hamiltonian (as required by the Keldysh method: see Sec. III B). Strictly speaking, this is not possible, since the Hamiltonian (1) conserves the total magnetization of the system and that conservation law prevents the magnetization of the system from relaxing. However, this conservation law is not physical (we have omitted, for example, spin-orbit terms in the leads); therefore, to allow the system to relax to its steady state, we introduce a coupling g that breaks the conservation laws. As a specific example, one could consider an anisotropic ($J_x\neq J_y\neq J_z$) Kondo model. As this extra coupling is relaxed to zero, the transient time taken to reach the steady state diverges but we make the assumption that the values of physical quantities in the steady state are smooth functions of g. Therefore, once we have taken the $t\to\infty$ limit, we can set $g=0$ in the equation which determines the steady state. The details of the derivation are as follows.

(1) Using the perturbative expansion, we establish the Dyson equation at finite times,

$$\int du\{G^{-1}_{\psi 0}(t,u)-\Sigma_{\mathrm{skel}}[G_\psi](t,u)\}*G_\psi(u,t') = \delta(t-t')\otimes 1, \tag{E1}$$

where Σ_{skel} is a functional of G_ψ defined by the skeleton expansion of the self-energy. The product should be understood as a matrix product in the LO basis. G_ψ is a function of two times and of J,g. To obtain this equation, we write the Dyson equation in the finite time diagrammatic expansion outlined in Sec. III B 4,[22] and use the definition of the skeleton diagrams.[26,27] In Eq. (E1), the times t,t' and u run from $-\infty$ to ∞, and the couplings J are time dependent: $J_\alpha(t)=J_\alpha\theta(t)$, i.e., we switch on the interaction suddenly at $t=0$.

(2) The assumption that the system relaxes to a nonequilibrium time translation invariant steady state (the coupling g to the relaxation bath is finite) is transcribed mathematically as the existence of the limit

$$\lim_{\substack{t,t'\to\infty\\ t-t'=\tau}} G(t,t')=G(\tau) \tag{E2}$$

(we shall denote it with the same function name). This assumption is not trivial. In particular, in Ref. 28 the existence of steady oscillating states has been suggested using a large-N slave-boson treatment. We exclude them here on physical grounds, since we do not expect such states to appear in the regime where the perturbation theory is applicable anyway (i.e., at high temperatures or at high magnetic fields in the Kondo problem). We can thus take the long time limit, and Fourier transform the Green's functions and self-energy in Eq. (E1) to obtain

$$G_\psi(J,s,g,\omega)=\{G^{-1}_{\psi 0}(\omega)-\Sigma_{\mathrm{skel}}[G_\psi](J,s,g,\omega)\}^{-1}. \tag{E3}$$

(3) Taking the $g\to 0$ limit as discussed above, we finally obtain the Dyson equation (33),

$$G_\psi(J,s,\omega)=\{G^{-1}_{\psi 0}(\omega)-\Sigma_{\mathrm{skel}}[G_\psi](J,s,\omega)\}^{-1}. \tag{E4}$$

APPENDIX F: COMPUTATION OF THE SELF-ENERGY DIAGRAM

In this appendix, we present the computation of $h_B(B)$ at order 0. The computation is in three steps. First, we compute the self-energy diagrams at second order as a function of a ''generic'' diagram. (This simplifies the problem by separating the Majorana and spin indices from the Keldysh structure.) Second, we compute this generic diagram. Finally, we solve the resulting implicit equation for $h_B(B)$. Throughout this appendix, we streamline our notation by omitting the temperature T; in the final formulas, therefore, B and V should be replaced by B/T and V/T, respectively.

The first part of the computation reduces the spin and lead indices, and thus expresses the self-energy diagrams as functions of the ''generic'' diagram presented in Fig. 4, where we allow any potentials V_1 and V_2 for the electrons and any field B for the internal Majorana line. The main formula is

$$\Sigma_B(\omega)=-\sum_{\substack{\alpha,\beta\in\{R,L\}\\ \sigma=+,-}}\frac{|J_{\alpha\beta}|^2}{4}[f(\omega,B_s,V_\alpha,\sigma,V_\beta,\sigma) + f(\omega,0,V_\alpha,1,V_\beta,-1)]. \tag{F1}$$

In this expression, the Keldysh structure is implicit and by convention, the Majorana line is an η^z line when $B_s=0$ and an f line otherwise. To establish Eq. (F1), we compute the spin and lead indices of Σ_{xx} and Σ_{xy}, which are given by the diagrams of Fig. 5 (with the Feynman rules given in Sec. III B), and we use Eq. (28).

We now compute the Keldysh structure of the generic diagram f (Fig. 6). We have

$$\begin{aligned} f_{+-}(\omega) &= i\pi \int dx dy\, \rho(x+\sigma_1 B_c - V_1) \\ &\times \rho(y+\sigma_2 B_c - V_2) n_F(x-V_1) \check{n}_F(y-V_2) \\ &\times [1+\tilde{h}(B)]\delta(\omega+y-x-B), \\ f_{-+}(\omega) &= -i\pi \int dx dy\, \rho(x+\sigma_1 B_c - V_1) \\ &\times \rho(y+\sigma_2 B_c - V_2) \check{n}_F(x-V_1) n_F(y-V_2) \\ &\times [1-\tilde{h}(B)]\delta(\omega+y-x-B), \end{aligned} \tag{F2}$$

where $\check{n}_F(x) \equiv n_F(-x)$, and the function $\tilde{h}(\omega)$ is defined by

$$\tilde{h}(\omega) = \begin{cases} h_B(\omega), & \omega \neq 0, \\ h_z(0), & \omega = 0. \end{cases} \tag{F3}$$

We then use the relations[22]

$$f^K = -(f_{+-} + f_{-+}), \tag{F4}$$

$$f^A - f^R = -(f_{+-} - f_{-+}). \tag{F5}$$

We can perform the integrals in the large bandwidth limit $D \to \infty$ using

$$\int dx\, n_F(x+A)\check{n}_F(x) = \frac{A}{e^A - 1},$$

and replacing the densities ρ by their finite value ρ_0 (we can take the limit $D \to \infty$ under the integral). Using the definition $V = V_2 - V_1$, we find

$$\frac{f^K}{2i\pi} = \frac{\rho_0^2}{2}(\omega - B + V)\left(1 - \frac{\tilde{h}(B)}{\tanh\dfrac{\omega - B + V}{2}}\right), \tag{F6}$$

$$\frac{(f^A - f^R)}{2i\pi} = -\frac{\rho_0^2}{2}(\omega - B + V)\left(\frac{1}{\tanh\dfrac{\omega - B + V}{2}} - \tilde{h}(B)\right). \tag{F7}$$

Using Eq. (F1), and introducing

$$\varphi(x) \equiv \frac{x}{\tanh\dfrac{x}{2}}, \tag{F8}$$

we now have

$$\begin{aligned} \sigma_B(\omega) &= \frac{|\rho_0 J_{RL}|^2}{4}[-h_B(B)(\omega - B + V) - h_z(0)(\omega + V) \\ &+ \varphi(\omega - B + V) + \varphi(\omega + V) + (V \to -V)] \\ &+ \sum_{\alpha = R,L} \frac{|\rho_0 J_\alpha|^2}{4}[-h_B(B)(\omega - B) \\ &- h_z(0)(\omega) + \varphi(\omega - B) + \varphi(\omega)], \end{aligned} \tag{F9}$$

$$\begin{aligned} \sigma_B^K(\omega) &= -\frac{|\rho_0 J_{RL}|^2}{4}\left[(\omega - B + V)\left(1 - \frac{h_B(B)}{\tanh\dfrac{\omega - B + V}{2}}\right)\right. \\ &\left. + (\omega + V)\left(1 - \frac{h_z(0)}{\tanh\dfrac{\omega + V}{2}}\right) + (V \to -V)\right] \\ &- \sum_{\alpha = R,L} \frac{|\rho_0 J_\alpha|^2}{4}\left[(\omega - B)\left(1 - \frac{h_B(B)}{\tanh\dfrac{\omega - B}{2}}\right)\right. \\ &\left. + \omega\left(1 - \frac{h_z(0)}{\tanh\dfrac{\omega}{2}}\right)\right]. \end{aligned} \tag{F10}$$

Using the definition of the Majorana Green's function $G_{ab}^K(t)$,

$$G_{ab}^K(t) = -i\langle[\eta_a(t), \eta_b(0)]\rangle, \tag{F11}$$

we have $G_{ab}^K(-t) = -G_{ba}^K(t)$. In the frequency representation, this reads $G_{ab}^K(\omega) = -G_{ba}^K(-\omega)$ and hence we infer that $G_{zz}^K(\omega)$ is odd in frequency. Consequently, given the form $G_{zz}^K = h_z(\omega)\delta(\omega)$ it is clear that

$$h_z(0) = 0.$$

Using this result together with Eq. (39) and denoting $x = -h_B(B)$, we have

$$x = \frac{|J_{RL}|^2[2x\varphi(V) + 2B] + (J_R^2 + J_L^2)(2x + B)}{|J_{RL}|^2[2\varphi(V) + \varphi(B+V) + \varphi(B-V)] + (J_R^2 + J_L^2)[2 + \varphi(B)]}.$$

Solving for x and substituting into Eq. (43) gives Eq. (4) of the text.

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