

Interplay between exchange interactions and charging effects in metallic grains

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Abstract. We study the effect of strong antiferromagnetic exchange interactions in metallic grains in the Coulomb blockade regime. These interactions can be large in superconducting systems or in metallic antiferromagnetic particles. We extend the standard description of the grain in terms of a single collective variable, the charge and its conjugated phase, to include the spin degree of freedom. The suppression of spin fluctuations enhances the tendency towards Coulomb blockade. The effective charging energy and conductance are calculated numerically in the regime of large grain-lead coupling.

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1 Introduction

Coulomb blockade in metallic grains is a well studied phenomenon [1,2]. The transport through a grain in the Coulomb blockade regime can be studied using rate equations when the coupling to the leads is weak [1,3]. The renormalization of the charging energy when the coupling to the leads is large is also well understood [3–6]. This strongly coupled regime is best studied by introducing a single collective degree of freedom, the phase, conjugated to the total number of electrons in the grain Q/e [7–9]. The use of this variable is justified when the separation between the electronic levels within the grain can be neglected, or, alternatively, when the conductance of the grain is large. In this limit, the interaction effects within the grain can be described by a simple Hamiltonian [10], expressed in terms of the total charge ($E_C(\hat{Q} - Q_0)^2$), the total spin ($J_S \mathbf{S}^2$) and the individual electronic degrees of freedom.

In the presence of attractive interactions in the grain a pairing term ($\lambda_{\text{BCS}} \hat{T}^+ \hat{T}$, where $\hat{T} = \sum_{\alpha} c_{\alpha\uparrow} c_{\alpha\downarrow}$) should also be included [11] that will drive the system towards superconducting state with energy gap Δ_{BCS} . In such case both the pairing λ_{BCS} and the exchange J_S will grow under renormalization group (integration of energies down from Thouless energy) to a scale much larger than the

bare one, which is initially of the order of the level spacing. Moreover due to the attractive character of the interaction, the spin susceptibility due to exchange will be positive, so that spin fluctuations get suppressed much in the same way as charge fluctuations do due to the charging energy. The essential distinction between the two will stem from the topological differences between spin group $SU(2)$ and charge group $U(1)$ in which their conjugate phases exist.

In the following, we will generalize the usual description of a small grain in the Coulomb blockade regime in terms of phase dynamics [7]. We include also the dynamics of the total spin of the grain in $SU(2)$, on the same footing as the total charge. We assume that the grain has a negligible level spacing and a finite positive renormalized susceptibility according to the arguments above.

The effects of a constant exchange term on the transport properties of a quantum dot has already been studied in the limit where the coupling to the leads is weak, using rate equations [12–14]. The present formalism goes one step beyond this by summing all processes up to cotunneling level. However we do not consider here the changes in the grain susceptibility induced by the spin-orbit coupling which has been considered by other authors [15].

The next section contains an analysis of systems where the regime studied in this paper can be achieved. Then we formulate the model to be studied. The model is analyzed using path integral methods, which are described in the

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next section. We then present the main results, and the last section summarizes the main conclusions.

2 Metallic grains with antiferromagnetic exchange

As mentioned in the introduction, the universal Hamiltonian proposed for small metallic systems in the diffusive regime [11] includes a ferromagnetic exchange term, whose magnitude is of the order of the level spacing within the grain. This contribution is a generalization of the direct exchange interaction in atoms and molecules. Our work here focuses on the collective properties of the grain, at scales larger than the level spacing. In the following, we discuss two situations where the fluctuations of the spin of the grain are determined by interactions which can be much larger than the level spacing.

2.1 Antiferromagnetic metallic grains

There are a variety of materials which exhibit a metallic antiferromagnetic phase at low temperatures. One of the most studied class of materials which exhibit this phase are the heavy fermion compounds [16], and the antiferromagnetic metallic — paramagnetic transition is a well-known example of quantum critical point [17].

The ground state of finite antiferromagnetic clusters is a singlet. Its excitation spectrum can be split into two regimes [18]: (i) at low energies, $\epsilon \ll J_{AF}(a/L)$, where J_{AF} is the antiferromagnetic coupling between neighboring spins, a is the lattice constant, and L is a length proportional to the dimension of the system, the spin excitations are global rotations of all the spins mutually locked into an antiferromagnetic configuration. The effective Hamiltonian at these scales is a quantum rotor, equivalent to the spin term to be studied in this article. The coupling which characterizes the stiffness of the total spin scales as $J_S \sim J_{AF}/N_s$, where N_s is the total number of spins. This coupling scales in a similar way as the level spacing, $\Delta \sim W/N_{el}$, where W is the width of the conduction band, and N_{el} is the total number of electrons in the conduction band; (ii) at higher energies, $\epsilon \gg J_{AF}(a/L)$ the excitations can be described as confined spin waves.

The total spin of the system can be considered a collective variable in regime (i). When $J_S \gg \Delta$, the fluctuations of the total spin take place at frequencies much larger than the level spacing, and the discussion in the following sections is applicable. In typical metallic antiferromagnets, $J_{AF} \leq W$, although this inequality can be reversed in heavy fermion metals, with a narrow resonance at the Fermi level. On the other hand, the number of spins is related to the number of magnetic ions in the system, while the total number of electrons is determined by all the bands at the Fermi level, in a system with many atoms in the unit cell. Hence, the inequality $N_S \ll N_{el}$ can be satisfied in a variety of systems.

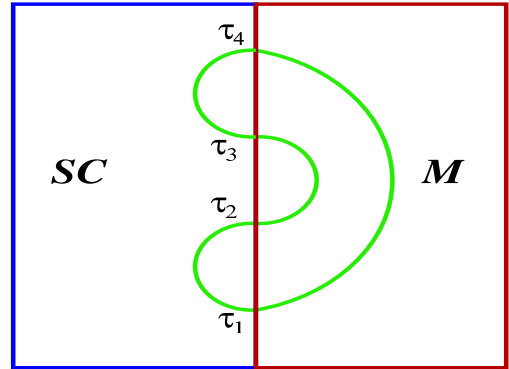


Fig. 1. Fourth order coupling term between a normal metal and a superconductor which describes Andreev processes [19] (see text for details).

2.2 Superconducting grains

As mentioned in the introduction, in a metallic grain with attractive interactions, the universal Hamiltonian contains a pairing term and a spin term with positive exchange, $J_S > 0$, favoring a singlet ground state. The integration of high energy electron-hole pairs required to define the Hamiltonian at low energies lead to the enhancement of these two terms. If we neglect momentarily the pairing contribution, we can define an effective Hamiltonian at temperatures near, or below, the bulk critical temperature, with $J_S \sim \Delta_{BCS} \gg \Delta$, where Δ_{BCS} is the bulk superconducting gap and Δ is the level spacing.

The effect of the pairing term is to reduce phase fluctuations, and to open a gap in the electronic spectrum. In a completely isolated grain, the quantum fluctuations of the collective phase are determined by the charging energy, which leads to the same effective model for a metallic or superconducting grain (see below). The spectrum of quasiparticle excitations determines the coupling to the external leads, and, in particular, the long time properties of the kernel which leads to the damping of charge fluctuations [7]. To second order in the hopping between the grain and the external lead, a gap in the spectrum of the grain leads to a short range kernel, with a typical decay on time scales of the order of the inverse of the superconducting gap. Thus, this contribution to the action is irrelevant for low frequency correlations at temperatures much below the gap, and will be ignored.

We consider here grains strongly coupled to the external leads. Then, at fourth order in the hopping between the grain and each individual channel in the leads, Andreev reflections lead to a long range (Ohmic) kernel at long times [19,20]. In the absence of the spin effects considered here, the effective description of a superconducting grain strongly coupled to a metallic lead is given by the same action considered for a single metallic grain [8,9]. The relevant diagram is shown in Figure 1. The quasiparticle Green's functions in the superconductor decay exponentially for $\tau_1 - \tau_2 \gg \Delta_{BCS}^{-1}$ and $\tau_3 - \tau_4 \gg \Delta_{BCS}^{-1}$. Then, at energy or temperature scales much lower than Δ_{BCS} , we can set $\tau_1 \approx \tau_2 \approx \tau$ and $\tau_3 \approx \tau_4 \approx \tau'$. In this

regime, the coupling between the grain and the leads is described by an effective action with an ohmic kernel, where the hopping amplitude entering the coefficient should be interpreted as the hopping of a Cooper pair.

Thus, when the conductance between the grain and the leads is determined by Andreev reflections, the analysis of the spin fluctuations in the grain described below is applicable. A similar situation can arise when leakage currents between the grain and the lead exist [21].

In the following, we assume that either the superconducting gap Δ_{BCS} is much smaller than the renormalized charging energy, $\Delta_{\text{BCS}} \ll E_C^*$, so that the dissipation looks ohmic in wide range of frequencies $\Delta_{\text{BCS}} \ll \omega \ll E_C^*$ (like for Al superconducting grains with radii below 100 nm), or that the coupling is sufficiently strong so that an ohmic current due to Andreev processes cannot be neglected.

3 The model

The Hamiltonian that we study is: $\mathcal{H} = \mathcal{H}_{\text{grain}} + \mathcal{H}_{\text{lead}} + \mathcal{H}_{\text{hop}}^1$, where

$$\begin{aligned} \mathcal{H}_{\text{grain}} &= \sum_{i,s} \epsilon_i d_{i,s}^\dagger d_{i,s} + E_C \hat{N}^2 + J_S \mathbf{S}^2 \\ \mathcal{H}_{\text{lead}} &= \sum_{k,s} \epsilon_k c_{k,s}^\dagger c_{k,s} \\ \mathcal{H}_{\text{hop}} &= -t \sum_{i,k,s} c_{k,s}^\dagger d_{i,s} + \text{H.c.} \end{aligned} \quad (1)$$

and \hat{N} and \mathbf{S} are the total number of electrons and the total spin of the grain, $\hat{N} = \sum_{i,s} d_{i,s}^\dagger d_{i,s}$ and $\mathbf{S} = 1/2 \sum_{i,s,s'} d_{i,s}^\dagger \boldsymbol{\sigma}_{s,s'} d_{i,s'}$, where $\boldsymbol{\sigma}$ denotes the Pauli matrices. The grain-dot conductance, in dimensionless units, can be approximated by $\alpha \approx t^2 \rho_{\text{grain}}(\epsilon_F) \rho_{\text{lead}}(\epsilon_F)$, where $\rho_{\text{grain/lead}}(\epsilon_F)$ is the density of states at the Fermi level of the grain and the lead. As mentioned above, we neglect the energy dependence of the density of states of the grain. The only interactions included in equation (1) are through the total spin and charge of the grain.

4 Path integral formulation

We can integrate out the fermionic degrees of freedom and obtain a description in terms of collective variables only by using the path integral formalism. The action is

¹ We do not include a possible superconducting pairing term for simplicity, since as we have argued in the introduction it gives rise only to a gapped kernel to second order in the single electron hopping, which we ignore. In fact, in the case of a superconducting grain one should consider the creation operators in the above model as corresponding to Cooper pairs.

$S = S_{\text{grain}}^0 + S_{\text{lead}} + S_{\text{hop}} + S_{\text{int}}$, where

$$\begin{aligned} S_{\text{grain}}^0 &= \int_0^\beta d\tau \sum_{i,s} \bar{d}_{i,s} (\partial_\tau + \epsilon_i - \mu_{\text{grain}}) d_{i,s} \\ S_{\text{lead}} &= \int_0^\beta d\tau \sum_{k,s} \bar{c}_{k,s} (\partial_\tau + \epsilon_k - \mu_{\text{lead}}) c_{k,s} \\ S_{\text{hop}} &= -t \int_0^\beta d\tau \sum_{i,k,s} \bar{d}_{i,s} c_{k,s} + \text{H.c.} \\ S_{\text{int}} &= E_C (\hat{N} - N_{\text{ext}})^2 + J_S (\mathbf{S} - \mathbf{S}_{\text{ext}})^2. \end{aligned} \quad (2)$$

We have included an offset electron number $N_{\text{ext}} = V_{\text{gate}}/eC_g$ induced by a gate voltage V_{gate} and an offset spin $\mathbf{S}_{\text{ext}} = \mathbf{H}_{\text{ext}}/2J_S$ induced by an external magnetic field \mathbf{H}_{ext} , that couples to the total spin.

We can now decouple the quartic interaction term S_{int} by means of a Hubbard-Stratonovich transformation $e^{-E_C(N - N_{\text{ext}})^2} \propto \int \mathcal{D}V e^{-V^2/4E_C - iV(N - N_{\text{ext}})}$, and similarly for \mathbf{S} , which introduces a new scalar field V for the total charge and a *vector* field \mathbf{H} for the total spin. We then have $S_{\text{int}} = S_0 + S_1$, with

$$\begin{aligned} S_0 &= \int_0^\beta d\tau \left(\frac{V^2}{4E_C} + \frac{\mathbf{H}^2}{4J_S} - iV N_{\text{ext}} - i\mathbf{H} \cdot \mathbf{S}_{\text{ext}} \right) \\ S_1 &= i \int_0^\beta d\tau \left(V \hat{N} + \mathbf{H} \cdot \mathbf{S} \right). \end{aligned} \quad (3)$$

We now perform a time dependent canonical transformation (a phase and spin rotation) on the electronic wavefunctions, in order to cancel the term S_1 in equation (3). This $U(1) \times SU(2)$ transformation can be written as:

$$\begin{aligned} d_{ks}(\tau) &\rightarrow U_{ss'}(\tau) d_{ks'}(\tau) \\ U(\tau) &= e^{i\phi(\tau)} e^{\frac{i}{2}\xi(\tau)\hat{n}(\tau)\cdot\boldsymbol{\sigma}}. \end{aligned} \quad (4)$$

The transformation is parametrized by the angles $\phi(\tau)$ and $\xi(\tau)$, and by the three dimensional unitary vector $\hat{n}(\tau)$. The requirement that S_1 in (3) is cancelled implies:

$$(\partial_\tau U) U^\dagger = iV + \frac{i}{2} \mathbf{H} \cdot \boldsymbol{\sigma} \quad (5)$$

so that $V = \dot{\phi}$ and

$$\mathbf{H} = \dot{\xi} \hat{n} + \sin \xi \dot{\hat{n}} + (1 - \cos \xi) \hat{n} \times \dot{\hat{n}}. \quad (6)$$

These identities provide an alternative and convenient parametrization of the auxiliary fields V and \mathbf{H} , represented now by the ϕ, ξ, \hat{n} fields, which will be used in the following. Note that the $U(1)$ gauge transformation needed to replace V by the phase ϕ leads to the standard description of charging effects in terms of phase fluctuations. It is interesting to note that equation (6) implies that \mathbf{H} is proportional to the angular momentum of a *sphere*, considered as a rigid body [22]. The periodicity in imaginary time of the arguments in the action implies that $U(0) = U(\beta)$. This constraint implies the usual quantization of the charge in the grain, and also of the spin (see below), due to the discreteness of transport events.

A more compact notation for the transformation in equation (4) can be given in terms of the following, τ dependent, two- and four-dimensional unit vectors:

$$\begin{aligned}\hat{u}_\tau &= (\sin \phi_\tau, \cos \phi_\tau) \\ \hat{v}_\tau &= \left(\hat{n}_\tau \sin \frac{1}{2} \xi_\tau, \cos \frac{1}{2} \xi_\tau \right).\end{aligned}\quad (7)$$

We can use these vectors to write S_0 as:

$$S_0 = \int_0^\beta d\tau \left\{ \frac{(\partial_\tau \hat{u})^2}{4E_C} + \frac{(\partial_\tau \hat{v})^2}{4J_S} \right\} \quad (8)$$

where external gates and fields have been taken as zero for the moment. The transformation in equation (4) modifies also the lead-grain coupling, S_{hop} :

$$S_{\text{hop}} = -t \int_0^\beta d\tau \sum_{i,k,s,s'} \bar{c}_{k,s} U_{s,s'} d_{i,s'} + \text{H.c.} \quad (9)$$

A final step to obtain the effective action for the rotor fields is to integrate out the fermionic fields to order t^2 , using $\langle e^{-S_{\text{hop}}} \rangle_0 = e^{-\frac{1}{2} \langle S_{\text{hop}}^2 \rangle_0 + \mathcal{O}(t^4)}$. One obtains the following dissipation term:

$$S_{\text{diss}} = -\frac{\alpha}{4} \int_0^\beta d\tau \int_0^\beta d\tau' K(\tau - \tau') \text{Tr} \left[U_\tau^\dagger U_{\tau'} + U_{\tau'}^\dagger U_\tau \right] \quad (10)$$

where $K(\tau) = -[G_{\text{lead}}(\tau)G_{\text{grain}}(-\tau)]/[\rho_{\text{lead}}(\epsilon_F)\rho_{\text{grain}}(\epsilon_F)] = (\pi T)^2 / \sin^2(\pi T\tau)$, G_{lead} and G_{grain} being the lead and grain unperturbed Green's functions in imaginary time. Recall here that the finite range that the superconducting gap could bring in is assumed larger than the decay time of the phase correlators which is of order E_C^* , so that the gapless $K(\tau - \tau')$ of the normal state yields equivalent results. S_{diss} may be finally recast as

$$S_{\text{diss}} = \alpha \int_0^\beta d\tau \int_0^\beta d\tau' K(\tau - \tau') \left[1 - (\hat{u}_\tau \cdot \hat{u}_{\tau'}) (\hat{v}_\tau \cdot \hat{v}_{\tau'}) \right]. \quad (11)$$

This term is sufficient to account for second order tunnelling processes, and in particular it can describe cotunnelling features. The derivation is valid when the conductance between the grain and the electrode *per channel* is small, and it can be used even if the total value is large.

The method leading to equation (11) can be easily generalized to the case with a spin dependent density of states in the grain or in the leads. The effective action will contain terms involving $\sin(\xi_\tau + \xi_{\tau'})$ which break the symmetry between the four components of the vector \hat{v}_τ . These terms are analogous to the Josephson term which arises in the charge dynamics when the leads, or the grain are superconductors [7].

The final action is $S_{\text{eff}} = S_0 + S_{\text{diss}}$, written in terms of the dynamical variables \hat{u}_τ and \hat{v}_τ only. In the limit $J_S = 0$, the field \hat{v}_τ can be taken as a constant, and the model reduces to the standard phase only model.

5 Results

It is instructive to analyze first the decoupled grain, described by S_0 in equation (8), to see where this spherical rotor description of the total spin comes from. As mentioned above, S_0 contains the usual phase term, which leads to the quantization of the charge, and a contribution which is equivalent to that of a rigid rotor, and which leads to the conservation of spin. The eigenvalues associated to S_0 can be written as $E_{N,S,S_z,K} = E_C N^2 + J_S S(S+1)$, where $N = 0, 1, 2, \dots$, $S = 0, 1, 2, \dots$, $-S \leq S_z \leq S$ and $-S \leq K \leq S$.² The degeneracy of a given state is $(2S+1)^2$ [22]. This degeneracy can be understood by noting that, in the limit studied here, the level spacing within the grain can be neglected. The grain energy is solely determined by the total charge and the total spin. Let us assume that, in the neutral dot, there are N_0 spin 1/2 electrons which contribute to the total spin. The number of states of total spin S (each with degeneracy $2S+1$) is:

$$C_S^{N_0} = \binom{N_0}{\frac{N_0}{2} - S} - \binom{N_0}{\frac{N_0}{2} - S - 1}. \quad (12)$$

In the limit of many electrons $N_0 \rightarrow \infty$, one obtains $\lim_{N_0/S \rightarrow \infty} C_S^{N_0} = (2S+1)C_{N_0}$, where $C_{N_0} = \frac{2^{N_0+3/2}}{\sqrt{\pi} N_0^{3/2}}$ is a constant independent of S . This means that the total degeneracy of a state composed of many 1/2 spins and given value of the total spin momentum $\langle \hat{S}^2 \rangle = S(S+1)$ is $C_{N_0}(2S+1)^2$, just as C_{N_0} rigid rotors with total angular momentum S . The existence of this degeneracy leads to a prefactor in the free energy which is independent of the angular momentum. This multiplicity, like similar degeneracies in the case of ordinary Coulomb blockade, does not affect the effects associated to the spin gap discussed in this paper.

The following calculations including the full action S_{eff} have been done by averaging over all paths in the unit circle parameterized by \hat{u} and the four-dimensional sphere which defines \hat{v} , using an extension of the Monte Carlo code developed earlier for related problems [23,6]. The effective charging energy is calculated by summing over winding numbers of the phase. The conductance between the grain and the electrode has been approximated by the expression $G(\beta/2)$ [23,24], valid at low temperatures, where G is the correlation function, in imaginary time, of the variable $\hat{v}_\tau \hat{u}_\tau$. The latter combination describes the transfer of a full electron to the grain. We calculate, separately, the correlations $G_u = \langle \hat{u}_\tau \cdot \hat{u}_{\tau'} \rangle$ and $G_v = \langle \hat{v}_\tau \cdot \hat{v}_{\tau'} \rangle$ which correspond to charge only and spin only currents.

The current correlation functions are shown in Figure 2. It is interesting to note that both $\langle \hat{u}_\tau \cdot \hat{u}_{\tau'} \rangle$ and $\langle \hat{v}_\tau \cdot \hat{v}_{\tau'} \rangle$ decay exponentially, while the composite correlation $\langle \hat{u}_\tau \cdot \hat{u}_{\tau'} \hat{v}_\tau \cdot \hat{v}_{\tau'} \rangle$ decays as $(\tau - \tau')^{-2}$, as required

² Note that the extra degenerate quantum number K respect to the usual spin S degeneracy is due to the fact that we are dealing with the orientations of a sphere, which are more numerous than the orientations of a spin (there are three Euler angles versus the two spherical coordinates of the Bloch sphere).

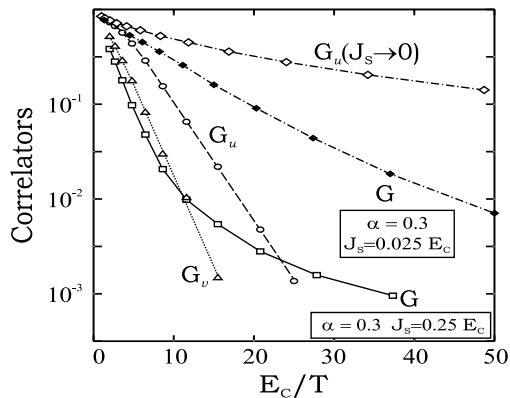


Fig. 2. Charge-charge, spin-spin and electron-electron current correlations (see text) versus inverse temperature for $\alpha = 0.3$ and different values of J_S .

by Griffith's inequality [25]. The differences between the phase-phase, "rotation-rotation" and current-current correlations is reminiscent of the behavior of a Luttinger liquid. It implies that the electron current cannot be factorized into its spin and charge components. The exponential decay of the correlations associated with the collective charge and spin degrees of freedom can be understood as the effect of a charge and spin gap in the grain. It can be obtained by making a mean field decoupling of the variables, in a similar way to the calculation for charging effects in coupled grains [26]. The $(\tau - \tau')^{-2}$ decay of the current-current correlation describes the cotunnelling processes at low temperatures.

The effective charging energies, as functions of α and J_S , are shown in Figures 3 and 4. The effect of a finite J_S on the renormalized charging energy is significant, even for small values of J_S . We can estimate analytically this effect, by assuming that when $J_S \rightarrow 0$ the fluctuations in the variable \hat{v}_τ are small. The effect of these fluctuations on the variable \hat{u}_τ can be approximated by replacing α in equation (11) by $\alpha \langle |\hat{v}|^2 \rangle$. Assuming that the fluctuations of \hat{v}_τ are harmonic, we find:

$$\langle |\hat{v}|^2 \rangle \approx 1 - \int_{E_C}^{\Lambda} \frac{d\omega}{\omega^2/2J_S} \approx 1 - \frac{J_S}{E_C} \quad (13)$$

where Λ is a high energy cutoff, comparable to the electronic bandwidth. Then, using the well-known expression for the renormalized charging energy for large values of α [3, 4]: $E_C^* \approx E_C \exp\{-2\pi^2\alpha(1 - J_S/E_C)\}$. This enhancement of the effective charging energy by a spin gap is another manifestation of the non-separability of charge and spin.

6 Conclusions

We have analyzed the influence of the exchange term in a small superconducting grain on the charging effects in the regime where the superconducting gap is smaller or comparable to the charging energy. The suppression of the spin susceptibility reduces large fluctuations in the spin of

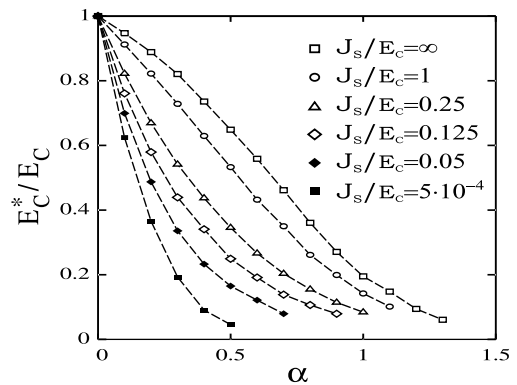


Fig. 3. Renormalized charging energy of the grain in the presence of a finite spin gap J_S , versus the dimensionless grain-lead coupling α . Note that the decay becomes less pronounced for growing spin gap.

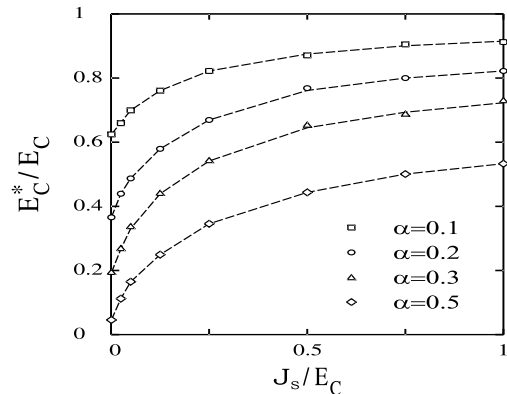


Fig. 4. Renormalized charging energy of the grain in the presence of a finite spin gap J_S , versus the value of the spin gap J_S . Note the saturation for large J_S .

the grain, and enhances the tendency towards Coulomb blockade. Our analysis integrates out the electronic degrees of freedom in the grain and in the external leads, and provides a simple description in terms of the charge and spin degrees of freedom of the grain only.

The effects of the exchange term have been analyzed for closed quantum dots, which are almost decoupled from the leads [12–14]. Our scheme provides a generalization which is non-perturbative in the coupling strength in the sense that one can recover exponential effects in the coupling, such as the renormalization of the charging energy, which cannot be derived from the addition of sequential processes.

A statistical approximation to the electron-electron interactions in a small dot predicts that the bare exchange term is negative and of the order of the separation between electronic levels [10, 27]. Our analysis, on the other hand, is valid only when the exchange term is positive and larger than the level spacing. This regime corresponds to systems with an attractive electron-electron interaction near a superconducting transition, when the exchange J is significantly enhanced [28], or, alternatively, to antiferromagnetic metallic grains. Spin fluctuations in grains in

this regime can therefore have a strong influence on charge fluctuations, restoring the system to a Coulomb blockade regime even when the coupling to the leads is strong.

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