Josephson versus Kondo coupling in a Quantum Dot Connected to Two Superconductors

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Abstract

We apply a Gutzwiller-like variational technique to study Josephson conduction across a quantum dot with an odd number of electrons connected to two superconducting leads. Our method projects out all states on the dot but the Kondo singlet and is valid when Kondo correlations are dominant and no Andreev bound states localized at the dot are available for Kondo screening. In these conditions superconducting pairing is a competing effect and the junction is $\pi$–like, to optimize antiferromagnetic correlations on the dot. As the superconducting gap increases, the Josephson current also increases, but its phase dependence becomes strongly non sinusoidal.

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Phase coherent transport in hybrid superconducting-semiconducting nanostructures is already extensively investigated. In these devices quite interesting and surprising features emerge, due to electron-electron interaction [1]. In this paper we study a Quantum Dot (QD) at Coulomb Blockade (CB) with an odd number of electrons $N$, connected to superconducting contacts [2]. Although such a kind of system has not been realized yet, it is very likely that continuous improvement of nanofabrication techniques will soon make it available. In particular, we address the question whether the formation of a Kondo singlet at the dot competes or cooperates with BCS s-wave pairing in the leads. We set up a variational non perturbative approach which can be adopted as long as $\Delta < k_B T_K$, where $T_K$ is the Kondo temperature of the dot and $\Delta$ is the superconducting gap. We show how the flow to the Kondo fixed point is affected by superconductivity because singlet pairing starts to compete with Kondo correlations. The system behaves as a $\pi$-Josephson junction [3], with a rather small critical Josephson current $I_J$. As $\Delta$ increases and becomes comparable with $k_B T_K$, there is a crossover to a regime in which Kondo correlations on the dot are very much affected. The junction is still $\pi$-like and the Josephson critical current increases but it becomes strongly non sinusoidal.

In our approach bound states localized within the dot area are responsible for the bare magnetic moment on the dot. They are only weakly modified when the contacts become superconducting, as they turn into Andreev bound states within the gap. As discussed in the conclusions, the fact that our model does not include other empty Andreev states hinders the possibility of screening out the dot spin completely, in the large Kondo limit.

We believe that this is the reason why the $\pi$-coupling always wins in our model, even when the ratio $\Delta/k_B T_K$ is quite small. Indeed $\pi$ coupling optimizes pair tunneling in the presence of strong antiferromagnetic correlations on the dot.

Coulomb Blockade has been widely investigated in Quantum Dots with normal leads [4]. If the coupling with the contacts is weak and the charging energy is higher than the thermal activation energy, DC conduction is strongly dependent on the gate voltage $V_g$. The DC conductance across the dot shows a sequence of peaks, occurring when $V_g$ provides the energy to add an extra electron to the dot, taken from the contacts at chemical potential $\mu$. As the dot is tuned in a “Coulomb Blockade” valley between two consecutive peaks, $N$ is fixed and the conductance is heavily suppressed.
Kondo conductance may be achieved in a QD with $N$ odd within a CB valley by increasing the coupling between dot and leads, provided the temperature $T < T_K$ [5]. At $T < T_K$, a strongly correlated state between dot and leads sets in and a resonance in the density of states of the QD opens up at $\mu$. Correspondingly, DC conductance across the dot increases, until it eventually reaches the unitarity limit at $T = 0$ [6, 7].

Since charge dynamics is “frozen out” by CB, the QD in this regime is usually modelized as a magnetic impurity with total spin $S = \frac{1}{2}$. A Schrieffer-Wolff transformation on the single level Anderson impurity model involving virtual zero or double occupancy of the impurity level leads to the effective Kondo Hamiltonian with antiferromagnetic coupling between the delocalized electrons of the leads and the impurity spin.

Magnetic impurities embedded in a bulk superconductor are known to strongly influence the superconducting critical temperature. Adding impurity states at energies below the superconducting gap can even give raise to gapless superconductivity [8].

One can extend all this to the case of superconducting contacts. The Schrieffer-Wolff transformation can be performed in the same way, provided $D >> \Delta$, where $D$ is the bandwidth of the itinerant electrons. However, the resulting model is not equivalent to a single magnetic impurity in a phase coherent superconductor. This is a crucial difference with respect to the case of normal contacts when the Kondo screening cloud shows that the phase coherence is established throughout the system.

On the contrary, in such a case the contacts retain their individual superconducting properties and their individual phase for the order parameter with a phase difference $\varphi$. The link between the two superconductors is offered by the dot and is tunnel-like. This gives rise to two important features:

1) Kondo phase coherence and superconducting phase coherence compete;
2) the system has the properties of a Josephson link between two different superconductors.

The reason is that strong on site Coulomb repulsion makes the phase breaking time induced by spin flip scattering processes much shorter than the Kondo resonance lifetime $\hbar/k_B T_K$ [2].

Indeed, tunneling across a magnetic impurity between superconductors with on site Coulomb repulsion has been recently revisited [9, 3, 10, 11]. If $\Delta \geq k_B T_K$, the system is unable to scale toward the strongly-coupled Kondo regime. Sub-gap Cooper pair tunneling is strongly suppressed by Coulomb
Figure 1: Sketch of cotunneling processes ((a) hole process, (b) particle process) in the perturbative regime, to move a Cooper pair from $L$ to $R$ across the QD. The sequence of steps $1 \to 2 \to 3$ is constrained by Pauli restrictions. Analogous processes not depicted here can take place, in which the spins are reversed.

repulsion, unless each cotunneling step is accompanied by a spin flip at the impurity. Pair tunneling can take place but it occurs in a three step sequence via virtual empty or doubly occupied intermediate dot states, because of Pauli exclusion principle, as depicted in Fig. 1. It has been proposed that such a mechanism may reverse the sign of the Josephson current through the dot so that the Josephson energy is at a minimum for $\varphi = \pi$, where $\varphi$ is the phase difference between the order parameters of the two superconductors attached to the impurity ($\pi$-junction) [3]. This is indeed what we find with our variational approach.

Fig. 1 mimicks Cooper pair tunneling via virtual breaking of a pair, one at a time, with one quasiparticle moving into and out of the dot. Particle and hole processes are allowed across the dot, with a mechanism similar to the one involved in the Schrieffer-Wolff transformation. Here we describe the hole process only; the particle one is fully analogous. Let us consider an $\uparrow$ electron localized at the dot. In order to move a pair from $L$ to $R$, we must first empty the dot. The sequence $1 \to 2 \to 3$ corresponds to an operator arrangement, $c_{L\uparrow}c_{R\downarrow}c_{L\downarrow}c_{R\uparrow}$, which has a minus sign with respect to the standard arrangement for Cooper pair tunneling: $c_{R\downarrow}c_{R\uparrow}c_{L\uparrow}c_{L\downarrow}$ [3]. This produces a phase difference of $\pi$ with respect to the usual Josephson coupling, which makes the system a "$\pi$-junction".
In Superconducting/Normal/Superconducting (SNS) structures, Cooper pair subgap tunneling current takes place via Andreev states localized in the normal region [12]. When the normal region is given by a QD, charge quantization at the dot is not spoiled as the contacts become superconducting [13]. The $I-V$ curve of a dot at CB between two superconductors has been derived in [14], showing an interplay between multiple Andreev reflection and electron–electron interaction at the QD.

We expect that fine tuning of the parameters of a S-QD-S device in an orthogonal magnetic field may allow for the investigation of the full range of physical conditions, from $\Delta \ll k_B T_K$ when the system can flow toward the strongly coupled Kondo regime prior to the onset of superconductivity in the leads, to $\Delta \gg k_B T_K$ when perturbation theory holds [11]. In fact, the ground state degeneracy required for Kondo coupling to take place can be obtained by driving the dot to a level crossing by means of an applied magnetic field [7, 15, 16, 17].

We study the $T = 0$ case with $\Delta < k_B T_K$ using a non perturbative variational technique. The fact that a global phase coherence cannot take place in the system justifies our variational approach outlined in the following. Hence, we add Kondo correlations to a state which is the superposition of two BCS states for the left and the right contact with different phases of the order parameter. Such a technique has already been applied to the case of a dot with normal contacts, and it has been shown to provide good qualitative results in the perturbative regime as well, where it reproduces the poor man’s scaling equation [17].

To construct the trial state, we start from the state $|\varphi, s\rangle$, given by the product of the left ($L$) condensate times the right ($R$) condensate, with the two order parameters having a phase difference $\varphi$, times the state of the dot $|s\rangle$:

$$|\varphi, s\rangle = |\text{BCS}, L\rangle \times |\text{BCS}, R, \varphi\rangle \times |s\rangle.$$  \hspace{1cm} (1)

Here $s$ is the spin component along the quantization axis of the dot spin $S_d = \frac{1}{2}$ at CB with odd $N$.

The minimal model for the Kondo interaction between electrons localized on the QD and electrons from the contacts is $H_K = J \vec{\sigma}(0) \cdot \vec{S}_d$. The spin density operator of the delocalized electrons, $\vec{\sigma}(0)$, at the position of the dot...
\( x = 0 \) along the vertical axis, is:

\[
\vec{\sigma}(0) = \frac{1}{2V} \sum_{q,q'} (c_{L,q,\sigma}^\dagger + c_{R,q,\sigma}^\dagger) \vec{\tau}_{\sigma,\sigma'} (c_{L,q',\sigma'} + c_{R,q',\sigma'}). 
\]

We have used the fermion operators \( c_{q,j} (c_{q,j}^\dagger) \), \((j = L, R)\) in the plane wave representation to describe the contacts particles and \( V \) is the normalization volume of the leads. We take the symmetric case in which hybridization of the dot with the \( L \) and \( R \) contacts, \( \Gamma \), is the same. Kondo coupling is antiferromagnetic (AF): \( J > 0 \).

The total Hamiltonian is \( H = H_S + H_K \), where \( H_S \), defined in eq. (5) below, is the Hamiltonian for the \( L \) and \( R \) superconducting contacts in the BCS approximation.

The correlated trial state is constructed by applying a Gutzwiller-like projector \( P_g \) to \( |\varphi, s\rangle \). As \( T \to 0 \), the system scales towards the strongly coupled, large \( J \) regime. Correspondingly, \( P_g \) gradually projects out the high-energy components of the trial state, so that eventually only a localized spin singlet survives at the QD.

The “projector” \( P_g \) is defined as [17]:

\[
P_g = \left( 1 - \frac{3}{4}g \right) + g(\vec{\sigma}(0))^2 - 4S_d \cdot \vec{\sigma}(0),
\]

where \( g \) is a variational parameter which ranges between \( g = 0 \) and \( g = 4/3 \). When \( g = 0 \) we have \( P_0 = 1 - 4S_d \cdot \vec{\sigma}(0) \); \( P_0 \) fully projects out the high energy localized spin triplet at \( x = 0 \). As \( g \) varies from 0 to 4/3 also the localized spin doublet state it increasingly projected out. Eventually, when \( g \) reaches the value \( g = 4/3 \), only the localized spin singlet is left over.

The trial state is defined as:

\[
|g, \varphi\rangle = P_g |\varphi, s\rangle.
\]

The value of \( g(J) \) is determined by finding the minimum of the energy functional \( E[g, J, \varphi, \Delta] \), defined as:

\[
E[g, J, \varphi, \Delta] = \frac{\langle g, \varphi | H | g, \varphi \rangle}{\langle g, \varphi | g, \varphi \rangle}. \tag{3}
\]

Eq. (3) can be calculated by first expressing the products \( P_g H P_g \) and \((P_g)^2\) in terms of the usual fermion quasiparticle operators \( \alpha_{j,q} (\alpha_{j,q}^\dagger), \beta_{j,q} (\beta_{j,q}^\dagger), (j = \)
which destroy (create) excitations on the BCS states of the L and R contact and then by normal-ordering the corresponding operator products.

The operators are [18]:

\[
\begin{align*}
\alpha_{j,q} &= u_q c_{j,q,\uparrow} - v_q e^{i\phi_j} c_{j,q,\downarrow} \\
\beta_{j,-q} &= u_q c_{j,-q,\downarrow} + v_q e^{i\phi_j} c_{j,q,\uparrow},
\end{align*}
\]

where \(u_q\) and \(v_q\) are the BCS coherence factors and \(\phi_R = \varphi\) while \(\phi_L = 0\).

The Hamiltonian for the contacts \(H_S\) is conveniently expressed in terms of these operators as:

\[
H_S = E_{BCS} + \sum_{q,j=L,R} E_q (\alpha_{j,q}^\dagger \alpha_{j,q} + \beta_{j,-q}^\dagger \beta_{j,-q}).
\]

Here \(E_{BCS}\) is the total ground state energy of the condensates and \(E_q = \sqrt{\xi_q^2 + \Delta^2}\) are the energies of the quasiparticle excitations with \(\xi_q = q^2/2m - \mu\) (we put \(\hbar = k_B = 1\) throughout the paper).

The variation of the energy [18] due to the AF coupling at the quantum dot in units of the bandwidth \(D\), \(\epsilon[\xi, j, \delta, \varphi] \equiv E[g, J, \Delta, \varphi]/D\), takes a simple form once expressed in terms of the parameters \(\xi = \frac{1}{2} \left( 1 - \frac{3}{4}g \right)\), \(j = 3J/4D\) and \(\delta = \Delta/D\):

\[
\epsilon[\xi, j, \delta, \varphi] = \frac{j}{2} \frac{1 - \frac{1}{2} \delta^2 (1 + \cos(\varphi))/N^2(0) \lambda^2}{(1 + \xi^2) + (\xi^2 - 1) \delta^2 (1 + \cos(\varphi))/2 (N(0) \lambda^2)} + \frac{(1 - \xi - \xi^2) \left[ \sqrt{1 + \delta^2 - \delta^2 \cos(\varphi)} / N(0) \lambda \right]}{(1 + \xi^2) + (\xi^2 - 1) \delta^2 (1 + \cos(\varphi))/2 (N(0) \lambda^2)}.
\]

Here \(\lambda\) is the BCS electron-electron interaction strength and \(N(0)\) is the normal phase density of states at the Fermi level for each spin polarization. We will take \(N(0) \lambda = 0.3\) throughout the paper. The first term is the expectation value of \(H_K\), while the second is the raise in the average value of the kinetic energy Hamiltonian, \(H_S\), due to the formation of the singlet between the QD and the contacts. The value of \(\xi_{\text{min}}\), at which the energy is at a minimum, measures how much higher-energy spin states are projected out: \(\xi_{\text{min}} = 0\) corresponds to full projection of states different from a localized spin singlet at the impurity.
Figure 2: $\epsilon/j$ vs. $\xi$ for different $j$ and $\delta = 0.06$ in the two cases $\varphi = 0$ (broken line) and $\varphi = \pi$ (full line). For large Kondo coupling $j$ the minimum of the curve moves toward $\xi = 0$.

At $\delta = 0$ the strong coupling fixed point is $j \to \infty$ and $\xi_{\text{min}}(j) \to 0$. In the flow to the fixed point, there is a large decrease of the Kondo energy:

$$\Omega_K \equiv \epsilon[\xi, j, 0, 0] = \frac{1 - \xi_{\text{min}} + (\xi_{\text{min}})^2 - \frac{j}{2}}{1 + (\xi_{\text{min}})^2}. \quad (7)$$

At $\delta \neq 0$, we follow the minimum of the energy of the correlated state, $\epsilon^*[j, \delta, \varphi] \equiv \epsilon[\xi_{\text{min}}, j, \delta, \varphi]$, in the flow $j \to \infty$. In Fig. 2 we plot $\epsilon/j$ vs. $\xi$ for different $j$ at $\delta = 0.06$. We see that the minimum of $\epsilon[\xi, j, \delta, \varphi]$ is located at smaller and smaller $\xi$ values the larger the Kondo correlations are ($j \to \infty$). However the zero value for $\xi_{\text{min}}$, corresponding to the full projection of states different from a localized spin singlet on the dot, is never reached. Energies corresponding to $\varphi = \pi$ (full line) are always lower than those corresponding to $\varphi = 0$ (broken line). This is a general feature of our results: the $\pi$-junction behavior is established throughout the whole range of $\delta$ values, until $\Delta$ reaches the Kondo scale $T_K$.

In Fig. 3 we plot $\xi_{\text{min}}$ vs. $\delta$ for $\varphi = 0, \pi$ and various $j$ values. In the case $\varphi = 0$, $\xi_{\text{min}}$ grows up abruptly for $\delta > 0.25$ what shows that the choice $\varphi = 0$
strongly disfavours the Kondo correlations, as soon as the superconducting pairing correlation length $\xi_S$ decreases. The dependence of $\xi_{\text{min}}$ on $\delta$ is much weaker when $\varphi = \pi$. Inspection of Fig. 5 shows that two regimes can be envisaged as $\delta$ increases:

a) $\Delta \ll T_K$: Superconducting pairing in the leads is a perturbation on the Kondo correlated state

For small values of $\delta$ the minimum of the energy can be attained both for $\varphi = 0$ and for $\varphi = \pi$. According to Fig. 3, there is a slight decrease of $\xi_{\text{min}}$ when $\delta$ increases for $\varphi = 0$. Hence, the superconducting singlet correlations seem to cooperate for small $\delta$'s in such a case. By contrast, $\xi_{\text{min}}$ increases steadily with $\delta$ for $\varphi = \pi$, what shows that singlet correlations on the impurity (with characteristic correlation length $\xi_K$) compete with singlet pairing but are much less disruptive. This is confirmed by Figs. 4a), 4b). The energy minimum is strongly affected when $\delta$ increases for $\varphi = 0$, while this is not the case for $\varphi = \pi$. Again, the case $\varphi = \pi$ is always favoured in energy for any $j$ value (see Fig. 4c)).

The energy of the correlated state $\epsilon^*[j, \delta, \varphi]$, to second order in $\delta$, is:

$$\epsilon^*[j, \delta, \varphi] = \Omega_K \left\{ 1 + \delta^2 \left( \frac{1}{2} + \frac{1 - (\xi_{\text{min}})^2}{(N(0)\lambda)^2} - \frac{\xi_{\text{min}}^2}{(N(0)\lambda)^2} \cos \varphi \right) \right\} +$$ $\frac{j}{2} \frac{\delta^2}{1 + (\xi_{\text{min}})^2} \left( \frac{1}{2} + \frac{1}{(N(0)\lambda)^2} \right).$  (8)

Because $\Omega_K < 0$ for large $j$, it is clear that $\varphi = \pi$ is favoured.

b) $\Delta \leq T_K$: Superconductivity strongly competes with Kondo ordering

Such a situation is better shown in Fig. 5a): when the phase difference between the two superconducting leads is $\varphi = 0$, the energy functional, as $\delta$ approaches the limit value 0.3, cannot gain a minimum. Indeed the system cannot develop a Kondo singlet on the impurity site. Conversely, Fig. 5b) shows that a minimum always exists when the phase difference is $\varphi = \pi$.

It is easy to derive the Josephson current, according to:

$$I_J = 2e \partial \epsilon^*[j, \delta, \varphi] / \partial \varphi.$$  

In regime a), $I_J$ it is still sinusoidal, of the form $I_J = 2e \frac{\Omega_K \delta^2 \xi_{\text{min}}^2}{(N(0)\lambda)^2} \sin \varphi$. The sign of such a current is reversed with respect to the conventional one.
Figure 3: $\xi_{\text{min}}$ vs. $\delta$ for various $j$, for $\varphi = 0$ (broken line) and $\varphi = \pi$ (full line). In the case $\varphi = 0$, for small values of $\delta$, $\xi_{\text{min}}$ is lowered, showing that a small amount of superconductivity singlet pairing favors Kondo correlations. On the other hand, for large $\delta$, $\xi$ increases abruptly, what signals the disruption of the Kondo correlations. By contrast, the weakening of the Kondo correlations is much steadier but less sharp when $\varphi = \pi$.

Figure 4: $\epsilon^*$ vs. $j$ for $\delta = 0.05, 0.15, 0.29$ in the two single cases $\varphi = 0$ (a) and $\varphi = \pi$ (b) and together (c) for a comparison.
Figure 5: $\epsilon$ vs. $\xi$ for $\delta = 0.05, 0.15, 0.20, 0.22, 0.25, 0.26, 0.27$ in the two cases $\varphi = 0$ (a) and $\varphi = \pi$ (b). (a) Deeper minima correspond to smaller $\delta$. For $\delta$ close to 0.3 the system will not be able to build up the Kondo singlet. (b) Deeper minima correspond to smaller $\delta$.

for Josephson systems, that is, the device behaves as a $\pi$-junction. In such a regime superconductivity starts to compete with Kondo correlations and the Josephson critical current comes out to be rather small, as we can see in Fig. 6a).

$\pi$-coupling wins in our model no matter how small the ratio $\Delta/k_B T_K$ is. This is counterintuitive because one expects that, when Kondo correlations are fully established and the magnetic moment at the dot is totally screened, the scattering of Cooper pairs should be potential-like only. This implies that no phase breaking takes place at the tunneling and the junction should behave as a conventional Josephson junction, with its energy minimum at $\varphi = 0$ [10]. This is not the case here because the density of states does not include extra subgap Andreev states localized at the dot which could provide the screening of the magnetic moment. We expect that, if Andreev bound states are accounted for properly in the model, they can provide the required full Kondo screening for vanishing $\Delta/k_B T_K$ ratio.

As $\delta$ increases, we enter a new regime in which superconductivity strongly competes with Kondo correlations. The Josephson critical current becomes now sizeable. When $\delta$ exceeds a threshold value (close to 0.3) the Josephson current develops strong non sinusoidal components (see Fig. 6b).

The regime $\Delta > T_K$, in which the superconducting order is dominant, cannot be described by our variational Ansatz. In this case, the Kondo inter-
action is better treated perturbatively. This regime was studied in ref. [10] using a NCA perturbative approach and in ref. [11]. Their results can be interpreted in terms of Andreev bound states in the normal region.

In the noninteracting limit the single particle energy spectrum for the S-N-S sandwich has two Andreev levels corresponding to states localized in the normal region with energy within the gap. Taking the Fermi level $\mu$ as the reference energy, the one of the two states with positive (negative) energy is mostly particle (hole)-like. In this case, Josephson conduction across the normal region involves both Andreev states. Because the levels are non-degenerate Kondo correlations cannot take place until $J$ is large enough. Kondo correlations require that the particle and hole-like states cross each other, so that screening of the impurity spin can take place at the dot site.

In conclusion, we have generalized the variational approach introduced in ref. [17] to study Josephson conduction through a quantum dot at Coulomb blockade connected to two superconducting leads. Within such an approach, we analyzed the region of parameters where the dot lays in the strongly coupled Kondo regime and showed that, in this regime, it behaves as a $\pi$-junction with a small Josephson current. Our formalism shows that the onset of $\pi$-junction regime takes place much before antiferromagnetic correlations at the dot can be treated perturbatively, that is, much before Kondo effect has been disrupted by superconductivity.
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