Influence of Paramagnetic and Kondo Impurities on Macroscopic Quantum Tunneling in SQUID's

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(Received 23 August 1988)

The possibility is investigated that impurity spins in SQUID tunnel junctions might have a strong dissipative effect on macroscopic flux tunneling. A microscopic theory is given which shows that for temperatures lower than the Kondo temperature T_K of the spins, there is a strong effect if $T_K \simeq \omega_J$, the Josephson plasma frequency. The connection with quantum measurement theory is discussed.

PACS numbers: 03.65.Bz, 74.50.+r

Much attention has been given recently to the possibility of macroscopic quantum tunneling (MQT) in SQUID systems.[1-3] Although MQT does not necessarily violate the Copenhagen interpretation of quantum mechanics,[4] its existence is still very surprising, since measurement theory indicates[5] that any interaction between the macroscopic tunneling coordinate (the flux) and the environmental coordinates, such that the environment registers (i.e., measures) the tunneling event, must suppress the tunneling. However, as discussed by Caldeira and Leggett,[1,2] and confirmed by various macroscopic analyses,[6] and by experiments,[3] the individual quasiparticle "environmental" modes are only very weakly perturbed by flux tunneling, thus rendering it observable. The fundamental consequences for quantum mechanics have been stressed by Leggett and coworkers.[1,2,7]

Nevertheless, suspicion remains—is no part of the environment able to register the tunneling? Consider, e.g., a two-state spin system; as is well known[4,5,7] (cf. Stern-Gerlach), such spins constitute almost ideal flux measurers, in properly designed experiments. Theory has so far ignored such "malevolent" coupling mechanisms to the environment, leaving room for skepticism. Moreover, the presence of paramagnetic impurities in most tunnel junctions makes such an investigation important in a practical sense. Here I give a detailed microscopic theory of the effect of paramagnetic and Kondo impurities, in a SQUID junction, on MQT; the conclusions are given below.

A fairly realistic model of such a junction is provided by the Appelbaum-Anderson Hamiltonian,[8] written as

$$\mathcal{H}_\tau = \sum_{pp'} \sum_{\alpha\beta} \{ T_0(pp') \psi^\dagger_{L\alpha}(p) \psi_{R\alpha}(p') + J_{pp'} T_J(pp') \hat{\mathbf{S}} \cdot [\psi^\dagger_{L\alpha}(p) \hat{\boldsymbol{\sigma}}^{\alpha\beta} \psi_{R\beta}(p')] + \text{H.c.} + J_{pp'} \hat{\mathbf{S}} \cdot [\psi^\dagger_{L\alpha}(p) \hat{\boldsymbol{\sigma}}^{\alpha\beta} \psi_{L\beta}(p') + \psi^\dagger_{R\alpha}(p) \hat{\boldsymbol{\sigma}}^{\alpha\beta} \psi_{R\beta}(p')] \} , \quad (1)$$

where the overlap between left (ψ_L) and right (ψ_R) wave functions gives both spin-assisted (via T_J) and ordinary tunneling (via T_0) as well as spin-flip reflection (via J).

The description of the system now requires a 4×4 Nambu Green's function[9]:

$$\hat{\mathbf{G}}(\mathbf{kk}';zz') = \hat{\mathbf{G}}_0(\mathbf{k},z)[\hat{\mathbf{I}} + \hat{\mathbf{t}}^s(\mathbf{kk}';zz')\hat{\mathbf{G}}_0(\mathbf{k},z)] , \quad (2)$$

$$\hat{\mathbf{G}}_0(\mathbf{k},z) = \frac{z + \hat{\tau}_3 \epsilon_\mathbf{k} + \hat{\tau}_2 \hat{\sigma}_y \Delta}{z^2 - \epsilon_\mathbf{k}^2 - \Delta^2} \equiv \begin{pmatrix} \mathcal{G}_0 & F_0 \\ F_0^\dagger & \tilde{\mathcal{G}}_0 \end{pmatrix}_{4\times 4} . \quad (3)$$

Here $\hat{\tau}_i$ and $\hat{\sigma}_j$ are Pauli matrices operating in particle-hole and spin spaces, respectively, and $\hat{\mathbf{t}}^s$ is the superconducting impurity T matrix, defined in terms of the normal-state $\hat{\mathbf{t}}^N$ by

$$\hat{\mathbf{t}}^s_{\mathbf{kk}'}(z,z') = \hat{\mathbf{t}}^N_{\mathbf{kk}'}(z,z') + \sum_{\mathbf{k}'',z''} \hat{\mathbf{t}}^N_{kk''}(z,z'')[\hat{\mathbf{G}}_0(k'',z'')\hat{\mathbf{g}}_0(k'',z'')]\hat{\mathbf{t}}^s_{\mathbf{k}''\mathbf{k}'}(z'',z') , \quad (4)$$

where $\hat{\mathbf{g}}_0(k,z) = (z - \hat{\tau}_3 \hat{\sigma}_0 \epsilon_k)^{-1}$ is the normal-state Green's function. All our problems arise from $\hat{\mathbf{t}}^N$, which contains the full complexity of the Kondo problem, and for which no general form is known. However, we can obtain results in three limits, viz. (i) $T \gg T_K$ (the Kondo temperature), (ii) $T \ll T_K$, and (iii) the classical limit $S \gg 1$ (with use of, e.g., a $1/N$ expansion, or otherwise). Results for the classical limit will be presented elsewhere—they are of no direct relevance here, since, as outlined in the introduction, we are interested in a two-state spin measuring system (i.e., $S = \frac{1}{2}$).

For the high- and low-temperature limits we can calculate all quantities in terms of the forward-scattering component $\hat{\mathbf{\Gamma}}_s$ of $\hat{\mathbf{t}}^s$, using dispersion relations following from unitarity.[10,11] For $T \gg T_K$ this has already been done[11]; assuming s-wave scattering (the classic Kondo problem), we have

$$\hat{\Gamma}_s(z) = \frac{3J^2}{8\pi}\int_{-\infty}^{\infty}\frac{d\epsilon}{z-\epsilon}\left[\hat{B}(\epsilon) - \frac{J}{\pi}\int_{-\infty}^{\infty} d\epsilon_2 \frac{\hat{B}(\epsilon_1)\hat{B}(\epsilon_2)}{(z-\epsilon_1)(\epsilon_1-\epsilon_2)}\tanh(\tfrac{1}{2}\beta\epsilon_2)\right], \tag{5}$$

$$\hat{B}(\epsilon) = \frac{\pi N(0)}{(\epsilon-\Delta^2)^{1/2}}(\hat{\tau}_0\epsilon + \hat{\tau}_2\hat{\sigma}_y\Delta)\theta(\epsilon^2-\Delta^2). \tag{6}$$

For the opposite limit $T \ll T_K$, no results appear to have been given. Calculating directly from (4), using Nozières Fermi-liquid theory,[12] for which

$$\hat{\mathbf{t}}^N(\omega+i\delta) = -[\hat{\tau}_3/\pi N(0)]e^{i\delta_0(\hat{\tau}_3\omega)}\sin\delta_0(\hat{\tau}_3\omega), \tag{7}$$

and assuming that $\delta_0(\omega) = \pi/2 - \omega/T_K$ [i.e., no potential scattering; this is consistent with Eq. (1)], we get (again using unitarity restrictions)

$$\mathrm{Im}\hat{\Gamma}_s(\omega+i\delta) = -\frac{1}{N}\frac{(\omega^2-\Delta^2)^{1/2}}{\pi N(0)\Delta}\theta(\omega^2-\Delta^2)\frac{\omega^2/(T_K^2+\omega^2)[\hat{\tau}_0\omega+\hat{\tau}_2\hat{\sigma}_y\Delta]}{\omega^2-\Delta^2[(T_K^2-\omega^2)/(T_K^2+\omega^2)]^2}, \tag{8}$$

where we assume N lattice sites per unit volume, and a Fermi surface density of states $N(0)$. Strictly speaking, (8) is only valid for $|\omega| \ll T_K$. Its use at higher frequencies amounts to use of the "resonance model" of the Kondo effect, which is known to give a good approximate description for $T \ll T_K$.[13]

To examine the MQT properties, we calculate the imaginary-time action ($t \to i\tau$) contribution S_τ arising from $\mathcal{H}_\tau$ in (1), to second order in $\mathcal{H}_\tau$. This is a generalization of previous work,[6] except we now need 8×8 matrices of the form

$$\hat{\mathbf{G}}_{\mathbf{kk}'}^{-1}(z,z') = \begin{pmatrix} \hat{\mathbf{G}}_L^{-1} & -\hat{\mathcal{T}} \\ -\hat{\mathcal{T}}^\dagger & \hat{\mathbf{G}}_R^{-1}\end{pmatrix}_{8\times 8} \equiv \hat{\mathbf{G}}_0^{-1}(\mathbf{k},\mathbf{k}';z,z') - \hat{\mathcal{T}}_{\mathbf{kk}'}(zz'), \tag{9}$$

where the left and right 4×4 matrices $\hat{\mathbf{G}}_{L,R}$ are given by (2). The result is

$$S_\tau = \frac{1}{\hbar}\int_0^{\hbar\beta} d\tau_1 \int_0^{\hbar\beta} d\tau_2 [\alpha(\tau_1-\tau_2)\cos\Psi(\tau_1,\tau_2) - \beta(\tau_1-\tau_2)\cos\Phi(\tau_1,\tau_2)], \tag{10}$$

where $\Psi(\tau,\tau') = \frac{1}{2}[\phi(\tau)-\phi(\tau')]$, $\Phi(\tau,\tau) = \frac{1}{2}[\phi(\tau)+\phi(\tau')]$, and $\phi(\tau)$ is the relative junction phase. $\alpha(\tau)$ and $\beta(\tau)$ are now complicated functions of $\hat{\Gamma}_s(z)$, whose general form is shown in Fig. 1. In addition to the terms $\alpha_0(\tau)$ and $\beta_0(\tau)$ of Refs. 7 [Fig. 1(a) shows $\beta_0(\tau)$], we have terms $\alpha_J(\tau)$, $\beta_J(\tau)$ [Fig. 1(b)] coming from $\mathcal{H}_\tau$ in (1); for a single impurity spin, we find the following, for $\hbar\beta > \tau > 0$ (and again using dispersion relations):

$$\alpha_J(\tau) = \frac{-2}{\hbar^2 N}\int_\Delta^\infty d\omega_1 \int_\Delta^\infty d\omega_2 \frac{\omega_1\omega_2 e^{-(\omega_1+\omega_2)\tau/\hbar}}{(\omega_1^2-\Delta^2)^{1/2}(\omega_2^2-\Delta^2)^{1/2}} n(\omega_2-\omega_1)[f(\omega_2)-f(\omega_1)]\pi N(0)$$
$$\times \mathrm{Im}\left\{|\tilde{T}_0|^2\left[\frac{\omega_1+\Delta}{\omega_1-\Delta}\Gamma_1^s(\omega_1+i\delta) + 2\omega_1\Delta\Gamma_2^s(\omega_1+i\delta)\right] + |\tilde{T}_J|^2\Gamma_2^s(\omega_1+i\delta)\right\}, \tag{11}$$

where we have written

$$\hat{\Gamma}_s(z) = \hat{\tau}_0\Gamma_1^s(z) + \hat{\tau}_2^s\hat{\sigma}_y\Gamma_2^s(z);$$

$|\tilde{T}_0|^2 \equiv N^2(0)\langle|T_0(\mathbf{p}\cdot\mathbf{p}')|^2\rangle$, and $f(\omega)$, $n(\omega)$ are Fermi and Bose functions. $\beta_J(\tau)$ is given by a similar formula, but with Δ^2 replacing the product $\omega_1\omega_2$ just after the integration symbols, and a minus sign in front of $|\tilde{T}_J|^2$.

Equation (11) is exact for $T \gg T_K$ and $T \ll T_K$, and gives us formulas for S_τ in the two limiting cases (a) $T \gg T_K$, T_K arbitrary, and (b) $T \ll T_K$ and $T_K \gg \Delta$ [this latter restriction because (8) is only strictly valid for $|\omega| \ll T_K$]. Unfortunately, these results are of no real interest, because in these regimes the Kondo resonance has little effect [in (a) it is completely smeared out (cf. Eq. (5)] while in (6) the scattering is essentially elastic and irrelevant to our problem. Moreover, Eq. (11) is $\sim O(1/N)$, and thus not important for any temperature; thus we reach the conclusion that a single junction impurity spin will have a negligible effect on MQT.

We now observe that any real junction will have many randomly distributed impurities, with a concentration c. The randomness is important; as is well known,[15] an average over the impurity positions then leaves only the forward-scattering part of $\hat{\mathbf{t}}^s$, provided $c \ll 1$. *Hence Eq. (11) is valid for all temperatures* (provided we replace $1/N$ by c) in a realistic type of situation. In the experimentally interesting situation where $kT \ll \Delta$, but T_K is arbitrary, we may obtain approximate formulas for α_J and β_J by using the "resonance" expression (8). The general result must be evaluated numerically,[16] but we

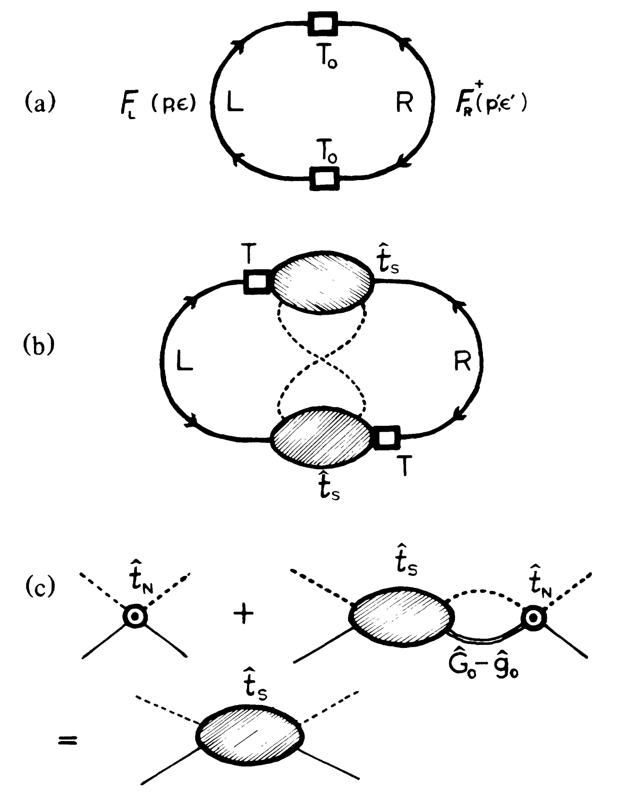

FIG. 1. The contributions to $\beta(\tau)$, represented diagrammatically. (a) The term $\beta_0(\tau)$ in the absence of spin impurities. (b) The impurity contribution $\beta_J(\tau)$ in terms of the superconducting T matrix, which satisfies (c) the integral equation; the Abrikosov diagrammatic convention (Ref. 14) is used. (b) is somewhat schematic, since there are several spin channels involved.

are really interested in any regime for which S_τ will be strongly affected. This requires knowing typical values of $\partial\phi/\partial\tau$ in (10), i.e., the typical (imaginary) frequencies involved in the tunneling process. These are given by 1/(bounce time), i.e., by the Josephson plasma frequency ω_J (typical values of which are $\hbar\omega_J \simeq \Delta/20$). Then detailed evaluation of (11) shows that while both $\alpha_J(\tau)$ and $\beta_J(\tau)$ decay over similar time scales to $\alpha_0(\tau)$ [i.e., time scales $\sim\hbar/\Delta$, arising from the $\theta(\omega^2-\Delta^2)$ functions], nevertheless if $T_K\sim\omega_J$, $\alpha_J(\tau)$ and $\beta_J(\tau)$ are multipled by large factors, whose final effect is to add a term

$$S_\tau^{(J)} = \frac{\hbar}{16} c \frac{\Delta^2}{\omega_J^2} \int_0^{\mathrm{tr}} d\tau_1 \left[\frac{\partial\phi}{\partial\tau_1}\right]^2 \int_0^{\mathrm{tr}\beta} d\tau_2\, \tau_2^2 \alpha_0(\tau_2) \qquad (12)$$

to the contribution already arising from ordinary tunneling; a similar term arises from $\beta_J(\tau)$, and there are nonlocal terms in $\tau_1\pm\tau_2$ which are omitted here for simplicity.[16] In this regime there is no contribution from T_J, and so we may simply absorb the effects of $S_\tau^{(J)}$ into a capacitance correction C_J:

$$C_J = \frac{3\pi\hbar}{128 R_N\Delta} c \left[\frac{\Delta^2}{\omega_J^2}\right] (1-\tfrac{1}{3}\cos\phi)\,, \qquad (13)$$

where $R_N^{-1} = 4\pi e^2 |\tilde{T}_0|^2/\hbar$ is the phenomenological junction conductance in the absence of the spin impurities; i.e., we have increased the "effective mass" of the instanton and thereby reduced the tunneling rate.

Since Δ^2/ω_J^2 is quite large, we see that the dissipative effect of even a low concentration of impurity spins should be very large (equivalent to the entire effect of the junction in their absence) provided that T_K is correctly tuned to ω_J. This would be a very interesting test of the above theory. In practice, typical values of ω_J are in the range 1-20 GHz (i.e., 0.05-1 K). Thus for a Nb point contact junction ($\Delta\sim 16$ K) with ω_J adjusted to resonate with Kondo impurities having $T_K\sim 0.05$ K, the effective capacitance of the junction will be doubled by an impurity concentration c of only 10^{-5}.

However, we have not achieved the complete suppression of tunneling that the simple-minded argument of the introduction implies (and certainly not for a single spin). Why not? The essential point[16] is that for a single spin to suppress tunneling, it must be coupled to a large external system (a "measuring apparatus") in such a way that this system also registers the tunneling. If the apparatus is sufficiently massive, it will give a further contribution to S_τ which will entirely suppress the tunneling, in line with the usual ideas[5] of measurement theory. We also see that the Kondo spins do not constitute "ideal measuring apparati," because they are coupled to other degrees of freedom apart from the fluxon. Thus they register the instanton passage in a fairly inefficient way. It is interesting to note that one way to increase both this efficiency and the spin contribution to S_{eff} would be to use very small junctions. In this way the spins will be coupled to the electronic charge in the vicinity of the junction (i.e., directly to the capacitance; this is the "Coulomb blockade"). It then becomes quite feasible that a single resonant level may be used to measure (and thereby entirely suppress) MQT in SQUID's. This and the effect of applied field will be discussed in more detail elsewhere.[16]

I would like to thank Dr. B. Barbara for encouragement during the period this work was done.

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[13]See, e.g., K. D. Schotte and U. Schotte, Phys. Lett. **55A**, 38 (1975); a more sophisticated version is in P. Schlottmann, Phys. Rev. B **25**, 4815, 4828, 4838 (1982). In fact, Eq. (8) has a much wider validity than is clear at first. Thus it is also exact in the limit $T_K \ll \Delta$, and $T \ll T_K$, for $\omega \sim \Delta$. This is because the energy of the bound state in (8) is forced up to Δ as $T_K \to 0$, and their energy difference is not affected in this limit by the high-ω behavior of $\hat{t}^N(\omega + i\delta)$. This is the region of validity we shall need to derive Eq. (12).

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[16]A much more detailed version of this work will be published elsewhere (including a discussion of other resonant scattering mechanisms).