Spin-dependent Josephson current through double quantum dots and measurement of entangled electron states

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We study a double quantum dot, each dot of which is tunnel coupled to superconducting leads. In the Coulomb blockade regime, a spin-dependent Josephson coupling between two superconductors is induced, as well as an antiferromagnetic Heisenberg exchange coupling between the spins on the double dot that can be tuned by the superconducting phase difference. We show that the correlated spin states—singlet or triplets—on the double dot can be probed via the Josephson current in a dc-superconducting quantum interference device setup.

INTRODUCTION

In recent years, electronic transport through strongly interacting mesoscopic systems has been the focus of many investigations.¹ In particular, a single quantum dot coupled via tunnel junctions to two noninteracting leads has provided a prototype model to study Coulomb blockade effects and resonant tunneling in such systems. These studies have been extended to an Anderson impurity² or a quantum dot coupled to superconductors.^{3–5} In a number of experimental³ and theoretical⁴ papers, the spectroscopic properties of a quantum dot coupled to two superconductors have been studied. Further, an effective dc Josephson effect through strongly interacting regions between superconducting leads has been analyzed.⁶⁻⁹ More recently, on the other hand, research on the possibility to control and detect the spin of electrons through their charges has started. In particular in semiconducting nanostructures, it was found that the direct coupling of two quantum dots by a tunnel junction can be used to create entanglement between spins,¹⁰ and that such spin correlations can be observed in charge transport experiments.¹¹

Motivated by these studies we propose in the present work a scenario for inducing and detecting spin correlations, viz., coupling a double quantum dot (DD) to superconducting leads by tunnel junctions as shown in Fig. 1. It turns out that this connection via a superconductor induces a Heisenberg exchange coupling between the two spins on the DD. Moreover, if the DD is arranged between two superconductors (see Fig. 1), we obtain a Josephson junction (*S*-DD-*S*). The resulting Josephson current depends on the spin state of the DD and can be used to *probe* the spin correlations on the DD.

MODEL

The double-dot (DD) system we propose is sketched in Fig. 1: Two quantum dots (a,b), each of which contains one (excess) electron and is connected to two superconducting leads (L,R) by tunnel junctions (indicated by dashed lines).¹² There is no direct coupling between the two dots. The Hamiltonian describing this system consists of three parts, $H_S+H_D+H_T \equiv H_0+H_T$. The leads are assumed to be

conventional singlet superconductors that are described by the BCS Hamiltonian

$$H_{S} = \sum_{j=L,R} \int_{\Omega_{j}} \frac{d\mathbf{r}}{\Omega_{j}} \left\{ \sum_{\sigma=\uparrow,\downarrow} \psi_{\sigma}^{\dagger}(\mathbf{r})h(\mathbf{r})\psi_{\sigma}(\mathbf{r}) + \Delta_{j}(\mathbf{r})\psi_{\uparrow}^{\dagger}(\mathbf{r})\psi_{\downarrow}^{\dagger}(\mathbf{r}) + \text{H.c.} \right\},$$
(1)

where Ω_j is the volume of lead j, $h(\mathbf{r}) = (-i\hbar\nabla + [e/c]\mathbf{A})^2/2m - \mu$, and $\Delta_j(\mathbf{r}) = \Delta_j e^{-i\phi_j(\mathbf{r})}$ is the pair potential. For simplicity, we assume identical leads with the same chemical potential $-\mu$, and $\Delta_L = \Delta_R = \Delta$. The two quantum dots are modeled as two localized levels ϵ_a and ϵ_b with strong on-site Coulomb repulsion U, described by the Hamiltonian

$$H_{D} = \sum_{n=a,b} \left[-\epsilon \sum_{\sigma} d^{\dagger}_{n\sigma} d_{n\sigma} + U d^{\dagger}_{n\uparrow} d_{n\uparrow} d^{\dagger}_{n\downarrow} d_{n\downarrow} \right], \quad (2)$$
$$a$$



FIG. 1. Upper panel: sketch of the superconductor-double quantum dot-superconductor (*S*-DD-*S*) nanostructure. Lower panel: schematic representation of the quasiparticle energy spectrum in the superconductors and the single-electron levels of the two quantum dots.

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where we put $\epsilon_a = \epsilon_b = -\epsilon$ ($\epsilon > 0$) for simplicity. *U* is typically given by the charging energy of the dots, and we have assumed that the level spacing of the dots is $\sim U$ (which is the case for small GaAs dots¹), so that we need to retain only one energy level in H_D . Finally, the DD is coupled *in parallel* (see Fig. 1) to the superconducting leads, described by the tunneling Hamiltonian

$$H_T = \sum_{j,n,\sigma} \left[t \exp\left(-\frac{i\pi}{\Phi_0} \int_{\mathbf{r}_n}^{\mathbf{r}_{j,n}} dl \cdot \mathbf{A}\right) \psi_{\sigma}^{\dagger}(\mathbf{r}_{j,n}) d_{n\sigma} + \text{H.c.} \right],$$
(3)

where $\mathbf{r}_{j,n}$ is the point on the lead *j* closest to the dot *n*. Here, $\Phi_0 = hc/2e$ is the superconducting flux quantum. Unless mentioned otherwise, it will be assumed that $\mathbf{r}_{L,a} = \mathbf{r}_{L,b} = \mathbf{r}_L$ and $\mathbf{r}_{R,a} = \mathbf{r}_{R,b} = \mathbf{r}_R$.

Since the low-energy states of the whole system are well separated by the superconducting gap Δ as well as the strong Coulomb repulsion $U(\Delta, \epsilon \ll U - \epsilon)$, it is sufficient to consider an effective Hamiltonian on the reduced Hilbert space consisting of singly occupied levels of the dots and the BCS ground states on the leads. To lowest order in H_T , the effective Hamiltonian is

$$H_{eff} = P H_T [(E_0 - H_0)^{-1} (1 - P) H_T]^3 P, \qquad (4)$$

where *P* is the projection operator onto the subspace and E_0 is the ground-state energy of the unperturbed Hamiltonian H_0 . (The second-order contribution leads to an irrelevant constant.) The lowest-order expansion (4) is valid in the limit $\Gamma \ll \Delta, \epsilon$ where $\Gamma = \pi t^2 N(0)$ and N(0) is the normal-state density of states per spin of the leads at the Fermi energy. Thus, we assume that $\Gamma \ll \Delta, \epsilon \ll U - \epsilon$, and temperatures that are less than ϵ (but larger than the Kondo temperature).

EFFECTIVE HAMILTONIAN

There are a number of virtual hopping processes that contribute to the effective Hamiltonian (4); see Fig. 2 for a partial listing of them. Collecting these various processes, one can get the effective Hamiltonian in terms of the gaugeinvariant phase differences ϕ and φ between the superconducting leads and the spin operators \mathbf{S}_a and \mathbf{S}_b of the dots (up to a constant and with $\hbar = 1$):

$$H_{eff} = J_0 \cos(\pi f_{AB}) \cos(\phi - \pi f_{AB}) + [(2J_0 + J)(1 + \cos \varphi)$$

$$+2J_{1}(1+\cos \pi f_{AB})][\mathbf{S}_{a} \cdot \mathbf{S}_{b}-1/4].$$
(5)

Here, $f_{AB} = \Phi_{AB}/\Phi_0$ and Φ_{AB} is the Aharonov-Bohm (AB) flux threading through the closed loop indicated by the dashed lines in Fig. 1. One should be careful to define *gauge-invariant* phase differences ϕ and φ in Eq. (5). The phase difference ϕ is defined as usual¹³ by

$$\phi = \phi_L(\mathbf{r}_L) - \phi_R(\mathbf{r}_R) - \frac{2\pi}{\Phi_0} \int_{\mathbf{r}_R}^{\mathbf{r}_L} dl_a \cdot \mathbf{A}, \tag{6}$$

where the integration from \mathbf{r}_R to \mathbf{r}_L runs via dot *a* (see Fig. 1). The second phase difference, φ , is defined by

$$\varphi = \phi_L(\mathbf{r}_L) - \phi_R(\mathbf{r}_R) - \frac{\pi}{\Phi_0} \int_{\mathbf{r}_R}^{\mathbf{r}_L} (dl_a + dl_b) \cdot \mathbf{A}.$$
(7)



FIG. 2. Partial listing of virtual tunneling processes contributing to H_{eff} (4). The numbered arrows indicate the direction and the order of occurrence of the charge transfers. Processes of type (a) and (b) give a contribution proportional to J_0 , whereas those of type (c) and (d) give contributions proportional to J. Other processes not listed here give negligible contributions in the energy regions of interest.

The distinction between ϕ and φ , however, is not significant unless one is interested in the effects of an AB flux through the closed loop in Fig. 1 (see Ref. 11 for an example of such effects). The coupling constants appearing in Eq. (5) are defined by

$$J = \frac{2\Gamma^2}{\epsilon} \left[\frac{1}{\pi} \int \frac{dx}{f(x)g(x)} \right]^2,$$

$$J_0 = \frac{\Gamma^2}{\Delta} \int \frac{dxdy}{\pi^2} \frac{1}{f(x)f(y)[f(x)+f(y)]g(x)g(y)}, \quad (8)$$

$$I_1 = \frac{\Gamma^2}{\Delta} \int \frac{dxdy}{\pi^2} \frac{g(x)[f(x)+f(y)]-2\zeta g(y)}{g(x)^2 g(y)[g(x)+g(y)][f(x)+f(y)]},$$

where $\zeta = \epsilon/\Delta$, $f(x) = \sqrt{1+x^2}$, and $g(x) = \sqrt{1+x^2} + \zeta$.

J

Equation (5) is one of our main results. A remarkable feature of it is that a Heisenberg exchange coupling between the spin on dot *a* and on dot *b* is induced by the superconductor. This coupling is antiferromagnetic (all *J*'s are positive) and thus favors a singlet ground state of spin *a* and *b*. This in turn is a direct consequence of the assumed singlet nature of the Cooper pairs in the superconductor.¹⁴ As discussed below, an immediate observable consequence of H_{eff} is a *spin-dependent* Josephson current from the left to right superconducting lead (see Fig. 1), which probes the correlated spin state on the DD.

The various terms in Eq. (5) have different magnitudes. In particular, the processes leading to the J_1 term involve qua-

siparticles only as can be seen from its AB-flux dependence, which has period $2\Phi_0$. In the limits we will consider below, this J_1 term is small and can be neglected.

In the limit $\zeta \ge 1$, the main contributions come from processes of the type depicted in Figs. 2(a) and 2(b), making $J_0 \approx 0.1(\Gamma^2/\zeta\epsilon) \ln \zeta$ dominant over J and J_1 . Thus, Eq. (5) can be reduced to

$$H_{eff} \approx J_0 \cos(\pi f_{AB}) \cos(\phi - \pi f_{AB}) + 2J_0 (1 + \cos\varphi) \left[\mathbf{S}_a \cdot \mathbf{S}_b - \frac{1}{4} \right], \tag{9}$$

up to order $(\ln \zeta)/\zeta$. As can be seen in Fig. 2(a), the first term in Eq. (9) has the same origin as that in the single-dot case:² Each dot separately constitutes an effective Josephson junction with coupling energy $-J_0/2$ (i.e., π junction) between the two superconductors. The two resulting junctions form a dc superconducting quantum interference device (SQUID), leading to the total Josephson coupling in the first term of Eq. (9). The Josephson coupling in the second term in Eq. (9), corresponding to processes of type Fig. 2(b), depends on the correlated spin states on the double dot: For the singlet state, it gives an ordinary Josephson junction with coupling $2J_0$ and competes with the first term, whereas it vanishes for the triplet states. Although the limit $\Delta \ll \epsilon \ll U - \epsilon$ is not easy to achieve with present-day technology, such a regime is relevant, say, for two atomic impurities embedded between the grains of a granular superconductor.

More interesting and experimentally feasible is the case $\zeta \ll 1$. In this regime, the effective Hamiltonian (5) is dominated by a single term (up to terms of order ζ),

$$H_{eff} \approx J(1 + \cos \varphi) \left[\mathbf{S}_a \cdot \mathbf{S}_b - \frac{1}{4} \right], \tag{10}$$

with $J \approx 2\Gamma^2/\epsilon$. The processes of type Figs. 2(b) and 2(c) give rise to Eq. (10). Below we will propose an experimental setup based on Eq. (10).

Before proceeding, we digress briefly on the dependence of *J* on the contact points. Unlike the processes of type Fig. 2(a), those of types Figs. 2(b), 2(c), and 2(d) depend on $\delta r_L = |\mathbf{r}_{L,a} - \mathbf{r}_{L,b}|$ and $\delta r_R = |\mathbf{r}_{R,a} - \mathbf{r}_{R,b}|$, see the remark below Eq. (3). For the tunneling Hamiltonian (3), one gets (putting $\delta r = \delta r_L = \delta r_R$)

$$J(\delta r) = \frac{8t^4}{\epsilon} \left| \int_0^\infty \frac{d\omega}{2\pi} \frac{F^R(\delta r, \omega) - F^A(\delta r, \omega)}{\omega + \epsilon} \right|^2, \quad (11)$$

where $F^{R/A}(\mathbf{r},\omega)$ is the Fourier transform of the Green's function in the superconductors,¹⁵

$$F^{R/A}(\mathbf{r},t) = \pm i\Theta(\pm t) \langle \{\psi_{\uparrow}(\mathbf{r},t),\psi_{\downarrow}(0,0)\} \rangle.$$

For example, in the limit $\varepsilon \ll \Delta \ll \mu$, we find $J(\delta r) \approx J(0)e^{-2\delta r/\xi} \sin^2(k_F \delta r)/(k_F \delta r)^2$ up to order $1/k_F \xi$, with k_F the Fermi wave vector in the leads. Thus, to have $J(\delta r)$ nonzero, δr should not exceed the superconducting coherence length ξ .



FIG. 3. dc-SQUID-like geometry consisting of the *S*-DD-*S* structure (filled dots at the top) connected in parallel with another ordinary Josephson junction (cross at the bottom).

PROBING SPINS WITH A dc SQUID

We now propose a possible experimental setup to probe the correlations (entanglement) of the spins on the dots, based on the effective model (10). According to Eq. (10) the *S*-DD-*S* structure can be regarded as a *spin-dependent* Josephson junction. Moreover, this structure can be connected with an ordinary Josephson junction to form a dc-SQUIDlike geometry, see Fig. 3. The Hamiltonian of the entire system is then given by

$$H = J[1 + \cos(\theta - 2\pi f)] \left(\mathbf{S}_a \cdot \mathbf{S}_b - \frac{1}{4} \right) + \alpha J(1 - \cos\theta),$$
(12)

where $f = \Phi/\Phi_0$, Φ is the flux threading the SQUID loop, θ is the gauge-invariant phase difference across the auxiliary junction (J'), and $\alpha = J'/J$ with J' being the Josephson coupling energy of the auxiliary junction.¹⁶ One immediate consequence of Eq. (12) is that at zero temperature, we can *effectively* turn on and off the spin exchange interaction: For half-integer flux (f=1/2), singlet and triplet states are degenerate at $\theta=0$. Even at finite temperatures, where θ is subject to thermal fluctuations, singlet and triplet states are almost degenerate around $\theta=0$. On the other hand, for integer flux (f=0), the energy of the singlet is lower by J than that of the triplets.

This observation allows us to probe directly the spin state on the double dot via a Josephson current across the dc-SQUID-like structure in Fig. 3. The supercurrent through the SQUID ring is defined as $I_S = (2 \pi c / \Phi_0) \partial \langle H \rangle / \partial \theta$, where the brackets refer to a spin expectation value on the DD. Thus, depending on the spin state on the DD we find

$$I_S/I_J = \begin{cases} \sin(\theta - 2\pi f) + \alpha \sin\theta & \text{(singlet)} \\ \alpha \sin\theta & \text{(triplets),} \end{cases}$$
(13)

where $I_J = 2eJ/\hbar$. When the system is biased by a dc current *I* larger than the spin- and flux-dependent critical current, given by $\max_{\theta}\{|I_S|\}$, a finite voltage *V* appears. Then one possible experimental procedure might be as follows (see Fig. 4). Apply a dc bias current such that $\alpha I_J < I < (\alpha + 1)I_J$. Here, αI_J is the critical current of the triplet states,



FIG. 4. Schematic representation of dc voltage V vs time when probing the spin correlations of the DD. The flux through the SQUID loop is switched from f=1/2 to f=0 at t=0. Solid line: $\tau_{sw} < \tau_{spin}$. Dashed line: $\tau_{sw} > \tau_{spin}$.

and $(\alpha + 1)I_J$ the critical current of the singlet state at f = 0, see Eq. (13). Initially prepare the system in an equal mixture of singlet and triplet states by tuning the flux around f=1/2. (With electron g factors $g \sim 0.5-20$ the Zeeman splitting on the dots is usually small compared with k_BT and can thus be ignored.) The dc voltage measured in this mixture will be given by $(V_0+3V_1)/4$, where $V_0(V_1)\sim 2\Delta/e$ is the (current-dependent) voltage drop associated with the singlet (triplet) states. At a later time t=0, the flux is switched off (i.e. f=0), with I being kept fixed. The ensuing time evolution of the system is characterized by three time scales: the time $\tau_{coh} \sim \max\{1/\Delta, 1/\Gamma\} \sim 1/\Gamma$ it takes to establish coherence in the S-DD-S junction, the spin relaxation time τ_{spin} on

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the dot, and the switching time τ_{sw} to reach f=0. We will assume $\tau_{coh} \ll \tau_{spin}, \tau_{sw}$, which is not unrealistic in view of measured spin decoherence times in GaAs exceeding 100 ns.¹⁷ If $\tau_{sw} \ll \tau_{spin}$, the voltage is given by $3V_1/4$ for times less than τ_{spin} , i.e., the singlet no longer contributes to the voltage. For $t > \tau_{spin}$, the spins have relaxed to their ground (singlet) state, and the voltage vanishes. One therefore expects steps in the voltage versus time (solid curve in Fig. 4). If $\tau_{spin} \ll \tau_{sw}$, a broad transition region of the voltage from the initial value to 0 will occur (dashed line in Fig. 4).¹⁸

To our knowledge, there are no experimental reports on quantum dots coupled to superconductors. However, hybrid systems consisting of superconductors (e.g., Al or Nb) and 2DES (InAs and GaAs) have been investigated by a number of groups.¹⁹ Taking the parameters of those materials, a rough estimate leads to a coupling energy J in Eq. (10) or Eq. (12) of about $J \sim 0.05 - 0.5$ K. This corresponds to a critical current scale of $I_I \sim 5-50$ nA.

In conclusion, we have investigated double quantum dots, each dot of which is coupled to two superconductors. We have found that in the Coulomb blockade regime the Josephson current from one superconducting lead to the other is different for singlet or triplet states on the double dot. This leads to the possibility to probe the spin states of the dot electrons by measuring a Josephson current.

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- ¹⁴If, instead we had assumed leads consisting of unconventional superconductors with triplet pairing, we would find a *ferromagnetic* exchange coupling favoring a triplet ground state of spin a and b on the DD. Thus, by probing the spin ground state of the dots (e.g., via its magnetic moment) we would have a means to distinguish singlet from triplet pairing. The magnetization could be made sufficiently large by extending the scheme from two to N dots coupled to the superconductor.
- ¹⁵We note that the phase difference φ in Eq. (5) should now be defined with respect to the phase $\phi_j(\mathbf{r}_{j,a}, \mathbf{r}_{j,b})$ of the function $F_j^R(\mathbf{r}_{j,a}, \mathbf{r}_{j,b}) F_j^A(\mathbf{r}_{j,a}, \mathbf{r}_{j,b})$ on the lead *j*, see the definition below Eq. (5).
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