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 which 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-SQUID setup.

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In recent years, electronic transport through strongly interacting mesoscopic systems has been the focus of many investigations. In particular, a single quantum dot coupled via tunnel junctions to non-interacting leads has provided a prototype model to study Coulomb blockade effects and resonant tunneling in such systems. These studies have been extended to Anderson impurity or a quantum dot coupled to superconductors in a number of experimental and theoretical papers, the spectroscopic properties of a quantum dot coupled to two superconductors have been studied. Further, an effective direct Josephson effect through strongly interacting regions between superconducting leads has been analyzed. More recently, on the other hand, research on the possibility to control and detect the spin states—singlet or triplets—on the double dot can be tuned by the superconducting phase difference. We show that the correlated spin states on the DD are induced, as well as an antiferromagnetic Heisenberg exchange coupling between the spins on the double dot.

**Model**—The double-dot (DD) system we propose is the prototype model to study Coulomb blockade effects and resonant tunneling in such systems. The leads are assumed to be conventional singlet superconductors that are described by the BCS Hamiltonian.

![Diagram of double quantum dot and superconducting leads](image)

**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.

The Hamiltonian describing this system-consists of three parts, \( H_S + H_D + H_T = H_0 + H_T \). The leads are assumed to be conventional singlet superconductors that are described by the BCS Hamiltonian

\[
H_S = \sum_{j=L,R} \int d^3 \mathbf{r} \frac{1}{\Omega_j} \left\{ \sum_{\sigma=\uparrow,\downarrow} \psi_\sigma^\dagger(\mathbf{r}) h(\mathbf{r}) \psi_\sigma(\mathbf{r}) \right. \\
+ \Delta_j(\mathbf{r}) \psi_\uparrow^\dagger(\mathbf{r}) \psi_\downarrow^\dagger(\mathbf{r}) + h.c. \right\}, \tag{1}
\]

where \( \Omega_j \) is the volume of lead \( j \), \( h(\mathbf{r}) = (-i\hbar \nabla + \frac{\hbar^2 \mathbf{A}^2}{2m} - \mu) \), 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 modelled as two localized levels \( \epsilon_a \) and \( \epsilon_b \) with strong on-site Coulomb repulsion \( U \), described by the Hamiltonian

\[
H_D = \sum_{n=a,b} \left[ -\epsilon_n \sum_{\sigma} d_{n\sigma}^\dagger d_{n\sigma} + U d_{n\uparrow}^\dagger d_{n\downarrow}^\dagger d_{n\downarrow} d_{n\uparrow} \right], \tag{2}
\]

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 dots \cite{1}), 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

$$H_T = \sum_{j,n,\sigma} \left[ t e^{-i \pi \sigma} \int_{r_n} r_{j,n} d \mathbf{A} \psi_{j,n,\sigma}^\dagger(\mathbf{r}_{j,n}) d_{n,\sigma} + h.c. \right],$$  

(3)

where $\mathbf{r}_{j,n}$ is the point on the lead $j$ closest to the dot $n$. Here, $\Phi_0 = \hbar c/2e$ 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 $\Delta$ 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

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

(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 2nd order contribution leads to an irrelevant constant.) The lowest-order expansion \cite{1} is valid in the limit $\Gamma \ll \Delta, \epsilon$ where $\Gamma = \pi \ell^2 N(0)$ 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 which are less than $\epsilon$ (but larger than the Kondo temperature).

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

$$H_{\text{eff}} = J_0 \cos(\pi f_{AB}) \cos(\phi - \pi f_{AB})$$  

$$+ \left[ (2J_0 + J)(1 + \cos \varphi) + 2J_1(1 + \cos \pi f_{AB}) \right] \times [\mathbf{S}_a \cdot \mathbf{S}_b - 1/4].$$

(5)

Here $f_{AB} = \Phi_{AB}/\Phi_0$ and $\Phi_{AB}$ 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 $\phi$ and $\varphi$ in \cite{4}. The phase difference $\phi$ is defined as usual \cite{3} by

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

(6)

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

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

(7)

The distinction between $\phi$ and $\varphi$, however, is not significant unless one is interested in the effects of an AB flux through the closed loop in Fig. 1 (see Ref. \cite{4} for an example of such effects). The coupling constants appearing in \cite{4} are defined by

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

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

$$J_1 = \frac{\Gamma^2}{\pi^2} \int \frac{dxdy}{g(x)^2g(y)[g(x) + g(y)][f(x) + f(y)]},$$

(8)

where $\zeta = \epsilon/\Delta$, $f(x) = \sqrt{1 + x^2}$, and $g(x) = \sqrt{1 + x^2 + \zeta}$. Eq. (8) 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 \cite{1}. As discussed below, an immediate observable consequence of $H_{\text{eff}}$ 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 \cite{4} have different magnitudes. In particular, the processes leading to the $J_1$ term involve

**FIG. 2.** Partial listing of virtual tunneling processes contributing to $H_{\text{eff}}$ \cite{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.
quasiparticles 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 $\zeta \gg 1$, the main contributions come from processes of the type depicted in Fig. 3 (a) and (b), making $J_0 \approx 0.1(\Gamma^2/\zeta \epsilon) \log \zeta$ dominant over $J$ and $J_1$. Thus, (8) can be reduced to

$$H_{\text{eff}} \approx J_0 \cos(\pi f_{AB}) \cos(\phi - \pi f_{AB}) + 2J_0(1 + \cos \varphi) \left[ S_a \cdot S_b - \frac{1}{4} \right],$$

up to order $(\log \zeta)/\zeta$. As can be seen in Fig. 3 (a), the first term in (8) has the same origin as that in the singlet-dot case [3]: Each dot separately constitutes an effective Josephson junction with coupling energy $-J_0/2$ (i.e., pairing junction) between the two superconductors. The two resulting junctions form a dc SQUID, leading to the total Josephson coupling in the first term of (9). The Josephson coupling in the second term in (9), corresponding to processes of type Fig. 3 (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 (8) is dominated by a single term (up to terms of order $\zeta$),

$$H_{\text{eff}} \approx J(1 + \cos \varphi) \left[ S_a \cdot S_b - \frac{1}{4} \right],$$

with $J \approx 2\Gamma^2/\epsilon$. The processes of type Fig. 3 (b) and (c) give rise to (10). Below we will propose an experimental setup based on (10).

Before proceeding, we digress briefly on the dependence of $J$ on the contact points. Unlike the processes of type Fig. 3 (a), those of types Fig. 3 (b), (c), and (d) depend on $\delta r_L = |r_{L,a} - r_{L,b}|$ and $\delta r_R = |r_{R,a} - r_{R,b}|$, see the remark below Eq. (8). For the tunneling Hamiltonian (8), one gets (putting $\delta r = \delta r_L = \delta r_R$)

$$J(\delta r) = \frac{8t^4}{e} \int_0^{\infty} \frac{d\omega}{2\pi} \left. \left| \frac{F_A^{R/A}(\delta r, \omega) - F_A^{R/A}(\delta r, \omega)}{\omega + \epsilon} \right|^2 \right|_{\omega = \epsilon},$$

where $F^{R/A}_A(r, \omega)$ is the Fourier transform of the Green’s function in the superconductors, $F^{R/A}_A(r, t) = \mp i\Theta(\pm t)\langle \psi(r, t), \psi^*(0, 0) \rangle$ [12]. E.g., in the limit $\epsilon \ll \Delta \ll \mu$, we find $J(\delta r) \approx J(0) e^{-\delta r / \xi} \sin^2(k_F \delta r)/(k_F \delta r)^2$ up to order $1/k_F \xi$, with $k_F$ the Fermi wave vector in the leads. Thus, to have $J(\delta r)$ non-zero, $\delta r$ should not exceed the superconducting coherence length $\xi$.

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 (11). According to (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-SQUID-like geometry, see Fig. 3. The Hamiltonian of the entire system is then given by

$$H = J[1 + \cos(\theta - 2\pi f)] \left( S_a \cdot S_b - \frac{1}{4} \right) + \alpha J(1 - \cos \theta),$$

where $f = \Phi/\Phi_0$. $\Phi$ is the flux threading the SQUID loop, $\theta$ 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 (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 $\theta$ 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 $I_S = 2\pi e/\Phi_0 \partial[H]/\partial \theta$, where the brackets refer to a spin expectation value on the DD. Thus, depending on the spin state on the DD we find

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

where $I_J = 2eJ/h$. 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, $\alpha I_J$ is the critical current of the triplet states, and $(\alpha + 1)I_J$ the critical current of the singlet state at $f = 0$, see (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
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 \(\tau_{\text{coh}} \sim \max\{1/\Delta, 1/\Gamma\} \sim 1/\Gamma\) it takes to establish coherence in the S-DD-S junction, the spin relaxation time \(\tau_{\text{spin}}\) on the dot, and the switching time \(\tau_{\text{sw}}\) to reach \(f = 0\). We will assume \(\tau_{\text{coh}} \ll \tau_{\text{spin}}, \tau_{\text{sw}}\), which is not unrealistic in view of measured spin decoherence times in GaAs exceeding 100 ns [17]. If \(\tau_{\text{sw}} < \tau_{\text{spin}}\), the voltage is given by \(3V_1/4\) for times less than \(\tau_{\text{spin}}\), i.e. the singlet no longer contributes to the voltage. For \(t > \tau_{\text{spin}}\) 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 \(\tau_{\text{spin}} < \tau_{\text{sw}}\), a broad transition region of the voltage from the initial value to 0 will occur (dashed line in Fig. 4) [8].

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 \([10]\) or \([12]\) of about \(J \sim 0.05-0.5 K\). This corresponds to a critical current scale of \(I_J \sim 5-50 \mu A\).

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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[12] One could also consider atomic impurities embedded between the grains of a granular superconductor.
[13] See, e.g., M. Tinkham, Introduction to Superconductivity, 2nd ed. (McGraw-Hill, New York, 1996).
[14] If, 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.
[15] We note that the phase difference \(\varphi\) in (5) should now be defined with respect to the phase \(\phi_j(r_{j,a}, r_{j,b})\) of the function \(F^{(j)}(r_{j,a}, r_{j,b}) = F_j(r_{j,a}, r_{j,b})\) on the lead \(j\), see the definition below (1).
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