First- and Second-Order Phase Transitions, Fulde-Ferrel Inhomogeneous State and Quantum Criticality in Ferromagnet/Superconductor Double Tunnel Junctions

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First- and second-order phase transitions, Fulde-Ferrel (FF) inhomogeneous superconducting (SC) state and quantum criticality in ferromagnet/superconductor/ferromagnet double tunnel junctions are investigated. For the antiparallel alignment of magnetizations, it is shown that a first-order phase transition from the homogeneous BCS state to the inhomogeneous FF state occurs at a certain bias voltage $V^*$, while the transitions from the BCS state and the FF state to the normal state at $V_c$ are of the second-order. A phase diagram for the central superconductor is presented. In addition, a quantum critical point (QCP), $V_{QCP}$, is identified. It is uncovered that near the QCP, the SC gap, the chemical potential shift induced by the spin accumulation, and the difference of free energies between the SC and normal states vanish as $|V - V_{QCP}|^{\nu}$ with the quantum critical exponents $\nu = 1/2, 1$ and 2, respectively. The tunnel conductance and magnetoresistance are also discussed.

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Introduction. —Spin-dependent transport plays an essential role in magnetic hybrid nanostructures in the field of spintronics (see, e.g. Refs. 1, 2, 3, 4, 5, 6, 7 for review). Among others, ferromagnet/superconductor (F/S) heterostructures have attracted much attention theoretically. 3, 4, 5, 6, 7, 8, 9 and experimentally. 10, 11, 12, 13, 14, 15 in recent years. For F/S/F double tunnel junctions, it has been observed that the superconductivity is suppressed by the injection of spin-polarized current (e.g. Refs. 5, 11, 12, 13, 14, 18, 20), that is due to the nonequilibrium spin accumulation. When spin-polarized electrons are injected into the superconductor, a spin density is accumulated near the interfaces owing to the spin imbalance, thereby giving rise to an equivalent, small magnetic field that acts as a pair-breaking field, which leads to a suppression of superconductivity. There has been a recent study15 showing that the homogeneous superconducting (SC) state is strongly suppressed with increasing the bias voltage and completely destroyed at a critical voltage by the nonequilibrium spin density in the antiparallel alignment of magnetizations. This study is qualitatively consistent with the experimental observation for high biases, but is inconsistent for low biases16.

On the other hand, about forty years ago, Fulde and Ferrel (FF) 22, and Larkin and Ovchinnikov (LO) 23 independently, found that the SC order parameter can be modulated in real space by a spin-exchange field of a ferromagnet. Later, such an inhomogeneous superconducting state has been extensively explored under various circumstances (e.g. 17, 24). As the nonequilibrium spin accumulation may lead to an equivalent magnetic field in the central superconductor, the FFLO state, which is simply omitted in the previous treatment15, might be inevitable in the F/S/F double tunnel junction.

To understand profoundly the spin-dependent transport properties, in this Letter, the F/S/F double tunnel junction shall be systematically revisited. It is shown that in the antiparallel alignment, a first-order phase transition from a homogeneous Bardeen-Cooper-Schrieffer (BCS) SC state to the inhomogeneous FF SC state occurs at a certain bias voltage, while the transitions from the BCS state and the FF state to the normal state at $V_c$ are of the second-order. A phase diagram for the central superconductor is presented. In addition, a quantum critical point (QCP) is specified at bias $V_{QCP}$, near which the SC order parameter, the chemical potential shift induced by the spin accumulation, and the difference of free energies between the SC and normal states vanish as $|V - V_{QCP}|^{\nu}$ with the quantum critical exponents $\nu = 1/2, 1$ and 2, respectively. The tunnel conductance and magnetoresistance are also obtained.

Model. —Consider a symmetric F/S/F double tunnel junction with the left and right ferromagnetic (FM) electrodes applied by bias voltages $-V/2$ and $V/2$, respectively. The two identical FM electrodes are separated from the central superconductor by two insulating thin films. The central superconductor is presumably described within the framework of BCS theory. Suppose that the energy relaxation time of quasiparticles is shorter than the tunneling time, while the latter is shorter than the spin relaxation time. As the resistance of this tunnel junction with insulating thin films is greater than that of a conventional metallic contact, the Andreev reflection effect can be reasonably ignored for simplicity.

From the standard tunneling Hamiltonian, and in light of the linear response theory, the tunneling current through the $j$th junction can be readily obtained by

$$I_{j\sigma} = 2\pi e |\vec{T}|^2 D_{j\sigma}[N - \eta_j (\sigma S + Q - \frac{N}{2})].$$

(1)

where $\vec{T}$ is the tunneling matrix element, $j = 1, 2$, $D_{j\sigma}$ is the subband density of states (DOS) in the $j$th FM electrode, $\sigma = \pm 1$ for spin up and down, respectively, and $\eta_1 = 1$, $\eta_2 = -1$. The quantities $S$, $Q$, $N$ and $\bar{N}$ are defined by
$S = \frac{1}{2} \sum_k (f_{\uparrow}^k - f_{\downarrow}^k), \quad (2)

Q = \frac{1}{2} \sum_k (w_{\uparrow}^k - w_{\downarrow}^k)(f_{\uparrow}^k + f_{\downarrow}^k), \quad (3)

N = \frac{1}{2} \sum_k [f_0(E_k - eV/2) - f_0(E_k + eV/2)], \quad (4)

\tilde{N} = \frac{1}{2} \sum_k (w_{\uparrow}^k - w_{\downarrow}^k)[f_0(E_k - eV/2) + f_0(E_k + eV/2)], \quad (5)

where $f_0(z)$ denotes the Fermi distribution function of thermal equilibrium in FM electrodes, $f_{\sigma k}$ is the nonequilibrium distribution function of quasiparticles with energy $E_k$ and spin $\sigma (\uparrow, \downarrow)$ in the central superconductor, $S$ and $Q$ represent the spin density and the quasiparticle charge density, describing the spin imbalance and quasiparticle charge imbalance in the central superconductor, respectively.

Fulde-Ferrel State. —As there appears the nonequilibrium spin accumulation near the interfaces of the tunnel junction, it is believed that the SC state would generally include the inhomogeneous FFLO state\cite{22,23} in addition to the homogeneous BCS state. Without loss of generality, we suppose that the SC order parameter takes the form of FF type\cite{22}:

$$\Delta(r) = \Delta_{\mathbf{q}} e^{i\mathbf{q} \cdot \mathbf{r}}$$

with $\Delta_{\mathbf{q}}$ the amplitude of the order parameter. When $\mathbf{q} = 0$, it recovers the homogeneous BCS state. The quasiparticle dispersion $E_k$, the coherence factors $w_{\sigma k}$ and $v_{\sigma k}$ are given by

$$E_k = \sqrt{\xi_k^2 + \Delta_{\mathbf{q}}^2} + (v_F q/2)x, w_{\sigma k} = \frac{1}{2}(1 + \xi_k/\sqrt{\xi_k^2 + \Delta_{\mathbf{q}}^2}),$$

$$v_{\sigma k} = \frac{1}{2}(1 - \xi_k/\sqrt{\xi_k^2 + \Delta_{\mathbf{q}}^2}),$$

respectively, where $\xi_k$ is the free electron energy relative to the chemical potential, $v_F$ the Fermi velocity and $x = \mathbf{k} \cdot \mathbf{q}/(q k)$ the cosine of the angle between $\mathbf{k}$ and momentum $\mathbf{q}$ of a Cooper pair. The amplitude $\Delta_{\mathbf{q}}$ is determined by the gap equation

$$1 = \frac{V_{BCS}}{2} \sum_k \frac{(1 - f_{\uparrow}^k - f_{\downarrow}^k)}{\sqrt{\xi_k^2 + \Delta_{\mathbf{q}}^2}}, \quad (6)$$

where $V_{BCS}$ is the BCS-type pair interaction. The values of $\mathbf{q}$ will be specified later.

Let us proceed to determine the nonequilibrium distribution function $f_{\mathbf{F} k}$. In the absence of spin-flip scattering, the spin up and down tunneling currents are independent, and should be conserved, i.e. $I_{1\sigma} = I_{2\sigma}$, yielding

$$S^F = 0 \quad \text{for the parallel alignment}, \quad (7)$$

$$S^A = PN^A \quad \text{for the antiparallel alignment}, \quad (8)$$

$$Q^F = Q^A = 0 \quad \text{for both alignments}, \quad (9)$$

where the superscripts $F$ and $A$ refer to the parallel and antiparallel alignments, respectively, and $F = |D_{j\uparrow} - D_{j\downarrow}|/(D_{j\uparrow} + D_{j\downarrow})$ is the spin polarization of the FM electrodes. These solutions show that the nonequilibrium spin accumulation exists only in the antiparallel configuration. In the above derivation, we have adopted the conventional constant DOS approximation, $}\sum_k (\cdots) \simeq N(0) f_{\mathbb{E}_D}^{\mathbb{E}_D} (\cdots) d\xi_k$, where $\mathbb{E}_D$ is the cut-off (Debye) energy, and $N(0)$ denotes the DOS of free electrons at the Fermi level. The quantity $\tilde{N}$ vanishes identically for both alignments of magnetizations since the integrand is an odd function of $\xi_k$. Eq. 8 requires that $f_{\mathbf{q} k}^F$ should differ from $f_{\mathbf{q} k}^A$ in the presence of the tunneling current. Following Ref. 3, we consider the solutions of the form:

$$f_{\mathbf{q} k}^A = f_0(E_k - \sigma \delta \mu), \quad (10)$$

$$f_{\mathbf{q} k}^F = f_{\mathbf{q} k}^0 = f_0(E_k), \quad (11)$$

where $\delta \mu$ is introduced as the chemical potential shift induced by the nonequilibrium spin accumulation, and plays essentially the same role as the spin-exchange field explored by FF in their seminal article\cite{22}. This kind of solutions may be applicable if the thickness of the central superconductor is much smaller than the spin diffusion length, and the spin relaxation time is sufficiently long. Eqs. 10 and 11 are the solutions of Eqs. 7 and 9. However, Eqs. 8 and 6 with Eqs. 10 should be solved in a self-consistent manner to specify $\delta \mu$ and $\Delta_{\mathbf{q}}$ as functions of the bias $V$, temperature $T$ and polarization $P$.

For these coupled equations, when the self-consistent multiple solutions corresponding to different $q$ appear, only the value of $q$ that leads to the lowest free energy of the system is retained. In an inhomogeneous superconducting state, the nonzero solution for $\Delta_{\mathbf{q}}$ implies only the local minimum of the free energy, which does not necessarily mean the stable state. In order to clarify this issue, one must compare the free energies of the homogeneous BCS, the inhomogeneous FF and normal states, as emphasized by Abrikosov\cite{23}. The free energy of the present system can be obtained by integrating the gap equation 26:

$$F_S^{F(A)} - F_N^{F(A)} = \frac{(\Delta_{\mathbf{q}}^{F(A)})^2}{V_{BCS}}$$

$$- \int_0^{\Delta_{\mathbf{q}}^{F(A)}} dz \left[ 1 - f_0(\xi_k^2 + z^2 + \frac{v_F q \cdot k}{2k}) - \delta \mu^{F(A)} \right]$$

$$- f_0(\xi_k^2 + z^2 + \frac{v_F q \cdot k}{2k} + \delta \mu^{F(A)}) \right] \right]$$

where $F_S^{F(A)}$ and $F_N^{F(A)}$ stand for the free energy of the SC state and the N state, respectively, and $\delta \mu^{F} = \delta \mu = \delta \mu$. It turns out that both the homogeneous BCS ($q = 0$) and the FF ($q \neq 0$) SC solutions are possible in the antiparallel alignment, while the former solution is always favorable in the parallel configuration.
Results. — In the parallel alignment of magnetizations, since there is no spin and charge accumulation in this circumstance, the SC order parameter does not depend on the bias voltage; while in the antiparallel configuration, the situation becomes complicated, as the nonequilibrium spin accumulation characterized by $S^A$ intervenes in. Figure 1 presents the bias voltage dependence of the SC order parameter and the chemical potential shift at $T/T_c = 0.2$, with $T_c$ the SC critical temperature, for the antiparallel alignment. It is observed that the order parameter $\Delta^A$ remains almost constant at low biases and is in the homogeneous BCS state till a specific bias voltage $V^* = 1.36\Delta_0/e$ at which $\Delta^A$ drops suddenly, where $P = 0.4$, and $\Delta_0$ is the BCS zero-temperature energy gap. Then, $\Delta^A$ goes into the inhomogeneous FF state with $q \neq 0$, decreases with the bias, and vanishes completely at $V = V_c$ where superconductivity is quenched, i.e. $\Delta_0(V_c) = 0$, as shown in Fig. 1(a). At $V = V^*$, there is a discontinuity for $\Delta^A$, implying that a first-order phase transition from the homogeneous BCS phase to the FF phase exists in the system. The present result is quite different from that given in Ref.[9] where the inhomogeneous FF state was simply ignored. The chemical potential shift grows in the homogeneous BCS state with increasing the bias, and exhibits a jump at $V^*$, then increases slowly in the FF state till $V_c$, and is linear for $V \geq V_c$ in the normal state, as shown in Fig. 1(b). We have found that the free energy of the inhomogeneous FF phase is lower than the homogeneous BCS phase for $V_c > V > V^*$, suggesting that the FF state is stable. At $V = V^*$, the free energy of the FF state coincides with that of the homogeneous BCS state, revealing that the FF state can coexist with the BCS state at $V^*$.

The magnitude of momentum $q$ of a Cooper pair is in general a function of bias voltage, as shown in Fig. 2. At $V < V^*$, the system is in the homogeneous BCS state, and thus $|q|$ is zero; while for $V^* \leq V < V_c$, $|q|$ corresponding to the lowest free energy varies nonmonotonically with the bias, implying that the momenta of Cooper pairs in the stable, inhomogeneous SC state are not fixed. Note that there is a discontinuity for $|q|$ at $V = V^*$, which is again the signature of the first-order phase transition.

Phase Diagram. — In the antiparallel configuration, a schematic phase diagram for the central superconductor in the $T - V$ plane could be obtained, as shown in Fig. 3 for $P = 0.4$. There exist three phases: the homogeneous BCS phase with $\Delta$ constant ($q = 0$) at low bias; the inhomogeneous FF phase with $q \neq 0$ for $V^* \leq V < V_c$ at low temperature; the normal phase for $V \geq V_c$. Along the phase boundary from B to C, the homogeneous BCS phase coexists with the spatially modulated FF phase; along the $V_c$ boundary where the SC gap closes, there are two transitions: one is from the homogeneous BCS state to the normal state, and the other is from the spatially modulated FF state to the normal state, which are of second-order. At point C, three phases meet, implying it can be viewed as a Lifshitz point $QCP$; at point QCP where $V_{QCP}$ satisfies $P e V_{QCP}/2\Delta_0 = 0.754 \approx \Delta_0(V_{QCP})$, a quantum phase transition (QPT) from the ordered FF state to the disordered normal state occurs at $T = 0$.

TMR. — The total current are given by $I_F = I_0 N F$, and $I^A = I_0 (1-P^2) N^A$, where $I_0 = 2\pi e | \bar{T} |^2 (D_{\uparrow\downarrow}+D_{\downarrow\uparrow})$, and $N^A$ is given by Eq. (4). The differential conductance $G^F(A)$ and the tunneling magnetoresistance $TMR$ are obtained by $G^F(A) = 4I_F^2/\pi e$ and $TMR = G^F/G^C - 1$. The bias dependence of the tunnel conductance and the $TMR$ is shown in Fig. 4. The behavior of $G^F$ is consistent with that of Ref.[8], but $G^A$ and $TMR$ differ from those in Ref.[8] owing to the intervention of the FF state, where the oscillating behaviors for $G^A$ and $TMR$ are
As $V$ approaches $V_{QCP}$, the amplitude of SC order parameter, the chemical potential shift induced by the spin accumulation, as well as the difference of free energies between the SC and normal states vanish as $\Delta_0/\Delta_0$, $[\delta \mu(V_{QCP}) - \delta \mu(V)]$, $F_S^A - F_N^A \sim |V - V_{QCP}|^{\nu}$ with the critical exponents $\nu = 1/2, 1$ and 2. Obviously, such a QPT is of second-order. The present system offers a nice example of the QPT.

In summary, we have revisited the spin-dependent transport in F/S/F double tunnel junctions by taking the proximity effect into account. It is found that in the antiparallel configuration, the first- and second-order phase transitions, the inhomogeneous FF state, and quantum criticality can be revealed simultaneously in the central superconductor. The present study rectifies the previous result [2] where the FF state was simply ignored.

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