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Tunneling, relaxation of spin-polarized quasiparticles, and spin-charge separation in superconductors

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The nonequilibrium spin imbalance created when electrons tunnel from a ferromagnet to a superconductor is calculated. Since the charge current in a superconductor is carried by the condensate within a penetration length of the surface and the spin current, which is carried by quasiparticles, can exist in the bulk, one may be able to directly demonstrate spin-charge separation in a superconductor using spin injection techniques. After estimating the spin-relaxation rates due to magnetic impurity and spin-orbit scattering, an explicit experimental geometry for demonstration of spin-charge separation is discussed.

I. INTRODUCTION

Spin-polarized electrons have been a subject of both experimental and theoretical investigation for many years. A common way to generate spin-polarized electrons in a metal or superconductor is to inject them from a ferromagnet. The earliest tunneling experiment in this context was done by Tedrow and Meservey,[1] who injected current from a ferromagnetic film to a superconductor. A static field was used to Zeeman split the quasiparticle density of states in the superconductor, and the asymmetry in the tunneling conductance showed the polarization of tunneling electrons. An alternative method is to use a normal metal-magnetic semiconductor-superconductor tunnel junction, where electrons with different spins see different tunneling barriers in the magnetic semiconductor.[2] Nonequilibrium spins injected into a superconductor or a metal have also been shown to have interesting effects on the nuclei which would be observable through electron spin resonance.[7]

Recently, in a series of papers,[3] Johnson and Silsbee have developed a spin injection and detection technique in which a dc current is driven through a ferromagnetic film into a bulk metal sample. The nonequilibrium spins created in the metal were detected by another ferromagnetic film. By flipping the direction of magnetization of the second ferromagnetic film, a small voltage signal proportional to the spin density can be measured between the second ferromagnetic film and the paramagnetic metal. Johnson and Silsbee attributed such a voltage signal to the different chemical potentials of the two spin species in the metal (paramagnet) caused by the injected current.

Although the junctions used in Ref. 3 were not tunnel junctions, this experiment can in principle also be done with tunnel junctions provided the voltage bias is large enough. In the case of superconductor–normal-metal (nonferromagnetic) tunneling junctions, this is the so-called *charge imbalance* in superconductors and has been extensively investigated.[4–6] For spin-polarized quasiparticle tunneling into superconductors, one can discuss charge imbalance and spin imbalance in a unified manner as we do in this paper.

In the following, we first present the calculation of the tunneling currents and the spin imbalance created by a ferromagnetic-superconductor (FS) tunnel junction under nonequilibrium conditions. The calculation closely follows those of Refs. 5 and 6. In Sec. II, we discuss the quasiparticle spin-relaxation processes. Finally in Sec. III, we formulate the quasiparticle charge and spin transport equations and discuss the concept of spin and charge separation in superconductors as well as a possible means for its verification.

II. TUNNELING RESULTS FOR SPIN-POLARIZED PARTICLES

We start with the tunneling Hamiltonian

$$H = \sum_{k,k',\sigma} \left[T_{kk'} \left(u_k \gamma^\dagger_{k\sigma} + \sigma v_k S^\dagger \gamma_{-k-\sigma} \right) c_{k'\sigma} + \text{H.c.} \right], \tag{1}$$

where $k,\ k'$ label electrons in the superconductor and the ferromagnet, respectively, $\sigma = \pm 1$ is the spin variable, $S^\dagger$ is the creation operator of a Cooper pair, and u_k and v_k are the usual BCS coherence factors

$$u_k^2 = \frac{1}{2}\left(1 + \frac{\xi_k}{E_k}\right), \quad v_k^2 = \frac{1}{2}\left(1 - \frac{\xi_k}{E_k}\right),$$

$$\xi_k = \epsilon_k - \mu, \quad E_k = (\xi_k^2 + \Delta^2)^{1/2}. \tag{2}$$

We have assumed that the tunneling matrix element $T_{kk'}$ is independent of spin. The spin dependence will be accounted for by the spin dependent density of states at the Fermi surface of the ferromagnet. In Eq. (1), we have replaced the electron creation operator in the superconductor by quasiparticle operators through the Bogoliubov-Valatin transformation

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$$\gamma_{k\sigma}^{\dagger} = u_k c_{k\sigma}^{\dagger} - \sigma v_k S^{\dagger} c_{-k-\sigma}. \tag{3}$$

There are four possible processes in the tunneling Hamiltonian of Eq. (1). For each of these processes we now use Fermi's golden rule to compute the spin and charge currents across the interface. Table I lists the contributions from each of these processes in the order: $\gamma_{k\sigma}^{\dagger} c_{k'\sigma}$, $\gamma_{-k-\sigma} c_{k'\sigma}$, $c_{k'\sigma}^{\dagger}\gamma_{k\sigma}$, $c_{k'\sigma}^{\dagger}\gamma_{-k-\sigma}^{\dagger}$. The first column is their contribution to tunneling probability computed from Fermi's golden rule. The Fermi function $f_\sigma(E)$ describes the electron distribution in the metal, while $f_{k\sigma}$ describes the quasiparticle distribution in the superconductor. When injecting electrons into the superconductor, a negative voltage $-V$ is applied on the ferromagnet. Consequently, the energy conservation conditions are $\epsilon_{k'} + eV = E_k$ for the $\gamma_{k\sigma}^{\dagger} c_{k'\sigma}$ and $c_{k'\sigma}^{\dagger}\gamma_{k\sigma}$ processes, and $\epsilon_{k'} + eV = -E_k$ for the $\gamma_{-k-\sigma} c_{k'\sigma}$ and $c_{k'\sigma}^{\dagger}\gamma_{-k-\sigma}^{\dagger}$ processes.

Since a quasiparticle has probability u_k^2 to be an electron and probability v_k^2 to be a hole, the quasiparticle charge is given by

$$q_k = u_k^2 - v_k^2 = \xi_k / E_k. \tag{4}$$

The part of charge carried by the condensate is $2v_k^2 = 1 - q_k$. The quasiparticle spin, on the other hand, is unity because $u_k^2 + v_k^2 = 1$.

Following Table I, the spin injection current is

$$I_s = \frac{2\pi}{\hbar}|T|^2 \sum_{k,\sigma} \sigma N_\sigma \left\{ \frac{1}{2}\left[f_\sigma(E_k - eV) - f_\sigma(E_k + eV)\right] - u_k^2 f_{k\sigma} + v_k^2 f_{-k-\sigma} \right\}, \tag{5}$$

where N_σ is the spin dependent density of states at the Fermi surface of the injecting ferromagnet. If we write the quasiparticle distribution $f_{k\sigma}$ in terms of its local equilibrium distribution $f(E_k)$ plus the deviation, i.e.,

$$f_{k\sigma} = f(E_k) + \delta f_{k\sigma}, \tag{6}$$

and define the quasiparticle spin and charge density as

$$\delta S_\sigma = \sum_k \delta f_{k\sigma}, \quad \delta Q_\sigma = \sum_k q_k \delta f_{k\sigma}, \tag{7}$$

we can write the spin injection current as

$$\begin{aligned} I_s &= \frac{d}{dt}(\delta S_\uparrow - \delta S_\downarrow) \\ &= \frac{\pi}{\hbar}|T|^2 \Big((N_\uparrow - N_\downarrow) \sum_k [f(E_k - eV) - f(E_k + eV)] \\ &\quad -(N_\uparrow + N_\downarrow)(\delta S_\uparrow - \delta S_\downarrow) \\ &\quad -(N_\uparrow - N_\downarrow)(\delta Q_\uparrow + \delta Q_\downarrow) \Big). \end{aligned} \tag{8}$$

The quasiparticle charge current is

$$\begin{aligned} I_q &= \frac{d}{dt}(\delta Q_\uparrow + \delta Q_\downarrow) \\ &= \frac{\pi}{\hbar}|T|^2 \Big((N_\uparrow + N_\downarrow) \sum_k q_k^2 [f(E_k - eV) \\ &\quad - f(E_k + eV)] - (N_\uparrow + N_\downarrow)(\delta Q_\uparrow + \delta Q_\downarrow) \\ &\quad -(N_\uparrow - N_\downarrow) \sum_k q_k^2 (\delta f_{k\uparrow} - \delta f_{k\downarrow}) \Big). \end{aligned} \tag{9}$$

The total charge current including the charge carried by the condensate is

$$\begin{aligned} I = \frac{\pi}{\hbar}|T|^2 \Big(&(N_\uparrow + N_\downarrow) \sum_k [f(E_k - eV) - f(E_k + eV)] \\ &-(N_\uparrow - N_\downarrow)(\delta S_\uparrow - \delta S_\downarrow) - (N_\uparrow + N_\downarrow)(\delta Q_\uparrow + \delta Q_\downarrow) \Big). \end{aligned} \tag{10}$$

As in charge imbalance experiments,[4] we expect the injection voltage to be much larger than the detection voltage, which is related to the nonequilibrium terms with δQ_σ and δS_σ. Thus in Eqs. (8)–(10), the first term dominates and the fraction of the total charge current which is injected as quasiparticle spin and charge is

$$\eta_s \equiv \frac{I_s}{I} \approx \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}, \tag{11}$$

$$\eta_q \equiv \frac{I_q}{I} \approx \frac{\sum_k q_k^2 [f(E_k - eV) - f(E_k + eV)]}{\sum_k [f(E_k - eV) - f(E_k + eV)]}. \tag{12}$$

For a large injection voltage, $eV \gg \Delta$, the quasiparticle charge is approximately one, $q_k \approx 1$, because most of the injected quasiparticles have energies larger than the gap. Thus, η_1 in Eq. (12) is approximately one, i.e., all the charge is injected as quasiparticle charge.[5,6]

To detect the nonequilibrium spin, one can use another ferromagnetic film to form a FSF tunnel junction.[3] This

TABLE I. The contribution to tunneling probability from the four processes in Eq. (1). The terms appear in the order $\gamma_{k\sigma}^{\dagger} c_{k'\sigma}$, $\gamma_{-k-\sigma} c_{k'\sigma}$, $c_{k'\sigma}^{\dagger}\gamma_{k\sigma}$, $c_{k'\sigma}^{\dagger}\gamma_{-k-\sigma}^{\dagger}$ from top to bottom.

Probability	Electrons added	Excitations added	Quasiparticle charge added	Condensate charge added	Spins added
$u_k^2(1 - f_{k\sigma}) f_\sigma(E_k - eV)$	1	1	q_k	$1 - q_k$	σ
$v_k^2 f_{-k-\sigma}[1 - f_\sigma(E_k + eV)]$	1	-1	$-q_k$	$1 + q_k$	σ
$u_k^2 f_{k\sigma}[1 - f_\sigma(E_k - eV)]$	-1	-1	$-q_k$	$-1 + q_k$	$-\sigma$
$v_k^2(1 - f_{-k-\sigma}) f_\sigma(E_k + eV)$	-1	1	q_k	$-1 - q_k$	$-\sigma$

is illustrated in Fig. 1 where the tunneling current is injected from the ferromagnetic film $F1$ and drained from the superconductor. A voltage signal is measured at the detecting ferromagnetic film $F2$. Equations (8)–(10) are also valid for the detecting junction. In charge imbalance and spin injection experiments, the voltage applied at the detecting junction is used to nullify the total current Eq. (10), i.e., the detecting junction is a voltage probe (see the illustration in Fig. 1). As we have mentioned, such voltages (typically of order of nanovolts), are much smaller than the injection voltage (typically of order of millivolts) and k_BT/e. Therefore we can expand the Fermi functions in Eq. (10),

$$2(N'_\uparrow + N'_\downarrow)N_0 g_{\rm ns} eV_1 - (N'_\uparrow - N'_\downarrow)(\delta S_\uparrow - \delta S_\downarrow) - (N'_\uparrow + N'_\downarrow)(\delta Q_\uparrow + \delta Q_\downarrow) = 0, \quad (13)$$

where $N_0 g_{\rm ns} = \sum_k[-\partial f(E)/\partial E]$ with N_0 being the normal density of states of one spin species in the superconductor and $g_{\rm ns}$ being a function of Δ/T, which is the tunnel conductance divided by its normal state value. N'_σ is the density of states in the detecting ferromagnetic film. When the magnetization direction of the detecting film is flipped, a different voltage V_2 is needed to nullify the total current. This voltage is given by an equation identical to Eq. (13) except that $N'_\uparrow$ and $N'_\downarrow$ are interchanged. Combining the two equations, we have

$$V_1 - V_2 = \frac{N'_\uparrow - N'_\downarrow}{N'_\uparrow + N'_\downarrow}\frac{\delta S_\uparrow - \delta S_\downarrow}{eN_0 g_{\rm ns}}, \quad (14)$$

$$V_1 + V_2 = \frac{\delta Q_\uparrow + \delta Q_\downarrow}{eN_0 g_{\rm ns}}. \quad (15)$$

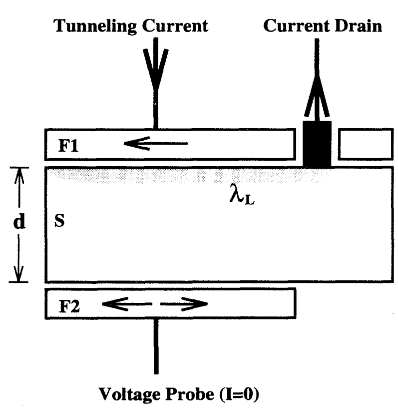

FIG. 1. A schematic setup for the tunneling spin injection and detection experiment. $F1$ is the current-injecting ferromagnetic film with the arrow inside representing the direction of magnetization. S is a superconductor. $F2$ is the detecting ferromagnetic film. When switching the direction of magnetization in $F2$, the voltage difference measured is proportional to the spin density at the junction. To check the idea of charge-spin separation in the superconductor, the penetration length (shaded region) should satisfy $\lambda_L \ll d \sim$ spin diffusion length λ_s.

Therefore the difference between the two voltages is a measure of the nonequilibrium spin density, while the sum is a measure of the nonequilibrium quasiparticle charge density in the superconductor. At a steady state, the nonequilibrium charge and spin are determined by the balance between injection rate and relaxation rate. Using Eqs. (11) and (12), we have

$$I_s = \left(\frac{dS}{dt}\right)_{\rm inj} = -\left(\frac{dS}{dt}\right)_{\rm coll} = \frac{\delta S_\uparrow - \delta S_\downarrow}{\tau_s} = \eta_s I, \quad (16)$$

$$I_q = \left(\frac{dQ}{dt}\right)_{\rm inj} = -\left(\frac{dQ}{dt}\right)_{\rm coll} = \frac{\delta Q_\uparrow + \delta Q_\downarrow}{\tau_q} = \eta_q I, \quad (17)$$

where τ_s, τ_q are the spin and charge relaxation time, respectively. Thus, the voltage signals can also be written as

$$V_1 - V_2 = \frac{N'_\uparrow - N'_\downarrow}{N'_\uparrow + N'_\downarrow}\frac{\eta_s \tau_s I}{eN_0 g_{\rm ns}}, \quad (18)$$

$$V_1 + V_2 = \frac{\eta_q \tau_q I}{eN_0 g_{\rm ns}}. \quad (19)$$

This shows that both the spin- and the charge-relaxation time can be measured in such an experiment, since all other quantities in Eqs. (18) and (19) are directly accessible to measurement.

In Eq. (16) we have neglected the spin current through the detecting junction. This current is small but finite in general. Using Eqs. (8), (10), and (14), when $I = 0$, we have

$$I'_s = \frac{4\pi|T|^2}{\hbar}\frac{N'_\uparrow N'_\downarrow}{N'_\uparrow + N'_\downarrow} eN_0 g_{\rm ns}(V_1 - V_2). \quad (20)$$

This current is due to a spin-up and a spin-down current, equal in magnitude but flowing in opposite directions so that the total charge current across the detection junction is zero.

III. QUASIPARTICLE CHARGE AND SPIN RELAXATION

As we discussed above, the measured charge and spin signals are directly related to the charge- and spin-relaxation time. Therefore it is necessary to study these relaxation processes. The charge relaxation can be due to inelastic electron-phonon scattering or to impurity elastic scattering in the presence of gap anisotropy. Such relaxation processes have been studied in detail by many authors.[5,6,8] In the presence of spin-flip scattering close to T_c, the charge-relaxation rate is[8]

$$\frac{1}{\tau_q} = \frac{\pi}{4}\frac{\Delta}{k_B T_c}\frac{1}{\tau_E}\left(1 + \frac{2\tau_E}{\tau_s}\right)^{1/2}, \quad (21)$$

where τ_E is the energy relaxation time and τ_s is the spin-relaxation time. The charge-relaxation time, τ_q, diverges as one approaches T_c. This is because in the normal state all the charge is carried by quasiparticles, and there is no cross-branch scattering or quasiparticle recombination to convert the quasiparticle charge to the condensate charge. The factor Δ/k_BT_c appears because the quasiparticles which are effective in relaxing the charge are those with q_k differing from unity, i.e., those with $|\xi| \leq \Delta$, whereas the thermally excited quasiparticles have $|\xi| \leq k_BT_c$. Thus, the fraction of processes that changes the quasiparticle charge significantly is of order Δ/k_BT_c.

A more complete picture of quasiparticle relaxation in charge imbalance experiments also involves the initial quasiparticle cooling process.[5] When quasiparticles are injected into a superconductor, they distribute roughly uniformly between Δ and $eV_{\rm inj}$. Inelastic scattering cools down the quasiparticle distribution towards its equilibrium form with the characteristic time $\tau_E \sim 10^{-10}$ sec.[5] Since the cooling does not involve branch crossing scattering (between $\xi_k > 0$ and $\xi_k < 0$), it has little effect on the charge relaxation.

The situation is quite different for spin relaxation. Spins can relax without branch crossing scattering and quasiparticle recombination. And as we will show, in some cases the initial cooling process may have a significant effect on spin relaxation because of the extremely nonequilibrium quasiparticle distribution. We now discuss two major processes: relaxation through magnetic impurity scattering and through spin-orbit scattering. For simplicity, we restrict our discussions to elastic processes.

A. Relaxation by magnetic impurity scattering

For relaxation by magnetic impurity scattering the Hamiltonian describing spin-flip scattering is

$$H' = \sum_{k_1,k_2,\sigma} V_{k_1,k_2} c^\dagger_{k_1\sigma} c_{k_2-\sigma}, \tag{22}$$

where V_{k_1,k_2} is roughly a constant, representing a short-range scattering potential. In the superconducting phase, the quasiparticle spin relaxation involves only the following part of the Hamiltonian:

$$H'' = \sum_{k_1,k_2,\sigma} V_{k_1,k_2}(u_{k_1}u_{k_2} + v_{k_1}v_{k_2})\gamma^\dagger_{k_1\sigma}\gamma_{k_2-\sigma}, \tag{23}$$

where we have expressed the electron operators in terms of quasiparticle operators using a Bogoliubov transformation. The relaxation due to spin-flip scattering is

$$\begin{aligned} \frac{df_{k\sigma}}{dt} = &-\frac{2\pi}{\hbar}\sum_{k'}|V_{k,k'}|^2(u_ku_{k'} + v_kv_{k'})^2 \\ &\times(f_{k\sigma} - f_{k'-\sigma})\delta(E_{k'} - E_k). \end{aligned} \tag{24}$$

If we define the spin-density distribution and the spin density as

$$\delta S_k = \delta f_{k\uparrow} - \delta f_{k\downarrow}, \quad \delta S = \sum_k \delta S_k, \tag{25}$$

then summing Eq. (24) over k for up spins and subtracting the corresponding quantity for down spins yields

$$\begin{aligned} \left(\frac{d\delta S}{dt}\right)_{\rm coll} &= -\frac{2\pi}{\hbar}\sum_{k,k'}|V_{k,k'}|^2\left(1 + \frac{\xi_k\xi_{k'} + \Delta^2}{E_{k'}E_k}\right)\delta S_k \\ &\quad\times\delta(E_{k'} - E_k) \\ &= -\frac{2\pi}{\hbar}\sum_{k,k'}|V_{k,k'}|^2\left(1 + \frac{\Delta^2}{E_{k'}E_k}\right)\delta S_k \\ &\quad\times\delta(E_{k'} - E_k), \end{aligned} \tag{26}$$

where the term $\xi_k\xi_{k'}$ drops out because the summation is over both $\xi > 0$ and $\xi < 0$. To proceed further, we have to know the distribution δS_k. We consider the following two cases.

(1) When the spin relaxes slower than the cooling process, i.e., $\tau_s \gg \tau_E$, the nonequilibrium spin distribution is

$$\delta S_k = -C\frac{\partial f(E_k)}{\partial E_k}. \tag{27}$$

The ratio of the normal to superconducting relaxation times is

$$\begin{aligned} \frac{\tau_s^{(n)}}{\tau_s} &= 2\int_{\Delta+\delta}^{\infty}\frac{E^2 + \Delta^2}{E^2 - \Delta^2}\left(-\frac{\partial f(E)}{\partial E}\right)dE \\ &\approx 2f(\Delta)\left[1 + \frac{\Delta}{k_BT}[1 - f(\Delta)]\ln\left(\frac{2\Delta}{\delta}\right)\right], \end{aligned} \tag{28}$$

where δ is a measure of gap anisotropy, which eliminates the logarithmic divergence of the integral at $E = \Delta$. For real superconductors δ is small. Equation (28) is very similar to that for nuclear spin lattice relaxation.[9] The spin-relaxation rate first increases below T_c and then decreases exponentially at even lower temperatures.

(2) When the spin relaxes faster than the cooling process, i.e., $\tau_s \ll \tau_E$, the nonequilibrium spin distribution is

$$\delta S_k = \begin{cases} C' & \text{for } \Delta < E_k < eV_{\rm inj}, \\ 0 & \text{otherwise.} \end{cases} \tag{29}$$

The ratio of the normal to superconducting relaxation times is

$$\frac{\tau_s^{(n)}}{\tau_s} = \frac{1}{eV_{\rm inj}}\int_{\Delta+\delta}^{eV_{\rm inj}}\frac{E^2 + \Delta^2}{E^2 - \Delta^2}dE \approx 1 + \frac{\Delta}{eV_{\rm inj}}\ln\frac{0.74\Delta}{\delta}. \tag{30}$$

In this case the spin-relaxation rate below T_c will increase and then saturate as the temperature is lowered.

B. Relaxation by spin-orbit scattering

For relaxation by spin-orbit scattering the Hamiltonian describing the spin-flip scattering is

$$H' = \sum_{k,k',\sigma,\sigma'} i\lambda(\vec{k} \times \vec{k'}) \cdot \vec{s}_{\sigma\sigma'} c^\dagger_{k\sigma} c_{k'\sigma'}, \tag{31}$$

where λ is a short-range potential and $\vec{s}$ is the electron spin operator. The part of the Hamiltonian which flips the quasiparticle spin in the superconducting phase is

$$H'' = \sum_{k,k',\sigma} i\lambda(\vec{k}\times\vec{k'})\cdot\vec{s}_{\sigma,-\sigma}(u_k u_{k'} - v_k v_{k'})\gamma^\dagger_{k\sigma}\gamma_{k'-\sigma}. \tag{32}$$

Notice that the coherence factor is different from that for magnetic impurity scattering. Going through similar calculations, the relaxation due to spin-flip scattering is

$$\left(\frac{d\delta S}{dt}\right)_{\rm coll} = -\frac{2\pi}{\hbar}\sum_{k,k'}|\lambda(\vec{k}\times\vec{k'})\cdot\vec{s}_{\uparrow\downarrow}|^2 \times\left(1-\frac{\Delta^2}{E_{k'}E_k}\right)\delta S_k\,\delta(E_{k'}-E_k). \tag{33}$$

To simplify the calculation, we replace the quantity in the absolute sign by its average over the Fermi surface. The ratio of the normal to superconducting relaxation times is then

$$\frac{\tau_s^{(n)}}{\tau_s} = \begin{cases} 2f(\Delta) & \text{for } \tau_s \gg \tau_E, \\ 1-\frac{\Delta}{eV_{\rm inj}} & \text{for } \tau_s \ll \tau_E. \end{cases} \tag{34}$$

Notice that in both cases the spin-relaxation rate decreases (relaxation time increases) below T_c. This is to the advantage of spin injection measurements.

IV. SEPARATION OF CHARGE AND SPIN TRANSPORT

In the above discussion we implicitly assumed that the thickness, d, of the superconducting layer is small compared to the charge and spin diffusion lengths, $\lambda_q = \sqrt{D\tau_q}$ and $\lambda_s = \sqrt{D\tau_s}$. ($D$ is the diffusion coefficient in the normal state.) For thicker samples, $d \geq \lambda_q, \lambda_s$, Eqs. (18) and (19) should be modified to account for diffusion effects. The situation is similar to the normal-metal–superconductor interface problem.[10] When quasiparticles are injected into the superconductor at the first junction, their charge and spin diffuse into the superconductor with characteristic times τ_q and τ_s. However, to keep charge neutrality in the superconductor, the condensate will adjust itself to screen the charge carried by quasiparticles. In the process of this adjustment, the condensate builds up a supercurrent inside the superconductor to cancel the quasiparticle current and a supercurrent at the surface (within the penetration length) to carry away the electric charge. However, the electric field (potential gradient) does leak into the superconductor over a distance of the quasiparticle charge diffusion length. This is due to the following reason: To screen a quasiparticle charge density δQ, the condensate must be reduced in charge density by the same amount, so that the number density must be reduced by $\delta Q/e$. This means that the Fermi level must be lowered by $\delta\mu_s = \delta Q/eN_0$. However, in a steady state, the electrochemical potential in a superconductor must be a constant due to the acceleration equation[11]

$$m\frac{\partial \vec{v}_s}{\partial t} = -\nabla(\mu_s + e\Phi). \tag{35}$$

Therefore the electrostatic potential Φ has a finite gradient over a quasiparticle charge diffusion length inside the superconductor. Such a potential gradient is responsible for the excessive electric resistance close to T_c at a normal-metal–superconductor interface.[10] The charge density associated with such a potential gradient is negligibly small [smaller than δQ by a factor of $(l_s/\lambda_q)^2$, where $l_s = (4\pi e^2 N_0)^{1/2}$ is the Thomas-Fermi screening length].

It is straightforward to make the above discussion quantitative. The linearized Boltzmann equation for quasiparticles in a superconductor is

$$\frac{\partial f_{k\sigma}}{\partial t} + \vec{v}_k\cdot\nabla\delta f_{k\sigma} - \frac{\partial f_{k\sigma}}{\partial E_k}\vec{v}_k\cdot\nabla\delta E_k = \left(\frac{df_{k\sigma}}{dt}\right)_{\rm coll}, \tag{36}$$

where $\vec{v}_k$ is the quasiparticle velocity ($|\vec{v}_k| = v_F|q_k|$) and the spatial variation of quasiparticle energy is

$$\delta E_k = \hbar\vec{k}\cdot\vec{v}_s - q_k(\delta\mu_s + e\delta\Phi) + \frac{\Delta}{E_k}\delta|\Delta|. \tag{37}$$

The deviation of the distribution function from local equilibrium is related to $\delta f_{k\sigma}$ by

$$\delta f^{\rm le}_{k\sigma} = \delta f_{k\sigma} - \frac{\partial f_{k\sigma}}{\partial E_k}\delta E_k. \tag{38}$$

Therefore Eq. (36) can be written more compactly as

$$\frac{\partial f_{k\sigma}}{\partial t} + \vec{v}_k\cdot\nabla\delta f^{\rm le}_{k\sigma} = \left(\frac{df_{k\sigma}}{dt}\right)_{\rm coll}. \tag{39}$$

Notice that the electrostatic potential does not appear in Eq. (36) directly. It comes in through its effect on the condensate [Eq. (35)] and the variation of quasiparticle energy δE_k. For a steady state, $\mu_s + e\Phi$ is constant due to Eq. (35). Even δE_k is independent of the electrostatic potential. This is a manifestation of the screening of quasiparticle charge by the condensate. On the other hand, Eq. (36) can be readily reduced to the normal state Boltzmann equation by setting $\vec{v}_s$, Δ, $\delta\mu_s = 0$.

To obtain the charge-relaxation equations, we multiply Eq. (39) by q_k and sum over the momentum and spin:

$$\nabla\cdot\vec{j}_Q = -\frac{\delta Q}{\tau_q}, \tag{40}$$

where the quasiparticle current is

$$\vec{j}_Q = \sum_{k\sigma} q_k\vec{v}_k\delta f^{\rm le}_{k\sigma}. \tag{41}$$

When T_c is approached, τ_q^{-1} goes to zero and Eq. (40) becomes the charge conservation condition in the normal state. Multiplying Eq. (39) by $q_k\vec{v}_k$ and summing over the momentum and spin yields

$$\nabla \left(\sum_{k\sigma} q_k^3 \frac{v_F^2}{3} \delta f_{k\sigma}^{\mathrm{le}} \right) = \left(\frac{d\vec{j}_Q}{dt} \right)_{\mathrm{coll}} . \quad (42)$$

In a relaxation time approximation

$$\left(\frac{d\vec{j}_Q}{dt} \right)_{\mathrm{coll}} = -\frac{\vec{j}_Q}{\tau}, \quad (43)$$

where $\tau \ll \tau_q$, τ is the impurity scattering time which determines the normal state resistivity, since Eq. (42) must reduce to Ohm's law for a metal when $\Delta \to 0$. Close to T_c one can also replace q_k^3 by q_k with an accuracy of Δ/k_BT_c. Equation (42) then becomes the diffusion equation

$$\vec{j}_Q = -D\nabla \delta Q, \quad (44)$$

where the diffusion constant is $D = v_F^2 \tau/3$. When the temperature is raised above T_c, the electrostatic potential shows up in Eq. (36). Going through similar calculations, one obtains the drift-diffusion equation in the normal state instead of Eq. (44).

In a similar manner we can derive the relevant equations for spin relaxation:

$$\nabla \cdot \vec{j}_S = -\frac{\delta S}{\tau_s}, \quad (45)$$

$$\vec{j}_S \equiv \sum_k \vec{v}_k (\delta f_{k\uparrow}^{\mathrm{le}} - \delta f_{k\downarrow}^{\mathrm{le}}) = -D\nabla \delta S. \quad (46)$$

Solving Eqs. (40), (44), (45), and (46), we see that the quasiparticle charge and spin densities decrease exponentially away from the injection junction

$$\delta Q \sim e^{-x/\lambda_q}, \quad \delta S \sim e^{-x/\lambda_s}, \quad (47)$$

where x is the direction into the superconductor. Therefore if the superconducting layer thickness, d, is larger than λ_q and λ_s, Eqs. (18) and (19) should be replaced by

$$V_1 - V_2 = \frac{N'_\uparrow - N'_\downarrow}{N'_\uparrow + N'_\downarrow} \cdot \frac{\eta_s \tau_s I}{e N_0 g_{\mathrm{ns}}} \, e^{-d/\lambda_s}, \quad (48)$$

$$V_1 + V_2 = \frac{\eta_q \tau_q I}{e N_0 g_{\mathrm{ns}}} \, e^{-d/\lambda_q}. \quad (49)$$

Now we discuss the implication of these transport equations and the idea of spin-charge separation in superconductors introduced in Ref. 12. At first look, such a separation is indicated by the two sets of transport equations for charge and spin characterized by the two different times, τ_q and τ_s. However, a closer look reveals more. As we have mentioned, for a steady superconducting state, because $\mu_s + e\Phi$ is constant, the electrostatic potential has no direct effect on the quasiparticles. If the gap is spatially uniform in the bulk of the superconductor, the third term in Eq. (36) drops out completely and the resulting equation resembles that of a neutral particle with spin. This resemblance has at least the following two meanings. First these quasiparticles do not interact with the electric field which is present inside the superconductor within the charge diffusion length. Second, there is no net electric current associated with them. Both are results of the condensate response to the injected quasiparticles. The net electric charge injected into a superconductor is actually carried away by a supercurrent on the surface of the superconductor.

The situation for spins is different. Quasiparticles carry a well-defined spin which is not screened by the condensate. Therefore spins can diffuse into the bulk of the superconductor and cause a bulk spin current. It was argued in Ref. 12 that such a separation of charge and spin transport is more generic and fundamental in the sense that Bogoliubov quasiparticles of a fully gapped superconductor in three dimensions should be neutral, spin-$\frac{1}{2}$ particles due to the perfect screening of charge and the Meissner effect in a bulk superconductor. The solution of the two sets of transport equations (40), (44), (45), and (46) gives two characteristic length scales λ_q and λ_s for δQ and δS. But due to screening, δQ does not represent the net charge transport. The net injected electric charge and current must be mainly confined to within a Thomas-Fermi screening length and a London penetration length of the surface, respectively. The spin density decreases over the spin diffusion length λ_s, which can be large in the superconducting phase as we have discussed in Sec. II. Therefore the charge and spin transport in a superconductor are separated in the following sense: the charge current is carried by the condensate and exists only within the penetration length of the surface; the spin current is carried by quasiparticles and can exist inside the bulk of a superconductor.

The separation of charge and spin transport in a superconductor can also be checked experimentally. The key is to verify the existence of a bulk spin current in a superconductor. We propose an experimental setup as in Fig. 1 with a thick superconductor layer sandwiched between two ferromagnetic films in a FSF tunnel junction. The thickness of the superconductor is chosen to be much larger than the London penetration length but of the same order as the spin diffusion length, i.e., $\lambda_L \ll d \sim \lambda_s$. Such a choice is possible because the spin-relaxation time usually increases at low temperature, except for case (2) of magnetic impurity scattering discussed in Sec. II. For the normal state spin diffusion lengths of order a micron are attainable.[3] On the other hand, typical London penetration lengths are of order of a few hundred Å. In the Fig. 1 setup, the tunneling current is injected at the $F1$-S junction and drained at the same side of the superconducting layer. The electric current must flow within a penetration length of the top surface of the superconductor (shaded region). When the direction of magnetization of $F2$ is switched, a voltage signal of Eq. (48) would indicate a nonvanishing spin density at the S-$F2$ junction, which in turn implies a spin current flowing through the superconductor. For the spin-relaxation mechanisms discussed in Sec. II, we plot in Fig. 2 the temperature dependence of $-(\tau_s^{(n)}/\tau_s)^{1/2}$, which according to Eq. (48) is roughly proportional to $\ln(V_1 - V_2)$. Notice however that these curves do not represent the absolute magnitude of the signal because the normal state relaxation

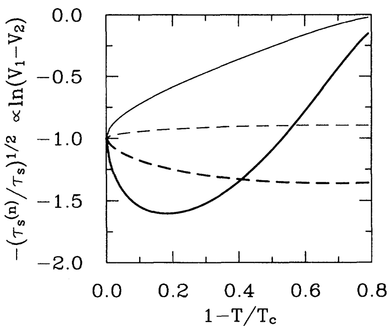

FIG. 2. The calculated temperature dependence of $-(\tau_s^{(n)}/\tau_s)^{1/2}$ which is proportional to the logarithm of the spin signal. Thick lines are for magnetic impurity scattering, and thin lines are for spin-orbit scattering. Solid lines are for the case when the spin-relaxation time τ_s much larger than the energy relaxation time τ_E, and dashed lines are for the case of $\tau_s \ll \tau_E$. The parameters used are $\delta/\Delta_0 = 10^{-2}$, $\Delta_0/eV_{\rm inj} = 0.2$, where δ is the gap anisotropy and $V_{\rm inj}$ is the injection voltage.

time can be very different for different spin-relaxation mechanisms.

The experimental evidence of charge and spin separation is still not clear. A recent experiment was done with a permalloy-Nb-permalloy sandwich.[13] The junctions were not tunnel junctions so the proximity effect may be important. A signal [Eq. (48)] was seen close to T_c; however, it was found to decrease rapidly when the temperature was lowered. An anomalously short spin diffusion length (of order 50 Å) was also obtained. This could be due to magnetic impurity scattering, since this is the only mechanism which gives an increasing spin-relaxation rate at low temperatures. The shortness of the spin diffusion length implies that case (2) applies. The anisotropy of Nb is also small because the intrinsic anisotropy in Nb is smaller than most of other superconductors and impurity scatterings will make it even less. Taking these factors into account, Eq. (30) can explain the experimentally observed $\ln(V_1-V_2) \sim -(1-T/T_c)^{1/4}$ temperature dependence of the signal. To experimentally check the idea of spin-charge separation, the following two aspects may be critical: First, one should use tunnel junctions to isolate the nonequilibrium region from the ferromagnetic material to eliminate the proximity effect and minimize magnetic impurity scattering. If the spin relaxation is due to spin-orbit scattering, then the rate will decrease at lower temperatures as we have calculated in Sec. II. Second, materials with longer spin-relaxation times in their normal state, such as Al, can be used to increase the spin voltage signal.

V. CONCLUSION

In this paper, we have performed the electron tunneling calculation of nonequilibrium spin injection and detection between ferromagnets and a superconductor. The spin-relaxation rate due to magnetic impurity scattering and spin-orbit scattering in a superconductor was also calculated. The nonequilibrium charge and spin injected into the superconductor obey diffusion equations. Under certain conditions, the charge and spin transport in a superconductor are separated in the sense that the charge current is carried by the condensate and exists only within the penetration length of the surface. The spin current is carried by quasiparticles and can exist inside the bulk of a superconductor. This can be experimentally checked by spin injection and detection technique with tunnel junctions in the geometry of Fig. 1.

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[1] P. M. Tedrow and R. Meservey, Phys. Rev. Lett. **26**, 192 (1971); Phys. Rev. B **7**, 318 (1973).
[2] X. Hao, J. S. Moodera, and R. Meservey, Phys. Rev. B **42**, 8235 (1990).
[3] M. Johnson and R. H. Silsbee, Phys. Rev. Lett. **55**, 1790 (1985); Phys. Rev. B **37**, 5312 (1988); **37**, 5326 (1988); M. Johnson, Phys. Rev. Lett. **70**, 2142 (1993).
[4] J. Clarke, Phys. Rev. Lett. **28**, 1363 (1972).
[5] M. Tinkham, Phys. Rev. B **6**, 1747 (1972).
[6] C. J. Pethick and H. Smith, Ann. Phys. (NY) **119**, 133 (1979).
[7] A. G. Aronov, JETP Lett. **24**, 32 (1976); Sov. Phys. JETP **44**, 193 (1976).
[8] A. Schmid and G. Schön, J. Low Temp. Phys. **20**, 207 (1975).
[9] L. C. Hebel and C. P. Slichter, Phys. Rev. **113**, 1504 (1959); L. C. Hebel, *ibid.* **116**, 79 (1959).
[10] A. B. Pippard, in *Nonequilibrium Superconductivity, Phonons, and Kapitza Boundaries*, Vol. 65 of *NATO Advanced Study Institute,* edited by K. E. Gray (Plenum, New York, 1981).
[11] J. R. Waldram, Proc. R. Soc. London, Ser. A **345**, 231 (1975).
[12] S. A. Kivelson and D. S. Rokhsar, Phys. Rev. B **41**, 11693 (1990).
[13] M. Johnson, Appl. Phys. Lett. **65**, 1460 (1994).

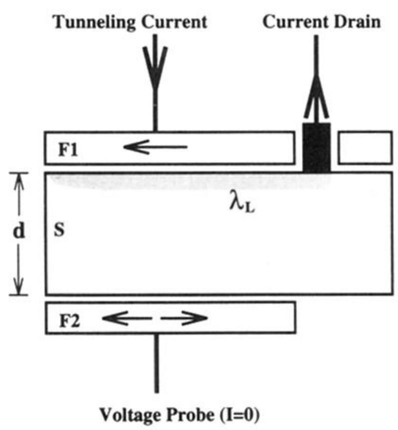

FIG. 1. A schematic setup for the tunneling spin injection and detection experiment. $F1$ is the current-injecting ferromagnetic film with the arrow inside representing the direction of magnetization. S is a superconductor. $F2$ is the detecting ferromagnetic film. When switching the direction of magnetization in $F2$, the voltage difference measured is proportional to the spin density at the junction. To check the idea of charge-spin separation in the superconductor, the penetration length (shaded region) should satisfy $\lambda_L \ll d \sim$ spin diffusion length λ_s.