Spin Hanle effect in mesoscopic superconductors

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We present a theoretical study of spin transport in a superconducting mesoscopic spin valve under the action of a magnetic field misaligned with respect to the injected spin. We demonstrate that the coherent rotation and relaxation of the spins is modified by superconductivity, and that the nonlocal magnetoresistance depends on whether the dominating spin relaxation mechanism is due to spin-orbit coupling or spin-flip scattering at impurities. We also predict a hitherto unknown subgap contribution to the nonlocal conductance in multiterminal superconducting hybrid structures.

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Effective control over spin-polarized transport is a cornerstone for many spintronics applications. 1 One way of implementing spin manipulation and control is to exploit the Hanle effect, i.e., the coherent rotation of a spin in an external magnetic field. Such rotation has been experimentally demonstrated in semiconducting nanosstructures, 2 graphene, 4 and normal metals. 5 In the latter case, however, the strong spin-relaxation mechanisms requires much larger magnetic fields to rotate the spin within a distance comparable to the spin coherence length. 6 This explains why in metallic systems only π spin rotation has been achieved experimentally in Al wires, 6, 7 in contrast to the 4π spin rotation observed in Si. 8

One possible alternative to tune the spin rotation in metals is to use superconductivity. 10 In such materials the spin carriers are the Bogoliubov quasiparticles, which move at the group velocity \( v_g \sim v_F \sqrt{\Delta^2 - \varepsilon^2} \) that tends to zero near the gap edge \( \varepsilon = \Delta \). Hence while travelling a fixed distance \( L \) they are exposed to the spin-rotating field for a longer time, which results in an enhanced spin precession. This also implies an increase of the spin relaxation due to the exchange interaction with magnetic impurities. 13, 15 However, besides generating the spin rotation and relaxation, the magnetic field induces a Zeeman splitting of quasiparticle states which can separate the contribution of spin and charge degrees of freedom. 10 and a drastic suppression of spin relaxation. 11, 12, 17 Thus, in principle the magnetic field can either enhance the spin relaxation due to the Hanle mechanism or suppress it by the polarization of the quasiparticles. Although some works have been devoted to the theory of spin relaxation in superconductors, 13–16, none of them have addressed the problem of noncollinear spin-splitting fields, essential to understand the Hanle effect in superconductors.

In this Letter we address this problem and present a full study of spin transport in a typical nonlocal measurement setup. We extend the existing spin transport theory in diffusive superconductors by taking into account noncollinear configurations of spin injector and detector electrodes and a finite external magnetic field in an arbitrary direction. We show that the nonlocal magnetoresistance depends crucially on the spin relaxation mechanism in the superconductor. If the latter is mainly due to an extrinsic spin-orbit coupling, the nonlocal spin signal in the superconducting state is suppressed by smaller fields as compared to the normal case, whereas the period of characteristic oscillations of the Hanle curve becomes smaller. In contrast, if the main source of spin relaxation is due to magnetic impurities (spin-flip scattering) the decay of the nonlocal spin signal with the applied field is larger in the superconducting state but its oscillation become less pronounced for \( T < T_c \). Our theory also predicts that the injection of spins noncollinear with the external field can generate in the superconductor a subgap pure spin imbalance that provides an additional, and hitherto unknown contribution to the subgap nonlocal conductance in multiterminal superconducting hybrid structures. 18–20

We consider the nonlocal spin valve shown in Fig. 1. A spin-polarized current is injected in the superconducting wire from a ferromagnetic electrode with magnetization \( P_I \). The detector is also a ferromagnet with a polarization \( P_D \) located at a distance \( L_D \) from the injector. Both the injector and the detector are coupled to the wire via tunnel contacts. A magnetic field \( B \) is applied in \( z \) direction. The current \( I_D \) at the detector is given by (below, we use \( h = e = k_B = 1 \))

\[
R_D I_D = \mu_0 + \mu \cdot P_D
\] (1)
where $R_D$ is the detector interface resistance in the normal state, $\mu_0$ is the the chemical potential of quasiparticles in the superconductor, and the last term describes the spin dependent contribution to the current which is proportional to the local spin accumulation $\mu$.

In a nonlocal measurement scheme, the spin accumulation is tested by measuring the voltage at the detector where no charge current flows $I_D = 0$, i.e., one sets in Eq. (1) $I_D = 0$. The spin-dependent voltage $V_S$ is defined as the difference of voltages measured in the parallel and anti-parallel configurations between the injector and detector. The nonlocal spin signal is determined by the ratio $R_S = V_S/I_{inj}$, where $I_{inj} = V\chi/R_I$ is the injected current, $R_I$ is the injector interface resistance, and $\chi = \int_0^\infty dz N_+ \frac{\partial n_+}{\partial \mu}$ is the "Yosida function" [15]. Here $N_+$ is the density of states (DOS) in superconductor near the ferromagnetic electrode and $n_0(\epsilon)$ is the Fermi function. As usual, we consider only the linear response limit $|V| \ll T$. The divergence of the spin signal in the low-temperature limit [21] is cut off by proximity-induced subgap contributions to $N_+$. The expression for the nonlocal resistance obtained from Eq. (1) reads

$$R_S = 2R_I(V\chi)^2\left<\mu \cdot P_D\right>.$$  

(2)

The local spin accumulation $\mu$ can be written in terms of the Keldysh quasiclassical Green function as $\mu = \int_0^\infty m(\epsilon) d\epsilon$, where $m(\epsilon) = \text{Tr} (\tau_3 \sigma g^K)/8$, $\tau_3$ is the third Pauli matrix in Nambu space, $g^K$ is the (2×2) matrix Keldysh component of the quasiclassical Green’s function matrix $\hat{g} = \begin{pmatrix} g^R & g^K \\ 0 & g^A \end{pmatrix}$, and $g^{RA}$ is the retarded (advanced) Green’s function. In the diffusive superconduction wire the matrix $\hat{g}$ obeys the Usadel equation [22]

$$D\nabla \cdot (\hat{g}D\nabla) + [\hat{\Lambda} + \hat{\Sigma}_{so} + \hat{\Sigma}_{sf}, \hat{g}] = 0.$$  

(3)

Here $D$ is the diffusion constant, $\hat{\Lambda} = i\epsilon\tau_3 - i(h \cdot S)\tau_3 - \Delta$, $\epsilon$ is the energy, $\hat{\Delta} = \Delta\tau_1$ the spatially homogeneous order parameter in the wire, $h = \mu_B B$ the Zeeman field, $\mu_B$ the Bohr magneton, and $S = (\sigma_1, \sigma_2, \sigma_3)$ the vector of Pauli matrices in spin space. The last two terms in Eq. (3), $\hat{\Sigma}_{so} = \tau_3^{-1}(\hat{S} \cdot \hat{g}S)$ and $\hat{\Sigma}_{sf} = \tau_3^{1/2}(S \cdot \tau_3 \hat{g}S)$, describe spin relaxation due to the spin-orbit scattering and exchange interaction with magnetic impurities, characterized by the relaxation times $\tau_{so}$ and $\tau_{sf}$ respectively. Equation (3) is complemented by the normalization condition $\hat{g}^R = \hat{g}^R f - f \hat{g}^A$, where $f$ is the distribution function with a general spin structure

$$f = f_L + f_T \tau_3 + (\sigma \cdot f_T) + (\sigma \cdot f_L) \tau_3.$$  

(4)

We assume that the transparencies of the detector and injector interfaces are small, so that up to leading order the spectral (retarded and advanced) Green functions obtain their bulk values in the presence of a Zeeman splitting field. In the present case $h = h z$, they read

$$g^R = g_{01}\tau_1 + g_{31}\tau_3 + g_{03}\tau_3 + g_{33}\tau_3$$  

and $g^A = -g_{33}g^R\tau_3$. While the terms diagonal in Nambu space ($\tau_3$) correspond to the normal GFs, the $g_{01}, g_{31}$ describe the singlet and zero-spin triplet anomalous components [22]. The coefficients in Eq. (5) are determined by solving the nonlinear equation $[\Lambda^R + \Sigma_{sf}^R + \Sigma_{so}^R, g^R] = 0$, where the spin-dependent scattering self-energies reduce the (self-consistent) spectral gap to $\Delta_0 < \Delta - h$ and smear the gap edge singularities [23], as shown in Fig. 2. Except of the temperature interval in the vicinity of $T_c$ the applied fields are much smaller than the paramagnetic Chandrasekhar-Clogston limit [24] $|h| \ll \Delta/\sqrt{2}$. However to describe the entire temperature range we calculate $\Delta$ self-consistently taking into account the effects of paramagnetic suppression and spin-scattering self-energies.
of the spectral spin polarization \( \mathbf{m} = (m_x, m_z) \), which is given by

\[
m_i = N_z f_i + h^{-1} \text{Im} g_{33}(f_i \times \mathbf{h}),
\]

where \( f_i = (f_{T1}, f_{T2}) \). The first term in Eq. (6) describes the quasiparticle contribution. It is proportional to the total DOS \( N_z = \text{Re} |g_{33}| \) modified by the Zeeman splitting and the spin dependent scattering mechanisms (see Fig. 2(b)). This contribution is only finite for energies above the spectral gap \( \varepsilon > \Delta_g \). The second term in Eq. (6), being proportional to \( \text{Im} g_{33} \), is nonzero only if the superconducting spectrum is spin-polarized. In contrast to the usual quasiparticle contribution, this term is not suppressed at low temperatures \( T \ll T_c \) since \( \text{Im} g_{33} \) is nonzero at subgap energies (dash-dotted curve in Fig. 2(b)). As we demonstrate below, this term leads to a finite subgap nonlocal conductance in the lowest order in transparency. This contribution only involves spin degrees of freedom, in contrast to previous works on the subgap charge transport [18,21].

From Eq. (4) we obtain an equation for \( f_i \) which can be written in a compact form

\[
\nabla \cdot \mathbf{j}_s = g_s m_i \times (\mathbf{h} + \mathbf{h}_s) + m_i / \tau_m, \tag{7}
\]

where we have introduced an electronic spin g-factor \( g_s = 2 \), the transversal spin current density \( \mathbf{j}_s = D \partial_s f_i / 2 \), and the diffusion tensor \( D = D_1 + i D_2 \sigma_2 \) with components defined via

\[
D_1 = D \left( 1 + |g_{02}|^2 - |g_{01}|^2 + |g_{31}|^2 - |g_{33}|^2 \right),
\]

\[
D_2 = 2D \text{Im} \left( g_{33} g_{03} - g_{31} g_{01} \right). \tag{8}
\]

In Eq. (7), \( \tau_m \) and \( \mathbf{h}_s \) are the spin relaxation time and the correction to the effective Zeeman field. In the superconducting state they are energy dependent [22], and defined as \( \tau_m^{-1} = 2h(H_2 S_2 + H_1 S_1) / (\beta^2 + H_2^2) \), \( \mathbf{h}_s = \mathbf{h}(H_2 S_2 - H_1 S_1) / (\beta^2 + H_2^2) \), where \( H_1 = 4h \text{Im} g_{33} \) and \( H_2 = 4h N_\gamma, S_1 = 16 \tau_\gamma^{-1} \left| \text{Re} g_{33} \right|^2 + \beta |\text{Im} g_{01}|^2 \), \( S_2 = 16 \tau_\gamma^{-1} \left| \text{Im} g_{33} \text{Re} g_{30} - \beta \text{Im} g_{01} \text{Re} g_{31} \right| \) with \( \tau_\gamma = (\tau_{so} + \tau_{sf}) / (\tau_{so} + \tau_{sf}) \). The parameter \( \beta = (\tau_{so} - \tau_{sf}) / (\tau_{so} + \tau_{sf}) \) characterizes the relative strength of spin-orbit and spin-flip scattering. For example, in Al wires used in the spin-transport experiments, the typical spin relaxation time is \( \tau_\gamma \approx 800 \text{ ps} \approx 40 / T_{c0} \) where \( T_{c0} \approx 1.6 \) K is the bare critical temperature of the superconductor in the absence of exchange field [13]. In Al \( \beta \approx 0.5 \) indicating the dominating spin-flip relaxation mechanism [15] while for Nb one expects spin-orbit as the main source of scattering [17].

The set of equations is completed by boundary conditions (BC) at the spin-polarized injector interface \( z = 0 \). We use the BC of Ref. [22] that generalizes the Kupriyanov-Lukichev [27] one to the case of spin-dependent barrier transmission. We find

\[
\mathbf{j}_s |_{z=0} = -D \kappa_T m_i |_{z=0}. \tag{9}
\]

Here \( \kappa_T = (\sigma_n R_{J\square})^{-1} \) is the injector transparency, where \( \sigma_n \) is the bulk normal state conductivity of the S wire and \( R_{J\square} \) is the barrier resistance per unit area. To evaluate the r.h.s. of BC (9) we use the Eq. (6) neglecting terms of second and higher order in \( \kappa_T \) so that \( f_i = P_i (n_0 (\varepsilon - V) - n_0 (\varepsilon + V)) / 2 \). This expression describes to the lowest order by transparency \( \kappa_T \) the injection of non-equilibrium electrons from the voltage-biased electrode through the spin-filtering ferromagnetic barrier. After substitution of Eq. (6) into Eq. (4) one can easily verify that there is a finite subgap contribution to the spectral spin current originating from the second term in r.h.s. of Eq. (4). It is important to emphasize that this subgap spin imbalance appears in linear order in \( \kappa_T \), and exists exclusively in the presence of a Zeeman field and noncollinear spin injection, \( \mathbf{h} \neq \mathbf{P}_1 \). Equation (7) can be solved analytically. The components of \( f_i \) have the form \( f_{T1} = - \text{Im} (\alpha e^{-k \varepsilon}) \) and \( f_{T2} = \text{Re} (\alpha e^{-k \varepsilon}) \), where \( \alpha \) is an integration constant determined by the BC, and

\[
k_T = \left[ \left( S_1 - H_1 - i(S_2 + H_2) / (D_1 - i D_2) \right) \right]^{1/2}, \tag{10}
\]

is the inverse length scale. Its (positive) real part determines the inverse (spectral) spin relaxation length whereas its imaginary part the precession of the spin of quasiparticles with energy \( \varepsilon \). As can be seen from Figs. 3(a),(b), the precession and relaxation lengths depend on the nature of the spin scattering mechanism. At intermediate temperatures below \( T_c \), the main contribution to the spin-dependent \( \mathbf{m} \) in Eq. (2) comes from energies close to the gap \( \Delta_g \). In the case of dominating spin-orbit scattering (\( \beta < 0 \)), one clearly sees in Figs. 3(a),(b) that while \( \kappa_T \) at \( \varepsilon \approx \Delta_g \) is larger than in the normal case (\( \varepsilon \gg \Delta_g \)), the imaginary part of \( \kappa_T \) has a peak. This results in a modified Hanle curve for temperatures below \( T_c \) [see Figs. 3(c),(e)] in which the suppression of the spin signal (\( R_{Sy} \) corresponding to the detector polarization \( P_D = P_I \mathbf{P}_y \) so that \( P_D \parallel P_I \perp \mathbf{h} \)) appears at smaller magnetic fields than in the normal state, while the oscillation becomes smaller. In contrast, if the spin-flip mechanism dominates (\( \beta > 0 \)), the imaginary part of \( \kappa_T \) is suppressed at \( \varepsilon \approx \Delta_g \) [Figs. 3(b)]. This leads to an increase of the oscillation period of \( R_{Sy}(h) \) by decreasing the temperature [Figs. 3(d),(f)]. The real part \( \kappa_T \) has a larger value than in the normal state [Fig. 3(a)]. Such an increase is mainly determined by the large renormalization of the spin relaxation in the superconducting state associated with spin-flip scattering [13,14]. In this case the spin relaxation length has a weaker dependence on external magnetic field than in the normal state. This explains the Hanle curves shown in Figs. 3(d),(f), where the decay scale of \( R_{Sy}(h) \) increase towards lower temperatures at \( T < T_c \).

At sufficiently low temperatures, the main contribution to the nonlocal resistance comes from energies below the gap in Eqs. (6,9), and the transport is dictated
by subgap tunneling processes. We find that in the non-collinear contribution the sub-gap processes are of the first order in the tunnelling parameter $\kappa_1$. For the particular orientation of the field $\mathbf{h} \perp \mathbf{P}_I$ we can neglect the previously discussed crossed Andreev reflection or elastic cotunneling processes \cite{20} provided $|\mathbf{h}| \gg \kappa_1 \sqrt{D/D_\Delta}$.

The subgap tunneling of quasiparticles between injector to the detector electrodes leads to a complete suppression of the Hanle effect. Indeed as shown in Figs. 3(a), (b), the inverse length $k_F$ for $\varepsilon < \Delta_g$ is real. Moreover, it can be shown to have only a very weak dependence on $h$. As a result, in the configuration $\mathbf{P}_D \parallel \mathbf{P}_I \perp \mathbf{h}$ both the precession and decay of the nonlocal signal disappears at $T \to 0$, as shown in Figs. 3(a), (c).

However, if instead of the above analyzed configuration one assumes that the three vectors ($\mathbf{P}_D$, $\mathbf{P}_I$, $\mathbf{h}$) are perpendicular to each other (e.g. $\mathbf{P}_D = P_D \mathbf{e}_x$, $\mathbf{P}_I = P_I \mathbf{y}$, $\mathbf{h} = h \mathbf{z}$), the subgap current is absent in the detector circuit and the corresponding spin signal $R_{sz}$ has a strong dependence on $h$ even in the limit $T \to 0$ (see Figs. 4(b), d). Note that $R_{sz}(h) = -R_{sz}(-h)$ and therefore, in contrast to the usual Hanle effect, this signal can be measured without changing the magnetization of electrodes $\mathbf{P}_D, \mathbf{P}_I$ but just by changing the sign of the external magnetic field $\mathbf{h}$.

In conclusion, we have developed a theoretical framework to study spin rotation and relaxation in superconductors in the case of noncollinear spin fields. We have analyzed the Hanle effect in a mesoscopic superconductor and demonstrated that the nonlocal magnetoresistance deviates from the one in the normal state. Moreover, we show that the Hanle curves depend on the nature of the spin scattering mechanism, either spin-orbit or spin-flip impurities. Our findings provide a way to identify these mechanisms by standard magnetoresistance measurements in nonlocal spin valves, and establish the fundamental physics underpinning the spin control and manipulation in superconducting devices.

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