Long-Range Spin Accumulation from Heat Injection in Mesoscopic Superconductors with Zeeman Splitting

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We describe far-from-equilibrium nonlocal transport in a diffusive superconducting wire with a Zeeman splitting, taking into account different spin relaxation mechanisms. We demonstrate that due to the Zeeman splitting, an injection of current in a superconducting wire creates spin accumulation that can only relax via thermalization. This effect leads to a long-range spin accumulation detectable in the nonlocal signal. Our model gives a qualitative explanation and provides accurate fits of recent experimental results in terms of realistic parameters.

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Hybrid ferromagnetic-superconducting (FS) structures reveal a rich physics originating from the interplay between magnetism and superconductivity [1,2]. While most of the research activity has been focused on the study and detection of proximity-induced triplet superconducting correlations in an equilibrium situation [2,3], more recent experiments addressed the problem of spin and charge accumulation in superconducting wires [4–11].

This Letter is motivated by a puzzling experimental finding that in superconductors with a strong Zeeman splitting, the spin accumulation has been detected at distances from the injector much larger than the spin-relaxation length in the normal state [6–8]. Moreover, the spin accumulation can be created by injecting the current from nonferromagnetic electrodes [8].

To explain this unusual behavior we develop a microscopic model based on the well-established Keldysh kinetic equations for superconductors extended to spin-dependent phenomena, and solve this puzzle. We demonstrate that the observed long-range spin accumulation can be understood as a thermoelectric effect for Bogolubov quasiparticles in a spin-polarized superconductor. Because of the Zeeman splitting of spin subbands, the heating of a superconducting wire originated, for example, by an injected current produces a spin accumulation that can be detected as an electric signal by a ferromagnetic detector.

Recent theoretical works have shown that the linear thermoelectric effect in superconductors with a Zeeman field is exponentially small at the temperatures well below the energy gap $k_B T \ll \Delta$ [12,13]. Hence, in order to explain the large electric signals observed in the experiments [6–8], it is necessary to consider nonlinear thermoelectric effects produced by the quasiparticles injected at

voltages exceeding the energy gap. The spin accumulation created in such a way can relax only due to the thermalization of injected quasiparticles, and therefore the spin relaxation length is determined by the inelastic electron-phonon and electron-electron scattering that can well exceed the usual spin diffusion length.

If the Zeeman splitting in a superconducting wire is induced by an applied external magnetic field, in addition to the splitting the magnetic field generates an orbital depairing effect. The depairing causes a strong suppression of superconductivity, and also provides the main source of charge imbalance relaxation in superconductors at low temperatures [14]. The different behaviors observed for the nonlocal conductance g_{nl} as a function of the injection voltage V_{inj} depend on the value of the orbital depairing parameter α_{orb} defined below. Taking this effect into account, our theory provides accurate fits of the experimental data [see Fig. 3].

The main physics can be understood by first considering the possible quasiparticle nonequilibrium modes that can be excited by current injection into a superconductor with Zeeman splitting [see Fig. 1(a)]. The spin states are split by the Zeeman energy $2\mu_B B$, where μ_B is Bohr magneton and B is the external magnetic field, which gives rise to four distinct quasiparticle branches, electron or hole and spin up or spin down. Generalizing the well-known description of nonequilibrium states in spin-degenerate superconductors [15], we introduce a four-component electron distribution function for the modes illustrated in Figs. 1(b)–1(e). It includes two modes with electron-hole branch imbalance: the charge f_T and spin-energy f_{L3} imbalance modes. The remaining modes are electron-hole symmetric describing the spin f_{T3} and energy f_L nonequilibrium.

FIG. 1 (color online). (a) Schematic density of states, illustrating the nonequilibrium population created by current injection from a voltage-biased normal electrode into a superconductor with Zeeman splitting. (b)–(e) Dispersion curves of quasiparticle excitation energies in a superconductor in the presence of Zeeman splitting. The occupation numbers, corresponding to different nonequilibrium modes, are indicated schematically: Small filled circles correspond to the occupied states and large open circles show an equilibrium population. (b) Charge imbalance f_T , (c) spin energy imbalance f_{L3} , (d) spin imbalance f_{T3} , and (e) energy imbalance f_L . Elastic relaxation processes towards equilibrium are shown by dashed arrows, in (b) charge imbalance relaxation and in (c), (d) spin imbalance relaxation due to the spin-orbital and spin-flip impurity scattering.

Electrical probes are sensitive to the total charge imbalance, and spin-selective probes (e.g., ferromagnets) to the total spin imbalance in the superconductor. Qualitatively, these can be found by counting the number of occupied states on the different branches [16]. The charge imbalance is determined by the difference between the number of occupied states in the electron and hole branches, i.e., by the f_T and f_{L3} components in Figs. 1(b) and 1(c). For the spin accumulation on the other hand, the relevant modes are f_{T3} and f_L .

The total charge imbalance and nonequilibrium spin accumulation in the superconductor can be characterized by the local quasiparticle electrostatic potential μ and the difference of potentials between spin-up and spin-down species μ_z , which can be related to the distribution functions via (here and below, $\hbar = k_B = 1$, the detailed derivation is given in the Supplemental Material [18])

$$\mu = -\frac{1}{2} \int_0^\infty d\varepsilon (N_+ f_T + N_- f_{L3}), \quad (1)$$

$$\mu_z = -\frac{1}{2} \int_0^\infty d\varepsilon [N_+ f_{T3} + N_- (f_L - n_0)], \quad (2)$$

where $N_+ = N_\uparrow + N_\downarrow$ is the total density of states (DOS), $N_- = N_\uparrow - N_\downarrow$ is the DOS difference between the spin subbands, and $n_0(\varepsilon) = \tanh(\varepsilon/2T)$ is the equilibrium distribution function.

According to Eq. (2) there are two contributions to the spin signal. One is described by the first term in the integrand of Eq. (2), which is finite even in the absence of Zeeman splitting. It can be generated by spin-polarized injection from a ferromagnetic electrode, but relaxes at a short length scale determined by the elastic scattering processes shown in Fig. 1(d). The second term is generated from the longitudinal component f_L , and is only finite in the presence of a Zeeman splitting of the DOS ($N_- \neq 0$). This contribution is independent of elastic spin scattering mechanisms: energy-conserving transitions do not result in relaxation of the f_L mode, which corresponds to equal populations of states at the same energy, as illustrated in Fig. 1(e). Relaxation of this mode can be only provided by inelastic processes, e.g., electron-phonon and electron-electron scattering.

The mechanism described above explains qualitatively recent experimental observations of a long-range spin accumulation in terms of excitation of the energy non-equilibrium mode f_L . Spin polarization is also expected to arise by injecting nonpolarized currents from nonferromagnetic electrodes. This is also in accordance with the experimental results [8]. Indeed, current injection at voltages exceeding the superconducting energy gap unavoidably generates energy imbalance simply by heating the quasiparticles in the superconducting wire. According to Eq. (2), it is then the Zeeman splitting in the superconductor which converts the energy imbalance into observed spin polarization.

To proceed beyond the above qualitative explanation of the long-range spin accumulation observed in recent experiments [6–8], we present a microscopic theory that provides a quantitative picture explaining accurately the experimental observations in terms of realistic parameters. For this purpose, we consider a typical geometry (see Fig. 2) used for nonlocal measurements to test the non-equilibrium spin polarization [7]. It consists of a nonlocal spin valve where the superconducting wire is driven to a nonequilibrium state by the injected current from the electrode, which in principle can be either ferromagnetic or nonferromagnetic [6–8]. To consider the particular experimental situation of Ref. [7], we assume that the detector is a ferromagnetic electrode that is used to measure the nonlocal differential conductance $g_{nl} = dI_{det}/dV_{inj}$,

FIG. 2 (color online). (a) Schematic view of the setup for nonlocal conductance measurements. Here we assume that the polarizations of the magnetic contacts are collinear with the magnetic field, $\mathbf{P}_{\text{inj}} \parallel \mathbf{P}_{\text{det}} \parallel \mathbf{B}$. (b) Schematic illustration of the thermoelectric effect at the interface between the ferromagnetic detector (FD) and the superconductor (S) with Zeeman splitting. The charge transfer is mediated by transitions of spin-down electrons between FD and S. In the presence of a Zeeman splitting in S the energy imbalance mode creates more occupied than empty states in the spin-down subband that in turn enhances the transmission of electrons from the S to the FD side than vice versa. This, in combination with the spin-filter effect at the interface, explains how the energy imbalance in the spin-polarized S is converted into an electric current.

where I_{det} is the current in the detector circuit at zero bias $V_{\text{det}} = 0$.

To calculate the tunneling current I_{det} measured by the detector we use a generalization of Ohm's law for the case of spin-dependent interface conductance $I_{\text{det}} = G_{\uparrow}(\mu + \mu_z) + G_{\downarrow}(\mu - \mu_z)$ where $G_{\uparrow}$ and $G_{\downarrow}$ are the conductances for the spin-up and spin-down electrons. Introducing the effective polarization [7] $P_{\text{det}} = (G_{\uparrow} - G_{\downarrow})/G_{\text{det}}$ and the total conductance $G_{\text{det}} = G_{\uparrow} + G_{\downarrow}$ we get

$$I_{\text{det}} = G_{\text{det}}(\mu + P_{\text{det}}\mu_z). \quad (3)$$

The first term in the rhs of Eq. (3) is the shift of quasiparticle chemical potential, determined by the charge imbalance [Eq. (1)], and the second measures the spin accumulation [Eq. (2)]. As illustrated in Fig. 2(b), energy nonequilibrium in a superconductor with Zeeman splitting, caused for example by heating, can produce a nonzero net current at the spin-polarized detector electrode.

In order to describe the various nonequilibrium modes and relaxation mechanisms relevant for a diffusive spin-polarized superconducting wire, we use the quasiclassical

Usadel-Keldysh theory [2]. We take into account the spin and charge imbalance relaxation due to the spin-orbit scattering, exchange interaction with magnetic impurities, and orbital magnetic depairing [14]. This results in a set of coupled diffusion equations for the four distribution function components. As shown in the Supplemental Material [18], if one neglects inelastic relaxation processes, the system of equations separates into two decoupled sets. This can be understood by considering again the generic picture of nonequilibrium modes in Figs. 1(b)–1(e). The elastic relaxation processes, represented by the dashed arrows, couple only the modes f_T and f_{L3} . An additional pairwise coupling between the other two components f_L and f_{T3} arises from the diffusive terms due to the difference in diffusion coefficients for spin-up and spin-down electrons, as demonstrated in the Supplemental Material [18]. Notice that our theory generalizes previous models by including the Zeeman splitting [22].

The solution of the diffusion equations for the components f_T and f_{L3} is given by a superposition of two exponentially decaying functions with different length scales. These functions describe the relaxation of the (coupled) charge and spin energy imbalance. The components f_L and f_{T3} on the other hand have a quite different behavior. While the function f_{T3} also decays exponentially on length scales determined by elastic scattering, the energy imbalance varies linearly and is limited only by boundary conditions. This long-ranged solution is in accordance with the qualitative picture illustrated by Fig. 1, which demonstrates that elastic scattering cannot relax the energy imbalance mode.

We obtain the relaxation length scales and amplitudes of the nonequilibrium modes by solving the Keldysh-Usadel equations with boundary conditions obtained from the general tunneling Hamiltonian approach [23]. Details of the kinetic theory are described in the Supplemental Material [18].

We have chosen realistic values of parameters corresponding to the diffusive Al wires used in the experiment [7]: diffusion constant $D = 40 \text{ cm}^2/\text{s}$, normal-state spin relaxation length and time $\lambda_{\text{sn}} = 350 \text{ nm}$, and $\tau_{\text{sn}} = 30 \text{ ps}$, respectively. We parameterize the spin-orbit (SO) and spin-flip (SF) scattering rates in the normal state as $\tau_{\text{SO}} = 2\tau_{\text{sn}}/(1 - \beta)$ and $\tau_{\text{SF}} = 2\tau_{\text{sn}}/(1 + \beta)$ where $\beta \in [-1, 1]$ describes their relative magnitude. To obtain the most accurate fits we put $\beta = -0.5$ implying the dominant role of the spin-orbital scattering [24]. Superconductivity modifies these rates drastically [9,18].

The sizable spin relaxation leads to a considerable suppression of the critical temperature [25]. Assuming that the real $T_c = 1.7 \text{ K}$ we obtain that in the absence of spin relaxation $T_{c0} = 2 \text{ K}$. The order parameter at zero field is given by $\Delta = 1.64T_{c0} = 3.28 \text{ K}$ so that the coherence length is $\xi = \sqrt{D/\Delta} \approx 95 \text{ nm}$. In our calculations we set the temperature to $T = 100 \text{ mK}$.

The orbital depairing rate can be written in the form $\tau_{\text{orb}}^{-1} = T_{c0}\alpha_{\text{orb}}(\mu_B B/T_{c0})^2$ where α_{orb} is the dimensionless parameter measuring the relative strength of orbital and paramagnetic effects. It can be estimated as [26] $\alpha_{\text{orb}} = T_{c0}\Delta/(\mu_B B_c)^2$, where $B_c = \sqrt{12}\phi_0/(\pi\xi W)$ is the critical field of a thin superconducting film of width W , and ϕ_0 is the magnetic flux quantum. The most accurate fits to the experimental curves are obtained for $\alpha_{\text{orb}} = 0.42$ which yields an effective film width $W \approx 4$ nm. The distance between injector and detector is the one of the experiment [7], $L_{\text{det}} = 3\lambda_{\text{sn}} \approx 1 \mu\text{m}$, as well as the polarizations $P_{\text{inj}} = P_{\text{det}} = 0.19$.

In Fig. 3 we show a comparison to the experimental results from Fig. 3(a) in Ref. [7] (blue lines), and the nonlocal conductance calculated from Eqs. (1), (2), (3) (red lines) for several values of the magnetic field. The experimental voltage is divided by $T_{c0} = 2 \text{ K} = 172 \text{ mV}$

FIG. 3 (color online). The nonlocal conductance as a function of the injecting voltage, $g_{\text{nl}}(V_{\text{inj}})$: Comparison between theory (red curves) and experiment (blue curves). Magnetic field in panels (a–f) increases from 0.25 T to 1.5 T with the step 0.25 T. Experimental curves are taken from Fig. 3(a) in Ref. [7]. The parameters of the theoretical model are the same as in the experiment and listed in the text.

in all curves and the vertical scale of theoretical curves is scaled by adjusting the detector conductance G_{det} , which is the same for all panels in Fig. 3.

There is an overall excellent agreement between the calculated and experimental curves that demonstrates the correctness of both, our qualitative explanation of the observed long-range spin accumulation and our microscopic theory. Note that in the experiment, the decay of the long-range spin imbalance is only limited by inelastic relaxation, which is not taken into account in our kinetic equations. The observed relaxation length $\lambda \sim 5\text{--}10 \mu\text{m}$ cannot be explained by electron-phonon scattering, which already in the normal state leads to a much larger value $\lambda_{\text{ph}} = \sqrt{\tau_{\text{ph}} D} \approx 20 \mu\text{m}$ [7,8]. Electron-electron scattering, on the other hand, can redistribute the total energy in the electron system and damp nonequilibrium components of the signal. In order to obtain the observed relaxation length $\lambda_{ee} \sim 5\text{--}10 \mu\text{m}$ one should assume that the e - e scattering time is $\tau_{ee} \sim 1\text{--}10$ ns, which can be achieved in bulk dirty Al [27] as well as in low-dimensional samples [28]. The e - e thermalization process as well as nonuniversal properties of the heat transport in real experimental setups could explain the suppression of the spin imbalance relaxation by the Zeeman field observed in Refs. [7,8].

The influence of e - e scattering likely explains the discrepancy between our theoretical model and the experimental data in the large $V_{\text{inj}} > 0$ region of Figs. 3(c)–3(f). The high-voltage tails of the experimental curves $g_{\text{nl}}(V_{\text{inj}})$ have an antisymmetric component, which is smaller in the theoretical results. Because of the thermalization by e - e interaction, the distribution of injected quasiparticles $f_L(\epsilon)$ acquires a smoothed-out part, which produces additional antisymmetric spin signal tails in $g_{\text{nl}}(V_{\text{inj}})$ at large voltages. Our preliminary results suggest this can improve the agreement, but the detailed investigation of the effects related to e - e interaction is beyond the scope of this Letter.

To conclude, we have developed a theoretical framework to study the transport properties of superconductors with a Zeeman splitting. We have demonstrated that the splitting field leads to a strong suppression of the relaxation of spin imbalance created by the injected current. In particular, the long-range spin accumulation observed in recent experiments is shown to be a manifestation of the nonlinear thermoelectric effect and it is only limited by the inelastic relaxation length which can be larger than the spin relaxation time in normal metals by several orders of magnitude. Our model gives a qualitative explanation for a wide range of experiments on SF nonlocal spin valves, and predicts a strong dependence of the nonlocal conductance on orbital depairing, characterized by α_{orb} . Besides explaining the properties of superconductor-ferromagnet structures, the approach may be straightforwardly extended for the general description of thermoelectric effects in far-from-equilibrium situations in terms of the well-established nonequilibrium quasiclassical theory.

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