

Proximity-effect–assisted decay of spin currents in superconductors

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Abstract – The injection of pure spin current into superconductors by the dynamics of a ferromagnetic contact is studied theoretically. Taking into account the suppression of the order parameter at the interfaces (inverse proximity effect) and the energy-dependence of spin-flip scattering, we determine the temperature-dependent ferromagnetic resonance linewidth broadening. Our results compare well with recent experiments in Nb|permalloy bilayers (BELL C. *et al.*, *Phys. Rev. Lett.*, **100** (2008) 047002).

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Cooper pairs in conventional superconductors are spin-singlet states and therefore cannot carry a spin current. Some aspects of the resilience of the superconducting state against spin-current injection have been experimentally demonstrated in hybrid ferromagnet-superconductor spin valves [1], switches [2], and π -junctions [3]. In these experiments, the spin current flow in the superconducting state can only be inferred via charge current measurements. This complicates the understanding of the spin current flow in superconductors.

In recent experiments by Bell *et al.* [4], a magnetization precessing under ferromagnetic resonance (FMR) conditions “pumps” spins into adjacent superconductors. We study such ferromagnet|superconductor structures theoretically, and show how spin pumping [5] leads to pure spin current injection into superconductors. The angular momentum loss of the magnetization can be observed directly in terms of an increased broadening of the FMR spectrum. The spin transport thus measured as a function of temperature and device/material parameters offers direct insight into spin-flip relaxation and the inverse

proximity effect in superconductors. Our theory agrees well with recent experimental results [4], and provides predictions for future experiments.

The theoretical challenge of spin-pumping into superconductors as compared to normal conductors is the strong energy dependence of quasiparticle transport properties around the superconducting energy gap [6]. A qualitative explanation of the effect is the following. Spin current in the singlet superconductor can only be carried by quasiparticle excitations. The low-energy density of quasiparticle states is suppressed by superconducting correlations, thus the spin transport resistivity is enhanced in the superconducting state. This results in reduced spin injection from the ferromagnet in comparison to the normal state, and is measured as a reduced damping of the magnetization dynamics.

Spin-flip processes enhance the measured magnetization damping by dissipating spin current [5]. In superconductors, the energy-dependent spin-flip scattering rates caused by spin-orbit coupling or magnetic impurities differ. Experiments that directly probe spin transport, such as ref. [4], therefore provide unique information about the spin-flip scattering mechanism. A complicating factor

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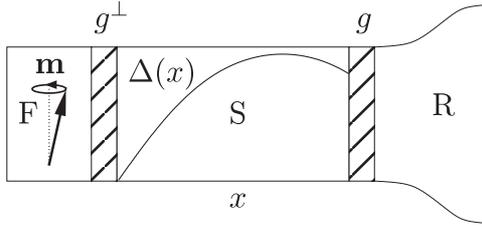


Fig. 1: The ferromagnet|superconductor|reservoir (F|S|R) structure. The precessing magnetization $\mathbf{m}(t)$ pumps spins into S which are dissipated in R or by spin-flip in S. The “spin-mixing” conductance of the F|S interface for spins polarized transverse to the magnetization direction is g^\perp and the S|R interface conductance is g . The superconducting gap $\Delta(x)$ is suppressed close to the interfaces by the inverse proximity effect.

is the inverse proximity effect [7] that suppresses the superconducting order parameter close to a metallic interface with ferromagnets like Ni, Co, and Fe. The resulting spatial dependence of the superconducting gap requires solution of the full transport equations in the entire superconducting layer. The temperature dependence of the FMR linewidth near and below the critical temperature therefore provides a wealth of information about superconducting correlations in magnetic heterostructures and spin-flip processes, with potential implications for different areas of mesoscopic physics.

In this letter we develop a theory of energy-dependent spin pumping at a ferromagnet|superconductor interface and calculate the resulting spectral spin current flow in the superconductor. We consider a diffusive metallic heterostructure consisting of a superconducting layer (S) with thickness L that is sandwiched by a ferromagnet (F) with thickness d and a reservoir (“spin sink”) (R), see fig. 1. The precessing magnetization $\mathbf{m}(t)$ emits a spin current that is transversely polarized with respect to the instantaneous magnetization direction [5]. The spin current that flows through S is immediately dissipated upon reaching R. R thus increases the sensitivity of the experiments to the spin transport through S. R can be realized either as a cap of an efficient spin-flip scattering material such as Pt or a large reservoir of a high-mobility metal [5]. We assume sufficient thermal anchoring such that any heating by microwave radiation can be disregarded. Further, we consider elastic transport since the inelastic scattering length exceeds the system length scales at the relevant quasiparticle energies.

The magnetization dynamics is determined by the generalized Landau-Lifshitz-Gilbert equation

$$\frac{d\mathbf{m}}{dt} = -\gamma \mathbf{m} \times \mathbf{H}_{\text{eff}} + \frac{G_0}{\gamma M_S} \mathbf{m} \times \frac{d\mathbf{m}}{dt} + \frac{\gamma}{M_S V} \mathbf{I}_s. \quad (1)$$

Here γ is the gyromagnetic ratio, \mathbf{H}_{eff} is the effective magnetic field, M_S is the saturation magnetization density, and V is the volume of the ferromagnet. The intrinsic dissipation in the bulk ferromagnet is parameterized by the

Gilbert damping constant G_0 . \mathbf{I}_s is the spin (*i.e.* angular momentum) current generated by the precessing ferromagnet. This loss of angular momentum is equivalent to an interface contribution to the magnetization damping and observable in terms of the enhanced FMR linewidth broadening. Our task is to evaluate the effect of superconducting correlations on \mathbf{I}_s .

To illustrate the physics, we first sketch the results for $\mathbf{m}(t)$ rotating in the xy -plane and in the absence of spin-flip scattering. The ferromagnet then emits a constant spin current polarized along the z -axis which can dissipate in R [5]. To determine \mathbf{I}_s we solve the energy-dependent transport through the heterostructure (see (6) below). Our result (2) can be summarized in terms of a spectral Ohm’s law for spin transport, *viz.* the energy-dependent resistance for spin transport, $R_{\text{eff}}^\perp(E)$, is governed by the spin transport resistance of S in series with the spin-mixing resistance $r^\perp = 1/g^\perp$ of the F|S interface and the conventional resistance $r = 1/g$ of the S|R interface,

$$R_{\text{eff}}^\perp(E) = \frac{r^\perp}{N(0, E)} + \int_0^L dx' \frac{\rho_L(x', E)}{A} + \frac{r}{N(L, E)}. \quad (2)$$

Here $\rho_L = 1/(\hbar N_0 D_L)$ is the effective spin resistivity of the superconductor in units of e^2/h , N_0 is the density of states at the Fermi level in the normal state, and $D_L(x, E)$ and $N(x, E)$ are the effective spin diffusion coefficient and the normalized density of state in S at position x and energy E , respectively [6].

At zero temperature, the relevant quasiparticle energy E is determined by the FMR frequency which is typically $f_{\text{FMR}} \sim 10$ GHz. For BCS superconductors $\hbar f_{\text{FMR}}/\Delta_0 \approx 0.3 K/T_c$ where Δ_0 is the bulk zero-temperature energy gap and T_c the critical temperature of the superconductor. For small-angle precession, the effective instantaneous “rotation” frequency can be introduced as $f \sim \phi f_{\text{FMR}}$, where ϕ is the angle of precession. Thus the relevant energy scale for FMR-generated excitations is in practice expected to be much smaller than $\hbar f_{\text{FMR}}$, and the characteristic energy of pumped electrons is set by the temperature, see eq. (3) below. R_{eff}^\perp depends on temperature through the local gap $\Delta(x, T)$ which determines $N(x, E)$ and $\rho_L(x, E)$. Close to the F|S interface $\Delta \approx 0$ due to the inverse proximity effect. The spin angular momentum loss of the ferromagnet determined by (2), is consistent with the Gilbert phenomenology in terms of an increased damping parameter G in (1),

$$G = G_0 + \frac{(g_L \mu_B)^2}{2\pi \hbar} \frac{1}{d} \int dE \frac{-df_{\text{FD}}(E)/dE}{AR_{\text{eff}}^\perp(E)} \quad (3)$$

Here g_L is the g -factor, μ_B is the Bohr magneton, A is the sample cross-section area, and f_{FD} is the Fermi-Dirac distribution function.

At temperatures $T \ll T_c$, $\Delta(x)$ as a function of the distance from the F|S interface approaches the bulk value on the scale of the bulk superconducting coherence length $\xi_0 = \sqrt{\hbar D/2\pi k_B T_c}$. Since the relevant spin resistivity

$\rho_L(x, E)$ and thus R_{eff}^\perp are very large for $E < \Delta$, ξ_0 sets the penetration length scale for spin current into the superconductor. At low temperatures and $L > \xi_0$ the Gilbert damping (3) will therefore be weakly enhanced, $G \approx G_0$. On the other hand, at $T \lesssim T_c$ the gap is suppressed throughout S and transport channels at energies $E \gtrsim \Delta$ become accessible. R_{eff}^\perp and the Gilbert damping then approach the normal-state values.

Spin-flip scattering in S dissipates the spin current emitted from F, and enhances G by suppressing the back-flow of spins into the ferromagnet. The spin-flip length in the normal state is given by $l_{\text{sf}} = \sqrt{D\tau_{\text{sf}}}$, where D is the normal-state diffusion coefficient. We take into account spin-flips caused by magnetic impurities as well as spin-orbit coupling at non-magnetic impurities in terms of the spin-flip rate $1/\tau_{\text{sf}} = 1/\tau_{\text{m}} + 1/\tau_{\text{so}}$ [6]. The spin-orbit coupling respects the symmetry of singlet Cooper pairs, whereas the pair-breaking scattering by magnetic impurities suppresses superconductivity and reduces T_c . Below T_c , the spin-flip rates in S depend on energy. For $E < \Delta$ spin-flip rates both due to spin-orbit coupling and magnetic impurities are suppressed. For $T \ll T_c$ and $L > \xi_0$, the Gilbert damping will therefore be weakly enhanced. For $E > \Delta$ the spin-flip rate due to magnetic impurities is enhanced whereas the spin-flip rate due to spin-orbit coupling is similar to that in the normal state. We therefore predict a non-monotonic temperature dependence of the Gilbert damping close to the critical temperature when spin-flip is dominated by magnetic impurities. Experimental data indicate that $l_{\text{sf}} > \xi_0$ for typical S. $l_{\text{sf}} = 48 \text{ nm}$ and $\xi_0 = 13 \text{ nm}$ has been reported for Nb [1] (which is used in ref. [4]) whereas $l_{\text{sf}} = 1.1 \mu\text{m}$ and $\xi_0 = 124 \text{ nm}$ for Al [8,9]. When $L \leq \xi_0$ spin-flip in S is therefore inefficient since $L \leq \xi_0 < l_{\text{sf}}$ in these materials and can be disregarded. When $L \gg l_{\text{sf}}$ the spin current never reaches R so that G is governed exclusively by spin-flip in S for all temperatures. In the interesting regime in which $l_{\text{sf}} \approx L$, the full theoretical treatment sketched in the following has to be invoked in order to compute the competing effects that determine G .

The enhanced magnetization damping from spin pumping in (1) is determined by the total spin current leaving F. This can be expressed as an energy integral over the pumped and back-flow currents $\mathbf{I}_s = \int dE (\mathbf{i}_s^{\text{inj}} - \mathbf{i}_s^{\text{back}})$. To obtain \mathbf{I}_s we must calculate the nonequilibrium spin transport throughout the heterostructure. Conservation of spectral spin current at the F|S and S|R interfaces give boundary conditions for the transport problem. The energy-dependent spin current injected into S is [5,10]:

$$\mathbf{i}_s^{\text{inj}}(E) = \frac{\hbar N(0, E)}{4\pi} \frac{f_{\text{FD}}(E - hf/2) - f_{\text{FD}}(E + hf/2)}{hf} \times \left(g_r^\perp \mathbf{m} \times \frac{d\mathbf{m}}{dt} + g_i^\perp \frac{d\mathbf{m}}{dt} \right), \quad (4)$$

where f is the instantaneous rotation frequency. Here, g_r^\perp and g_i^\perp are the real and imaginary parts of spin-mixing

conductance. For metallic interfaces, $g_r^\perp \gg g_i^\perp$ [11]. We therefore disregard the ‘‘effective field’’ g_i^\perp in (4), although it contributes to the interface boundary conditions discussed below. The magnetization damping that follows from (4) is frequency dependent beyond the Gilbert phenomenology. We have checked numerically that the f -dependent terms contribute weakly to the damping even when $hf \lesssim \Delta_0$ for the parameters studied. We therefore restrict attention to the linear response regime in which the Fermi-Dirac functions in (4) can be expanded to first order in hf . This leads to frequency-independent enhanced Gilbert damping in (1). The spectral back-flow of spin current into F induced by the spin accumulation on the S side of the interface is

$$\mathbf{i}_s^{\text{back}}(E) = -\frac{N(0, E)}{4\pi} g_r^\perp \mathbf{h}_{\text{TS}}(0, E). \quad (5)$$

where $\mathbf{h}_{\text{TS}}(x, E)$ is the nonequilibrium spin distribution function. The total spin current inside the S bulk is given the gradient of this distribution, $\mathbf{I}_s(x) = \hbar AN_0 \int_{-\infty}^{\infty} dE D_L(E, x) \partial_x \mathbf{h}_{\text{TS}}(x, E)/2$. The Keldysh transport theory determines the diffusion equation for $\mathbf{h}_{\text{TS}}(x, E)$ [6],

$$\left(N \partial_t + \partial_x D_L \partial_x - \frac{\alpha_{\text{TSTS}}^{\text{m}}}{\tau_{\text{m}}} - \frac{\alpha_{\text{TSTS}}^{\text{so}}}{\tau_{\text{so}}} \right) \mathbf{h}_{\text{TS}} = 0. \quad (6)$$

Diffusion through S is taken to be instantaneous on the scale of the FMR frequency as long as $f < D/L^2$ and/or $f \ll 1/\tau_{\text{sf}}$ so that \mathbf{h}_{TS} in (6) becomes time-independent. Spectral transport properties in (6) are determined by renormalization factors for the spin-flip rates due to magnetic impurities (spin-orbit coupling) $\alpha_{\text{TSTS}}^{\text{m(so)}} = [\text{Re} \cosh \theta]^2 + (-)[\text{Re} \sinh \theta]^2$, and the spin diffusion coefficient $D_L/D = \alpha_{\text{TSTS}}^{\text{so}}$. These functions are parametrized by $\theta(x, E)$ which we must determine from the Usadel equation for the retarded Green function $\hat{G}^{\text{R}} = \hat{\tau}_3 \cosh \theta + i\hat{\tau}_2 \sinh \theta$,

$$\frac{\hbar D}{2} \frac{\partial^2 \theta}{\partial x^2} = i\Delta \cosh(\theta) - iE \sinh(\theta) + \frac{3}{8} \frac{\hbar}{\tau_{\text{m}}} \sinh(2\theta), \quad (7)$$

to be solved with the BCS gap equation $\Delta = N_0 \lambda \int_0^{E_{\text{D}}} dE \tanh(E/2k_{\text{B}}T) \text{Re} \sinh(\theta)/2$ [6]. Here, E_{D} is the Debye cut-off energy and λ the interaction parameter.

Conservation of spin current at the interfaces represents boundary conditions for the diffusion equation (6), *e.g.* at $x = 0$, $\hbar AN_0 D_L \partial_x \mathbf{h}_{\text{TS}}/2 = \mathbf{i}_s^{\text{inj}} - \mathbf{i}_s^{\text{back}}$. For (7) we use boundary conditions derived in ref. [12] at the S|R interface. At the F|S interface we must take into account the inverse proximity effect. Effects from nontrivial interplay of superconductivity and magnetism can occur in structures with weak ferromagnets or opaque interfaces [13]. However, in the systems considered here we assume complete suppression of superconducting correlations close to the good metallic F|S contact ($\theta(x = 0, E) = 0$) for the following reasons. The large exchange energy in

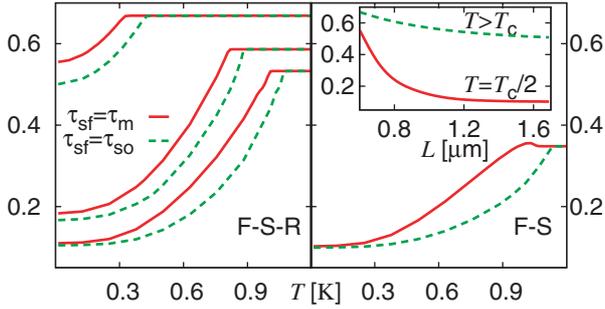


Fig. 2: (Colour on-line) Calculated $G - G_0$ [10^8 s^{-1}] (same ordinate scale in all plots). Red solid (green dashed) lines for system where $\tau_{sf} = \tau_m$ ($\tau_{sf} = \tau_{so}$). Left panel: F|S|R system with L [nm] from top to bottom: 600, 900, 1300. Right panel: F|S system (no R) with $L = 900$ nm. Inset: L dependence [μm] of $G - G_0$ for $T > T_c$ (green dashed line) and $T \ll T_c$ (red solid line).

transition metal ferromagnets completely suppress superconducting correlations. Additionally, spin-dependent interface scattering at the S side [14] induces an effective pair-breaking exchange field, which we estimate as $B_{\text{eff}} = \hbar g_{\uparrow}^{\perp} / e^2 g_L \mu_B N_0 A \xi_0$ [15]. Here, $N_0 A \xi_0$ is the number of states at the Fermi energy within ξ_0 from the interface. With $g_{\uparrow}^{\perp} \approx 0.05 g_{\text{Sh}}$, where g_{Sh} is the Sharvin conductance [11], and approximating N_0 by the free-electron value, $\mu_B B_{\text{eff}}$ is comparable to Δ_0 , *e.g.*, $\mu_B B_{\text{eff}}(\text{Nb}) \sim 0.56 \text{ meV}$, $\mu_B B_{\text{eff}}(\text{Al}) \sim 69 \mu\text{eV}$. The bulk F exchange splitting and the induced B_{eff} by spin-dependent interface scattering leads to a vanishing gap (and θ) at the F|S interface [16,17].

The spin diffusion equation (6) can be solved analytically in the absence of spin-flip, proving (2). We now use the full machinery sketched above to make contact with experimental results for a F|S device (without R) similar to sample C in ref. [4]. Numerically computing \mathbf{I}_s including spin-flip caused by magnetic impurities [18], we obtain the enhanced Gilbert damping G from (1). In the experiment, F is a permalloy layer with $d = 2 \text{ nm}$, and $g_L = 2.1$. S is Nb with $L = 70 \text{ nm}$, bulk critical temperature $T_{c0} = 8.91 \text{ K}$, $l_{\text{sf}} = 48 \text{ nm}$, and $D = 5.41 \text{ cm}^2 \text{ s}^{-1}$ [1,19]. For the interface resistances we use $Ar = 3 \text{ f}\Omega\text{m}^2$ [20]. We find $G - G_0 = 0.777 \times 10^8 \text{ s}^{-1}$ at $T_c/2 = 3.6 \text{ K}$ and $1.19 \times 10^8 \text{ s}^{-1}$ in the normal state. When the inhomogeneous linewidth broadening is small, the width of the FMR spectra are proportional to G and the experimental data gives $[G(T > T_c) - G(T = T_c/2)] / G(T > T_c) \approx 21\%$. Using $G_0 = 0.7 \times 10^8 \text{ s}^{-1}$ [5], we obtain 22%. The measured reduction of the Gilbert damping upon cooling the sample from above T_c to $T_c/2$ agrees *quantitatively* with our calculation.

We can make additional predictions for the Gilbert damping in F|S|R systems, focusing on Al as S since its spin-flip length is much longer than that of Nb, and as a weak-coupling superconductor is better described by BCS theory. The Al material parameters are $T_{c0} = 1.26 \text{ K}$, $l_{\text{sf}} = 1.1 \mu\text{m}$, and $D = 160 \text{ cm}^2 \text{ s}^{-1}$. In the left panel of fig. 2 we show the temperature dependence of $G - G_0$ for three

different thicknesses L when spin-flip is induced exclusively by either magnetic disorder or spin-orbit coupling to impurities. In contrast to spin-orbit scatterers, magnetic impurities reduce T_c due to the pair-breaking term in (7). For $L > l_{\text{sf}}$ and $T \ll T_c$, as well as for $T > T_c$, the results do not depend on the nature of the spin-flip scattering. In general, we observe that T_c strongly depends on L due to the inverse proximity effect. We also note that the difference in damping between the normal state and the superconducting state is small when $L \sim \xi_0$ since only a small gap develops.

The experiments of ref. [4] probed the regimes $L \ll \xi_0$ as well as $L \gg \xi_0$. We also present results for arbitrary L/ξ_0 . In the normal state, G decreases with increasing L due to increasing bulk spin transport resistance, which limits relaxation in R, until L reaches the value of l_{sf} where R becomes irrelevant (inset fig. 2). When $T \ll T_c$, on the other hand, the relevant length scale for spin penetration into S is ξ_0 . This explains the more rapid decay of $G - G_0$ as a function of L in the superconducting state. When $L > \xi_0$, the spin-current absorption is completely determined by the inverse proximity effect: Spin dissipation in R by transport through S is suppressed by the superconducting gap, and, furthermore, spin relaxation deep in S is suppressed by the superconductivity. However, some spin dissipation results from the inverse proximity effect which enhances the low-energy density of states and spin-flip scattering rates close to the F|S interface.

When $L < l_{\text{sf}}$, the results depend strongly on the S|R contact described by g . In the right panel of fig. 2, we show the temperature dependence of $G - G_0$ for $L = 900 \text{ nm}$ in an F|S system (no R or $g = 0$). At $T > T_c$, the damping is much smaller in the F|S system (the right panel) than in the F|S|R system with the same L (the middle pair of curves in the left panel). T_c is also higher since there is no inverse proximity effect at $x = L$. At very low temperatures, $T \ll T_c$, $G - G_0$ saturates at the same value for the F|S system as the F|S|R system with the larger thickness, $L = 1300 \text{ nm}$. For such thick S, T_c is unaffected by R and spins cannot diffuse through S and dissipate in R, so that the resulting damping is the same as in the F|S system. We also see from the right panel of fig. 2 that when $T \lesssim T_c$ the enhanced Gilbert damping can be somewhat larger than above T_c when spin-flip is induced by magnetic impurities, because the induced spin accumulation of quasiparticles with energy $k_B T \gtrsim \Delta$ experiences an enhanced spin-flip rate through α_{TSTS}^m . In the F|S|R system, this effect is overwhelmed by spin dissipation in R.

In conclusion, our theory quantitatively reproduces the measured FMR linewidth broadening in ferromagnet|superconductor structures. We make additional predictions for varying system sizes and temperatures, and the nature and strength of spin-flip scattering. We hope to stimulate more experiments studying the strong inverse proximity effect and energy-dependent spin-flip scattering in these systems.

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