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Controlling spin supercurrents via nonequilibrium spin injection

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We propose a mechanism whereby spin supercurrents can be manipulated in superconductor/ferromagnet proximity systems via nonequilibrium spin injection. We find that if a spin supercurrent exists in equilibrium, a nonequilibrium spin accumulation will exert a torque on the spins transported by this current. This interaction causes a new spin supercurrent contribution to manifest out of equilibrium, which is proportional to and polarized perpendicularly to both the injected spins and the equilibrium spin current. This is interesting for several reasons: as a fundamental physical effect; due to possible applications as a way to control spin supercurrents; and timeliness in light of recent experiments on spin injection in proximitized superconductors.

In the field of superconducting spintronics, a key objective is to study the interactions between superconductors (S) and ferromagnets (F)^{1–4}. These interactions produce new types of Cooper pairs $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ with a net spin polarization, enabling the use of S/F systems for dissipationless spin transport. From a fundamental physics perspective, such an interplay between different types of quantum order is expected to give rise to interesting physics to explore. Ultimately, the ambition is to exploit these phenomena in devices related to e.g. supercomputing and ultrasensitive detection of heat and radiation.

One particular area that has attracted attention is how one may generate controllable spin supercurrents in equilibrium, either via inhomogeneous magnetism^{5–10} or spin-orbit coupling^{11–14}. In the magnetic case, it has been shown that two layers with noncollinear magnetic moments \mathbf{m}_1 and \mathbf{m}_2 give rise to an equilibrium spin supercurrent $\mathbf{j}_s^{\text{eq}} \sim \mathbf{m}_1 \times \mathbf{m}_2$ ¹⁵. While most work so far relies on magnetic control of spin supercurrents via rotation of \mathbf{m}_1 relative to \mathbf{m}_2 , it would be interesting to determine if a spin supercurrent can be controlled via electronic spin injection instead. Such a mechanism might be more beneficial for coupling superconducting and nonsuperconducting spintronics devices. Note that this is different from many previous works on spin injection in superconductors, which were largely explained in terms of quasiparticles and not a spin-triplet condensate^{16–22}.

Recently, there has been a renewed interest in using nonequilibrium spin injection as a means to manipulate spin supercurrents. This is largely due to a recent spin-pumping experiment²³, where microwaves were used to excite spins in the ferromagnetic layer of an N/S/F/S/N junction. This experiment showed that the pumped spin current could increase below the critical temperature T_c of the S layers. It has been proposed that this effect was assisted by a Cooper-pair spin supercurrent¹³, although alternative explanations have been proposed²⁴.

In this manuscript, we consider how an injected nonequilibrium spin accumulation in general affects an existing spin supercurrent (see Fig. 1). We show that such a spin injection actually produces new terms in the equations for the spin supercurrent itself. These terms have a natural interpretation in the form of the injected spin accumulation ρ_s exerting a torque on an equilibrium spin supercurrent \mathbf{j}_s^{eq} , thus giving rise to a new component $\mathbf{J}_s^{\text{neq}} \sim \rho_s \times \mathbf{J}_s^{\text{eq}}$ perpendicular to both. Although this term occurs out-of-equilibrium, it shares the property of an equilibrium spin supercurrent that it does not require gradients in any chemical potential. Therefore, it is legitimate to refer to the new term $\mathbf{j}_s^{\text{neq}}$ as a supercurrent flowing without dissipation, as there is no energy loss associated with a spatially varying chemical potential. Our result is different from e.g. ref. 13, which proposed that *equilibrium* spin accumulation might produce spin supercurrents in some materials.

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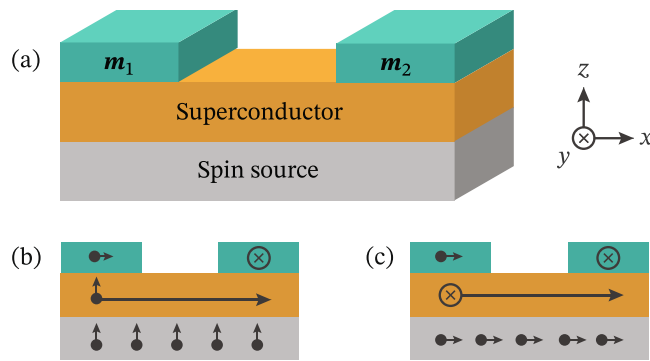


Figure 1. (a) Magnetic insulators with magnetizations \mathbf{m}_1 and \mathbf{m}_2 on a superconductor. In equilibrium, this yields a spin supercurrent $\mathbf{J}_s^{\text{eq}} \sim \mathbf{m}_1 \times \mathbf{m}_2$. A spin source injects a spin accumulation ρ_s , which exerts a torque on the spins transported by the equilibrium current, resulting in a new contribution $\mathbf{J}_s^{\text{neq}} \sim \rho_s \times (\mathbf{m}_1 \times \mathbf{m}_2)$. (b) If the magnets are magnetized in the x - and y -directions, an equilibrium spin- z supercurrent arises. Injection of spin- z particles does not affect its polarization. Note that a spin supercurrent is in general a rank-2 tensor, encoding both a polarization (short arrow) and transport direction (long arrow). (c) If spin- x particles are injected, however, a new spin- y supercurrent component is generated. Similarly, spin- y injection would produce a spin- x component. We model this setup as a 1D system, where the magnetic insulators connect to the superconductor at the sides; but in the diffusive limit, this should yield physically equivalent results to the setup depicted in this figure.

Results

Analytical results. Let us first consider a material with a spin-independent density of states $N(\epsilon)$, where ϵ is the quasiparticle energy. The nonequilibrium spin accumulation ρ_s can then be related to a spin distribution function \mathbf{h}_s ,

$$\rho_s = -\frac{\hbar}{2} \int_0^\infty d\epsilon N(\epsilon) \mathbf{h}_s(\epsilon), \tag{1}$$

where \mathbf{h}_s describes the imbalance between spin-up and spin-down occupation numbers. We define \mathbf{h}_s as a vector that points in the polarization direction of the spins, while its magnitude can be described in terms of a spin voltage V_s ,

$$h_s(\epsilon) = \{ \tanh[(\epsilon + eV_s)/2T] - \tanh[(\epsilon - eV_s)/2T] \} / 2, \tag{2}$$

where $e < 0$ is the electron charge and T is the temperature. The spin voltage is defined as $V_s := (V_\uparrow - V_\downarrow)/2$, where V_σ are the effective potentials seen by spin- σ quasiparticles^{25–27}, and \mathbf{h}_s defines the spin-quantization axis.

For a normal metal at $T = 0$, the density of states $N(\epsilon) = N_0$ is flat, while the spin distribution $|\mathbf{h}_s| = 1$ for $|\epsilon| < eV_s$. This results in a spin accumulation $|\rho_s| = (\hbar/2)N_0eV_s$ that increases linearly with V_s . This gives a simple interpretation of V_s as a control parameter: if the spin source in Fig. 1 is a nonsuperconducting reservoir, then the spin voltage V_s is directly proportional to the spin accumulation in the reservoir.

Similarly to the above, the excitation of quasiparticles from the Fermi level is described by an energy distribution $h_0(\epsilon)$,

$$h_0(\epsilon) = \{ \tanh[(\epsilon + eV_s)/2T] + \tanh[(\epsilon - eV_s)/2T] \} / 2. \tag{3}$$

At low temperatures, this shows that a spin voltage V_s also excites quasiparticles in a region of width $2eV_s$ around the Fermi level $\epsilon = 0$. For a more in-depth discussion of the nonequilibrium distribution function, see refs 20,25,26.

Spin supercurrents can in general be expressed as energy integrals of spectral spin supercurrents,

$$\mathbf{J}_s = -\frac{\hbar}{2} N_0 \int_0^\infty d\epsilon \text{Im}[\mathbf{j}_s]. \tag{4}$$

In equilibrium, the spectral current \mathbf{j}_s^{eq} is given by^{3,10}

$$\mathbf{j}_s^{\text{eq}} = (\mathbf{g}_t \times \nabla \mathbf{g}_t - \mathbf{f}_t \times \nabla \tilde{\mathbf{f}}_t) h_0. \tag{5}$$

Here, \mathbf{g}_t describes the spin-polarization of the density of states, while \mathbf{f}_t describes spin-triplet correlations³. The cross products should be taken between the orientations of the vectors \mathbf{g}_t and \mathbf{f}_t . Since the gradient of a vector is a rank-2 tensor, the spin supercurrent is such a tensor, enabling it to encode both the spin polarization and the transport direction. However, in effectively 1D systems like Fig. 1, we can let the position derivative $\nabla \rightarrow \partial_x$. In other words, in systems with 1D transport, the spin supercurrent reduces to a vector that describes spin polarization. Note that the result depends only on the energy distribution h_0 , which is the only part of the distribution function which remains finite in equilibrium.

Outside equilibrium, the spin distribution \mathbf{h}_s can become finite, and the spectral current gains an additional contribution:

$$\mathbf{j}_s^{\text{neq}} = (\mathbf{g}_t \times \nabla \mathbf{g}_t - \mathbf{f}_t \times \nabla \tilde{\mathbf{f}}_t) \times i\mathbf{h}_s. \quad (6)$$

The full derivation of this result is included in the Supplementary Information (Sec. II). The structure of Eq. (6) is very reminiscent of Eq. (5), since both depend on $\mathbf{g}_t \times \nabla \mathbf{g}_t - \mathbf{f}_t \times \nabla \tilde{\mathbf{f}}_t$. However, its cross product structure generates a spin current *perpendicular* to the one in Eq. (5). We also see that it contains an extra factor i ; since the distribution functions h_0 and \mathbf{h}_s are both real, this causes Eq. (4) to extract the real and not imaginary part of $\mathbf{g}_t \times \nabla \mathbf{g}_t - \mathbf{f}_t \times \nabla \tilde{\mathbf{f}}_t$. This comparison shows that the nonequilibrium contribution can be summarized as

$$\mathbf{j}_s^{\text{neq}} = \mathbf{j}_s^{\text{eq}} \times (i\mathbf{h}_s/h_0). \quad (7)$$

So long as $\mathbf{g}_t \times \nabla \mathbf{g}_t - \mathbf{f}_t \times \nabla \tilde{\mathbf{f}}_t$ is a complex number—which it in general is—it produces an equilibrium spin supercurrent \mathbf{j}_s^{eq} according to Eqs (4) and (5), which combined with a finite spin distribution \mathbf{h}_s immediately produces the new supercurrent term $\mathbf{j}_s^{\text{neq}}$ according to Eq. (7). This suggests an intuitive interpretation of the effect: injected spins described by \mathbf{h}_s exert a torque on the spins transported by the equilibrium component \mathbf{j}_s^{eq} , producing a nonequilibrium component $\mathbf{j}_s^{\text{neq}}$ perpendicular to both. It also suggests that the nonequilibrium spin supercurrent should increase linearly with the equilibrium spin supercurrent and the injected spin accumulation. Thus, an equilibrium spin supercurrent gains a new component when propagating through a region with spin accumulation ρ_s . All these predictions that arise from Eq. (7) are confirmed numerically later in this paper.

Let us now consider the setup in Fig. 1. In equilibrium, the x - and y -polarized magnets give rise to a z -polarized spin supercurrent $\mathbf{j}_s^{\text{eq}} \sim \mathbf{z}$. A generic spin source then introduces a spin imbalance in the superconductor, which we describe via a nonzero spin distribution \mathbf{h}_s . If these spins are polarized in the z -direction, meaning that $\mathbf{h}_s \parallel \mathbf{j}_s^{\text{eq}}$, then the nonequilibrium contribution $\mathbf{j}_s^{\text{neq}} = 0$. On the other hand, if these spins are polarized in the x -direction, so that $\mathbf{h}_s \perp \mathbf{j}_s^{\text{eq}}$, then the nonequilibrium contribution $\mathbf{j}_s^{\text{neq}} \sim \mathbf{j}_s^{\text{eq}} \times \mathbf{h}_s$ obtains a y -polarized component proportional to the spin imbalance. Similarly, if one had injected spin- y particles instead, a spin- x supercurrent would appear in the superconductor. In our calculations, we will for simplicity use an effective 1D model where the magnetic insulators are connected to the superconducting region at its sides rather than deposited on top of the superconductor. In practice, this has essentially no consequence since we are considering the diffusive limit of transport. The reason for this is that the spin supercurrent flow through the superconductor arises regardless of the exact spatial point at the edge of the superconductor (on top or at its side) where the magnetic insulators couple to the Cooper pairs, and in the diffusive limit the randomized motion of charge carriers further makes the precise coupling point irrelevant. Thus, our calculations should correspond well to the suggested setup in Fig. 1.

To summarize, for the geometry in Fig. 1, the analytical results suggest that we should expect a spin- y supercurrent proportional to the spin- x voltage, while the spin supercurrent should remain unchanged for a spin- z voltage. In the following sections, we compare these expectations to numerical results.

Numerical results. The spin supercurrent in the model considered here is conserved throughout the superconductor. In fact, we have checked both analytically and numerically that the spin supercurrent remains conserved in the presence of spin-flip and spin-orbit impurities, thus extending the equilibrium results from ref. 28 to this particular nonequilibrium situation. The analytical proof is straight-forward: the argument in ref. 28 shows that $\nabla \cdot \mathbf{j}_s^{\text{eq}} = 0$ as long as h_0 is position-independent. Since the new contribution proposed in this paper $\mathbf{j}_s^{\text{neq}} = \mathbf{j}_s^{\text{eq}} \times (i\mathbf{h}_s/h_0)$, we conclude that $\nabla \cdot \mathbf{j}_s^{\text{neq}} = 0$ if \mathbf{h}_s is position-independent. However, if either h_0 or \mathbf{h}_s becomes inhomogeneous, this argument breaks down, and the spin supercurrent is no longer conserved.

In Fig. 2, we show the spin supercurrent in the superconductor as a function of spin voltage at a low temperature $T = 0.01T_c$. Up until $eV_s \approx \Delta_0/2$, where Δ_0 is the bulk superconducting gap at zero temperature, these results are in perfect agreement with the analytical predictions. More precisely, we see that a spin- z injection (Fig. 2(a)) has no effect on the spin supercurrent, while a spin- x injection (Fig. 2(b)) leads to a spin- y supercurrent. The spin- y supercurrent increases linearly with the spin voltage, again in agreement with the predictions. Remarkably, the spin- z supercurrent does not decrease as the spin- y supercurrent increases, in contrast to what one might intuitively expect.

At low temperatures, we also see that there is a bistable regime at high spin voltages $eV_s > \Delta_0/2$. This means that both a superconducting and normal-state solution exist, which both correspond to local minima in the free energy. Depending on the dynamics of the system, this can either lead to hysteretic behaviour, or a first-order phase transition. This first-order phase transition was discussed already in the 1960s by Chandrasekhar and Clogston^{29,30}, while the possibility of hysteretic behaviour was suggested more recently in refs 31,32. Precisely where in the bistable region the thermodynamic transition point occurs is however difficult to predict within the Usadel formalism, as it is not straight-forward to explicitly evaluate the free energy²⁵.

Within the bistable regime, there is a point where the spin- z supercurrent reverses direction as a function of the spin voltage. This behaviour can be understood³³ as a spin equivalent of the S/N/S transistor effect^{34,35} where, according to Eq. (3), the energy distribution h_0 is also modulated by a spin voltage, and may therefore tune the equilibrium contribution in Eq. (5).

Since the spin- y supercurrent remains positive for all spin voltages, there exists a point where we get a pure spin- y supercurrent. In other words, there is a particular spin voltage that causes a 90° rotation of the spin supercurrent polarization compared to equilibrium. The fact that the spin- y supercurrent can remain finite while the

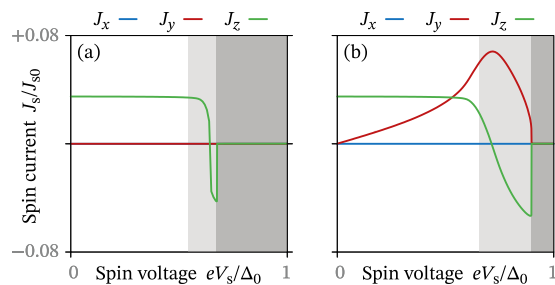


Figure 2. Spin supercurrent J_s as a function of spin voltage V_s . The spin voltage corresponds to injected (a) spin-z or (b) spin-x accumulation. The light shaded regions show where the system is bistable, and the dark ones where superconductivity vanishes.

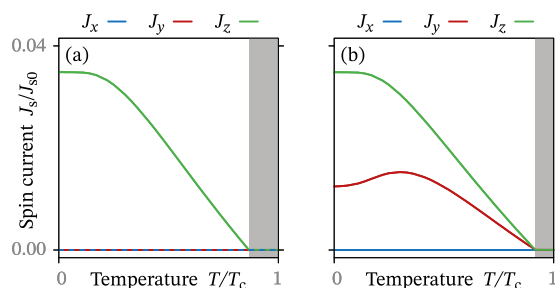


Figure 3. Spin supercurrent J_s as a function of temperature T for a fixed spin voltage $eV_s = \Delta_0/4$ in the superconductor. This spin voltage corresponds to injected (a) spin-z or (b) spin-x accumulation. Superconductivity vanishes in the shaded region.

spin-z supercurrent goes to zero might at first seem contradictory to our previous explanation $\mathbf{j}_s^{\text{neq}} \sim \mathbf{j}_s^{\text{eq}} \times (i\mathbf{h}_s/h_0)$. However, it is the *energy-integrated* spin currents $\mathbf{J}_s \sim \int d\epsilon \text{Im}[\mathbf{j}_s]$ that are plotted in Fig. 2. The spin-y current is generated from the *spectral* spin-z current, which remains finite even though the *total* spin-z current is zero.

In Fig. 3, we show how the spin supercurrent varies as a function of temperature for a fixed spin voltage $eV_s = \Delta_0/4$. Curiously, we find that the spin current increases *linearly* with decreasing temperature in a relatively large parameter regime. That the spin-y current decreases at the same rate as the spin-z current seems reasonable in light of the equation $\mathbf{j}_s^{\text{neq}} \sim \mathbf{j}_s^{\text{eq}} \times \mathbf{h}_s$: if \mathbf{j}_s^{eq} decreases linearly, then $\mathbf{j}_s^{\text{neq}}$ should do so as well. The most important message from Fig. 3 is perhaps that the nonequilibrium contribution $\mathbf{j}_s^{\text{neq}}$ to the spin supercurrent remains significant all the way up to the critical temperature of the junction. This means that relevant experiments can be performed at any temperature where superconductivity exists.

Discussion

In the previous sections, we have shown that injection of a nonequilibrium spin accumulation can be used to generate new spin supercurrent components. The results are especially encouraging since the nonequilibrium contribution to the spin supercurrent can even be made larger than the equilibrium contribution, and we found that it persists all the way up to the critical temperature of the junction. Both these features should make it a particularly interesting effect for experimental detection and future device design. However, there are some questions that we have not addressed yet.

The first question is how the spin source in Fig. 1 works. So far, we have simply treated it as a generic device that manipulates the spin distribution \mathbf{h}_s inside the superconductor directly. One alternative is to use a normal metal coupled to a voltage-biased ferromagnet¹⁸ or half-metallic ferromagnet³⁶. In that case, the polarization of the magnets enable a charge-spin conversion, thus translating an electric voltage into a spin voltage. Another possibility would be spin-pumping experiments, where it is a microwave signal that is translated to a spin voltage²³. In the limit of weak superconductivity, an expression for the distribution function of a spin-pumped ferromagnet was derived in ref. 37. When the precession frequency Ω and cone angle α are sufficiently small, the result is just a spin voltage $eV_s = \Omega/2$ along the magnetization direction of the ferromagnet. This is the relevant limit for the experiment in ref. 23: superconductivity inside $\text{Ni}_{80}\text{Fe}_{20}$ is weak, $\Omega \approx 0.5$ meV is much smaller than its magnetic exchange field, and $\alpha \approx 1^\circ$ should be small enough to use the leading-order expansions $\sin\alpha \approx \alpha$ and $\cos\alpha \approx 1$.

In all these cases, the spin source necessarily contains magnetic elements, and one challenge would be how to prevent the spin source from affecting the *equilibrium* spin current. One solution might be to embrace the existing magnets in Fig. 1: one could use the same magnets to generate the equilibrium spin supercurrent and for spin injection. This spin injection may then be performed either using spin pumping—or if the magnets are

sufficiently thin for electron tunneling—by placing voltage-biased contacts on top of the magnets. One complication with this strategy is that since the resulting spin accumulation will necessarily be inhomogeneous, both spin supercurrents and resistive spin currents have to coexist.

How to directly measure a spin supercurrent is an open question, although suggestions have recently been proposed³⁸. Indirect measurements of spin supercurrents, on the other hand, have already been performed experimentally. Most of these rely on measuring dissipationless charge currents through strongly polarized materials^{39–46}. Since only $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ pairs can penetrate over longer distances, and the polarization breaks the degeneracy between them, one can infer the existence of spin supercurrents from the measured charge supercurrents.

One solution to the measurement problem might be to look for an *inverse* effect. We have shown that spin injection into a superconductor results in a torque on the spins transported by the equilibrium spin supercurrent. However, this interaction should cause a reaction torque on the spin source, which might be possible to detect. For instance, in a setup similar to ref. 18, this reaction torque might directly affect the nonlocal spin conductance. Similarly, in a spin-pumping setup, this might affect the FMR linewidths. In both cases, this reaction torque should only exist when there is an equilibrium spin supercurrent $\mathbf{j}_s^{\text{eq}} \sim \mathbf{m}_1 \times \mathbf{m}_2$ to interact with, so it should depend on the magnetic configuration of the device.

We have shown analytically and numerically that if a system harbors a spin supercurrent \mathbf{j}_s^{eq} in equilibrium, then a spin injection \mathbf{h}_s creates a new component $\mathbf{j}_s^{\text{neq}} \sim \mathbf{j}_s^{\text{eq}} \times \mathbf{h}_s$. This effect can be intuitively understood as the injected spins exerting a torque on the spins transported by the equilibrium spin supercurrent, generating a component that is perpendicular to both. These results have implications for the control of spin supercurrents in novel superconducting spintronics devices.

Methods

In this section, we briefly describe how our numerical results were obtained. For more details about the numerical model, see the Supplementary Information (Sec. III). The numerical implementation is available at <https://github.com/jabirali/GENEUS>.

The numerical calculations were performed using the Usadel equation^{26,47–50}, which provides a good description of superconducting systems in the quasiclassical and diffusive limits. Within this formalism, physical observables are described via 8×8 quasiclassical propagators in Keldysh \otimes Nambu \otimes spin space,

$$\check{g} = \begin{pmatrix} \hat{g}^R & \hat{g}^K \\ 0 & \hat{g}^A \end{pmatrix}. \quad (8)$$

These matrix components are related by the identities $\hat{g}^K = \hat{g}^R \hat{h} - \hat{h} \hat{g}^A$ and $\hat{g}^A = \hat{\tau}_3 \hat{g}^R \hat{\tau}_3$. Here \hat{h} is a 4×4 distribution function, which in systems with spin accumulation can be written

$$\hat{h} = h_0 \hat{\sigma}_0 \hat{\tau}_0 + \mathbf{h}_s \cdot \hat{\boldsymbol{\sigma}} \hat{\tau}_3, \quad (9)$$

where h_0 and \mathbf{h}_s were introduced earlier. The $\hat{\tau}_n$ are $\hat{\sigma}_n$ are Pauli matrices in Nambu and spin space, respectively. As for the retarded component \hat{g}^R , we analytically use the parametrization³

$$\hat{g}^R = \begin{pmatrix} (\mathbf{g}_s + \mathbf{g}_t \cdot \boldsymbol{\sigma}) & (\mathbf{f}_s + \mathbf{f}_t \cdot \boldsymbol{\sigma}) i \sigma_2 \\ -i \sigma_2 (\tilde{\mathbf{f}}_s - \tilde{\mathbf{f}}_t \cdot \boldsymbol{\sigma}) & -\sigma_2 (\tilde{\mathbf{g}}_s - \tilde{\mathbf{g}}_t \cdot \boldsymbol{\sigma}) \sigma_2 \end{pmatrix}, \quad (10)$$

while we numerically use the Riccati parametrization⁵¹. General equations for calculating spin supercurrents and spin accumulations from these quasiclassical propagators are derived and presented in the Supplementary Information (Sec. I).

To determine the propagators above for the setup in Fig. 1, we have to simultaneously solve the Usadel equation,

$$i \xi^2 \nabla (\check{g} \nabla \check{g}) = [\hat{\Delta} + \epsilon \hat{\tau}_3, \check{g}] / \Delta_0, \quad (11)$$

and a selfconsistency equation for the gap Δ which depends on \hat{g}^K ⁵². The other quantities are the dirty-limit coherence length ξ and bulk gap Δ_0 . The magnetic insulators in Fig. 1 are modelled as spin-active interfaces^{53–56}.

We assume a fixed distribution function \hat{h} , and do not solve any kinetic equation^{20,21,25,26,47,48,57}. This approximation is valid when the superconducting layer is thin compared to its spin relaxation length. Thus, there is no resistive spin current flowing in the superconductor, as $\nabla \mathbf{h}_s = 0$ ensures that there is no gradient in the spin accumulation.

Finally, we briefly summarize our parameter choices. The superconductor was taken to have a length $L = 1.5 \xi$. The magnetic insulators were described with $G_\varphi / G_N = 0.6$, where G_N is the bulk normal-state conductance of the superconductor, and G_φ describes the spin-dependent phase-shifts obtained by quasiparticles reflected at a magnetic interface^{53–56}. Finally, we assumed a constant spin voltage V_s throughout the entire superconductor, instead of explicitly modelling the details of the spin source in Fig. 1. Thus, the junction is treated as a 1D superconductor with magnetic boundary conditions. Our results are not qualitatively sensitive to these parameter choices. The main constraints are that superconductivity collapses if L/ξ is too low and G_φ / G_N too high, while spin supercurrents become vanishingly small in the opposite limits.

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Author Contributions

J.A.O. performed all the analytical and numerical calculations, and wrote the initial draft for the manuscript. All authors contributed to the ideas behind the project, and to discussions and revisions of the manuscript.

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