Cooper-Pair Spin Current in a Strontium Ruthenate Heterostructure

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It has been recognized that the condensation of spin-triplet Cooper pairs requires not only broken gauge symmetry but also spin ordering as well. One consequence of this is the possibility of a Cooper-pair spin current analogous to the magnon spin current in magnetic insulators, the analogy also extending to the existence of the Gilbert damping of the collective spin-triplet dynamics. The recently fabricated heterostructure of the thin film of the itinerant ferromagnet $SrRuO_3$ on bulk Sr_2RuO_4, the best-known candidate material for a spin-triplet superconductor, offers a promising platform for generating such spin current. We show how such heterostructure allows us to not only realize the long-range spin valve but also electrically drive the collective spin mode of the spin-triplet order parameter. Our proposal represents both a novel experimental realization of superfluid spin transport and a transport signature of the spin-triplet superconductivity therein.

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Introduction.—Harnessing spin rather than charge in electronic devices has been a major topic in solid-state physics; it has not only been utilized for various memory devices but is also expected to play a key role in processing quantum information [1]. In order for various spin devices to function robustly, long-range spin transport needs to be achieved. Metallic wires, however, typically do not transport spins beyond the spin-diffusion length due to single electron spin relaxation [2].

In recent years, it has been shown that exponential damping can be circumvented in the spin transport via collective magnetic excitations. For example, easy-plane (ferromagnetic and antiferromagnetic) magnetic insulators are analogous to the conventional superfluid in being characterized by the U(1) order parameter [3–5]. As Fig. 1(a) illustrates, planar spiraling of the order parameter in such magnetic insulators gives rise to a spin supercurrent, just as the phase gradient of a conventional superfluid gives rise to a mass supercurrent; in this sense these magnetic insulators can be regarded as spin superfluids [6].

Interestingly, there exists a class of superfluids and superconductors which can support both mass and spin supercurrent. Such superfluids and superconductors should break both spin-rotational symmetry and gauge symmetry. Examples include the spin-1 boson condensate [7], the ^{3}He superfluid [8,9], and the spin-triplet superconductor [10,11]; in the two latter cases, the dissipationless spin current would be carried by Cooper pairs. While the vortices with spin supercurrent circulation have been observed in all theses systems [12,13], the bulk spin supercurrent has not been detected in the superconductor.

In this Letter, we will show how spin superfluidity in a spin-triplet superconductor leads not only to long-range spin current but also electrical excitation of a spin wave in the bulk. For realizing these phenomena, we propose a two-terminal setup with voltage bias between ferromagnetic metal leads in contact with a spin-triplet superconductor. While the static order-parameter case [14] essentially reduces to the Blonder-Tinkham-Klapwijk-type formalism [15] for interfacial transport, here we complement it with the appropriate equations of motion for collective spin dynamics in a superconductor. Recently, a thin film of the itinerant ferromagnet $SrRuO_3$ has been epitaxially deposited on bulk Sr_2RuO_4, the best-known candidate material for a spin-triplet superconductor [16], yielding, due to their structural compatibility, an atomically smooth and highly conductive interface [17] with a strong Andreev

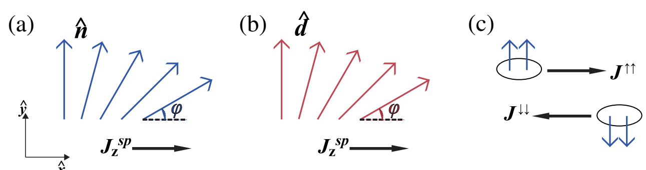

FIG. 1. Schematic illustration of the analogy between a magnetic insulator and a spin-triplet superconductor. (a) The planar spiraling of the magnetic order parameter $\hat{\mathbf{n}}$ leads to spin current. (b) The same phenomena occurs for that of a spin component $\hat{\mathbf{d}}$ of a spin-triplet superconductor order parameter, (c) the dual picture of which is the counterflow of the spin up-up and down-down Cooper pairs.

conductance [18]. This makes Sr_2RuO_4 and $SrRuO_3$ the primary candidate materials for the bulk and the leads, respectively, of our setup [19]. For the remainder of this Letter, we will first show how the simplest effective spin Hamiltonian for a spin-triplet superconductor and the resulting spin dynamics are analogous to those of an antiferromagnetic insulator; then, we will discuss the magnetoresistance for the dc bias voltage and the coupling between the ac bias voltage and the spin wave.

General considerations.—We first point out the close analogy between a spin order parameter of an antiferromagnet and a spin-triplet superconductor. Defined as [20]

$$i(\mathbf{d}\cdot\boldsymbol{\sigma})\sigma_y = \begin{bmatrix} -d_x + id_y & d_z \\ d_z & d_x + id_y \end{bmatrix} \equiv \begin{bmatrix} \Delta_{\uparrow\uparrow} & \Delta_{\uparrow\downarrow} \\ \Delta_{\downarrow\uparrow} & \Delta_{\downarrow\downarrow} \end{bmatrix}, \quad (1)$$

the $\mathbf{d}$ vector of a spin-triplet pairing, whose direction $\hat{\mathbf{d}}$ parametrizes the Cooper-pair spin state, behaves similarly under spin rotations to the Néel order parameter of an antiferromagnet, i.e., $[S_i(\mathbf{r}), d_j(\mathbf{r}')] = i\hbar\epsilon_{ijk}\delta(\mathbf{r}-\mathbf{r}')d_k(\mathbf{r})$ and $[d_i, d_j] = 0$ for the condensate spin $\mathbf{S}$ (unlike the magnetization, neither the Néel order parameter nor the $\mathbf{d}$ vector generate spin rotation in themselves) [8,9,11]. Given that, in both cases, $\mathbf{S}\times\hat{\mathbf{d}}$ is the conjugate momentum to $\mathbf{d}$ by the commutation relations, it is natural that the simplest effective Hamiltonian for a spin-triplet superconductor $\hat{\mathbf{d}}$ vector,

$$H = \frac{1}{2}\int d\mathbf{r}[A(\nabla\hat{\mathbf{d}})^2 + K\hat{d}_z^2 + \gamma_e^2\mathbf{S}^2/\chi], \quad (2)$$

where γ_e is the electron gyromagnetic ratio, A the $\hat{\mathbf{d}}$-vector stiffness, and χ the magnetic susceptibility, should be equivalent to that of the antiferromagnet Néel order parameter, once we identify the $\hat{\mathbf{d}}$ vector with the Néel order parameter [4]. This Hamiltonian, which can be constructed from the phenomenological approach of Ref. [21], applies to electron pairing respecting spin-rotational symmetry. Assuming a rigid $\mathbf{k}$-space configuration of $\hat{\mathbf{d}}$ at low energy, the low-energy manifold of the theory is parametrized by $\hat{\mathbf{d}}(\mathbf{r})$, associated with smooth spatial variations of the triplet order. An easy-plane anisotropy K for planar spin dynamics can be induced perpendicular to an applied magnetic field, analogously to the spin-flop transition in antiferromagnets [22]. In the case of an antiferromagnet, the (xy) planar texture of the orientational order parameter $\hat{\mathbf{n}} \to (\cos\phi, \sin\phi, 0)$ is associated with a collective (z-polarized) spin current $J_z \propto \mathbf{z}\cdot(\hat{\mathbf{n}}\times\partial_i\hat{\mathbf{n}}) \to \partial_i\phi$ flowing in the ith direction. While this extends directly to our spin-triplet case, Eq. (1) gives the intuitive dual picture of Fig. 1(c) for planar spiraling of the $\mathbf{d}$ vector, i.e., $\hat{\mathbf{d}} = (\cos\alpha, \sin\alpha, 0)$. Namely, as the phase of $\Delta_{\uparrow\uparrow}$ ($\Delta_{\downarrow\downarrow}$) is given by $\phi_c \mp \alpha$ (where ϕ_c is the overall phase of the superconductor), spiraling of the $\mathbf{d}$ vector on the xy plane as shown in Fig. 1(b), or the gradient of α, would imply counterflow of the spin up-up and down-down pairs. The resultant (z-polarized) spin current is $\propto -\nabla\alpha$ [23]. Given the same commutation relation and the same effective Hamiltonian, it is natural that, in the absence of dissipation, the equations of motion for these two cases, the Leggett equations, the $\hat{\mathbf{d}}$ vector [8,9,24], and the Landau-Lifshitz-type equation for the Néel order parameter are identical.

We further argue that both cases have the same phenomenological form of dissipation as well. For the case of the Néel order parameter $\hat{\mathbf{n}}$, such energy dissipation, at the rate $\propto \alpha(\partial_t\hat{\mathbf{n}})^2$ for low frequencies, known generally as Gilbert damping for collective magnetic dynamics, has been understood phenomenologically [4,25–27]. That such dissipation has not been featured in the ^{3}He superfluid literature is due not to the intrinsic nature of the spin-triplet pairing but rather to the ^{3}He spin-orbit coupling originating from the very weak nuclear dipole-dipole interaction [8]. In contrast, electrons in Sr_2RuO_4 are subject to Ru atomic spin-orbit coupling [28] estimated to be ~0.1 eV [29]. In this work, we will consider the decay rate of $\alpha n\hbar\gamma_e^2/\chi$ for the condensate spin, the addition of which makes the Leggett equations of motion for spin [30] equivalent to Landau-Lifshitz-Gilbert-type equations for antiferromagnets:

$$\partial_t\hat{\mathbf{d}} = -\hat{\mathbf{d}}\times\frac{\gamma_e^2}{\chi}\mathbf{S},$$
$$\partial_t\mathbf{S} = \hat{\mathbf{d}}\times(A\nabla^2\hat{\mathbf{d}} - K\hat{d}_z\hat{\mathbf{z}} - \alpha n\hbar\partial_t\hat{\mathbf{d}}), \quad (3)$$

where α is the dimensionless Gilbert damping parameter and n the Cooper-pair density. Through this set of equations, we can obtain the local $\hat{\mathbf{d}}$-vector dynamics, e.g., the spin-wave excitation and the collective dissipation, starting from the effective Hamiltonian of Eq. (2).

For the boundary conditions, at the interface between a ferromagnetic lead and a spin-triplet superconductor, we consider a two-channel interface conductance due to the spins aligned or antialigned to the lead magnetization. We note that the $SrRuO_3$ thin film has a 50% transport spin polarization [34–36] with the magnetization enhanced in the heterostructure [17], promising a much higher spin injection and detection efficiency compared to graphene-based devices used in a recent long-range spin transport experiment [37]. In this Letter, we shall consider only the simple case of the collinear lead magnetizations. Furthermore, the $\mathbf{d}$ vector of a bulk spin-triplet superconductor will be taken to be perpendicular to the lead magnetization; i.e., the Cooper pairs are equal-spin paired along the magnetization direction. For the Sr_2RuO_4 superconductor, the c-axis magnetic field of 200 G reportedly suffices for the $\mathbf{d}$ vector to flop into the ab plane [38]. This interpretation is based on the model of time-reversal symmetry broken p-wave superconductivity [11], which we will follow for the details of our experimental

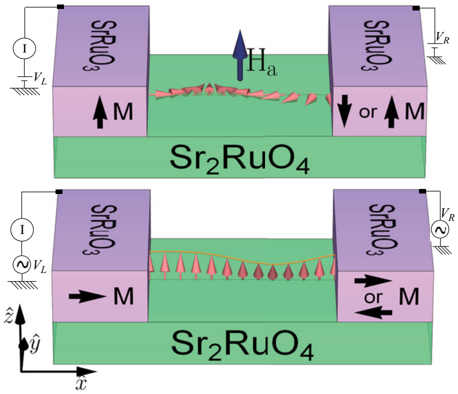

FIG. 2. The setup for the dc voltage bias for the spin valve (upper) and the ac bias voltage for the spin-wave detection (lower), where $\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}}$ coincide with the crystalline a, b, c axes, respectively. For the upper illustration, the lead magnetization is along the c axis, with the applied magnetic field $H_a \geq 200$ G along the c axis giving us the easy-plane $\mathbf{d}$-vector configuration on the ab plane, hence the spiraling in the ab plane. For the lower illustration, the lead magnetization is along the a axis; as the easy-axis $\mathbf{d}$-vector anisotropy favors alignment along the c axis, in the absence of an applied field, the ac bias voltage gives us the low-frequency standing wave of the $\mathbf{d}$ vector oscillating around the c axis in the bc plane.

proposals; however, the phenomena we predict can arise in any spin-triplet superconductor close to the SO(3) Cooper-pair spin-rotational symmetry.

Long-range spin valve.—The simplest physics that can arise in our two-terminal setup is spin-valve magnetoresistance due to the lead magnetization alignment shown in Fig. 2. We consider the case where the spin-triplet superconductor has easy-plane anisotropy, that is, $K > 0$ in Eq. (2) (for which a ≥ 200 G field is applied along the c axis), with the lead magnetization perpendicular to this plane as in the Fig. 2 upper panel. In this case, we can take $\hat{d}_z$ to be a small parameter in $\hat{\mathbf{d}} = \left(\sqrt{1-\hat{d}_z^2}\cos\phi_z, \sqrt{1-\hat{d}_z^2}\sin\phi_z, \hat{d}_z\right)$ and $|S_{x,y}| \ll |S_z|$. In such a case, $[\phi_z(\mathbf{r}), S_z(\mathbf{r}')] = i\hbar\delta(\mathbf{r}-\mathbf{r}')$ gives us the conjugate pair, leading to

$$\partial_t\phi_z = \frac{\gamma_e^2}{\chi}S_z, \qquad \partial_t S_z = A\nabla^2\phi_z - \alpha n\hbar\partial_t\phi_z, \tag{4}$$

where the first equation is a spin analogue of the Josephson relation and the second is the spin continuity equation with a relaxation term. One confirms the condensate spin imbalance relaxation time to be $\chi/\alpha n\hbar\gamma_e^2$ from Eq. (4) through deriving $\partial_t S_z + \mathbf{\nabla}\cdot\mathbf{J}_z^{sp} = -\alpha n\hbar\gamma_e^2 S_z/\chi$, where $\mathbf{J}_z^{sp} = -A\mathbf{\nabla}\phi_z$. The parameters K, A, and α of Eq. (4) are effectively renormalized by the Abrisokov vortex lattice in the spin-triplet superconductor due to averaging over the macroscopic length scale.

We consider the spin-up current and the spin-down current to be independent at the interface:

$$I_{L,R}^{\sigma} = \pm g_{L,R}^{\sigma\sigma}(V_{L,R} - \hbar\partial_t\varphi_\sigma/2e), \tag{5}$$

where $g_{L,R}^{\sigma\sigma}$'s are the conductances for the σ spin, $I_{L,R}$ the σ-spin current into (out of) the left (right) lead, and $V_{L,R}$ the bias voltage of the left (right) lead; this is due to the spin-triplet superconductor having an equal-spin pairing axis collinear with the lead magnetization and, hence, $g^{\uparrow\downarrow} = 0$. In the paragraph of Eqs. (1) and (2), we have shown that the overall (or charge) phase of a superconductor is given by the average of the spin up-up and the spin down-down condensate phase, $\phi_c = \sum_\sigma \varphi_\sigma/2$, while ϕ_z of Eq. (4) is given by $\phi_z = \sum_\sigma \sigma\varphi_\sigma/2$. We are interested here in the steady-state solution, i.e., $\partial_t\varphi_\sigma = \text{const}$, for which we define the constant precession rate of $\omega_c \equiv \sum_\sigma \partial_t\varphi_\sigma/2$ for the overall phase ϕ_c and $\Omega_s \equiv \sum_\sigma \sigma\partial_t\varphi_\sigma/2$ for ϕ_z. For such a solution, the following continuity conditions can be applied to the charge and spin supercurrents, respectively:

$$\sum_\sigma (I_L^\sigma - I_R^\sigma) = 0, \qquad \sum_\sigma \sigma(I_L^\sigma - I_R^\sigma) = 2\alpha ne\Omega_s SL \tag{6}$$

(S is the bulk cross section area and L the spacing between the two leads [39]), the former from the charge conservation and the latter from applying the steady-state condition on Eq. (4), along with the spin current loss $\propto \alpha L$ in the superconductor.

The current through Sr_2RuO_4 bulk can be obtained from the interface boundary conditions and the continuity conditions above, with the larger magnitude for parallel magnetization than for antiparallel magnetization. We define the total conductance $g_{L,R} \equiv \sum_\sigma g_{L,R}^{\sigma\sigma}$ and the conductance polarization $p_{L,R} \equiv \sum_\sigma \sigma g_{L,R}^{\sigma\sigma}/g_{L,R}$, which defines the relevant transport spin polarization. Applying the continuity conditions Eq. (6) on the interface boundary conditions Eq. (5) and setting $V_L = -V_R = V/2$, we obtain

$$\begin{pmatrix} g_L + g_R & p_L g_L + p_R g_R \\ p_L g_L + p_R g_R & g_L + g_R + g_\alpha \end{pmatrix}\begin{pmatrix}\omega_c \\ \Omega_s\end{pmatrix} = \frac{eV}{\hbar}\begin{pmatrix} g_L - g_R \\ p_L g_L - p_R g_R\end{pmatrix}, \tag{7}$$

where $g_\alpha \equiv (4\alpha ne^2 SL/\hbar)$. We can now obtain the dependence of charge current on conductance polarization:

$$I^c = \sum_\sigma I^\sigma = I_0\left(1 - \frac{g_L g_R (p_L - p_R)^2}{(g_L + g_R)(g_L + g_R + g_\alpha) - (p_L g_L + p_R g_R)^2}\right), \tag{8}$$

where $I_0 \equiv g_L g_R V/(g_L + g_R)$. Note that I^c is maximized at $p_L = p_R$, when the steady-state angle ϕ_z remains static. Different spin polarizations at the two ends, on the other hand, would trigger spin dynamics and result in a nonzero dissipation rate of $R = \frac{1}{2}\alpha n\hbar\Omega_s^2 = R_0(1 - I^c/I_0)^2/(p_L - p_R)^2$ per volume of the superconducting bulk, where $R_0 = 8\alpha n(eV)^2/\hbar$. Given that $p_{L,R}$ change sign on magnetization reversal, the above results effectively give us the spin-valve magnetoresistance of our heterostructure, i.e., a larger conductance for parallel magnetizations than for antiparallel magnetizations. Any effect that spin-triplet pairing may have on magnetization, hence conductance polarization [40], can be ignored when the Curie temperature of $SrRuO_3$ (~160 K) [41] is 2 orders of magnitude higher than the superconducting critical temperature (~1.5 K) Sr_2RuO_4.

We emphasize that the above magnetoresistance result is obtained solely for the current carried by Cooper pairs. At a finite temperature, the quasiparticle contribution would generally result in an exponentially decaying magnetoresistance, negligible for the lead spacing beyond the spin-diffusion length. By contrast, the current of Eq. (8), which is carried by Cooper pairs, gives us the $\sim 1/L$ magnetoresistance for the large spacing limit. Therefore, any magnetoresistance beyond the quasiparticle spin-diffusion length should arise only below the superconducting transition at T_c, upon the emergence of a Cooper-pair condensate. For our $Sr_2RuO_4/SrRuO_3$ heterostructure, detection of magnetoresistance in the superconducting state for the lead spacing larger than the Sr_2RuO_4 spin-diffusion length can be taken as a transport evidence for spin-triplet superconductivity. The value of the spin-diffusion length itself can be extracted by measuring the exponential decay of the (normal) magnetoresistance, both above and below the transition.

Electrically driven spin collective mode.—For the case of easy-axis anisotropy of the **d** vector, hence, $K < 0$ in Eq. (2), the spin collective excitation of the Cooper pairs [8,9,42,43] will modify the supercurrent transport under the ac bias voltage. We shall still continue to consider the case where Eq. (5) would be valid, i.e., the equal-spin pairing axis of a spin-triplet superconductor collinear to the lead magnetizations. One way to satisfy this condition would be to have the lead magnetizations collinear to the a axis, with no applied magnetic field as in the Fig. 2 lower panel; that would leave the a axis as the equal-spin pairing axis, with the **d** vector moving on the bc plane. The equations of motion, corresponding to spin injection polarized along the x direction, are then modified to

$$\partial_t \phi_x = \frac{\gamma_e^2}{\chi} S_x,$$
$$\partial_t S_x = A\nabla^2\phi_x - \omega_0^2 \frac{\chi}{\gamma_e^2}\cos\phi_x \sin\phi_x - \alpha n\hbar\partial_t\phi_x, \quad (9)$$

where ϕ_x is conjugate to S_x and $\omega_0^2 \equiv |K|\gamma_e^2/\chi$ is the spin-wave energy gap. For the ac voltage bias $V = V_0 \exp(-i\omega t)$

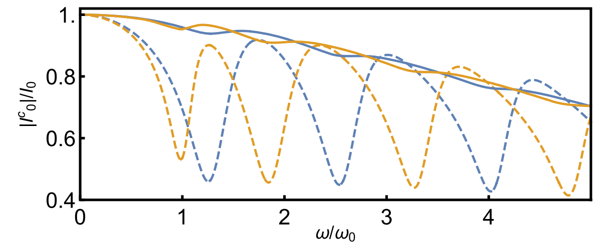

FIG. 3. Charge current versus frequency plotted for $\tilde{g} = 0.5$, $\tilde{L} = 2$, $\Gamma/\omega_0 = 0.1$, and $\tilde{A} = 0.2$, with the orange curve representing $p_L = p_R = p$ and the blue curve $p_L = -p_R = p$. Note that $p = 0.2, 0.8$ for the solid and dashed lines, respectively.

at frequencies far below the plasma frequency, the steady-state solution for the spin phase $\phi_x(x,t) = f(x)\exp(-i\omega t)$ and the charge phase $\phi_c(x,t) = g(x)\exp(-i\omega t)$ behave differently. Hence, the spin equations of motion Eq. (9) give us $f(x) = C_+ \cosh\kappa x + C_- \sinh\kappa x$, where $v^2\kappa^2 = \omega^2 - \omega_0^2 - i\omega\Gamma$, with $v \equiv \gamma_e\sqrt{A/\chi}$ [the $\hat{\mathbf{d}}$-vector stiffness A defined in Eq. (2)] being the spin-wave velocity and $\Gamma \equiv \alpha n\hbar\gamma_e^2/\chi$ the damping rate. By contrast, the charge current $J^c(x,t) = -\rho\partial_x\phi_c$, where ρ is the ϕ_c stiffness, should be uniform, which means we can set $\phi_c(x,t) = \text{const} - x(J_0^c/\rho)\exp(-i\omega t)$, with a constant J_0^c. By imposing consistency between the current obtained from the boundary conditions of Eq. (5) and the dynamics of Eq. (9), we can solve for J_0^c and $C_\pm$; Fig. 3 shows the numerical results for $I_0^c \equiv J_0^c S$ for the case of both $p_L = p_R$ and $p_L = -p_R$.

Our numerical results show that magnetoresistance becomes significant at $\omega \gtrsim \omega_0$, where the collective spin mode of the Cooper pairs is activated. For simplicity we have set $g_L = g_R = g$ and used the dimensionless parameters $\tilde{g} \equiv g\hbar v/2eA$, $\tilde{L} \equiv \omega_0 L/2v$, and $\tilde{A} = A/\rho$. For $\omega < \omega_0$, in addition to barely noticeable magnetoresistance, the charge current amplitude does not oscillate with frequency; it remains close to the dc value I_0, unlike the complete transport suppression in the magnetic insulator [3]. In contrast, for $\omega > \omega_0$, we see an oscillation with the ω/ω_0 period of about $\pi/\tilde{L}$, where the current amplitude maxima for the antiparallel lead magnetization occur at the current amplitude minima for the parallel lead magnetization and vice versa. As in the ferromagnetic insulator [3], we expect that for $\tilde{L} \ll 1$, i.e., much shorter than the d-vector relaxation length [44], the magnetoresistance of Eq. (8) is recovered for the static bias, i.e., $\omega \to 0$.

We point out that the detection of the oscillation shown in Fig. 3 would determine the yet-unknown energy parameters for spin-triplet pairing of Sr_2RuO_4. From the effective Hamiltonian of Eq. (2), if we had known accurately the field H_c along the c axis that would exactly restore the **d**-vector isotropy, the gap frequency ω_0 should be just the electron Larmor frequency of this field from the spin equations of motion of Eq. (9). However, we know no more than the upper bound $H_c < 200$ G, hence only $\omega_0 < \gamma_e \times 200$ G $= 3.5$ GHz, while the ac bias experiment, as shown in Fig. 3, would allow us to definitely identify the spin collective mode gap.

Conclusion and discussion.—We have studied the dc and ac current transport between itinerant ferromagnetic leads with collinear magnetizations through a spin-triplet superconductor. We showed here that magnetoresistance can arise for both cases due to Cooper-pair spin transport. For the dc bias, the persistence of magnetoresistance for the lead spacing larger than the quasiparticle spin-diffusion length can be taken as a transport evidence for spin-triplet pairing. For the ac bias, the activation of magnetoresistance and frequency-dependent oscillation above the threshold frequency will allow us to determine the spin anisotropy energy scale. All together, our work shows both a novel experimental realization of superfluid spin transport and a transport signature of spin-triplet superconductivity. The recently fabricated $SrRuO_3/Sr_2RuO_4$ heterostructure provides a promising experimental setup.

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