Nonlocal spin correlations mediated by a superconductor

Taewan Noh,[1] Manuel Houzet,[2] Julia S. Meyer,[2] and Venkat Chandrasekhar[1]

[1]*Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208, USA*

[2]*SPSMS, UMR-E 9001 CEA/UJF-Grenoble 1, INAC, Grenoble F-38054, France*

Nonlocal charge correlations induced in two normal metals contacted separately to a superconductor have been studied intensively in the past few years. Here we investigate nonlocal correlations induced by the transfer of pure spin currents through a superconductor on a scale comparable to the superconducting coherence length. As with charge currents, two processes contribute to the nonlocal spin signal: crossed Andreev reflection (CAR), where an electron with spin-up injected from one normal metal into the superconductor results in a hole with spin-down being injected into the second normal metal, and elastic cotunneling (EC), where the electron with spin-up injected from the first normal metal results in an electron with spin-up being injected into the second normal metal. Unlike charge currents, however, the spin currents associated with CAR and EC add due to the fact that the bulk superconductor cannot sustain a net spin current.

Electrons in two spatially separated normal metals in contact with a superconductor show nonlocal correlations that are mediated by their mutual interaction with the superconductor.[1–4] Two processes are responsible for these correlations. In the first process, called crossed Andreev reflection (CAR), an electron with one spin orientation, e.g., spin-up, is injected from the first normal metal (N) into the superconductor (S), resulting in a hole with spin-down being injected from the superconductor into the second normal metal, with a concomitant generation of a Cooper pair in the superconductor. In the second process, elastic cotunneling (EC), the spin-up electron injected from the first normal metal gives rise to a spin-up electron being injected into the second normal metal. Both processes are exponentially suppressed with the distance between the two NS interfaces on a length scale of the order of the superconducting coherence length ξ_S (Ref. 5). In the case of *charge* currents, the net current injected into the second normal metal in response to the drive current injected from the first normal metal is the *difference* of the two contributions CAR and EC. As the relative amplitude of CAR and EC is predicted to depend on the transparency of the interface,[6] the effect of electron-electron interactions,[7] and so forth, the sign of the net current injected into the second normal metal may be positive or negative. Experimentally, nonlocal correlations due to CAR and EC in charge transport have been verified by many different groups.[8–10]

In this Rapid Communication, we study theoretically nonlocal correlations induced in a *NSN* structure in response to a pure spin current. As with charge currents, both CAR and EC contribute to the resulting nonlocal spin signal. However, due to the fact that one cannot inject a pure spin current into the bulk of a *s*-wave superconductor (which is the case of interest here), we find that the resulting nonlocal spin current is the *sum* of CAR and EC contributions: Injecting a spin-down hole (CAR) or injecting a spin-up electron (EC) into the second normal metal, both correspond to injecting a net up-spin. Consequently, with a combination of charge and spin transport measurements on the same device, one should, in principle, be able to separate the contributions of CAR and EC, which has not been possible to do so far.

In addition to CAR and EC, there are additional processes that may contribute to the nonlocal signals in *NSN* or *FSF* structures. These are charge[11–15] and spin[16,17] imbalance, associated with the injection of quasiparticles, with energies larger than the superconducting gap Δ, into the superconductor. In contrast to these studies, the proposed experiment addresses the regime of subgap transport. Thus, no quasiparticle is injected above the gap, and the effect does not depend on the long spin relaxation times recently observed in the nonlocal measurements of Refs. 16 and 17 at applied bias voltage larger than Δ/e.

Figure 1 shows the schematic view of the device geometry that we consider here. Our device is similar to the usual *NSN* hybrid structure, except that there are two additional ferromagnets, F_1 and F_2, placed in contact with the two normal metals, N_1 and N_2, respectively. One of the normal-metal/ferromagnet junctions (N_1F_1) serves as a spin injector. If one drives a bias charge current I from F_1 to N_1, as shown in Fig. 1, spin accumulates at the N_1F_1 interface. The resulting spin imbalance leads to spin diffusion in all possible directions in N_1. Since the charge current is drained from the left side of N_1, the right side of N_1 carries a *pure* spin current with no net flow of charge. The second NF interface (N_2F_2) is used to detect the spin current flowing through N_2 by measuring the nonlocal voltage $V_{\rm nl}$ that establishes itself between N_2 and F_2. If F_1 and F_2 are designed to have different coercive fields, it is possible to realize both parallel and antiparallel magnetization directions by applying an external magnetic field. Below, we determine the nonlocal spin signal by computing the nonlocal resistance $R_{\rm nl} = V_{\rm nl}/I$.

In the following, we assume that the normal metals are oriented along the x axis, whereas the ferromagnets are oriented along the y axis (see Fig. 1). To simplify the notation, we use two different coordinate systems for N_1 and N_2 with origins at the respective NF interface and the x axis directed toward the superconductor; i.e., the respective NS interfaces are at $x = L_i$ ($i = 1,2$).

Let us first consider the processes that occur at the N_1SN_2 interfaces in more detail. As shown in Fig. 2, an incoming electron with subgap energy may undergo four possible
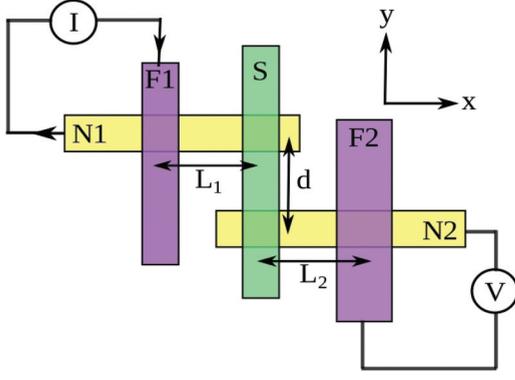
FIG. 1. (Color online) Schematic diagram of the device. The bias current is applied from a ferromagnet F_1 to a normal metal N_1 and nonlocal voltage is measured between F_2 and N_2.

processes at the $N_1 S$ interface: local Andreev reflection (AR), normal reflection (NR), CAR, and EC. Only the nonlocal processes (CAR and EC) contribute to the nonlocal signal. If the length of the superconducting link between N_1 and N_2 is much longer than ξ_S, these processes are suppressed. In that limit, only AR and NR occur at the $N_1 S$ interface, and thus the detector voltage between N_2 and F_2 is zero. In the opposite regime, when d is shorter than ξ_S, however, nonlocal processes such as CAR and EC can occur and generate a spin current in N_2. The resulting spin accumulation at the $N_2 F_2$ interface yields a finite voltage difference between N_2 and F_2. It is important to note here that, in the subgap regime, the superconductor cannot support spin accumulation: The spin of an electron injected into the superconductor at the $N_1 S$ interface is transferred to N_2, either via a hole with the opposite spin (CAR) or via an electron with the same spin (EC), thus leaving no net spin in the superconductor. The spin current injected from N_1 into S, $I_{N_1 S}^s$, equals the spin current injected from S into N_2, $-I_{N_2 S}^s$. Here $I_{N_i S}^s = I_{N_i S}^\uparrow - I_{N_i S}^\downarrow$ is the difference of spin-up and spin-down currents, with a fixed spin quantization axis collinear with the magnetizations in $F_{1/2}$. As a consequence, as can be seen schematically in Fig. 2, both CAR and EC contribute to a spin current in N_2 in the same direction, such that their contributions to the spin signal add.
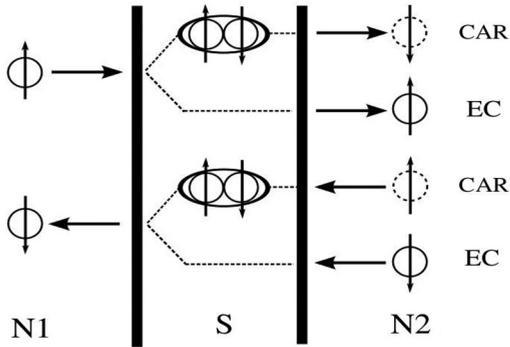

FIG. 2. Nonlocal correlations near the $N_1 S N_2$ structure. Pure spin current is injected from N_1 and a finite spin current is generated in N_2 by CAR and EC. One should note that the direction of the spin currents attributed to CAR and EC are in the same direction and there is no net spin accumulation in the superconductor.

This is completely different from the case of charge current, where one measures the difference between CAR and EC. The spin-resolved injected currents from N_1 into S are given by

$$I_{N_1 S}^\sigma = \frac{G_{\text{CAR}}}{2e} \left[\mu_{N_1}^\sigma(L_1) + \mu_{N_2}^{\bar{\sigma}}(L_2) - 2\mu_S \right]$$
$$+ \frac{G_{\text{EC}}}{2e} \left[\mu_{N_1}^\sigma(L_1) - \mu_{N_2}^\sigma(L_2) \right], \qquad (1)$$

where $\sigma = \uparrow, \downarrow$ and $\bar{\sigma} = \downarrow, \uparrow$. Here we set $[\mu_{N_i}^\uparrow(L_i) + \mu_{N_i}^\downarrow(L_i)]/2 = \mu_S$, since there is no charge current flowing into the superconductor. Overall, the spin current through the superconductor, $I_S^s = I_{N_1 S}^s = -I_{N_2 S}^s$, is related to the difference between the spin imbalances $\delta\mu_{N_i}(L_i) = [\mu_{N_i}^\uparrow(L_i) - \mu_{N_i}^\downarrow(L_i)]/2$ at the two interfaces,

$$I_S^s = \frac{G_S^+}{e} \left[\delta\mu_{N_1}(L_1) - \delta\mu_{N_2}(L_2) \right]. \qquad (2)$$

Here $\mu_{N_i}^{\uparrow/\downarrow}$ are the spin-resolved electrochemical potentials. Note that $\mu_{N_1}(L_1) = \mu_{N_2}(L_2)$, with $\mu_{N_i}(L_i) = [\mu_{N_i}^\uparrow(L_i) + \mu_{N_i}^\downarrow(L_i)]/2$, in the absence of a charge current. Furthermore, $G_S^+ = G_{\text{CAR}} + G_{\text{EC}}$, where $G_{\text{CAR/EC}}$ are the conductances due to CAR and EC, respectively.

To describe spin accumulation and spin transport at the NF interfaces, we use the model introduced by Takahashi *et al.*[18] The current density $j_\alpha^\sigma = -(\sigma_\alpha^\sigma/e)\nabla\mu_\alpha^\sigma$ for spin $\sigma = \uparrow, \downarrow$ is related with the spin-resolved conductivity σ_α^σ and electrochemical potential μ_α^σ in material $\alpha = F_i, N_i$. The continuity equation for the charge current density $j_\alpha = j_\alpha^\uparrow + j_\alpha^\downarrow$ reads

$$\nabla j_\alpha = 0, \qquad (3)$$

while the relaxation of spin imbalance is described by the phenomenological equation

$$\nabla^2 \delta\mu_\alpha - (1/\lambda_\alpha^2)\delta\mu_\alpha = 0, \qquad (4)$$

where λ_α is the spin diffusion length. The spin current density is $j_\alpha^s = j_\alpha^\uparrow - j_\alpha^\downarrow$.

The general solution of Eqs. (3) and (4) in F_i reads

$$\mu_{F_i}^{\uparrow/\downarrow}(y) = \mu_{F_i}(0) + \left(p_{F_i} \pm \frac{2\sigma_{F_i}^{\downarrow/\uparrow}}{\sigma_F} e^{-|y|/\lambda_F} \right) \delta\mu_{F_i}(0)$$
$$+ \frac{eI_i y}{\sigma_F A_F} \Theta(y), \qquad (5)$$

with $I_1 = I$ and $I_2 = 0$. $\Theta(y)$ is the Heaviside function. Note that the chemical potentials in the ferromagnets are defined in the same way as in the normal metals, i.e., $\mu_{F_i}^{\uparrow/\downarrow}(y) = \mu_{F_i}(y) \pm \delta\mu_{F_i}(y)$. For simplicity, we assume identical cross sections A_F, spin-diffusion lengths λ_F, and total conductivities $\sigma_F = \sigma_{F_i}^\uparrow + \sigma_{F_i}^\downarrow$ in the wires F_1 and F_2, while $p_{F_i} = (\sigma_{F_i}^\uparrow - \sigma_{F_i}^\downarrow)/\sigma_F$. In particular, p_{F_1} and p_{F_2} have the same modulus p_F, while their sign is determined by the magnetization direction in F_i, $p_{F_i} = \pm p_F$, depending on whether electrons with spin-up/down correspond to majority/minority $(+)$ or minority/majority $(-)$ electrons, respectively. Using Eq. (5) to compute the current densities at $y = 0^\pm$, we can relate the spin imbalance at the FN interfaces to the drive current and the spin currents $I_{F_i N_i}^s$ injected from F_i to N_i through the $F_i N_i$

interface,

$$\delta\mu_{F_i}(0) = \tfrac{1}{2}eR_F\left(p_{F_i}I_i - I^s_{F_iN_i}\right), \quad (6)$$

where $R_F = \lambda_F/[A_F\sigma_F(1 - p_F^2)]$.

The currents through the F_iN_i interface may also be expressed in terms of the potential drops across the interface,

$$I_i = \frac{G_{T_i}}{e}\left(\mu_{F_iN_i} + p_{T_i}\delta\mu_{F_iN_i}\right), \quad (7a)$$

$$I^s_{F_iN_i} = \frac{G_{T_i}}{e}\left(p_{T_i}\mu_{F_iN_i} + \delta\mu_{F_iN_i}\right), \quad (7b)$$

where $G_{T_i} = G^{\uparrow}_{T_i} + G^{\downarrow}_{T_i}$ and $p_{T_i} = (G^{\uparrow}_{T_i} - G^{\downarrow}_{T_i})/G_{T_i}$ are related with the (tunnel) conductances $G^{\uparrow/\downarrow}_{T_i}$ of the N_iF_i interface for spin-up and spin-down electrons. Furthermore, $\mu_{F_iN_i} = \mu_{F_i}(0) - \mu_{N_i}(0)$ and $\delta\mu_{F_iN_i} = \delta\mu_{F_i}(0) - \delta\mu_{N_i}(0)$. The sign of p_{T_i} again depends on the magnetization direction of the ferromagnet. Note that spin-flip scattering at the interface has been neglected in Eqs. (7) for simplicity. Inverting these equations, we obtain

$$\mu_{F_iN_i} = eR_{T_i}\left(I_i - p_{T_i}I^s_{F_iN_i}\right), \quad (8a)$$

$$\delta\mu_{F_iN_i} = eR_{T_i}\left(-p_{T_i}I_i + I^s_{F_iN_i}\right), \quad (8b)$$

where $R_{T_i} = 1/[G_{T_i}(1 - p_{T_i}^2)]$.

The general solution of Eqs. (3) and (4) in the normal metals N_i reads

$$\mu_{N_i}(x) = \mu_{N_i}(0) - \frac{eI_ix}{\sigma_NA_N}\Theta(-x), \quad (9a)$$

$$\delta\mu_{N_i}(x) = \frac{1}{2}eR_N\left(I^s_{F_iN_i}e^{-\frac{|x|}{\lambda_N}} - I^s_{N_iS_i}e^{-\frac{|L_i-x|}{\lambda_N}}\right), \quad (9b)$$

with $R_N \equiv 1/G_N = \lambda_N/\sigma_NA_N$. As for the ferromagnets, here we also assume identical cross sections A_N and spin-diffusion lengths λ_N and conductivities σ_N in the wires N_1 and N_2. Furthermore, $\sigma^{\uparrow}_N = \sigma^{\downarrow}_N = \sigma_N/2$. Using these equations, we find, in particular,

$$\delta\mu_{N_i}(0) = \tfrac{1}{2}eR_N\left(I^s_{F_iN_i} - I^s_{N_iS_i}e^{-L_i/\lambda_N}\right), \quad (10a)$$

$$\delta\mu_{N_i}(L_i) = \tfrac{1}{2}eR_N\left(I^s_{F_iN_i}e^{-L_i/\lambda_N} - I^s_{N_iS_i}\right). \quad (10b)$$

Combining Eqs. (1) with (2), we can eliminate the spin currents and imbalances at the NS interfaces to obtain

$$\delta\mu_{N_i}(0) = \frac{eR_N}{2}\left\{I^s_{F_iN_i}\left[1 - \frac{G^+_S}{2(G^+_S + G_N)}e^{-2L_i/\lambda_N}\right] + I^s_{F_{\bar{i}}N_{\bar{i}}}\frac{G^+_S}{2(G^+_S + G_N)}e^{-(L_1+L_2)/\lambda_N}\right\}, \quad (11)$$

where we used the notations $\bar{1} = 2$ and $\bar{2} = 1$.

The nonlocal voltage V_{nl} between F_2 and N_2 is given by $\mu_{N_2}(+\infty) - \mu_{F_2}(-\infty)$. Using Eqs. (5) and (9), we find $V_{nl} = [\mu_{F_2N_2} + p_{F_2}\delta\mu_{F_2}(0)]/e$, which, using Eqs. (6) and (8a), may be rewritten as $V_{nl} = -(p_{F_2}/p_F)R_{NF_2}I^s_{F_2N_2}$ where we defined $R_{NF_i} = p_FR_F/2 + |p_{T_i}|R_{T_i}$ and used $p_{T_i}/|p_{T_i}| = p_{F_i}/p_F$. Finally, combining Eqs. (6), (8b), and (11), we determine the

spin current $I^s_{F_2N_2}$ as a function of the injection current I to obtain the nonlocal spin resistance,

$$R_{nl} = \pm\frac{R_{NF_1}R_{NF_2}R_0e^{-(L_1+L_2)/\lambda_N}}{R_{NS_1}R_{NS_2} - R_0^2e^{-2(L_1+L_2)/\lambda_N}}, \quad (12)$$

where $R_0 = G^+_S/[4G_N(G^+_S + G_N)]$ and $R_{NS_i} = R_N/2 + R_F/2 + R_{T_i} - R_0e^{-2L_i/\lambda_N}$. The overall sign of R_{nl} depends on whether the ferromagnets are aligned parallel $(+)$ or antiparallel $(-)$.

Equation (12) is the central result of this Rapid Communication: It predicts a finite nonlocal resistance with no charge current injected into the superconductor. The exponential dependence $\propto e^{-(L_1+L_2)/\lambda_N}$ of R_{nl} on the lengths of the normal wires clearly shows that it is due to the spin transport through the structure. In the case of fully polarized ferromagnets, where $|p_{T_i}| = p_F = 1$ and $R_F/2 + R_{T_i} \to \infty$, the result (12) simplifies to $R_{nl} = \pm R_0e^{-(L_1+L_2)/\lambda_N}$.

A nonlocal spin signal is possible also in the absence of superconductivity. Note that, at $G^+_S \gg G_N$, when the contribution of the superconducting element to the nonlocal signal is negligible, Eq. (12) is similar to the result of Ref. 18 up to factors due to a different geometry of the normal part of the FNF spin valve. In our geometry, a conventional spin valve is realized at temperatures above the superconducting transition temperature T_c. We expect the decay lengths of the nonlocal signal within the superconductor to be quite different below and above the transition. At $T \ll T_c$, our results yields $R_{nl} \propto e^{-d/\xi_S}$ since $G_{CAR/EC}$ decay exponentially on the scale ξ_S. In contrast, above T_c, the nonlocal resistance should be proportional to e^{-d/λ_S}, where λ_S is the spin diffusion length of the superconductor in the normal state. Typically, λ_S for Al is $\sim$500–1000 nm and ξ_S is $\sim$100–300 nm. Thus, one would expect an abrupt change in the nonlocal resistance when the superconductor transitions to its zero-resistance state. This effect is possible to demonstrate in experiments similar to Ref. 8.

The magnitude of the nonlocal resistance that is induced by the injection of pure spin current may be estimated from the nonlocal signal measured in the case of charge current injection. In that case, based on the formalism suggested by Falci et al.,[3] the nonlocal resistance can be written as $R^{NSN}_{nl} = (G_{EC} - G_{CAR})/G^2_{AR}$, where the conductance due to AR at a single NS interface $G_{AR} \gg G_{CAR}, G_{EC}$ is assumed to be the same for the N_1S and N_2S interfaces. Using the measured R^{NSN}_{nl} as well as estimated values of G_{AR} from Cadden-Zimansky et al.[10] yields a rough estimate of $G_{EC} - G_{CAR} \sim 0.5 \, \Omega^{-1}$. A lower bound for the nonlocal spin resistance can be obtained if we assume that $G_{EC} \gg G_{CAR}$. For copper wires with a spin diffusion length $\lambda_N \sim 1 \, \mu$m and cross section $A_N = 100 \times 50 \, $nm^2, corresponding to $G_N \approx 0.3 \, \Omega^{-1}$, the factor R_0 in Eq. (12) would be of the order of 0.5 Ω in this case. If G_{CAR} and G_{EC} are of the same order of magnitude, the nonlocal resistance would likely be much larger.

The advantage of the device suggested in the Rapid Communication is that it is possible to perform multiple measurements on the same device. First, one can perform the nonlocal measurement similar to that reported earlier[10] on NSN structure of the device by biasing charge current

from N_1 to S and measuring the nonlocal voltage between N_2 and S. The nonlocal resistance measured in this scheme is attributed to the difference between the contribution of CAR and EC. By combining with the nonlocal resistance that arises in response to pure spin current injection described earlier in the Rapid Communication, it should be possible to separate CAR and EC. Second, if one biases charge current from F_1 to S, the charge current flowing through N_1 is spin polarized. Since the nonlocal voltage between F_2 and S would exhibit additional effect due to spin polarization on the nonlocal voltage induced by charge current injection that can be picked up by the nonlocal voltage between N_2 and S, the effect of spin polarization can be investigated by comparing the the nonlocal voltages between N_2/S and F_2/S.

In summary, we have investigated the nonlocal signal that arises from the injection of pure spin current into a superconductor. We have shown that a finite electrical resistance may be generated entirely due to the nonlocal correlations mediated by a superconductor. In contrast to other recent work, this nonlocal signal arises from spin transport at energies far below the superconducting gap. Measurements of the nonlocal resistance resulting from charge and spin injection on the same sample would allow independent determination of the contributions due to CAR and EC.

[1] J. M. Byers and M. E. Flatté, Phys. Rev. Lett. **74**, 306 (1995).

[2] G. Deutscher and D. Feinberg, Appl. Phys. Lett. **76**, 487 (2000).

[3] G. Falci, D. Feinberg, and F. W. J. Hekking, Europhys. Lett. **54**, 255 (2001).

[4] G. Bignon, M. Houzet, F. Pistolesi, and F. W. J. Hekking, Europhys. Lett. **67**, 110 (2004).

[5] D. Feinberg, Eur. Phys. J. B **36**, 419 (2003).

[6] R. Mélin and D. Feinberg, Eur. Phys. J. B **26**, 101 (2002).

[7] A. Levy Yeyati, F. S. Bergeret, A. Martín-Rodero, and T. M. Klapwijk, Nat. Phys. **3**, 455 (2007).

[8] D. Beckmann, H. B. Weber, and H. V. Löhneysen, Phys. Rev. Lett. **93**, 197003 (2004).

[9] S. Russo, M. Kroug, T. M. Klapwijk, and A. F. Morpurgo, Phys. Rev. Lett. **95**, 027002 (2005).

[10] P. Cadden-Zimansky and V. Chandrasekhar, Phys. Rev. Lett. **97**, 237003 (2006).

[11] J. Clarke, Phys. Rev. Lett. **28**, 1363 (1972).

[12] M. Tinkham and J. Clarke, Phys. Rev. Lett. **28**, 1366 (1972).

[13] J. Brauer, F. Hübler, M. Smetanin, D. Beckmann, and H. V. Löhneysen, Phys. Rev. B **81**, 024515 (2010).

[14] F. Hübler, J. Camirand Lemyre, D. Beckmann, and H. V. Löhneysen, Phys. Rev. B **81**, 184524 (2010).

[15] K. Yu. Arutyunov, H.-P. Auraneva, and A. S. Vasenko, Phys. Rev. B **83**, 104509 (2011).

[16] F. Hübler, M. J. Wolf, D. Beckmann, and H. V. Löhneysen, Phys. Rev. Lett. **109**, 207001 (2012).

[17] C. H. L. Quay, D. Chevallier, C. Bena, and M. Aprili, Nat. Phys. **9**, 84 (2013).

[18] S. Takahashi and S. Maekawa, Phys. Rev. B **67**, 052409 (2003).