Tunable Pure Spin Supercurrents and the Demonstration of Their Gateability in a Spin-Wave Device

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(Received 10 January 2020; revised 6 April 2020; accepted 27 May 2020; published 27 July 2020)

Recent ferromagnetic resonance experiments and theory of Pt/Nb/Ni₈Fe₂ proximity-coupled structures strongly suggest that spin-orbit coupling (SOC) in Pt in conjunction with a magnetic exchange field in Ni₈Fe₂ are the essential ingredients to generate a pure spin supercurrent channel in Nb. Here, by substituting Pt for a perpendicularly magnetized Pt/Cu/Pt spin sink, we are able to demonstrate the role of SOC and show that pure spin supercurrent pumping efficiency across Nb is tunable by controlling the magnetization direction of Co. By inserting a Cu spacer with weak SOC between Nb and Pt/(Co/Pt) spin sink, we also prove that Rashba-type SOC is key for forming and transmitting pure spin supercurrents across Nb. Finally, by engineering these properties within a single multilayer structure, we demonstrate a prototype superconductor spin-wave device in which lateral spin-wave propagation is gateable via the opening or closing of a vertical pure spin supercurrent channel in Nb.

I. INTRODUCTION

Spin-triplet Cooper pairs carry a net spin in addition to charge and are therefore key to the development of superconducting spintronics [1–3], underlying a future revolution in energy-efficient computing. It is established that spin-polarized triplet pairs are generated via spin-mixing and spin-rotation processes at magnetically inhomogeneous superconductor-ferromagnet (SC-FM) interfaces [1–3]. Recently, theoretical [4–8] and experimental studies [9–13] have been dedicated to an alternative mechanism for triplet pair creation involving spin-orbit coupling (SOC) in combination with a magnetic exchange field $h_{ex}$. In such systems, triplet pair creation depends on the commutation relationship [4–7] between SOC and $h_{ex}$.

The latter mechanism via SOC in conjunction with $h_{ex}$ offers a conceptually novel approach to tune superconducting spin currents, as we demonstrate here using ferromagnetic resonance (FMR) spin pumping [9,14]. When a perpendicularly magnetized Pt/Co/Pt spin sink is proximity coupled to Nb (singlet SC) [Fig. 1(a)], the Co thickness $t_{co}$-dependent magnetization anisotropy [15,16] changes its effective tilt angle $\theta_{co}$ under in-plane (IP) FMR of the IP magnetized Ni₈Fe₂ [Fig. 1(b)]. This alters the degree of orthogonality between $h_{ex}$ and SOC at the interface of Nb and Pt/(Co/Pt) spin sink. Manipulating $\theta_{co}$ determines the efficiency with which spin-zero ($S = 1$, $s_z = 0$) triplets [converted from spin singlets ($S = 0$) by the presence of $h_{ex}$] rotate to form equal-spin ($S = 1$, $s_z = ±1$) triplets [4–6]. This enables orthogonality tuning of spin-angular-momentum transfer from the precessing Ni₈Fe₂ through the proximity-induced equal-spin triplets into singlet Nb layers, which we call superconducting pure spin currents [9] (see Sec. II of Supplemental Material for...
the calculated spatial dependence of the equal-spin triplets [17]). Such transmitted spin currents to Pt/Co/Pt spin sinks result in the enhanced spin pumping or transfer which is then probed by measuring the FMR linewidth broadening (Gilbert damping increase) of the middle Ni₈Fe₂ layer [9,14].

To demonstrate our approach, we perform a series of FMR measurements on Pt/Co/Pt/Nb/Ni₈Fe₂/Nb/Pt/Co/Pt multilayers [Fig. 1(a)]. The ultrathin (≤1.5 nm) perpendicularly magnetized Co layers serve as an internal source of \( h_{\text{ex}} \) to the neighboring (inner) Pt layers, supplying spontaneous spin splitting [18,19] with out-of-plane (OOP) polarization [Fig. 1(a)]. The outer Pt layers boost the perpendicular anisotropy of the Co as well as the total effective spin conductance of Pt/Co/Pt trilayers [20] while suppressing the emergence of a noncollinear magnetic ground state.
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(e.g., magnetic Skrymion) owing to the structural symmetry and cancellation of top and bottom Dzyaloshinskii-Moriya interactions (DMIs) [21]. A weak DMI and thereby the absence of magnetic Skrymions in our Pt/Co/Pt symmetric structures are confirmed by magnetic force microscopy (MFM) (see the Appendix), which is in good agreement with previous experimental reports [22,23]. By inserting a thin Cu spacer with weak SOC at the interface between Nb and Pt/(Co/Pt) layers [Fig. 1(a)], we are able to separate the contribution of interfacial Rashba-type SOC at the Nb/Co to the \( \theta_{Co} \)-dependent superconducting spin-pumping efficiency from other contributions such as stray fields and to compare it with the prediction from spin-triplet proximity theory [4–6,8].

II. RESULTS AND DISCUSSION

We first measure the \( T_c \) dependence of the superconducting transition \( T_c \) [Fig. 1(c)] for a series of multilayers with and without Cu spacers. \( T_c \) decreases rapidly with increasing \( t_{Co} \) until it reaches about 1.5 nm, where it slightly increases. No significant change in \( T_c \) (\( t_{Co} \)) appears with the addition of the Cu spacer, consistent with its long (thermal) coherence length of several hundred nanometers [3]. In analogy with the original consideration on the nonuniform superconducting state [24,25], such nonmonotonic \( T_c \) behavior has been discussed based on a spatial modulation of the superconducting order parameter due to Cooper pairs acquiring a nonzero net momentum in the presence of \( h_{ex} \), in particular, for SC-FM multilayers or SC-FM bilayers with FM thickness \( t_{FM} \) of the order of the coherence length \( \xi_{FM} \), which leads to a damped oscillatory behavior of the order parameter [26]. A quantitative analysis (see the Appendix) of the \( T_c \) data [black lines in Fig. 1(d)] gives an effective \( \xi_{FM} \) of 1.4–1.6 nm and interface transparency \( \gamma_B = 0.18–0.20 \) for our samples, which are in reasonable agreement with those obtained from Nb/FM [27] bilayers and Nb/Cu/FM trilayers [28] with strong FMs.

The \( t_{Co} \)-dependent magnetization anisotropy of the Pt/Co/Pt spin sinks can be independently characterized by static magnetometry measurements on Pt/Co/Pt/Nb-only films with different \( t_{Co} \). Figure 1(e) shows the typical magnetization hysteresis \( m(H) \) curves obtained at 8 K by applying the external magnetic field \( \mu_0 H \) parallel and perpendicular to the film plane. At low \( t_{Co} \) (\( \leq 0.8 \) nm), the easy axis of the Co magnetization \( M_{Co} \) is OOP, indicating that the ultrathin Co sandwiched between two Pt layers has well-established perpendicular magnetization anisotropy (PMA), as expected for the Pt \( 5d \) – Co \( 3d \) orbital hybridization at either Pt/Co interface plus SOC [15]. As \( t_{Co} \) approaches 1.5 nm, the predominant magnetization anisotropy changes from OOP to IP, exhibiting the reorientation transition [16]. Using the relationship [16] \( \mu_0 H_{an} M_s/2 = K_{eff} \), where \( \mu_0 H_{an} \) is the anisotropy field and \( M_s \) is the saturation magnetization, the effective PMA energy \( K_{eff} \) is estimated for \( t_{Co} \leq 0.8 \) nm to be \( \sim 1 \) MJ m\(^{-3}\), comparable to typical values of the perpendicularly magnetized Pt/Co/Pt trilayers [29].

Assuming coherent rotation of \( M_{Co} \) from OOP under the application of IP resonance fields \( \mu_0 H_{res} \) for the middle Ni\(_8\)Fe\(_2\), the effective \( K_{eff} \) can be estimated using the simple Stoner-Wohlfarth model where \( \theta_{Co} = \arccos[M(\mu_0 H_{res})/M_s] \). We then achieve discrete tilt states of the Pt/Co/Pt spin sinks from OOP to IP [Fig. 1(f)], which are systematically controllable by varying \( t_{Co} \). Note that from a MFM study (Fig. 5), the typical dimension of Co magnetic domains (a few microns) in our structure is found to be approximately 2 orders of magnitude larger than both the superconducting coherence length of Nb thin film (\( \leq 40 \) nm) and the domain wall width \( \Delta_{DW} \) of the perpendicularly magnetized Co layer (8.6 nm at 300 K) [30]. In addition, given that \( \Delta_{DW} \) is inversely proportional to \( K_{eff} \) and \( M_s \) [30,31], \( \Delta_{DW} \) is expected to narrow even further at a lower \( T \). This rules out any possible contribution of domain walls and associated magnetic inhomogeneities to the superconducting-state FMR damping enhancement. One can thus assume that within the coherence length which determines the active regime of the triplet proximity effect, the Co magnetization is homogeneous and it rotates coherently under the application of an IP \( \mu_0 H_{res} \).

We next show the influence of the tilt states on the superconducting spin-pumping efficiency, namely that the associated orthogonality between \( h_{ex} \) and SOC at the Nb/Co/Pt interface strongly modifies the spin-angular-momentum transfer in the superconducting state. Figures 2(a) and 2(b) show the microwave frequency \( f \) dependence of FMR data for the Cu-absent (Cu-present) samples, taken above and below \( T_c \) of the Nb layers. From this, we extract the effective Gilbert damping \( \alpha \), which provides a measure [9,14,19] of the net spin current flow for the Ni\(_8\)Fe\(_2\) and the effective saturation magnetization \( \mu_0 M_s \) (see the Appendix).

The extracted \( \alpha \) and \( \mu_0 M_s \) values are plotted as a function of \( t_{Co} \) in Fig. 2(c). In the normal state (\( T/T_c > 1 \), \( \alpha \) is almost \( t_{Co} \) independent for both sample sets and there is a small decrease in the magnitude by introducing the Cu spacers. This means that the presence of ultrathin Co (\( \leq 2 \) nm) and Cu (5 nm) layers hardly changes the normal-state spin-pumping behavior, as expected from their small spin conductances [20] relative to Pt, and the three layers (Co, Cu, Pt) are all approximately spin transparent [33] with each other due to their similar crystal and electronic structures.

In the superconducting state (\( T/T_c < 1 \)), a significant \( t_{Co} \)-dependent enhancement of \( \alpha \) appears and is strongly affected by the addition of Cu. For the Cu-absent multilayers, as \( t_{Co} \) increases, the superconducting-state damping enhancement (indicating the enhanced spin flow or transfer mediated most likely by equal-spin triplet pairing) [8,14,19] rapidly rises until reaching 0.8 nm and then slowly decreases for thicker Co layers, resulting in a
FIG. 2. Correlation of Co tilt angle with superconducting spin-pumping efficiency. (a) Microwave frequency \( f \) dependence of ferromagnetic resonance (FMR) absorption for symmetric Pt(2.0 nm)/Co(\( t_{Co} \))/Pt(1.7 nm)/Nb(30 nm)/NiFe(6 nm)/Nb(30 nm)/Pt(1.7 nm)/Co(\( t_{Co} \))/Pt(2.0 nm) samples with various Co thicknesses, taken above and below \( T_c \) of the couple Nb. From this, one can extract the (effective) Gilbert-type damping \( \alpha \) and the (effective) saturation magnetization \( \mu_0 M_s \). (b) Data equivalent to (a) but for symmetric Pt(2.0 nm)/Co(\( t_{Co} \))/Pt(1.7 nm)/Cu(5 nm)/Nb(30 nm)/NiFe(6 nm)/Nb(30 nm)/Cu(5 nm)/Pt(1.7 nm)/Co(\( t_{Co} \))/Pt(2.0 nm) samples. Note that in any case, the zero-frequency line broadening \( \mu_0 \Delta H_0 \) due to long-range magnetic inhomogeneities is less than 0.5 mT and the FMR linewidth \( \mu_0 \Delta H \) scales linearly with \( f \), indicating the high quality of the samples and the absence of two-magnon scattering [32]. Extracted \( \alpha \) (c) and \( \mu_0 M_s \) (d) values as a function of \( t_{Co} \) for the samples with and without the Cu spacer. The dashed lines are guide to the eyes. (e) Damping difference across \( T_c \), denoted as \( |\alpha_{2K} - \alpha_{6K}|/2\Delta_{2K} \), where \( \Delta_2 \) is the superconducting gap at 2 K calculated from the measured \( T_c \) [Fig. 1(d)], as a function of the (effective) Co tilt angle \( \theta_{Co} \). The black solid (dashed) line is a fit from spin-triplet proximity theory [4–6,8] for the Cu-absent (Cu-present) samples (Secs. 1 and 2 of Supplemental Material [17]). (f) Interfacial SOC contribution \( \Delta H_{SOC, 2K} \) separated by taking the difference between the \( |\alpha_{2K} - \alpha_{6K}|/2\Delta_{2K} \) data (e) with and without the Cu spacer. The black solid line is a theoretical fit based on Rashba-type SOC-induced triplet paring [6,8] (Secs. 1 and 2 of Supplemental Material [17]). Here, the amplitude and component of Rashba SO field and the exchange field strength are only adjustable parameters to get to the theoretical fit. The inset of (e) and (f) shows \( |\alpha_{2K} - \alpha_{6K}|/2\Delta_{2K} \) data as a function of Cu spacer thickness \( t_{Cu} \) for the \( t_{Co} = 0.8 \) nm samples (Supplemental Material, Sec. IV [17]). The red and blue symbols in (c) and (d) represent independent sets of the samples grown each in a single deposition run.
maximum at $t_{Co} \approx 0.8$ nm. For the Cu-present samples, the overall amplitude of damping enhancement diminishes compared with the Cu-absent samples and the maximum moves to a lower value of $t_{Co}$ (0.4 nm). Since this nontrivial enhancement of $\alpha(t_{Co})$ occurs in the ultrathin regime ($t_{Co} \leq 2$ nm, about one order of magnitude smaller than the spin diffusion length [34]) only for the superconducting state, it must reflect how the tilt states of the Pt/Co/Pt spin sinks correlate with the superconducting spin transport.

To elucidate this, we have plotted the damping difference across $T_c$, defined as $[\alpha_{2K} - \alpha_{Sk}] / 2\Delta_{2K}$ where $2\Delta$ is the superconducting gap at 2 K calculated from the measured $T_c$ [Fig. 3(d)], with and without the Cu versus the effective $\theta_{Co}$ [Fig. 2(e)]. In the absence of the Cu, $[\alpha_{2K} - \alpha_{Sk}] / 2\Delta_{2K}$ rapidly rises with increasing $\theta_{Co}$ from 0° to 56° followed a fall for a higher angle. However, this characteristic angular dependence vanishes when the Cu spacer (with weak SOC) is present: the damping difference increases monotonically and slowly up to the highest angle and saturates to a value similar to the Cu-absent $\theta_{Co} \approx 76°$ ($t_{Co} = 0.4$ nm) sample.

There are, in principle, two different sources of proximity-induced triplet pairing which can contribute to the characteristic angular dependence observed in our experimental setup. First, it is well known that magnetization noncollinearity (or inhomogeneity) [1–3,35] between two FMs separated by a SC with a thickness of the order of the coherence length can generate equal-spin triplets through the entire structure. The equal-spin triplet density is then ascribed to the relative magnetization angle $\theta$ between the two FMs [35]: $\propto \mathbf{M}_{Co} \times \mathbf{M}_{Py} \propto \sin(\theta)$ (Py is Ni$_8$Fe$_2$). This explains why our $\theta_{Co} \approx 76°$ ($t_{Co} = 0.4$ nm) samples show larger enhancements than the $\theta_{Co} \approx 90°$ ($t_{Co} = 2.0$ nm) samples [Fig. 2(e)]. Second, even for a single magnetically homogeneous FM, the equal-spin triplet correlation is generated by introducing a strongly SO coupled interface (e.g., Pt) between the FM and SC [4–6,8]. In this case, the singlet-triplet conversion efficiency is predicted to scale with the degree of orthogonality between SOC and $h_{ex}$, or equivalently, the cross product of the SO vector operator $[A_z, \mathbf{h}^a\sigma^a]$ and the exchange field operator $\mathbf{h}^a\sigma^a$. Here $\mathbf{A}_{k=x,y,z}$ is the vector potential describing the form of the SOC, for instance, the Rashba constant $\alpha_R$ (Dresselhaus constant $\beta_D$) due to the interface (bulk) inversion asymmetry. $\sigma^a(h^a)$ with $a = x, y, z$ is the vector of Pauli matrices (exchange field).

For a metallic vertical structure with atomically flat interfaces, the vector potential can be approximated as $\hat{A}_z \approx 0, \hat{A}_y \approx -\beta_D\sigma^y + \alpha_R\sigma^z, \hat{A}_x \approx \beta_D\sigma^x - \alpha_R\sigma^y$ [4,5]. With finite Rashba ($\alpha_R \neq 0$) and zero Dresselhaus ($\beta_D = 0$) contributions to the SOC [6,8], as relevant to our experimental setup, a sinusoidal maximum of the equal-spin triplet correlation is expected when the canting angle between IP and OOP components of $h_{ex}$ becomes 45°. In such a case, the overall triplet density is quadratic in $\alpha_R$ and very sensitive to details of the spin-orbit coupled interface. The addition of a thin Cu spacer layer [36] at the spin-orbit coupled interface is sufficient to quench the interfacial Rashba-type SOC and provide the key test experiment for the mechanism responsible here [see inset of Figs. 2(e) and 2(f)].

We emphasize that for the $t_{Co} = 0.8$ nm sample set with various Cu spacer thicknesses $t_{Cu}$ prepared in a single deposition run [red symbols in the inset of Figs. 2(e) and 2(f)], FMR damping of the middle Ni$_8$Fe$_2$ layer is $t_{Cu}$ independent in the normal state (see Fig. S4 in Supplemental Material [17]). This proves that the addition of Cu has no measurable effect on the normal-state FMR. However, there is a dramatic decrease by a factor of 2 in the superconducting spin-pumping efficiency with increasing $t_{Cu}$, meaning that the presence of the Cu spacer strongly modifies the superconducting-state FMR response due to the quenching of the Rashba SOC at the interface between Nb and Pt/(Co/Pt) layers. Furthermore, the differences in the superconducting FMR response between sample sets with versus without a Cu spacer cannot be justified on the basis of slight variations in the Co static magnetization as such variations would also affect the normal-state FMR.

We note that nonvanishing of $\hat{A}_x(\neq 0)$, as would be expected from nonideal interfaces where the OOP component of the Rashba SO field with respect to the local interface plane survives on a scale of the coherence length [7], allows the equal-spin triplet to be generated locally even with a purely IP magnetized FM ($h^x = 0$). Each triplet channel is then able to transport spin angular momentum from the precessing FM (Ni$_8$Fe$_2$) through a singlet SC (Nb) to a spin dissipative bath (Pt spin sink) independently even if the spatial average of net polarization of total triplet channels over the entire interface plane becomes zero. This is a likely mechanism for our previous FMR experiments [9,19] and for the $t_{Co} = 0$ samples [Fig. 2(e)]. When the Pt spin sink is substituted for the perpendicularly magnetized Pt/Co/Pt spin sink, a global triplet channel opens in addition to the local channels, maximizing the overall superconducting spin-pumping efficiency at $\theta_{Co} \approx 45°$ [Fig. 2(e)].

By taking all these effects together, we can arrive at good fits to $[\alpha_{2K} - \alpha_{Sk}] / 2\Delta_{2K}$ versus $\theta_{Co}$ data for both sample sets [black solid and dashed lines in Fig. 2(e); see Secs. 1 and 2 of Supplemental Material [17]], thereby reasonably reproducing the experimental results and capturing the underlying physics. To focus on the second SOC mechanism, in particular for the interfacial contribution, we take the difference between the data with and without the Cu spacer [Fig. 2(f)]. We then find an approximately sinusoidal maximum at $\theta_{Co} \approx 45°$, which is in good agreement with the Rashba SOC-induced triplet pairing [6,8] described above. The data described above provide a proof-of-concept result demonstrating the orthogonality tuning of superconducting spin currents.

To understand better the FMR absorption data of symmetric structures [Fig. 2], we also measure the $t_{Co}$...
dependence of spin-pumping-induced inverse spin Hall effect (ISHE) [37,38] for the additional sets of asymmetric Pt/Co/Pt/Nb/Ni$_8$Fe$_2$ structures with and without Cu spacers (Fig. 6). This provides direct evidence for spin transport in the normal state. Figure 3(a) [Fig. 3(b)] displays the transverse dc voltage signals versus external magnetic field, $\mu_0 H$ (mT) at various temperatures for asymmetric Pt/Co/Pt/Nb/Ni$_8$Fe$_2$ samples with and without Cu spacers, respectively. The black solid lines are fits to Lorentzian functions (see the Appendix). Figure 3(b) shows the signal difference caused by the Cu spacer addition whereas the right inset in (c) exhibits the Cu spacer thickness dependence of ISHE for the $t_{Co} = 0.8$ nm samples (see Supplemental Material, Sec. V, for details [17]). The dashed lines in (c) are guide to the eyes, whereas the black solid (dashed) line in (d) is a fit to $\cos^2(\theta_{Co})$ for the Cu-absent (Cu-present) samples. The red and blue symbols in (c) and (d) represent independent sets of the samples grown each in a single deposition run.
FIG. 4. Experimental realization of superconductor spin-wave (SW) devices. (a) Spin-wave transmission $\Delta S_{12}$ as a function of frequency $f$ for the Nb(30 nm)/Ni$_8$Fe$_2$(6 nm)/Nb(30 nm) device with a different distance $d$ (10–25 μm) between two separate antennas. These spectra are obtained under application of a fixed external magnetic field $\mu_0 H = 70$ mT above and below $T_c$ of the coupled Nb. In each panel, the red, blue, and black curves represent, respectively, the real, imaginary, and absolute of the frequency $\Delta S_{12}$. (b) Data equivalent to (a) but for the Pt(2.0 nm)/Co(0.8 nm)/Pt(1.7 nm)/Nb(30 nm)/Ni$_8$Fe$_2$(6 nm)/Nb(30 nm)/Pt(1.7 nm)/Co(0.8 nm)/Pt(2.0 nm) device. (c) Normalized intensity of the real part of $\Delta S_{12}$ across $T_c$ for the Pt/Co(0.8 nm)/Pt-absent device with $d = 10–25$ μm. (d) Data equivalent to (c) but for the Pt/Co(0.8 nm)/Pt-present device. Each inset shows the associated $d$ dependence of the signal intensity above and below $T_c$. The dashed lines in (c) and (d) are guide to the eyes whereas the solid lines in each inset are fits to an exponential decay function to estimate the SW attenuation length $\lambda_{\text{eff}}$ [40,41] (see the Appendix).

IP $\mu_0 H$ for the Cu-absent (Cu-present) samples at $f = 5$ GHz, taken above and below $T_c$ (see the Appendix). Under IP FMR of the Ni$_8$Fe$_2$, a clear Lorentzian peak emerges in the dc voltage only in the normal state for both sample sets, which can be explained [38] by the strong decay of the quasiparticle charge imbalance relaxation time immediately below $T_c$. Importantly, the polarity of the Lorentzian peak is identical (opposite) to that of Pt/Ni$_8$Fe$_2$ (Nb/Ni$_8$Fe$_2$) bilayers [38], where the Pt (Nb) spin sink is known to have a positive (negative) spin Hall angle $\theta_{\text{SH}}$ [20,38]. This indicates that the pumped spin currents from the precessing Ni$_8$Fe$_2$ pass through the Nb (30 nm) layer to a large extent to the (Cu)/Pt/Co/Pt spin sinks and the overall ISHE in our structures is dominated by the (Cu)/Pt/Co/Pt (rather than the Nb).

For a quantitative analysis, we plot the ISHE voltage divided by sample resistance $V_{\text{ISHE}}/R$ versus $t_{\text{Co}}$ [Fig. 3(c)] and $\theta_{\text{Co}}$ [Fig. 3(d)]. In these plots, we can see that there is a clear decrease in the ISHE signal by the addition of Cu and its magnitude is strongly $\theta_{\text{Co}}$ dependent, which can be described by the rapid spin precession and dephasing of transverse spins [39] around $\hbar_{\text{ex}}$ of the Co layer: $\cos^2(\theta_{\text{Co}})$ [black lines in Fig. 3(d)]. Note that the signal difference caused by the addition of 5 nm of Cu [insets in Figs. 3(c) and 3(d)] is nearly $\theta_{\text{Co}}$ independent. These results taken together support our argument that Cu spacers weaken the interfacial SOC strength and it is the Co tilt state that then plays a dominant role in the spin transport process.

Finally, we progress to show the potential to harness these effects in a proof-of-principle prototype SC-based spin-wave (SW) device (Fig. 4). The idea behind this is that lateral SW propagation [40,41] in our proximity-engineered structure (e.g., $\alpha_{2K} - \alpha_{8K} \approx 0.005$ for the $t_{\text{Co}} = 0.8$ nm sample) between microwave injector and detector
antennas is readily altered by opening or closing the vertical spin transport channel via the proximity creation of triplet pairing. Figures 4(a) and 4(b) show the $f$-dependent SW transmission $\Delta S_{12}$ of two types of the SW devices with and without Pt/Co(0.8 nm)/Pt spin sinks, obtained above and below $T_c$ at the fixed external IP $\mu_0 H = 70$ mT in the magnetostatic surface wave (MSSW) geometry [40,41] (see the Appendix and Sec. VI of Supplemental Material [17] for details). The observed spectra containing two major peaks in the low $f$ (<7 GHz) regime and satisfying the SW dispersion relationship (Videos 1–4 and Sec. VI in Supplemental Material [17]) and their exponential decay in the intensity with increasing the distance $d$ between the two separate antennas [Figs. 4(c) and 4(d)] indicate the propagating SWs [40,41]. Notably, the absence of characteristic dips [42] in the SW spectra (Videos 1–4 and Sec. VI of Supplemental Material [17]) indicates no significant nucleation or pinning of (OOP) Abrikosov vortices in our device structure.

The most noteworthy aspect in this demonstration is that on entering the superconducting state, the intensity of the lateral SW transmission signal rises (decays) when the Pt/Co(0.8 nm)/Pt spin sinks are removed (added) [Fig. 4(c)] [Fig. 4(d), and see also Videos 1–4 in Supplemental Material [17]], and the degree of this change becomes pronounced with increasing $d$. This is because SWs experience weaker (stronger) effective attenuation during lateral propagation if spin angular momentum is less (more) transmitted across the adjacent superconducting Nb to the spin loss regimes in the vertical direction. Note that the SW attenuation increases proportionally to the total FMR damping of the system [40,41].

With the Pt/Co(0.8 nm)/Pt spin sinks, we are able to modulate the lateral SW transmission intensity up to about 40% by proximity generating the vertical triplet spin-transport channel. This result is encouraging and may provide a new type of SW logic functionality [43] activated in the superconducting state.

III. CONCLUSIONS AND OUTLOOK

We have shown that when a perpendicularly magnetized Pt/Co/Pt spin sink is proximity coupled to Nb, superconducting spin-pumping efficiency can be tuned by controlling the effective $\theta_{\text{Co}}$, i.e., by tuning the degree of orthogonality between the SOC and $\hbar_{\text{ex}}$ at the Nb/Pt/(Co/Pt) interface [4–6,8]. We have also found that by comparison with the Cu-present samples, the $\theta_{\text{Co}}$-dependent superconducting spin-pumping efficiency reflects characteristic features of Rashba SOC-induced triplet pairing [4–6,8]. Our results provide a timely step toward understanding key interfacial properties for tuning superconducting spin transport mediated via equal-spin triplet states in a spin-singlet superconductor. The approach developed here can be used to explore and characterize triplet pair generation in SC-FM heterostructures with Rashba SOC by the application of superconducting charge currents and magnetic fields [44]. Our finely proximity-engineered structures enable experimental realization of a prototype SC-based SW device. This concept can be extended to any Rashba system [45,46] for the development of superconducting spin-logic devices [1] in which SOC is gate tunable [45], leading to a superconducting spin-based transistor.

The data used in this paper can be accessed here [47].

ACKNOWLEDGMENTS

This work was funded by the EPSRC Programme Grant “Superspin” (No. EP/N017242/1) and EPSRC International Network “Oxide Superspin” (No. EP/P026311/1). The work was in-part funded by a Leverhulme Trust Research Project Grant No. RPG-2016-306.

APPENDIX: EXPERIMENTAL METHODS

1. Sample growth

Symmetric Pt/Co/Pt/Nb/Ni$_8$Fe$_2$/Nb/Pt/Co/Pt and asymmetric Pt/Co/Pt/Nb/Ni$_8$Fe$_2$ multilayers, with and without Cu spacer layers, were grown on 5 × 5 mm$^2$ thermally oxidized Si substrates by dc magnetron sputtering in an ultrahigh vacuum chamber [9,19]. The symmetric and asymmetric structures were prepared, respectively, for the ferromagnetic resonance (FMR) absorption [9,19] and inverse spin Hall effect (ISHE) (or transverse dc voltage) [38] measurements. All layers were grown in situ at room temperature. Ni$_8$Fe$_2$, Nb, Co, and Cu are deposited at an Ar pressure of 1.5 Pa and Pt at 3.0 Pa. The typical deposition rates were 5.1 nm/min for Ni$_8$Fe$_2$, 21.1 nm/min for Nb, 6.0 nm/min for Co, 9.7 nm/min for Cu, and 7.6 nm/min for Pt. The thicknesses of Ni$_8$Fe$_2$, Nb, inner (outer) Pt, and Cu layers were kept constant at 6, 30, 1.7 (2.2), and 5 nm, respectively, while the thickness of the Co layer varied from 0 to 2 nm to investigate the variation of FMR damping as a function of $t_{\text{Co}}$ (or the Co tilt angle $\theta_{\text{Co}}$) through the superconducting transition temperature $T_c$ of the coupled Nb. Note that for all samples, the Nb (inner Pt) thickness is fixed at 30 (1.7) nm where the Pt/Co/Pt spin sink was proximity coupled through the Nb layer to the precessing Ni$_8$Fe$_2$ layer and the largest enhancement of spin pumping in the superconducting state was achieved in our prior FMR experiments [9,19].

2. Magnetization characterization

The static magnetization hysteresis curves were measured on 5 × 5 mm$^2$ samples using a Quantum Design magnetic property measurement system at 8 K, immediately above the superconducting transition temperature $T_c$. The external magnetic field was applied parallel and perpendicular to the film plane direction. Moreover, we carried out magnetic force microscopy (MFM) measurements on Pt/Co/Pt/Nb-only films (Fig. 5) to check local
magnetic domain patterns and to assure the absence of a noncollinear magnetic ground state (e.g., magnetic Skyrmion) [21–23]. For the microscopic characterization, we used a room temperature DI3100 magnetic force microscope operated under ambient conditions. We used a high moment CoCr cantilever (Bruker MESP-HM). For the measurement, we operated a dual mode (DI Lift mode) where the topography (not shown) was obtained in tapping mode and the MFM image (Fig. 5) was obtained at a tip-sample distance of 30 nm.

3. Superconducting transition measurement
dc electrical transport measurements were conducted on (unpatterned) 5 × 5 mm$^2$ samples using a custom-built dipstick probe in a liquid helium Dewar with a four-point current-voltage method. The resistance $R$ (of a sample) versus temperature $T$ curves were obtained at the applied current $I$ of ≤0.1 mA while decreasing $T$. From the $T$ derivative of $R$, $dR/dT$, $T_c$ was defined as the $T$ value that exhibits the maximum of $dR/dT$.

We analyzed our $T_c(t_{Co})$ data [Fig. 1(d)] using the following approximate formula [26]:

$$\ln \left[ \frac{T_c^*}{T_c} \right] \approx \Psi \left( \frac{1}{2} \right) - \text{Re} \left\{ \Psi \left[ \frac{1}{2} + \frac{2T_c^*}{T_c^* - \frac{1}{2} \cosh(1 + i T_{FM})} \right] \right\},$$

where $T_c^* = T_c(t_{FM} = 0)$, $\Psi$ is the digamma function, $\frac{1}{2} \cosh(1 + i T_{FM}) = (1/4\pi T_c)(D_{SC}/t_{SC} \xi_{FM}) (\rho_{SC}/\rho_{FM})$, $D_{SC}$ is the diffusion coefficient of the Nb (10 cm$^2$/s at 8 K), $t_{SC}$ is the Nb thickness (30 nm), and $\rho_{SC}$ ($\rho_{FM}$) is the conductivity of the Nb (Co) [7 (30) $\mu$Ω cm at 8 K], $\tilde{\gamma} = \gamma_B (\xi_{SC}/\xi_{FM})$, where $\gamma_B$ is the interface transparency and $\xi_{SC}$ is the (dirty-limit) coherence length of the Nb (16–18 nm at 2 K) [9]. Note that in this formula, only the influence of $h_{ex}$ on the order parameter is taken into account [26].

When the out-of-plane (OOP) component of magnetic fluxes (e.g., stray fields from the OOP-magnetized Co layer) exists, unintentional Abrikosov vortex nucleation [48] can suppress the pair potential, the superconducting volume, and thus the singlet Cooper pair density of Nb that is the underlying source of proximity-induced triplet pairing. To take this detrimental effect into account in Figs. 2(e) and 2(f), we normalized the damping difference across $T_c$ by the calculated superconducting gap $2\Delta$ at 2 K from the measured $T_c$ data [Fig. 1(d)], which is directly proportional to the singlet pair density:

$$\Delta(T) \approx 1.76 k_B T_c \tanh \left[ 1.74 \sqrt{1 - \frac{T}{T_c}} \right],$$

where $k_B$ is Boltzmann’s constant.

4. Broadband FMR absorption and ISHE measurements
We measured the FMR response of the sample attached on a broadband coplanar waveguide (CPW) with either dc field or rf pulse modulation [9,19]. To obtain each FMR spectrum, the microwave power absorbed by the sample was measured while sweeping the external static magnetic field $\mu_0 H$ at the fixed microwave frequency $f$ of 5–20 GHz. At the beginning of each measurement, we applied a large IP $\mu_0 H$ (0.5 T) to fully magnetize the Ni$_8$Fe$_2$ layer, after...
which the field was reduced to the range of FMR. Once the 
f-dependent FMR measurements (from high to low $f$) were 
complete, the field was returned to zero to cool the system 
down further for a lower $T$ measurement. For all FMR 
absorption measurements, the microwave (MW) power was 
set to 10 dBm where the actual microwave power absorbed 
in the sample is a few milliwatt that has no measurable 
effect on $T_c$ of the Nb layer [9]. Based on our previous 
ISHE experiment (Fig. S1 of Ref. [38]), it is reasonable to 
assume that unintentional heating at a higher power 
($\geq 50$ mW in our setup) reduces profoundly the real super-
conducting volume, the effective pair potential, and thus the 
overall singlet pair density of Nb layers, which is the 
underlying source of proximity-induced triplet pairing. 
Note also that the fixed thickness (30 nm) of Nb layers 
studied here is much less than the magnetic penetration 
depth in the superconducting state ($\geq 100$ nm in thin Nb 
films), and so there is no considerable effect of Meissner 
screening on the local (dc or rf) magnetic field experienced 
by Ni$_8$Fe$_2$ below $T_c$, as supported by the insensitivity of 
the resonance field $\mu_0H_{\text{res}}$ across $T_c$ [Figs. 2(a) and 2(b)]. We 
employed a vector field cryostat from Cryogenic Ltd. that 
was employed for the measurements of $\Delta \mu_0H$ and the 
resonance field $\mu_0H_{\text{res}}$: 

$$
\frac{d\chi'}{dH} \propto A \left[ \frac{(\Delta H_{\text{HWHM}})^2 (H - H_{\text{res}})}{((\Delta H_{\text{HWHM}})^2 + (H - H_{\text{res}})^2)^2} \right]
+ B \left[ \frac{(\Delta H_{\text{HWHM}})^2 (H - H_{\text{res}})^2 - (\Delta H_{\text{HWHM}})^3}{((\Delta H_{\text{HWHM}})^2 + (H - H_{\text{res}})^2)^2} \right],
$$

where $A$ ($B$) is the amplitude of the field derivative 
of the symmetric (antisymmetric) Lorentzian function, 
$\mu_0H$ is the external dc magnetic field, and $\mu_0\Delta H_{\text{HWHM}} = 
(\sqrt{3}/2)\mu_0\Delta H$ is the half width at half maximum (HWHM) 
of the imaginary part $\chi''$ of the magnetic susceptibility. 

From the linear scaling of $\mu_0\Delta H$ with $f$ [Figs. 2(a) 
and 2(b)], we calculated the effective Gilbert-type damping 
constant $\alpha$: $\mu_0\Delta H(f) = \mu_0\Delta H_0 + (4\pi\alpha f/\sqrt{3})$, 
where $\mu_0\Delta H_0$ is the zero-frequency line broadening. We also 
estimated the effective saturation magnetization $\mu_0M_s$ (of the 
Ni$_8$Fe$_2$) from the dispersion relation of $\mu_0H_{\text{res}}$ with $f$ 
inset of Figs. 2(a) and 2(b) using Kittel’s formula, 
$f = (\gamma/2\pi) \sqrt{[\mu_0(M_{\text{eff}} + M_{\text{sat}})H_{\text{res}}]}$, where $\gamma = g_L\mu_B/h$ 
is the gyromagnetic ratio ($1.84 \times 10^{11}$ T$^{-1}$ s$^{-1}$), $g_L$ is 
the Landé $g$ factor (taken to be 2.1), $\mu_B$ is the Bohr magneton, 
and $h$ is Plank’s constant divided by $2\pi$. 

For the ISHE (or transverse dc voltage) measurement 
(Fig. 6) [38], the sample was attached face down on the 
CPW by using an electrically insulating high-vacuum 
grease. A microwave signal was passed through the 
CPW and excited FMR of the Ni$_8$Fe$_2$ layer; a transverse 
dc voltage as a function of $\mu_0H$ was measured between two 
Ag-paste contacts at opposite ends of the sample. In these 
measurements, the microwave frequency was fixed at 
5 GHz and the microwave power at the CPW at approxi-
ately 150 mW (for $T = 2$ and 8 K), which yields 
measurable signals ($\geq 5$ nV) in our setup. 

The measured dc voltage [Figs. 3(a) and 3(b)] can be 
decomposed into symmetric and antisymmetric Lorentzian 
functions with respect to $\mu_0H_{\text{res}}$, with weights of $V_{\text{sym}}$ and 
$V_{\text{asy}}$, respectively [38]: 

$$
V(H) - V_0 = V_{\text{sym}} \left[ \frac{(\Delta H)^2}{((\Delta H')^2 + (H - H_{\text{res}})^2)} \right]
+ V_{\text{asy}} \left[ \frac{(\Delta H')(H - H_{\text{res}})}{((\Delta H')^2 + (H - H_{\text{res}})^2)} \right],
$$

where $V_0$ is a background voltage and $\mu_0\Delta H'$ is the HWHM 
of the dc voltage. We attributed $V_{\text{sym}}$ to the ISHE signal $V_{\text{ISHE}}$. If the Co thickness in the Pt/Co/Pt spin sink is larger 
than its spin dephasing length (a few angstroms) [39], 
$V_{\text{ISHE}}(\theta_{\text{Co}})$ is simply proportional to $\cos^2(\theta_{\text{Co}})$ [Fig. 3(d)]. 

5. SW device fabrication 

To fabricate the standard SW devices [40,41] displayed in 
Fig. 7 and Sec. VI of the Supplemental Material [17], the 
Hall bar-type structures with an active SW track of $50 \times 
50$ $\mu$m$^2$ were patterned into the in situ grown Nb/ 
Ni$_8$Fe$_2$/Nb films with and without Pt/Co (0.8 nm)/Pt spin 
sinks by using optical lithography and Ar-ion beam 
etching. After depositing AlN (40 nm) for dc electrical 
isolation by reactive sputtering, coplanar waveguides
6. Propagating SW spectroscopy

A pair of antennas of the SW device were connected to ports 1 and 2 of a vector network analyzer (VNA, Rohde & Schwarz, 100 MHz–20 GHz) by multiple wire bonding to a precalibrated sample holder (having the 50 \( \Omega \) impedance) via phase-stable coaxial cables. The \( f \)-dependent forward complex transmission coefficient (e.g., scattering parameter \( S_{12} \); the MW power received at port 1 relative to the power conveyed to port 2) was measured in the variable temperaturer electron-beam lithography and lift-off of sputtered Cu layers. When the middle Ni\(_8\)Fe\(_2\) layer is (in-plane) magnetized along the positive \( y \) direction, the SW \( (k_{SW} \neq 0) \) driven by the microwave excitation propagates along the positive \( z \) direction via a collective precession of magnetization \( M(z, t) \).

(CPWs or MW antennas) with various interspacing of 10–25 \( \mu m \) were patterned on top of the SW track using electron-beam lithography and lift-off of sputtered Cu(100 nm)/Ti(5 nm) layers. Two identical CPWs consist of a MW signal line (2 \( \mu m \) wide) and two ground lines (1 \( \mu m \) wide) with an intraseparation of 2 \( \mu m \), which preferentially excites or detects the SWs with a number \( k_{SW} \) in the range of 0.9 ± 0.6 \( \mu m^{-1} \) (see Sec. VI of the Supplemental Material [17]).

\[
S_{12}(f, \mu_0 H) = \frac{S_{12}(f, \mu_0 H_{ref}) - S_{12}(f, \mu_0 H_{ref})}{S_{12}(f, \mu_0 H_{ref})}.
\]

The SW dispersion in the MSSW mode for symmetric sample structures is given by [40,41]

\[
f_{SW} \approx \frac{\gamma}{2\pi} \sqrt{\frac{\mu_0(H_{res} + M_{eff})\mu_0 H_{res} + \left(\frac{\mu_0 M_{eff}}{2}\right)^2 [1 - \exp(-2k_{SW}t)]}{\left[H_{res} + \frac{\pi(v_g a)^{-1}}{2\pi} M_{eff}\right]}},
\]

where \( t \) is the Ni\(_8\)Fe\(_2\) thickness (6 nm). By fitting the SW resonance, corresponding to the peak in the absolute of \( \Delta S_{12} \) [insets of Figs. 4(a) and 4(b)], to this dispersion relationship, we extracted the \( k_{SW} \) and \( \mu_0 M_{eff} \) values (Sec. VI of Supplemental Material [17]) for the Ni\(_8\)Fe\(_2\) layer. In addition, we deduced the SW attenuation length \( \lambda_{SW} \) from the fact [40,41] that the SW intensity, defined as the maximum peak-to-valley height of the real part of \( \Delta S_{12} \) [insets of Figs. 4(c) and 4(d)] exponentially decays with increasing \( d: \exp[-(d/\lambda_{SW})] \). Here, \( \lambda_{SW} = v_g \tau_{res} \), \( v_g = 2\pi(\partial f_{SW}/\partial k_{SW}) \) is the group velocity, and \( \tau_{res} = (\alpha\mu_0(2H_{res} + M_{eff}))^{-1} \) is the magnetization precession time.

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