Controlling spin pumping into superconducting Nb by proximity-induced spin-triplet Cooper pairs

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Supplementary Information (SI)

Supplementary note 1. Magnetization measurements on Nb/Cr/Fe/Cr/Nb and Nb/Fe/Nb samples at room temperature.

Magnetic field loops (M-H loops) of the samples were measured with an external dc magnetic field ($H_{ext}$) applied in-plane to the samples at room temperature (300 K). To investigate the anisotropy of the samples, $H_{ext}$ is applied in four orientations 90° to each other. M-H loops are shown in Supplementary Figure 1. All six samples show uniaxial anisotropy to varying strengths.
Supplementary Figure 1. Magnetic field loops (M-H loops) in four orientations with respect to the applied in-plane field measured at 300 K for films (a) 32Cr (b) 32NCr (c) 31Cr and (d) 31NCr (e) 30Cr (f) 25Cr.
Supplementary note 2. Microwave frequency dependence of FMR spectra for Nb/Cr/Fe/Cr/Nb and Nb/Fe/Nb structures

FMR measurements were performed in the same PPMS system as the ac susceptibility measurements, using a broadband coplanar waveguide (CPW) and ac-field modulation technique. The samples were placed face down on top of the CPW where an insulating tape is used for electrical insulation. The external dc field is applied along the in-plane direction of the samples, and the rf field perpendicular to the external dc field. See Supplementary Figure 2 for a schematic of the set up. To measure the effect of in-plane dc field direction, the samples are lifted from the CPW and rotated 90° before placing back on the CPW. The external dc field and rf field remain fixed with respect to each other and the CPW.

Supplementary Figure 2. Schematic ferromagnetic resonance set up The sample is on a coplanar waveguide with two ground conductors (G) and one signal conductor (S). The green arrow indicates the direction of the external dc field $H_{ext}$, and the red arrow indicates the alternating rf field $h_{rf}$. 
Supplementary Figure 3 displays typical FMR spectra attained from the samples with Nb/Cr/Fe/Cr/Nb and Nb/Fe/Nb at 16 K from which the data shown in Fig. 3 were extracted. To accurately determine the resonance magnetic field $\mu_0 H_{\text{res}}$ and the FMR (peak-to-peak) linewidth $\mu_0 \Delta H$, we fitted all FMR spectra with a Lorentzian derivative:

$$\frac{dP}{dH} = -V_{\text{sym}} h_{\text{ac}} \frac{2(H_{\text{ext}} - H_{\text{res}}) \Delta H^2}{[(\Delta H^2 + (H_{\text{ext}} - H_{\text{res}})^2)]^2} - V_{\text{asym}} h_{\text{ac}} \frac{\Delta H((H_{\text{ext}} - H_{\text{res}})^2 - \Delta H^2)}{[(\Delta H^2 + (H_{\text{ext}} - H_{\text{res}})^2)]^2}, \tag{S1}$$

where $V_{\text{sym}}$ and $V_{\text{asym}}$ denote the symmetric and antisymmetric Lorentzian components, $h_{\text{ac}}$ is the small modulation field provided by the Helmholtz coils. The temperature dependence of $\mu_0 H_{\text{res}}$ for samples 31Cr and 31NCr are shown in Supplementary Figure 4(a), and the difference in resonance fields $\mu_0 \Delta H_{\text{res}}$ between when $H_{\text{ext}}$ lies along easy and hard directions is shown in Supplementary Figure 4(b). Similar to samples 31Cr and 31NCr (Fig. 2(c) and 2(d)), the samples with Cr demonstrate a higher $\mu_0 H_{\text{res}}$ and $\mu_0 \Delta H_{\text{res}}$. This further confirms the spin misorientation due to the presence of Cr next to Fe that survives up to $\mu_0 H_{\text{res}}$. 
**Supplementary Figure 3.** Ferromagnetic resonance (FMR) spectra FMR absorption power derivative ($dP/dH$) measured at 16 K and 12 GHz for samples (a) 32Cr (b) 32NCr (c) 31Cr (d) 31NCr (e) 30Cr and (f) 25Cr.
Supplementary Figure 4. Temperature dependence of the resonance magnetic field

Resonance magnetic field ($\mu_0 H_{\text{res}}$) as a function of temperature extracted from FMR measurements for films (a) 31Cr and 31NCr (c) 30 Cr and (e) 25Cr. The temperature dependence of the difference in $\mu_0 H_{\text{res}}$ between the hard and easy orientations $\mu_0 \Delta H_{\text{res}}$ for films (b) 31Cr and 31NCr (d) 30Cr and (f) 25Cr.
Supplementary note 3. Frequency dependence of the linewidth

The microwave frequency $f$ dependence of the linewidth $\mu_0 \Delta H$ extracted from the FMR spectra, at temperatures above $T_c$ and below $T_c$, for samples 32NCr and 31Cr are shown in Supplementary Figure 5. These are fitted to Equation 2 (shown in the main text) and show the clear greater gradient ($\alpha$) for samples with Cr when cooled below the samples’ $T_c$. $\alpha$ is extracted as the gradient of these fits and plotted as a function of $T$ for samples 32Cr, 32NCr, 30Cr and 25Cr in Supplementary Figure 6. Similar to the 31 series samples, the samples with Cr have a higher average $\alpha$ and only the samples with Cr show an apparent rise in $\alpha$ below the samples’ $T_c$. The spin misorientation layer provides an additional spin relaxation channel, leading to the observed $\alpha(T)$ relationship. Further evidence of the spin misorientation layer is given by the magnetic inhomogeneity ($\mu_0 \Delta H_0$), extracted from Equation 2 and are shown for all samples as a function of $T$ in Supplementary Figure 7. The samples containing Cr demonstrate a higher $\mu_0 \Delta H_0$. 32Cr and 31Cr both have $\mu_0 \Delta H_0 \sim 22$ mT while 32NCr and 31NCr have $\mu_0 \Delta H_0 \sim 12$-15 mT. This indicates that the addition of Cr creates a spin misorientation layer and therefore increases $\mu_0 \Delta H_0$. Only the samples containing Cr (32Cr and 31Cr) show the possibility of an increased $\alpha$ below $T_c$ due to the additional spin relaxation channel.
Supplementary Figure 5. Frequency dependence of the linewidth. Microwave frequency dependence of the linewidth ($\mu_0 \Delta H$) for sample (a) 32Cr (b) 32NCr (c) 31Cr (d) 31NCr (e) 30Cr (f) 25Cr with the field applied in the hard axis orientation, above (8 K in black) and below (2 K in red) $T_c$. Solid lines are fits to equation 2.
Supplementary Figure 6. Gilbert damping $\alpha$ of samples $31\text{Cr}$, $31\text{NCr}$, $30\text{Cr}$ and $25\text{Cr}$

Temperature dependence of the Gilbert damping ($\alpha$) for (a) $31\text{Cr}$ (b) $31\text{NCr}$ (c) $30\text{Cr}$ and (d) $25\text{Cr}$, with $\chi'_{ac}$ in red.
Supplementary Figure 7. Magnetic inhomogeneity for the four samples. Temperature dependence of the magnetic inhomogeneity ($\mu_0\Delta H_0$) for samples (a) 32Cr, (b) 32NCr, (c) 31Cr, (d) 31NCr, (e) 30Cr and (f) 25Cr with the external magnetic field applied in the hard (black) and easy (blue) directions.
Supplementary note 4. Superconducting transition temperature $T_c$ for Nb/Cr/Fe/Cr/Nb and Nb/Fe/Nb structures.

The full set of critical temperature ($T_c$) curves for each sample structure are presented in Supplementary Figure 2, measured by surface contact resistivity in Supplementary Figure 2(a), and by bulk ac magnetic susceptibility with no dc field in Supplementary Figure 2(b). In-phase susceptibility ($\chi'_{ac}$) is shown. To show that both measurements are in good agreement, they are compared in Supplementary Figure 2(c) with the resistivity temperature offset by -1 K due to a difference in thermometer calibration.

Supplementary Figure 8. Critical temperature measurements of superconducting samples (a) The temperature ($T$) dependence of the resistivity ($\rho$) for 32Cr (black) and 32NCr (red). (b) $T$ dependence of the normalised in-phase component of the ac magnetic susceptibility ($\chi'_{ac}$) for all six samples. (c) Comparison of the $T$ dependence of $\rho$ and $\chi'_{ac}$ for 32Cr. (d) $T$ dependence of $\chi'_{ac}$ with various dc fields applied in-plane along the hard axis orientation for 32Cr. The inset shows $\chi'_{ac}(T)$ for 32Cr with a 500 mT field applied in-plane along the hard (solid line) and easy (dashed line) orientations.
The ac magnetic susceptibility measurements were performed in the same QuantumDesign Physical Property Measurement System® (PPMS®) as ferromagnetic resonance (FMR) measurements, and so we take the $T_c$ value from those measurements.

To characterize the $T_c$ transition of these samples in external in-plane dc fields of typical magnitudes applied in FMR measurements, ac susceptibility was measured at various $H_{ext}$ of 0 mT, 50 mT, 100 mT, and 300 mT. These measurements are shown in Supplementary Figure 2(d) for 32Cr. The application of $H_{ext}$ up to 300 mT causes minimal change in the $T_c$ transition, only broadening in field by a maximum of 0.2 K. The $T_c$ transition is also independent of the field orientation, as shown in the inset of Supplementary Figure 2(d). In both the easy and hard axis case for each sample, $\chi'_ac$ begins to decrease at the same $T$ indicating the onset of superconductivity. From the $T$ derivative of $\chi'_ac$, $T_c$ is taken to be the value of $T$ that gives a maximum slope.

In both the easy and hard axis case, the Gilbert damping $\alpha$ is constant at temperatures where the normalised $\chi'_ac$ value is 1. Only when $\chi'_ac$ starts to decrease, indicating the onset of superconductivity, does $\alpha(T)$ increase. This provides further evidence that the superconducting Nb affects the magnetisation dynamics of the system. In the easy axis case, $\alpha(T)$ increases sharply around the same temperature that $\chi'_ac$ decreases. $\alpha(T)$ then decreases within the $T_c$ transition width. This reflects the picture of enhanced availability of quasiparticle states as the superconducting gap opens [S1], but gradually decreases with $T$ as the quasiparticle states freeze out due to the superconducting gap opening further [S1, S2]. In the hard axis case, $\alpha(T)$ begins to increase at the same temperature as $\chi'_ac$ decreases and continues to increase beyond the $T_c$ transition width. This shows that $\alpha(T)$ increases below $T_c$ and well after the opening of a superconducting gap. Therefore, it cannot be explained by the quasiparticle spin transport, but can be attributed to long-range proximity induced spin-triplets.

**Supplementary note 5. Frequency dependence of the resonance field**

The $f$ dependence of $\mu_0H_{res}$ can be fitted to the modified Kittel formula, taking into consideration the magnetic anisotropy field contribution [S3]:
\[ f^2 = \left( \frac{\gamma \mu_0}{2\pi} \right)^2 \left[ \left\{ M_{\text{eff}} + H_{\text{res}} + H_k \sin^2 (\phi - \phi_0) \right\} \times \left\{ H_{\text{res}} - H_k \cos 2(\phi - \phi_0) \right\} \right], \] (S2)

where \( \gamma \) is the gyromagnetic ratio, \( M_{\text{eff}} \) is the effective saturation magnetisation, \( \phi \) is the angle of the film’s magnetisation with respect to the easy axis direction, \( \phi_0 \) is the angle of the hard axis direction with respect to the easy axis direction, and all other symbols take their usual definitions. In the case of the superconducting state \((T < T_c)\), it may be necessary to include a superconducting shift term \( \mu_0 H_{\text{shift}}^{SC} \) [S4]:

\[ f^2 = \left( \frac{\gamma \mu_0}{2\pi} \right)^2 \left[ \left\{ M_{\text{eff}} + H_{\text{res}} + H_k \sin^2 (\phi - \phi_0) + H_{\text{shift}}^{SC} \right\} \times \left\{ H_{\text{res}} - H_k \cos 2(\phi - \phi_0) + H_{\text{shift}}^{SC} \right\} \right], \] (S3)

This term arises from the onset of superconductivity, the Meissner effect, and vortices of pinned flux contributing to internal dc magnetic fields [S4]. Flux pinning forms in type II superconductors, such as Nb, when an external magnetic field is applied and trapped as quantized flux pins within the SC layer. The magnetic field strength must be between the superconductor’s lower and upper critical field \((H_{c1} < H_{\text{ext}} < H_{c2})\). The radius of each vortex is equivalent to the London magnetic penetration depth \((\lambda_L)\). Therefore, we would not expect flux pinning to form in our samples with \( H_{\text{ext}} \) applied in-plane where the Nb layers are thinner than the penetration depth of Nb thin films \((\lambda_{\text{Nb}} > 100 \, \text{nm})\) [S5, S6]. However, we observe anomalous \( M_{\text{eff}}(T) \) behaviour that can be explained by the addition of a \( H_{\text{shift}}^{SC} \) term.
Supplementary Figure 9. Resonance field at different frequencies Microwave frequency dependence of the resonance field ($\mu_0H_{res}$) for sample 31Cr, fitted to the Kittel formula (a) without and (b) with the $\mu_0H_{shift}^{SC}$ term.

The frequency dependence of $\mu_0H_{res}$ for sample 32Cr is shown in Supplementary Figure 9, fitted without $\mu_0H_{shift}^{SC}$ in Supplementary Figure 8(a), and fitted with $\mu_0H_{shift}^{SC}$ in Supplementary Figure 9(b). The values of $\mu_0M_{eff}$ determined from Supplementary Figure 8 using Equations S2 and S3 for each sample between 16 K and 2K are summarized in Supplementary Figure 10. The extracted values of $\mu_0M_{eff}$ show an anomalous increase below $T_c$ when fitted without $\mu_0H_{shift}^{SC}$, which is corrected when $\mu_0H_{shift}^{SC}$ is considered. This is similar to [S4] and demonstrates the need for the $\mu_0H_{shift}^{SC}$ term even in our samples of considerably thinner layers of superconducting Nb, well below the London penetration depth of Nb thin films. This indicates that the applied external field is not perfectly in-plane with the sample, but that there is a small out-of-plane component which can be trapped as quantized flux.
Supplementary Figure 10. Effective magnetisation $M_{\text{eff}}$ for the four samples Effective saturation magnetization extracted from Equation S3 as a function of temperature $T$ for various samples from 16 K to 2 K for (a) 32Cr (b) 32NCr (c) 31Cr (d) 31NCr (e) 30Cr and (f) 25Cr, with (blue triangles) and without (black squares) $\mu_0 H_{\text{shift}}^{\text{SC}}$.

It has been reported [S4] that Meissner screening may lead to linewidth broadening and enhancement of the inhomogeneous broadening $\mu_0 \Delta H_0$ due to Meissner fields perturbing the magnetisation precession. However, we only observe linewidth
broadening and damping enhancement in samples containing Cr unlike in [S4]. Jeon et al., in a study where controlled vortices have been introduced [S4], observe linewidth broadening in S/F/S structures i.e. without a spatially inhomogeneous magnetic layer contrary to our observations. Furthermore, we would expect trapped flux to occur independent of field orientation. Therefore, if the rise in Gilbert damping were due to trapped flux, we would also expect to observe the rise regardless of field orientation. However, this is not the case in all four samples that exhibit damping enhancement (only in the hard orientation). Note that Jeon et al. [S4] show that the broadening enhancement is not associated with Gilbert damping enhancement.

**Supplementary note 6. S parameter**

The S parameter vs magnetic anisotropy K for the samples measured with the applied magnetic field in the easy axis orientation is shown in Supplementary Figure 11. The S parameter values are zero or below, highlighting that there is no enhancement of the Gilbert damping when the field is applied in the samples’ magnetic easy axis regardless of the presence of Cr.

![Supplementary Figure 11](image-url)  
Supplementary Figure 11. Anisotropy constant K dependence of the S parameter defined as $S = [\alpha(0.2t) - \alpha(2t)]/\alpha(2t)$ for the Nb/Cr/Fe/Cr/Nb samples (in blue squares) and Nb/Cr/Nb samples (yellow circles).
Supplementary note 7. Measuring different regions of the samples

To ensure reproducibility and homogeneity on a mm length scale, the samples are measured in FMR across different regions with the microwave strip line placed in three different locations, labelled as top, middle, and bottom in Supplementary Figure 12. $\alpha$ and $\mu_0\Delta H_0$ for each region has been extracted and examples for sample 32Cr are shown in Supplementary Figure 13. It can be seen that the temperature dependence of $\alpha$ and $\mu_0\Delta H_0$ show consistent trends within experimental errors, indicating reproducibility and homogeneity in the samples (on a mm scale). $\alpha$ presented in the main text (Fig. 3) are taken from the averages of the linewidths at different sample regions.

Supplementary Figure 12. Schematic of the three waveguide positions to investigate sample homogeneity in the mm scale.
Supplementary Figure 13. Extracted values of the Gilbert damping $\alpha$ and inhomogeneous broadening $\mu_0 \Delta H_0$ measured at different regions of the sample 32Cr. (a), (b), Temperature dependence of $\alpha$, $\mu_0 \Delta H_0$, respectively, with the applied external field aligned with the sample hard axis. (c), (d), Temperature dependence of $\alpha$, $\mu_0 \Delta H_0$, respectively, with the applied external field aligned with the sample easy axis.

Supplementary Note 8. Investigating the source of anisotropy - Nb(3)/Fe(t)/Nb(3) films

In order to further study the origin of uniaxial anisotropy in our samples, additional films of Nb(3 nm)/Fe(t)/Nb(3 nm) were prepared (3 sets of each), where $t = 6$ or $12$ nm. Using VSM characterisation on all samples and FMR on one sample of each, all 6-nm-thick Fe films showed the uniaxial magnetic anisotropy feature (with a spread of anisotropy) and all 12 nm films did not. Example room temperature M-H loops for both structures are shown in Supplementary Figure 14, as well as example f v $\mu_0 \Delta H_{\text{res}}$ shown in Supplementary Figure 15. From the frequency dependence
of \( \mu_0 \Delta H_{\text{res}} \), we fitted the modified Kittel formula (equation 1) and extract the anisotropy constant by comparing \( \mu_0 \Delta H_{\text{res}} \) for the easy and hard axes. We find that for the sample where \( d = 6 \) (12) nm, \( K = 1860 \pm 280 \approx 0 \) Jm\(^{-3}\). A summary of the anisotropy values for all measured samples is shown in Supplementary Table 1. The lack of observable anisotropy in the thicker Fe sample strongly suggests that the source of anisotropy is interfacial.

**Supplementary Figure 14.** Magnetic field loops \((M-H \text{ loops})\) with the external in-plane field applied in four orientations as indicated for samples Nb(3 nm)/Fe(\(t\))/Nb(3 nm) with (a) \( d = 6 \) nm and (b) \( t = 12 \) nm.

**Supplementary Figure 15.** Microwave frequency dependence of the resonance field for samples of Nb(3 nm)/Fe(\(d\))/Nb(3 nm) with (a) \( d = 6 \) nm and (b) \( d = 12 \) nm. Solid lines are the modified Kittel fits to the experimental data.
### Supplementary Table 1

Fe thin film samples and their magnetic property values. $K_{VSM}$ and $K_{FMR}$ are the anisotropy constant values obtained from the vibrating sample magnetometer and the ferromagnetic resonance measurements respectively. $\mu_0 H_c$, $M_r$, $M_s$ are the coercive magnetic field, the remanent magnetization and the saturation magnetization respectively. The error in $\mu_0 H_c$ is ± 0.1 mT from measurement precision. The error in $M_s$ is from propagating errors in measuring the Fe volume and saturation moment. The error in $K_{FMR}$ is from the error in the Kittel fits to the frequency dependent $\mu_0 H_{res}$.

| Sample Structure | Ref | Easy axis $\mu_0 H_c$ (mT) | Hard axis $\mu_0 H_c$ (mT) | Easy axis $M_r/M_s$ | Hard axis $M_r/M_s$ | $M_s$ ($10^3$ A/m) | $K_{VSM}$ (J/m$^3$) | $K_{FMR}$ (J/m$^3$) |
|------------------|-----|---------------------------|---------------------------|---------------------|---------------------|-------------------|-------------------|-------------------|
| Nb(3)/Fe(6)/Nb(3) | F6A | 4.9 | 3.9 | 0.83 ± 0.03 | 0.58 ± 0.01 | 1200 ± 42 | 2230 ± 380 | 1890 ± 280 |
|                  | F6B | 4.1 | 1.8 | 0.82 ± 0.03 | 0.22 ± 0.02 | 870 ± 36 | 3235 ± 489 | n/a |
|                  | F6C | 4.6 | 3.3 | 0.74 ± 0.02 | 0.39 ± 0.01 | 700 ± 33 | 2710 ± 432 | n/a |
| Nb(3)/Fe(12)/Nb(3) | F12A | 7.1 | 0.58 ± 0.01 | 1120 ± 38 | ~0 | ~0 |
|                  | F12B | 6.5 | 0.22 ± 0.02 | 940 ± 32 | ~0 | n/a |
|                  | F12C | 4.6 | 0.39 ± 0.01 | 780 ± 24 | ~0 | n/a |
Supplementary Figure 16. Atomic force microscopy (AFM) images of the Fe thin film samples. AFM image of (a) Nb(3 nm)/Fe(6 nm)/Nb(3 nm) with RMS roughness of 0.670 nm and (b) Nb(3 nm)/Fe(12 nm)/Nb(3 nm) with RMS roughness of 0.590 nm.

In-plane uniaxial anisotropy has been observed in Fe films sputtered on SiO2/Si by Komogortsevab et al. [S7]. They attribute the source of anisotropy due to a sputtering angle relative to the Si crystal plane creating structural anisotropy. The authors used atomic force microscopy (AFM) to show anisotropy in the in-plane roughness with root-mean-squared (RMS) roughness of ~3 nm. We can rule out this structural anisotropy as the source of our observed uniaxial anisotropy as our films are expected to be nanocrystalline and produced by continuous rotation of the substrate during growth. Furthermore, we perform similar (AFM) measurements on our Nb(3)/Fe(d)/Nb(3) films, shown in Supplementary Figure 16, and observe similar RMS roughness between d = 6 nm and d = 12 nm of ~0.6 nm.

From the VSM and FMR measurements, our observed uniaxial anisotropy is interfacial. It is likely then that the source of the anisotropy is due to an interface alloy formed at the Nb/Fe and Nb/Cr/Fe interfaces. It is a well-known phenomenon that the magnetic field of the magnetron sputtering source can produce partial atomic pair ordering in the deposition process for various Fe alloys [S8, S9], and that atomic pair ordering gives rise to magnetic uniaxial anisotropy.

Taking into consideration the AFM, VSM and FMR measurements on the F series films, it is highly likely that interfacial layers exist in our samples that are the cause for the observed magnetic in-plane uniaxial anisotropy. It is well known that Fe can alloy with Nb [S9], and Cr interdiffuses into Fe during the sputtering growth process [S11]. These Fe alloys grown under an external magnetic field will exhibit atomic pair ordering which leads to magnetic uniaxial anisotropy.
Supplementary note 9. Theory

Supplementary Figure 17. Theoretical damping $\alpha$ compared with experimental damping

(a) Damping extracted from the theoretical mode versus reduced temperature. The angle $\theta$ is the angle of the magnetization of the F2 layer with respect to the fixed magnetization of the F1 layer. (b) Experimental damping versus reduced temperature with the external field applied in the hard (black) and easy (blue) axis.

We can compare our findings with theoretical prediction that resembles our experimental layout. In order to do this, we model the Fe/Cr/Nb systems by a F1/F2/S trilayer with a ferromagnetic layer F1, a ferromagnetic layer F2 (which has its spin magnetic moment misaligned with respect to F1 at a fixed angle) and a superconducting layer S. We treat only the simplest model, which in contrast to the experiment assumes that F2 precesses together with F1, allowing for an efficient numerical modelling of the space-time dependence following the treatment of Houzet [S12]. We assume that the precession in the F2 layer occurs at a relative angle compared to than in the F1 layer. This angle difference can be explained by different interaction and crystalline structure in both F1 and F2 layers. In the theoretical calculation, the misalignment of spin at the Fe/Cr/Nb interface is modelled by a single unique misalignment parameter $\theta$. The theory takes into account the transparency of the interface ($t$), the strength of the exchange field ($J$) and the suppression of the superconducting gap energy at the interface ($\Delta$). It does not take into account the quasiparticle temperature dependence (so it does not capture the coherence peak feature), interface roughness, or the dependence of the spin current on momentum direction. A full description of this theoretical approach is given in [S13].
Supplementary Figure 17 sets out the theoretical prediction of $\alpha$ as a function of $\theta$ for optimum values of the parameters of $t$, $J$, $\Delta$. The Gilbert damping is normalized here by the value of the Gilbert damping above $T_c$ with $\theta = \theta_0$. In the normal state $\alpha/\alpha_{T>T_c}$ increases as $\theta$ increases, indicative of an additional loss channel when the interfacial spins are misaligned to the precessing ferromagnet F1. This scenario can be mapped across to the experimental observation of ‘easy’ and ‘hard’ magnetic directions that we observe in the FMR resonance field.

At the lowest temperatures, although the theory prediction and the experimental observation are in qualitative agreement there is a discrepancy in the magnitude of the effect by a factor of 6 in $S$ parameter defined in equation 3, $S = 0.06$ theoretically and $0.3$ experimentally. The most significant aspect that the theory does not consider is the fact that in the experiment the second ferromagnetic layer has a static magnetization, which in combination with the precessing magnetization in the first ferromagnet leads to a time-dependent Gilbert damping. The additional production of spin waves in the F2 layer likely leads to an underestimation of the Gilbert damping due to the fact that spins are pumped back from F2 to F1, a process that is absent in the experimental setup. A full calculation would require a numerical study of space-time-dependent solutions which is left for future work. Further aspects that could be improved on are taking into account a dependence of the spin current on momentum direction, and taking into account interface roughness and crystallinity. It is possible therefore that the experimental situation with static magnetization in F2, polycrystalline films, a finite degree of interfacial roughness and a range of misalignment angles optimises the strength of the spin triplet current beyond the scope of the current theoretical predictions. The parallels between theoretical and experimental behaviour are encouraging as they suggest that strong superconducting spin channels can be created under realistic experimental conditions.

**Supplementary note 10. Theory Method**

We calculate non-equilibrium spin current in a F1/F2/S trilayer where F1 is a precessing ferromagnetic, F2 is a misaligned ferromagnetic layer with misalignment angle $\theta$, and S is a superconductor of thickness $d_s$. The misalignment between the magnetizations of the F1 and F2 layers induces equal-spin Cooper pairs across the entire trilayer. We consider
a spin precession occurring in the x-y plane while the magnetization is tilted in the y-z plane. The trilayer is stacked along the x axis and we assume that the layers extend infinitely in the y-z plane. In the following, we focus on the stationary regime where both F1 and F2 layer magnetizations exhibit the time-dependency [S12]:

\[ J_{Fi} (t) = |J_{Fi}| (sin \theta_{Fi} sin \Omega t, sin \theta_{Fi} cos \Omega t, cos \theta_{Fi}) \]  

(S4)

Where i=1,2, \( J_{Fi} \) is the Fi layer exchange-field strength, and \( \Omega \) is the precession frequency. We assume that the precession frequency is the same in both Fi layers. To model non-equilibrium and non-stationary properties in diffusive superconductors, we should use time-dependent non-equilibrium Usadel equations [S12] which are derived from quasiclassical equations [S13]. The exchange field time dependency described above allows to transform non-stationary Usadel equations in the laboratory frame into stationary Usadel equations in the rotating frame [S11]. Stationary non-equilibrium Usadel equations in the rotating frame write [S11, S13]:

\[ \frac{D}{\pi} \partial_x (\tilde{G} \partial_x \tilde{G}) + \left[ E \tilde{\tau}_3 - \frac{\Omega'}{2} \sigma^z - \tilde{\Sigma}, \tilde{G} \right] = 0 \]  

(S5)

where \( D \) is the diffusion coefficient, \( \sigma^z (r_3) \) the third Pauli matrix acting on the spin (particle-hole) subspace, \( [ , ] \) is the commutator and \( \tilde{G} \) and \( \tilde{\Sigma} \) describes the normal and anomalous Green’s function (self-energies) in the Keldysh space [S12, S14].

We rotate the axis such that the F1 layer magnetization points in the z-axis direction implying a reduced non-equilibrium exchange field \( \Omega \rightarrow \Omega' = \Omega cos (\theta_{Fi}) \) [S12]. For notational simplicity, we do not explicitly state identity matrices. For inner interfaces, we assume (current conserving) Nazarov boundary conditions [S13]:
\[ \sigma_l \tilde{G}_l \partial_x \tilde{G}_l = \sigma_r \tilde{G}_r \partial_x \tilde{G}_r \]

\[ \sigma_l \tilde{G}_l \partial_x \tilde{G}_l = \frac{1}{SR_b} \frac{2\pi^2 \tau \{ \tilde{G}_l, \tilde{G}_r \}}{4\pi^2 - \tau (\{ \tilde{G}_l, \tilde{G}_r \}) + 2\pi^2} \]  

(S6)

Where \( G^{l(r)} \) is the Green’s function on the left(right) side of the interface, \( \sigma \) is the normal state electrical conductivity, \( S \) is the area of the junction, \( R_b \) is the interface resistivity and \( 0 < \tau < 1 \) the interface transparency [49]. For the outer F1 interface, we impose the Green’s function to be the one for a bulk ferromagnetic material with a spin-resolved non-equilibrium distribution function \( f_{1(1)} = f_{FD}(E + (-\frac{\alpha}{2}) \Omega) \) with \( f_{FD} \) the Fermi Dirac distribution. At the outer S interface \((x=L)\), we impose a vanishing-current boundary condition \( \partial_x \tilde{G} \big|_{x=L} = 0 \) with \( L \) the trilayer thickness.

For the spatially varying superconducting order parameter we solve the self-consistency equation

\[ \Delta(x) = \frac{\int_{-c}^{c} \frac{dE}{4\pi} f_s^K(E, x)}{\int_{-c}^{c} \frac{dE}{2E} \tanh \left( \frac{E}{2T} \right) + \ln \left( \frac{T}{T_c} \right)} \]  

(S7)

where \( f_s^K \) is the singlet part of the Keldysh anomalous Green function [S12, S14].

Green’s functions are calculated by solving numerically the Usadel equations together with the boundary conditions and the superconducting self-consistency equation. The Green’s functions are then used to calculate spin currents. Spin currents is obtained from the relation [S12]:

\[ I_s = I_s^0 \int_{-\infty}^{+\infty} dE \text{Tr} \left[ \hat{e}_3 \sigma (\tilde{G} \partial_x \tilde{G})^K \right] \]

(S8)
with $I_s^0 = \frac{\hbar e N_0 D}{16 \pi^2}$, $N_0$ the density of state at the Fermi energy, $e$ the electrical charge, $\hbar$ the reduced Planck constant, and $\sigma=(\sigma^x, \sigma^y, \sigma^z)$ the spin Pauli matrix vector. The spin current vector is given by $I_s = (I_s^x, I_s^y, I_s^z)$ in terms of the spin Pauli matrix basis.

We solve the Usadel equation for a precessing F1 and F2 layers and we calculate the spin current $I_s^Z$ for all temperatures. Then, we calculate the Gilbert damping $\alpha_t$. The Gilbert damping $\alpha_t$ can be expressed in the form $\alpha_t = \alpha_0 + \alpha = \alpha_0 + \beta I_s^Z$ where $\alpha_0$ describes an intrinsic Gilbert damping independent of temperature and superconducting properties, and $\alpha$ describes the additional Gilbert damping due to spin injection; the latter is proportional to the dissipative part of the spin current [S15], which in itself is proportional to $I_s^Z$ in the F1 layer with a coefficient depending on the tip angle. The quantity $\alpha/\alpha_t = \alpha_0 = I_s^Z/I_s^Z$ is independent of the tip-angle dependent quantity $\beta$ and therefore quantifies the additional Gilbert damping in our system.

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