Giant Transition-State Quasiparticle Spin-Hall Effect in an Exchange-Spin-Split Superconductor Detected by Nonlocal Magnon Spin Transport

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ABSTRACT: Although recent experiments and theories have shown a variety of exotic transport properties of nonequilibrium quasiparticles (QPs) in superconductor (SC)-based devices with either Zeeman or exchange spin-splitting, how a QP interplays with magnon spin currents remains elusive. Here, using nonlocal magnon spin-transport devices where a singlet SC (Nb) on top of a ferrimagnetic insulator (Y3Fe5O12) serves as a magnon spin detector, we demonstrate that the conversion efficiency of magnon spin to QP charge via inverse spin-Hall effect (iSHE) in such an exchange-spin-split SC can be greatly enhanced by up to 3 orders of magnitude compared with that in the normal state, particularly when its interface superconducting gap matches the magnon spin accumulation. Through systematic measurements by varying the current density and SC thickness, we identify that superconducting coherence peaks and exchange spin-splitting of the QP density-of-states, yielding a larger spin excitation while retaining a modest QP charge-imbalance relaxation, are responsible for the giant QP iSHE. The latter exchange-field-modified QP relaxation is experimentally proved by spatially resolved measurements with varying the separation of electrical contacts on the spin-split Nb.

KEYWORDS: nonlocal magnon spin transport, exchange-spin-split superconductor, quasiparticle spin-Hall effect, resonant absorption of magnon spin, exchange-field-frozen QP relaxation

Over the past decade, it has been shown that the combination of superconductivity with spintronics leads to a variety of phenomena that do not exist separately.1−8 In particular, recent discovery and progress in the proximity generation and control of spin-polarized triplet Cooper pairs1−3 at carefully engineered superconductor (SC)/ferromagnet (FM) interfaces in equilibrium allow for the development of nondissipative spin-based logic and memory technologies.

Besides triplet Cooper pairs, nonequilibrium quasiparticles (QPs) in a spin-split SC4−6 have also raised considerable interest. This is because their exotic properties resulting from the mutual coupling between different nonequilibrium imbalances of spin, charge, heat, and spin-heat can greatly enhance spintronics functionality.5 For example, the coupling of spin and heat imbalances gives rise to long-range QP spin signals as observed in Al-based nonlocal spin valves8−10 with a Zeeman spin-splitting field. In addition, a temperature gradient between a normal metal (NM) and a spin-split SC separated by a tunnel barrier induces a pure QP spin current12 without an accompanying net charge current, analogous to the spin-dependent Seebeck tunneling.13,14 Substituting the NM by a FM, one can achieve large (spin-dependent) thermoelectric currents15,16 far beyond those commonly found in all-metallic structures.

Magnon spintronics17−19 has been an emerging approach toward computing devices in which magnons, the quanta of spin waves, are used to carry, transport, and process spin information instead of conduction electrons. Especially in the low-damping ferrimagnetic insulator yttrium–iron–garnet...
A magnon-carried spin current can propagate over extremely long distances (centimeters at best), and it is free from ohmic dissipation due to the absence of electrons in motion. However, the conversion efficiency changes dramatically when turning superconducting due to the development of quasiparticle (QP) density-of-states with exchange spin-splitting $\Delta E_{ex}$. Note that in contrast to spin-singlet ($S = 0$) Cooper pairs in a coherent ground state, the excited QPs can carry spin angular momentum in the superconducting state. (b) Optical micrographs of the fabricated devices with and without a 10-nm-thick Al$_2$O$_3$ spin-blocking layer. (c) In-plane (IP) magnetization hysteresis $m(H)$ curves of a bare YIG film, measured at a temperature $T$ of 2−300 K. The inset summarizes the $T$ dependence of the saturation magnetic moment. (d) IP magnetic-field-angle $\alpha$ dependence of nonlocal total voltages $\Delta V_{nl}$ measured with the Pt detector at $I_{dc} = \pm 1.0$ mA at 300 K, for the $t_{Nb} = 15$ nm device. From these, electrically ($\Delta V_{nl,el}$) and thermally ($\Delta V_{nl,th}$) driven magnon components are separated (see main text). Black solid lines in e and f correspond respectively to $\sin^2(\alpha)$ and $\sin(\alpha)$ fits. The estimated magnitude of $\Delta V_{nl,el}$ ($\Delta V_{nl,th}$) is plotted as a function of $|I_{dc}|$ in the inset of e (f), where the black solid line represents a linear fit (quadratic fit). (g–i) Data equivalent to d–f but for the control device with the Al$_2$O$_3$ spin-blocking layer.
RESULTS AND DISCUSSION

The nonlocal magnon spin-transport devices (Figure 1b) we study consist of two identical Pt electrodes and a central Nb layer on top of 200-nm-thick YIG films, which are liquid-phase epitaxially grown on a (111)-oriented single-crystalline gadolinium gallium garnet (Gd$_3$Ga$_5$O$_{12}$; GGG) wafer (see Methods). Control devices, in which a 10-nm-thick Al$_2$O$_3$ spin-blocking layer is inserted between Nb and YIG in an otherwise identical structure, are also prepared for comparison (Figure 1b). Here, we send a dc current $I_{dc}$ through one Pt (using leads 1 and 2 in Figure 1b) while measuring the in-plane (IP) magnetic-field angle $\alpha$ dependence of the nonlocal open-circuit voltages $V_{nl}^{\text{el}}(\alpha)$, $V_{nl}^{\text{th}}(\alpha)$ using both the other Pt (leads 7 and 8) and the central Nb (leads 3 and 4). Note that we apply an external in-plane magnetic field $\mu_0 H_{\text{ext}}$ of 5 mT, larger than the coercive field of YIG (Figure 1c), to fully align its magnetization $M_{\text{YIG}}$ along the field direction. $\alpha$ is defined as the relative angle of $\mu_0 H_{\text{ext}}$ to the long axis of two Pt electrodes, which are collinear.

As schematically illustrated in Figure 1a, the right Pt acts as a NM injector of magnon spin current across the Pt/YIG interface via either electron-mediated SHE (charge-to-spin conversion) or spin Seebeck effect (SSE) (heat-to-spin conversion) due to the accompanying Joule heating $[\Delta T \propto I_{dc}^2]$. The left Pt serves as an NM detector of the magnon spin current, diffusing through a YIG channel, via electron-mediated iSHE (spin-to-charge conversion) or spin Seebeck effect, whereas in the same device, the middle Nb functions as an exchange-spin-split SC detector of the diffusive magnon current via QF-mediated iSHE below $T_c$.

The total voltage measured across the detector is given by $V_{nl} = \Delta V_{\text{el}} + \Delta V_{\text{th}} + V_{\text{ext}}$, where $\Delta V_{\text{el}}$ and $\Delta V_{\text{th}}$ are proportional to the magnon spin current and accumulation created electrically (SHE $\propto I_{dc}$) and thermally (SSE $\propto I_{dc}^2$), respectively. These electrical and thermal magnon currents can be separated straightforwardly by reversing the polarity of $I_{dc}$, allowing us to determine the magnitude of each component based on their distinctive angular dependences. The relative angle of $\mu_0 H_{\text{ext}}$ to the long axis of two Pt electrodes, which are collinear.

![Figure 2. Temperature dependence of nonlocal signals measured by the Pt detector. (a) Electrically driven nonlocal voltages $[\Delta V_{nl}^{\text{el}}(\alpha)]^\text{Pt}$ as a function of IP field angle $\alpha$ for the $T_{\text{base}} = 300$ K devices with and without the Al$_2$O$_3$ layer, taken at various base temperatures $T_{\text{base}}$. The black solid lines are $\sin^2(\alpha)$ fits. (b) Data equivalent to a but for thermally driven nonlocal voltages $[\Delta V_{nl}^{\text{th}}(\alpha)]^\text{Pt}$, along with $\sin(\alpha)$ fits (black solid lines). In these measurements, $I_{dc}$ is fixed at 0.5 mA and the magnetic field $\mu_0 H_{\text{ext}}$ at 5 mT. (c) Nb resistance $R_{\text{Nb}}$ versus $T_{\text{base}}$ plots for the Al$_2$O$_3$-absent and Al$_2$O$_3$-present devices, measured using a four-terminal current–voltage method (using leads 3, 4, 5, 6 in Figure 1b) while applying $I_{dc} = 0.5$ mA to the Pt injector. A strong suppression of the superconducting transition temperature $T_c$ of the Nb of the Al$_2$O$_3$-absent device. Extracted magnitudes of $[\Delta V_{nl}^{\text{el}}]^{\text{Pt}}$ (d) and $[\Delta V_{nl}^{\text{th}}]^{\text{Pt}}$ (e) as a function of $T_{\text{base}}$ for the Al$_2$O$_3$-absent and Al$_2$O$_3$-present devices. In the inset of e, $\Delta V_{nl}^{\text{el}}(T_{\text{base}})$ [$\Delta V_{nl}^{\text{th}}(T_{\text{base}})$] with $\Delta V_{nl}^{\text{el}}(T_{\text{base}})$ [$\Delta V_{nl}^{\text{th}}(T_{\text{base}})$] is also shown. (f) $[\Delta V_{n}^{\text{el}}]^{\text{Pt}}$, $[\Delta V_{n}^{\text{th}}]^{\text{Pt}}$ with $\Delta V_{nl}^{\text{el}}$ as a function of $T_{\text{base}}$ and $T_{\text{base}}/T_c$ (inset).](https://dx.doi.org/10.1021/acsnano.0c07187)
Figure 3. Giant enhancement of nonlocal signals in the transition state of the Nb detector. (a) Thermally driven nonlocal voltages $\Delta V_{nl}^{\text{th}}(\alpha)$ as a function of IP field angle $\alpha$ for the $t_{\text{Nb}} = 15$ nm devices with and without the Al$_2$O$_3$ layer, taken at $I_{\text{dc}} = 0.5$ mA around the $T_c$ of the Nb. The black solid lines are $\sin(\alpha)$ fits. (b and c) Data equivalent to a but at $I_{\text{dc}} = 0.10$ mA (b) and $I_{\text{dc}} = 0.60$ mA (c), respectively, for the Al$_2$O$_3$-absent device. (d) Normalized Nb resistance $R_{\text{Nb}}/R_{\text{FMI}}^{20K}$ versus $T_{\text{base}}$ for the Al$_2$O$_3$-absent device, measured using a four-terminal current--voltage method (using leads 3, 4, 5, 6 in Figure 1b) with varying $T_{\text{base}}$ and it almost vanishes for $T_{\text{base}} \approx 10$ K, whereas $\Delta V_{nl}^{\text{th}}$ increases at low $T_{\text{base}}$. Such distinct $T_{\text{base}}$-dependencies are in line with previous experiments$^{23,24}$ and theoretical considerations$^{25,26}$ that the injection mechanisms for electrical and thermal magnons across the Pt/YIG interface (parametrized by the effective spin conductance and the interface spin Seebeck coefficient, respectively) differ fundamentally. Furthermore, the energy-dependent magnon diffusion and relaxation of the YIG channel may play a role in the transport process.

We below focus on the nonlocal signal from the thermally generated magnons ($\Delta V_{nl}^{\text{th}}$) since it remains sufficiently large at low $T_{\text{base}}$ for allowing a reliable analysis across $T_c$. In Figure 2f, we first plot the $T_{\text{base}}$ dependence of $\Delta V_{nl}^{\text{th}}$ without the Al$_2$O$_3$ layer normalized by that with the Al$_2$O$_3$ layer; $\Delta V_{nl}^{\text{th}}$ $\text{Pt,} \text{no Al}_2\text{O}_3$/$\Delta V_{nl}^{\text{th}}$ $\text{Pt,with Al}_2\text{O}_3$. This value reflects how much the magnon spin current is absorbed by the Nb layer. Notably, $\Delta V_{nl}^{\text{th}}$ $\text{Pt,} \text{no Al}_2\text{O}_3$/$\Delta V_{nl}^{\text{th}}$ $\text{Pt,with Al}_2\text{O}_3$ drops abruptly right below $T_c$ (extracted from the Nb resistance $R_{\text{Nb}}$ versus $T_{\text{base}}$ plot of Figure 2c), and then it rises progressively as the Nb enters deep into the superconducting state, resulting in a downturn at $T_{\text{base}}/T_c \approx 0.95$ (inset of Figure 2f). Such a nontrivial behavior is compatible with recent theoretical predictions$^{29,30}$ and experimental reports$^{31,32}$ on ferromagnetic insulator (FMI)/SC structures, where (spin-singlet) Cooper pairs from the SC cannot leak into the FMI even if the
Figure 4. Nb thickness dependence of the giant transition-state enhancement. Representative nonlocal signals \([\Delta V_{nl}^{Nb}(\alpha)]\) as a function of IP field angle \(\alpha\) for the Al2O3-absent devices with different \(t_{Nb}\) of 10 (a and b), 20 (c and d), and 35 nm (e and f), taken above (yellow background) and immediately below (blue background) \(T_c\) of the Nb layer. \([\Delta V_{nl}^{Nb}]/[\Delta V_{nl}^{Pt,Al2O3}]\) versus \(T_{base}/T_c\) plots for \(t_{Nb}\) = 10 nm (g), \(t_{Nb}\) = 20 nm (h), and \(t_{Nb}\) = 35 nm (i). In the insets of g–i, the associated \(R^{Nb}/R^{Pt,Al2O3}\) and \([\Delta V_{nl}^{Nb}]/[\Delta V_{nl}^{Pt,Al2O3}]\) are plotted as a function of \(T_{base}/T_c\) (j) \(t_{Nb}\)-dependent peak amplitude, width (inset), and position (inset).
reach up to 3 orders of magnitude at the smallest $t_{th} = 0.11\, \text{mA}$ ($J_{dc} = 0.61\, \text{MA/cm}^2$). With increasing $t_{th}$, its peak amplitude decays rapidly, the full-width-at-half-maximum (fwhm) broadens, and the peak is positioned farther away from $T_c$ (inset of Figure 3f). These results ensure that the depressed superconductivity with increasing the heating power has a negative effect on the transition-state enhancement of the QP iSHE.

We perform similar measurements on an additional set of devices with different $t_{Nb}$ (Figure 4a–f), comparable to or smaller than the superconducting coherence length $\xi_{SC}$ and thereby strong $t_{Nb}$-dependent superconducting properties (e.g., QP band structure and DOS). Since thin Nb films usually contain a larger amount of grain boundaries, defects, and disorders from the structural inhomogeneity near the growth interface than thick bulk Nb,42,43 the associated scattering effectively weakens electron–electron and electron–phonon interactions and therefore the smearing-out effect of the QP DOS around the gap edge.44 One would predict a greater enhancement of the QP iSHE if the Nb detector is thicker.

However, experiments give a very different result (Figure 4g–i). As $t_{Nb}$ increases, the peak amplitude of $[\Delta \rho_{qp}]_{t_{Nb}}$/$[\Delta \rho_{qp}]_{t_{th}=0}$ rises reaching 15 nm and then drops strongly for thicker Nb detectors, leading to a maximum at $t_{Nb} = 15\, \text{nm}$ (Figure 4i). The width and position of the transition-state enhancement, on the other hand, behave as expected for highly and quickly developed coherence peaks in the QP DOS of thick Nb when $T_c$ is crossed: a progressive narrowing of fwhm and a peak shift closer to $T_c$, respectively, with the increase of $t_{Nb}$ (inset of Figure 4f). The nontrivial $t_{th}$-dependent enhancement (Figure 4j) indicates that there is another key ingredient that controls the enhancement amplitude, that is to say, the exchange spin-splitting field,4,6 which has turned out to considerably modify the QP spin relaxation mechanism via a freezing out of elastic/intravalley spin-flip scattering.4,6 Below, we discuss how this exchange-field-frozen spin-flip scattering4,6 is linked to and modifies the QP charge relaxation.

To theoretically describe our results, we first calculate the excited QP spin current density $j_{SP}$ at the YIG/superconducting Nb interface as a function of the normalized temperature $T/T_c$ for different values of the magnon spin accumulation $\Delta \mu_{\parallel}$ relative to the zero-$T$ energy gap $2\Delta_0^{\parallel}$. (Figure 5a and b). For this calculation, we employ the recent models29,30 that explicitly take the superconducting coherence factor into account (see Supplementary Section 4 for full details). Note that the characteristic energy of incoherent magnons which excite spin-polarized QPs in the Nb detector is...
set by $\Delta \mu_m$ and the $T_c$ (or $2\Delta_0^{SC}$) suppression at a larger $\Delta \mu_m$ is inferred from our data set (Figures 3 and 4). For a quantitative comparison, $J_{s0}$qp is normalized to its normal state value $J_{s0}$. The calculated $J_{s0}$qp/$J_{s0}$ increases largely near $T_c (0.8T_c - 0.9T_c)$, and it decreases exponentially when $T < 0.8T_c$, reflecting the singularity behavior in a nonequilibrium population of spin-polarized QPs. In addition, the peak amplitude of $J_{s0}$qp/$J_{s0}$ is inversely proportional to $\Delta \mu_m/2\Delta_0^{SC}$ (inset of Figure 3a and b), explaining qualitatively the heating power dependence of the transition-state enhancement (Figure 3f). Nonetheless, this analysis based on the superconducting coherence factor does not capture the mechanism behind the nontrivial $t_{Nb}$ dependence (Figure 4). We next consider the QP resistivity $\rho_{SC}^{qp}$ (Figure 5c and d) and the volume fraction of QP charge imbalance $v_Q$ (Figure 5e and f), which together determine the effective resistivity $\rho_{SC}^{*} (=\rho_{SC}^{QP}v_Q$, inset of Figure 5e and f) of the superconducting Nb. Here $v_Q = \left(2T_0/l_i\right)\tanh\left(l_i/2T_0\right)$, where $l_i$ is the spin-active length of the Nb detector, given approximately by the sum of the length of the Pt injector $l_{Pt}$ and $l_{sd}$ in our device geometry, and $\lambda_Q$ is the QP charge-imbalance relaxation length. $\rho_{SC}^{QP}$ and $\rho_{SC}^{*}$ are normalized by their normal-state ones $\rho_0$ and $\rho_0^*$, respectively. We note that if the SC thickness is comparable to or smaller than the QP spin transport length, as relevant to our system, the QP-mediated iSHE voltage $V_{iSHE}^{QP}$ in the SC can be approximated as...
The most salient aspect of the calculations is that in the vicinity of $T_\text{c}$, $\tau_{q}\text{fi}$ dominates the $T$-dependent $\rho_{\text{SC}}$ over $\rho_{\text{QP}}$, resulting in $V_{\text{SH}}^{\text{QP}} \propto \lambda_{q} \rho_{\text{SC}}$ for given $f_{\text{fi}}$ and $l_{q}$ values. This signifies that the QP charge imbalance relaxation is likely responsible for the nontrivial $T_{\text{f}}$-dependent transition-state enhancement (Figure 4j) observed in our system.

We thus propose the following mechanism. If QP charge relaxes through the spin-flip scattering $1/\tau_{q}\text{fl}$ and the inelastic scattering $1/\tau_{\text{in}}$ and $1/\tau_{q}\text{fl} > 1/\tau_{\text{in}}$, the effective relaxation time $\tau_{q}\text{fi}$ for the QP charge imbalance \cite{foot25} is given by $\tau_{q}\text{fi}^* = \frac{4k_{B}T}{\pi \xi^{2}} \cdot \frac{\varepsilon_{0}}{\varepsilon_{N}^{0}}$, where $k_{B}$ is the Boltzmann constant. Based on the exchange-field-frozen spin-flip scattering\cite{foot20} and its proximity nature\cite{foot22} in an FMI/SC system, one can reasonably assume $\tau_{q}\text{fl} \propto \Delta_{q}\text{fi} \propto \frac{1}{l_{q}}$. This leads to $\lambda_{q} \propto \frac{\varepsilon_{0}}{l_{q}}$ and $V_{\text{SH}}^{\text{QP}} \propto \frac{\rho_{\text{SC}}}{l_{q}^{4}}$. Qualitatively, we can understand the $T_{\text{f}}$-dependent transition-state enhancement (Figure 4j) in the following manner. When $T_{\text{f}} \ll \xi_{N}^{0}$, the superconducting coherence is too weak to inject/excite large QP spin currents across the YIG/Nb interface. In contrast, for $T_{\text{f}} > \xi_{N}^{0}$, the exchange spin-splitting-field cannot propagate over the entire depth of such thick Nb and hence the converted QP charge relaxes faster primarily via the spin-flip scattering process. Overall, these two competing effects control the amplitude of the transition-state enhancement by which one would expect a maximum at the intermediate $T_{\text{f}}^{*} \approx \xi_{N}^{0}$ (around 15 nm for Nb thin films). Note also that the enhancement width and position are determined by $f_{\text{fi}} \propto \rho_{\text{SC}}$.

To check the validity of this proposal, we experimentally investigate how the transition-state enhancement scales with the separation distance $d_{\text{f}}$ between Au/ Ru electrical contacts on the exchange-spin-split Nb layer (Figure 6a, see Supplementary Section S). Importantly, while the peak position and width of the transition-state enhancement are almost independent of $d_{\text{f}}$ (Figure 6b and c), the peak amplitude increases quasi-exponentially with the increase of $d_{\text{f}}$ (inset of Figure 6c), reflecting the characteristics of the QP charge-imbalance relaxation effect (see Supplementary Section S). From the $d_{\text{f}}$-dependent $[\Delta V_{\text{SH}}^{\text{QP}}]^{\text{NB}}$ (Figure 6e), we are able to estimate $\lambda_{q}$ in the vicinity of $T_{\text{c}}$, ($T_{\text{base}}/T_{\text{c}} = 0.94 – 0.98$) for the spin-split Nb to be around 90 μm. This is surprisingly a few orders of magnitude larger than either commonly assumed\cite{foot25} or hitherto reported in Nb films without the presence of spin-splitting fields\cite{foot26} and thereby should indicate the significantly exchange-field-modified QP relaxation in our system.

Finally, we briefly mention other relevant experiments. It has been previously shown that in all-metallic nonlocal spin-Hall devices,\cite{foot27} the giant iSHE (∼2000 times at most) is created by electrical spin injection from Ni$_{2}$Fe$_{2}$ through Cu into superconducting Nb/Nb far below $T_{\text{c}}$, ($T_{\text{base}}/T_{\text{c}} = 0.3$) and attributed to the exponentially increasing QP resistivity at a lower T. By contrast, a recent experiment has reported that for a YIG/NbN vertical junction\cite{foot28} the 2–3 times enhanced iSHE voltage by local SSE is measurable only in a limited T range right below $T_{\text{c}}$ ($T_{\text{base}}/T_{\text{c}} = 0.96$). In this work, the superconducting coherence factor is pointed out as a main source for such an enhancement, and a quantitative description of the data is also provided. In metallic/conducting Nb/Ni$_{2}$Fe$_{2}$ bilayers,\cite{foot29} a monotonic decay of spin-pumping-induced iSHE appears across $T_{\text{c}}$ indicating no superconducting coherence effect detectable.

**CONCLUSIONS**

The key findings of our study that help understand these puzzling results are as follows. The spin-to-charge conversion mediated by QPs is substantially enhanced in the normal-to-superconducting transition regime, where the interface superconducting gap matches the magnon spin accumulation. The conversion efficiency and characteristics depend crucially on the driving/heating power and the SC thickness, which is understood based on the two competing effects: the superconducting coherence\cite{foot30} and the exchange-field-modified QP relaxation.\cite{foot31} The validity of these competing mechanisms is experimentally confirmed by spatially resolved measurements with varying the separation of electrical contacts on the spin-split Nb layer. A quantitative reproduction of the results remains an open question for a theory. The coupling between different nonequilibrium imbalances (magnon, spin, charge, heat, magnon-heat, and spin-heat)\cite{foot32} with exchange spin-splitting and the nonlinear kinetic equations\cite{foot33} in the superconducting state should be taken into account rigorously. Moreover, how the magnetic-field-induced screening currents in a spin-split SC contribute to the QP spin-to-charge conversion when coupled with these nonequilibrium modes remains to be addressed. We speculate that the giant transition-state QP SHE is generic in any FMI/SC system, and its efficiency gets even larger especially with two-dimensional (2D) SCs\cite{foot34} where the exchange spin-splitting can readily proximity-penetrate the entire depth of the 2D SCs. We also anticipate that such a giant spin-to-charge conversion phenomenon (involving nonequilibrium QPs) can be used as an extremely sensitive probe of spin currents in emergent quantum materials.\cite{foot35}

**METHODS**

**Device Fabrication.** We fabricated the magnon spin-transport devices (Figure 1b) based on 200-nm-thick single-crystalline YIG films (from Matesy GmbH) by repeating a sequence of optical lithography, deposition, and lift-off steps. Note that these YIG films exhibited a very low Gilbert damping of 0.6 × 10$^{-4}$ at room temperature, determined *via* ferromagnetic resonance line width measurements (by Matesy GmbH, https://www.matesy.de/en/products/materials/yig-single-crystal). We first defined the central Nb detector with a lateral dimension of 9 × 90 μm$^{2}$, which was grown by accelerated Ar-ion beam sputtering at a working pressure of 1.5 × 10$^{-4}$ mbar. For the control device, a 10-nm-thick Al$_{2}$O$_{3}$ spin-blocking layer was *in situ* deposited prior to the Nb deposition. We then defined a pair of Pt electrodes of 1.5 × 50 μm$^{2}$, which were deposited by dc magnetron plasma sputtering at an Ar pressure of 4 × 10$^{-3}$ mbar. These Pt electrodes are separated by a *center-to-center* distance $d_{\text{Pt}}$ of 15 μm, which is comparable to the typical $t_{\text{SC}}$ of single-crystalline YIG films\cite{foot36} and also to the estimated values from our Pt-only reference devices with different $d_{\text{Pt}}$.\cite{foot37} (Supplementary Section 1). The Nb thickness ranges from 10 to 35 nm, whereas the Pt thickness is fixed at 10 nm. Finally, we defined Au(80 nm)/Ru(2 nm) electrical leads and bonding pads, which were deposited by the Ar-ion beam sputtering. Before depositing the Au/Ru layers, the Nb and Pt surfaces were gently Ar-ion beam etched for transparent electrical contacts between them.
Nonlocal Measurement. We measured the nonlocal magnon spin-transport (Figure 1a and b) in a quantum design physical property measurement system at a temperature varying between 2 and 300 K. A dc current \( I \) in the range of 0.1 to 1 mA was applied to the first Pt using a Keithley 6221 current source, and the nonlocal voltages \( V_0^a(a), V_0^b(a) \) across the second Pt and the central Nb are simultaneously recorded as a function of in-plane magnetic-field-angle \( \alpha \) by a Keithley 2182A nanovoltmeter. \( \alpha \) is defined as the relative angle of \( \mu_b H_{ext} (\langle /M_{S0} \rangle) \) to the long axis of two Pt electrodes that are collinear.

ASSOCIATED CONTENT

4 Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nano.0c07187.

Estimation of the magnon spin-diffusion length of YIG, quantification of spin currents leaking into the central Nb at room temperature, first-order estimate of the YIG-induced internal field at the Nb/YIG interface, theoretical description of the conversion efficiency of magnon spin to QP charge in the superconducting Nb, spatially resolved measurements by varying the separation of electrical contacts on the spin-split Nb layer (PDF).

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Author Contributions
K.-R.J. conceived and designed the experiments. The magnon spin-transport devices were fabricated by K.-R.J. with help from J.-C.J., X.Z., and A.M. The nonlocal transport measurements were carried out by K.-R.J. with the help of J.Y. and J.-C.J. K.-R.J. performed the data analysis and model calculation. S.P.P.P. supervised the project. All authors discussed the results and commented on the manuscript, which was written by K.-R.J.

Notes
The authors declare no competing financial interest.

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