Spin-polarized quasiparticle control in a double spin-filter tunnel junction

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Spin-polarized quasiparticles can be easily created during spin-filtering through a ferromagnetic insulator (FI) in contact with a superconductor due to pair breaking effects at the interface. A combination FI-N-FI sandwiched between two superconductors can be used to create and analyze such spin-polarized quasiparticles through their nonequilibrium accumulation in the middle metallic (N) layer. We report spin-polarized quasiparticle regulation in a double spin-filter tunnel junction in the configuration NbN-GdN1-Ti-GdN2-NbN. The middle Ti layer provides magnetic decoupling between two ferromagnetic GdN and a place for nonequilibrium quasiparticle accumulation. The two GdN(1,2) layers were deposited under different conditions to introduce coercive contrast. The quasiparticle tunneling spectra has been measured at different temperatures to understand the tunneling mechanism in these double spin-filter junctions. The conductance spectra were found to be comparable to an asymmetric SINIS-type tunnel junction. A hysteretic R-H loop with higher resistance for the antiparallel configuration compared to parallel state was observed asserting the spin-polarized nature of quasiparticles. The hysteresis in the R-H loop was found to disappear for sub-gap bias current. This difference can be understood by considering suppression of the interlayer coupling due to spin-polarized quasiparticle accumulation in the Ti layer.

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I. INTRODUCTION

In superconductors, below the critical temperature $T_C$ the electrons with opposite momentum and spin are bound in (singlet) Cooper pairs, therefore, they can transport only charge but not spin. At finite temperature a fraction of Cooper pairs is broken into excited states called (Bogoliubov) quasiparticles which is capable of transporting both charge and spin. Quasiparticles can be created inside a superconductor while injecting current through a tunnel barrier or by irradiating electromagnetic radiation with energy, $h\nu \gg \Delta$, where $\nu$ is the frequency of radiation and $\Delta$ is the superconducting energy gap (binding energy of Cooper pairs) [1]. Eventually with time the quasiparticles recombine to from Cooper pairs after emitting a phonon maintaining equilibrium. In the presence of extra disturbances quasiparticle concentration can be increased and driven out of equilibrium which follow non-Fermi Dirac distribution function. The number and dynamics of these nonequilibrium quasiparticles has been the subject of intense research lately as they are primary source of decoherence in almost all superconducting electronics [2]. However, these nonequilibrium quasiparticles can be very advantageous for spintronics purposes as they have very large mean free path ($\lambda_Q$) compared to ordinary electrons [3].

Quasiparticle spintronics is not new and has been there since 1970s. Meservay and Tedrow have shown that spin polarization of various ferromagnets can be detected by injecting spin-polarized quasiparticles from a ferromagnet into a superconductor [3]. Recently, quasiparticle spintronics have got renewed interest and most of the study has been focused on spin transport inside superconductors through quasiparticle excitations [4-10]. It is now believed that spin and charge are transported by separate quasiparticle excitations in a superconductor [4-17]. Signatures of spin transport over distances up to several $\mu m$ has been observed in Zeeman split superconductors in proximity to a ferromagnetic insulator [11-15]. Many spintronics phenomena like quasiparticle mediated spin Hall effect (SHE) [16], Seebeck effect induced by spin-polarized quasiparticles [19], quasiparticle spin resonance [20], etc., has been experimentally observed. However, many fundamental aspects of the Quasiparticle spintronics remains poorly understood. Most interesting prospect of quasiparticle spintronics would be to explore possibility to take quasiparticles out of superconductor into a normal metal and introduce spintronics functionality. One obvious system for this type of study is double spin-filter device of the type S-FI-N-FI-S (Here FI is ferromagnetic insulator, N is normal metal and S is the superconductor) which is analogous to conventional SINIS-type devices [8-21]. Double barrier superconducting tunnel junctions of the S-I-N(s)-I-S structure have been extensively studied to cool down the electron in the normal metal (N) from 300 to 100 mK or to enhance superconductivity in the middle $s$ layer [22-24]. Operation of these devices are based on the modification of the quasiparticle distribution function in the N region of the junction which can have non-Fermi Dirac form leading to measurable out come. Blamire et. al. have observed enhancement in the superconductivity of Al up to 4 K in a symmetric Nb-AlO$_x$-Al-AlO$_x$-Nb double barrier junction [25]. Enhancing superconductivity by means of nonequilibrium effects got substantial theoretical interest.
Spin-filter tunnel junction comprising superconductors produces a great amount of spin-polarized quasiparticles by enforcing Cooper pairs to split while tunneling. Therefore double spin-filter devices of the type S-FI-N-FI-S provide unique opportunity to explore quasiparticle spintronics through nonequilibrium quasiparticle accumulation in the middle N layer. In this kind of devices when the two spin-filter layers are parallel to each other no spin accumulation happens as the number of injected spin-up electrons in the N layer is same as the number of spin-up electrons leaving it. Whereas in the antiparallel case finite nonequilibrium spin accumulation in the middle layer is expected which relaxes through spin-flip processes. Double spin-filter device with superconductivity-induced nonequilibrium has been predicted to show huge TMR \( \sim 10^2\)-% which can be tuned with biasing voltage and temperature.

In this paper, we report fabrication of double spin-filter devices in which a metallic Ti layer is symmetrically connected to two identical superconductors through ferromagnetic (GdN) tunnel barriers. We present quasiparticle tunneling spectra measurements on the NbN-GdN-NbN, NbN-Ti-GdN-NbN and NbN-GdN1-Ti-GdN2-NbN tunnel junctions measured at different temperatures. We explore possibility of creating nonequilibrium quasiparticle accumulation in the Ti layer and its effect on the magnetic coupling between the two GdN layers. The R-H loops of the double spin-filter tunnel junctions were measured at different bias currents and temperature to explore these effects.

II. EXPERIMENTAL

Multilayer structures NbN-GdN1-Ti-GdN2-NbN were grown by DC sputtering in an ultrahigh vacuum (UHV) chamber at room temperature. The NbN and GdN layers were deposited under similar conditions as described in the references. It has been observed that the magnetic and electrical property of GdN is sensitive to deposition condition and can be tuned by changing different Ar and N\(_2\) gas mixture and deposition power. The two GdN(1,2) layers were grown with different gas mixture in order to introduce coercive contrast. The GdN1 and GdN2 layers were deposited with 8 % and 4% Ar - N\(_2\) gas mixture, respectively. The Ti layer was grown in a pure Ar gas environment with a pressure 1.5 Pa and sputtering power of 40 W. The thickness of top and bottom NbN layers were kept fixed at 50 nm while thickness of GdN and Ti layers were varied in different depositions. Eight multilayer stack with different thickness of Ti were grown in the same deposition in the sequence NbN-GdN1-Ti-GdN2-NbN from left to right.

The double junctions were fabricated in a mesa structure in which junction area \((7 \, \mu m \times 7 \, \mu m)\) was defined by CF\(_4\) plasma etching and Ar-ion milling. The fabrication process is similar to described in the references.

except these devices were Ar-ion milled for 14 min instead of 4 min to ensure complete milling of Ti till bottom NbN layer. Fig. 2(d) shows schematic of the double tunnel junction in the mesa structure with measurement scheme. The electrical characterization of the devices up to 4.2 K were done in a custom made dip-stick. For 300 mK measurements a He-3 sorption insert form Cryogenics Lmt. was used. The differential conductance \(dI/dV\) of the junctions at 300 mK were obtained by numerically differentiating measured I-V curves. Conductance spectra at 4.2 K were obtained with standard lock-in technique. The \(R-H\) loops were measured with a DC current source and nanovoltmeter. In this report we show the results of one representative double junction. Measurements done on other junctions on the same chip and devices with different thickness of Ti are shown in the supplementary material. All the data reported in the manuscript were found to be extremely reproducible as shown in supplementary figures.

III. RESULTS AND DISCUSSION

![Graph](Figure 1: (Color online) Temperature dependence of resistance of the NbN(50nm) - GdN(2nm-8%) - Ti(8nm) - GdN(2nm-4%) - NbN(50 nm) double spin-filter tunnel junction. The measurement was done using a current \(I = 10 \, \mu A\). The upper inset shows \(R(T)\) in the range 0.3 to 1.3 K. Lower inset shows \(R(T)\) close to \(T_C\).]

Figure 1 shows temperature dependence of resistance of a double spin-filter tunnel junction with 8 nm thick Ti spacer. A semiconducting behavior can be seen till 35 K and metallic-like behavior below it due to onset of spin filtering at the Curie temperature, \(T_{Curie} \approx 35\) K of GdN layers. The \(R(T)\) is similar to a single NbN-GdN-NbN spin-filter tunnel junction. The superconducting transition of NbN can be seen to start at \(T_C \approx 13\) K. The transition was found to be broad with a width of \(\sim 1.7\) K as shown in the lower inset of Fig. 1. This is due to the difference in the \(T_C\) of top and bottom NbN in the double tunnel junction. The \(R(T)\) of some
other double tunnel junctions are shown in the supplementary material (SFig. 3). For measurements done with a bias voltage smaller than gap voltage, i.e., $eV < 2\Delta$, the resistance was found increase rapidly below $T_C$ of NbN. The electrical transport below $T_C$ is determined by quasiparticles. For bias voltage in the sub-gap region the tunneling current is weakly dependent on bias voltage and scales with temperature dependent quasiparticle density $n(T) \propto \sqrt{T} e^{-E_g/nk_BT}$. Therefore, temperature dependence of sub-gap resistance follows an exponential dependence, $R(T) \propto e^{-\Delta/k_BT}$, with a constant parallel leakage resistance $^\text{11}$. The upper inset in Fig. 1 shows $R(T)$ in the range 1.3 to 0.3 K. Bulk Ti is known to be a superconductor with $T_C \sim 0.49$ K. However, we could not observe any superconducting transition of Ti in our devices till 0.3 K. This might be due to large suppression of $T_C$ of the thin Ti layer sandwiched between two magnetic GdN layers.

A. Tunneling behavior

A double tunnel junction is essentially made of two tunnel junction in series. In our NbN-GdN1-Ti-GdN2-Nb double tunnel junction devices there are two tunnel junctions NbN-GdN1-Ti (Jn1) and Ti-GdN2-NbN (Jn2) in series. As the two tunnel junctions are deposited with opposite sequence they most likely have different resistances; $R_{Jn1}$ and $R_{Jn2}$. Besides NbN-GdN interface is expected to be more resistive than Ti-GdN interface due to different Schottky barrier height: $\Phi_{Sh} = W - E_g^{GdN}$. Where $E_g^{GdN}$ is the band gap of GdN and $W$ is the work function of the metal. As work function of NbN $\sim 4.7$ eV is larger than that of Ti $\sim 4.3$ eV, NbN-GdN interface have lower transparency than Ti-GdN interface. Therefore, the double tunnel junction NbN-GdN1-Ti-GdN2-NbN is most likely to develop asymmetry even with ideal interface without considering fabrication issues. Traditionally double tunnel junction with superconductors has been studied with structure Nb-Al$_2$O$_3$-Al-Al$_2$O$_3$-Nb or Nb-NbO$_x$-Al-AlO$_x$-Nb or Nb-NbO$_x$-Al.AlO$_x$-Nb. The Al spacer is most popular due to its tendency to form high quality pin-hole free native oxide. In this kind of tunnel junctions AlO$_x$ provides a large barrier height $\sim 1.7$ to $2.5$ eV which makes it possible to create potential well and observe fascinating effects like resonant tunneling in double barrier tunnel junctions. But in the case of GdN the barrier height is usually small $\sim 10$-100 meV, therefore, more transparent tunnel barrier is expected.

The I-V and $dI/dV - V$ measurements were done at different temperatures to understand tunneling nature of the double junctions. Fig. 2 shows conductance spectra $G(V) = dI/dV$ normalized to its value at $V = 0$ measured at different temperatures above the $T_C$ of NbN. Parabolic conductance spectra suggest tunneling type transport in these devices. For the $dI/dV$ measurements at 20 K deviation from parabolic behavior above $\pm 10$ mV is probably due to exchange splitting of the GdN tunnel barrier below the $T_{Curie}$. A small asymmetry can also be seen in the conductance spectra which suggest the two tunnel junctions involved in the double tunnel junction have different resistances; $R_{Jn1}$ and $R_{Jn2}$. The conductance spectra of the same junctions were also measured below $T_C$ of NbN. The normalized $dI/dV$ spectra measured in the temperature range 1.7 to 11 K are shown in the Fig. 2(c). Clear appearance of superconducting gap validate a tunneling type transport in these double tunnel junctions. Fig. 2(b) shows IV and $dI/dV$ measurement done on the same junction at 300 mK. Two conductance peaks separated by $4\Delta \sim 3.3$ meV can be observed. The superconducting gap $\Delta$ of NbN is suppressed along with smearing of gap edges probably due to magnetic GdN. In SINIS tunnel junctions nonequilibrium effects usually leads to sub-gap step structures whose position and amplitude strongly depends on the temperature. In some cases much sharper gap-edge structure is considered as an evidence of the nonequilibrium effects. However, none of these features can be seen in the conductance spectra as shown in Fig. 2(b,c). The reason for this is discussed below.

Below the $T_C$ of NbN the conductance spectra of our devices can be understood in terms of an asymmetric SINIS tunnel model with an asymmetry parameter $a_s = 2\pi/\Delta$ (1 $\leq a_s \leq 2$) with $x = R_{Jn1}/R_{Jn2}$.

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\[ 50 \]
FIG. 3: (Color online) (a) I-V and normalized conductance spectra $G(V)/G_N$ of NbN-GdN(2.3 nm)-NbN tunnel junction. The red solid line represents fitting to the S-I-S tunneling model with fitting parameter $\Delta = 1.415$ meV and $\gamma = 0.195$. (b) I-V and normalized conductance spectra $G(V)/G_N$ of NbN-Ti(9 nm)-GdN(2.6 nm)-NbN tunnel junction. The red solid line represents fitting to the N-I-S tunneling model with fitting parameter $\Delta = 1.38$ meV and $\gamma = 0.325$. (c) I-V and normalized conductance spectra $G(V)/G_N$ of NbN-GdN(2 nm)-Ti(8 nm)-GdN(2 nm)-NbN tunnel junction. The red solid line represents fitting to the S-I-N-I-S tunneling model with asymmetry parameter $a_s = 2$ (red) and 1.85 (black).(d) Temperature evolution of the fitting parameters $\Delta$ and $\gamma$ found from fitting Eq. (1) to the conductance spectra shown in Fig. 2(c). The red ($a_s = 2$) and blue ($a_s = 1.85$) solid lines are the fitting to the BCS type temperature dependence; $\Delta(T) = \Delta(0)\tanh(1.74\sqrt{(T_C - T)/T})$ with $T_C = 12.11$ K. Black solid line is the fitting to an exponential of the form; $\gamma \propto e^{-c/T}$ [49].

$$\frac{G(V)}{G_N} = \frac{1}{a_s} \int_{-\infty}^{+\infty} N_S(E) \left[ f_N(E - a_s eV/2) - f_N(E + a_s eV/2) \right] dE,$$

where $f_N(E)$ is the non-equilibrium distribution function inside the Ti layer and can be expressed as:

$$f_N(E) = \frac{N_s(E - a_s \sqrt{2}) f_0(E - a_s \sqrt{2}) + N_s(E - a_s \sqrt{2}) f_0(E - a_s \sqrt{2}) + \zeta/E}{N_s(E - a_s \sqrt{2}) + N_s(E - a_s \sqrt{2}) + \zeta/E}.$$

Here $f_0(E, T) = \frac{1}{1 + \exp(E/k_BT)}$ is the Fermi-Dirac function at temperature $T$. Superconducting quasiparticle density of state with Dynes parameter $\gamma$ is given by $N_S(E) = N(0) \text{Re} \left( \frac{E/\Delta - i\gamma}{\sqrt{(E/\Delta - i\gamma)^2 - 1}} \right)$. Here $\gamma$ incorporates finite life-time of quasiparticles in the superconductor. In Eq. (2), $\tau_\Phi$ is the relaxation time representing time scale for interchange of energy between the quasiparticle and the rest of the system. This energy relaxation rate is determined by electron-electron interactions, electron-phonon interactions and magnetic impurities which can induce decoherence. Here $\Gamma^{-1}$ refers to the mean residency time of quasiparticles inside Ti layer and is given by; $\Gamma = 2\zeta / N_N(E_F)R_NA_Le^2$. Here $N_N(E_F)$ is the normalized density of states of Ti, $R_N$ is the normal state resistance of Ti, $A$ and $L$ are the cross-section area and length of the normal metal (Ti), respectively. Clearly, the residency time and thereby influence of non-equilibrium processes grows with decreasing tunnel junction volume and tunneling resistance.

Now we discuss conditions for nonequilibrium. The distribution function of electron inside the normal metal is mainly determined by the ratio of the relaxation rate and rate of injection the electron into it. Usually for
\[ \tau_E \Gamma >> 1 \] (injection rate exceed relaxation rate) the distribution function in the normal metal deviates from the thermal equilibrium Fermi distribution function \( f_0(E, T) \) and if \( \tau_E \Gamma << 1 \) (equilibrium), the normal metal follows a Fermi distribution function. The conditions \( \tau_E \Gamma \rightarrow 0 \) and \( \tau_E \Gamma \rightarrow \infty \) correspond to complete equilibrium and nonequilibrium, respectively.

Although intuitively it seems in low resistance GdN tunnel barrier the nonequilibrium effects will be enhanced. But when the barrier transparency is decreased or effective life time of the quasiparticle in the interlayer is increased, the amount of quasiparticles that is scattered inelastically also increases due to magnetic nature of the tunnel barriers. Therefore, driving the middle Ti layer far from equilibrium in double spin-filter tunnel junction is not trivial like a SINIS-tunnel junction of the tunnel barriers. Therefore, driving the middle Ti layer far from equilibrium in double spin-filter tunnel junction is not trivial like a SINIS-tunnel junction with nonmagnetic elements. Our double spin-filter tunnel junctions can be reasonably assumed as a series connection of SIN and NIS junctions where the energy distribution function in the interlayer is the equilibrium Fermi distribution function, i.e., \( f_N(E, T) \approx f_0(E, T) \).

Figure 3(a,b,c) shows IV and conductance spectra of different type of tunnel junction measured at 4.2 K in the same experimental set-up (measured with lock-in technique). In a typical NbN-GdN-NbN tunnel junction conductance spectra shows a superconducting gap \( \Delta (4.2 \text{ K}) \sim 1.4-1.5 \text{ meV} \) depending on the tunnel barrier thickness and transparency. See supplementary material (SFig. 13) for conductance spectra of NbN-GdN-NbN tunnel junctions with different thickness of GdN. Fig. 3(a) shows conductance spectra of a NbN-GdN (2.3 nm)-NbN tunnel junction measured at 4.2 K. The red solid line is the fit to the typical SIS tunneling model with fitting parameter \( \Delta = 1.415 \text{ meV} \) and \( \gamma = 0.195 \) (See supplementary material for SIS tunnel model used for fitting). Fig. 3(b) shows conductance spectra of a NbN-Ti(9 nm)-GdN(2.6 nm)-NbN tunnel junction. As the thickness of the Ti (~9 nm) in this type of device is larger than both the superconducting coherence length \( \xi_{NbN} \sim 4.1 \text{ nm} \) and \( \xi_{Ti} \sim 3.6 \text{ nm} \) (\( \xi_{Ti} \); Normal state coherence length of Ti), this type of tunnel junction can be considered as NIS-type tunnel junction. The red solid line shows fitting of the NiS tunnel model to the conductance spectra with fitting parameter \( \Delta = 1.38 \text{ meV} \) and \( \gamma = 0.325 \) (see supplementary material for more detailed study of NIS-type tunnel junctions).

B. Spin-valve behavior

In a SINIS-type tunnel junction when the bias voltage \( eV \) exceeds \( 2\Delta \), quasiparticle current is produced from the energy gained primarily from applied bias voltage. Besides even for voltages less than \( 2\Delta \) at a finite temperature thermally excited quasiparticles above the gap are present whose number exponentially reduces as temperature is lowered below \( T << T_C \). However, in a double spin-filter tunnel junction additional spin-polarized quasiparticles are present due to pair breaking processes which equally populate electron and hole-like excitation spectrum. In double spin-filter tunnel junction the spin-polarized quasiparticle current can be turned ON and OFF by reorienting magnetization of the two spin-filter barrier parallel and antiparallel with respect to each other, respectively.

The presence of the superconducting gap in the conductance spectra induces an energy selectivity of quasiparticle tunneling. Therefore, in double spin-filter devices the number and the energy of quasiparticles can be drastically altered if a bias voltage above and below the gap is applied. The R-H loops measured above and below
the gap voltage can provide valuable information about non-equilibrium quasiparticle accumulation in the middle metallic (Ti) layer\[^{55}\]. Fig. 4(a-e) shows R-H loops of the double spin-filter tunnel junction measured at 300 mK with different bias current. The I-V curve measured at the same temperature is shown in the Fig. 4(f). The conductance spectra gap edges can be seen at \(\pm 1.68\) meV which corresponds to a bias current \(I = 99\) \(\mu\)A. At bias current \(I = 200\) \(\mu\)A a clear hysteretic R-H loop with resistance peaks near \(\pm 10\) mT can be observed. As coercive field of a single GdN layer is typically \(\sim 5\) mT\[^{35, 56}\], the hysteretic R-H loop observed in these double tunnel junctions is due to relative magnetization orientation of the two GdN(1,2) layers. A broad switching is observed in this case due to multi-domain nature of GdN layers.

One striking thing to note is that the hysteresis in the R-H loops was found to disappear as current is decreased from 200 to \(1\) \(\mu\)A. However, an overall high resistance state can be seen in the magnetic field range \(\pm 15\) mT when the two magnetic GdN(1,2) layers are not parallel to each other. The number of charge carriers (quasiparticles) that can transport charge through the S-FI-N-FI-S structure is reduced drastically as the bias voltage is reduced below gap-voltage. This can be seen as increase in the resistance of the double spin-filter tunnel junction from 12 to \(81\) \(\Omega\) as bias current is reduced from \(200\) to \(1\) \(\mu\)A. The R-H loops were also measured at different temperatures and similar behavior was found at all temperatures below \(T_C\) of NbN. The hysteresis in the R-H loop was found to disappear above \(15\) K (See supplementary figure SFig. 4). Although, \(T_C\) of GdN \(\sim 35\) K absence of hysteretic R-H loop above \(15\) K suggest absence of well established parallel and antiparallel state. A linear decrease in resistance with magnetic field can still be observed above \(15\) K confirming magnetic nature of individual GdN layers above \(15\) K.

The switching behavior can be understood by considering spin-polarized quasiparticle accumulation and relaxation inside the Ti layer. A finite spin-polarized quasiparticle accumulation is expected inside Ti layer when the two GdN(1,2) layers are antiparallel to each other. Therefore conductance is reduced and the resistance for the antiparallel state is expected to be higher than that for the parallel configuration. Also spin-polarized quasiparticle accumulation can modify interlayer exchange coupling. The absence of hysteretic R-H loop at sub-gap bias current is most likely due to the suppression of interlayer exchange coupling between two GdN(1,2) layers. This is expected as magnetic coupling is usually suppressed in F-S-F trilayer system below the critical temperature \(T_C\) of the superconductor\[^{59, 64}\]. Suppressed magnetic coupling has been observed in Fe\(_4\)N-NbN-Fe\(_4\)N\[^{57}\], (100)-oriented GdN/W/NbN/W multilayers\[^{58}\] and GdN-NbN-GdN trilayers\[^{61}\]. Recently, a different kind of interlayer exchange coupling mechanism in GdN-Nb-GdN has been proposed\[^{59}\]. Interlayer exchange coupling between ferromagnetic metal layers separated by superconducting spacer has been investigated extensively in many systems and a detailed discussion is beyond the scope of this paper\[^{59, 62, 63}\]. A more detailed experimental study with different thickness of the normal-metal spacer is needed to understand the interlayer exchange mechanism in presence of nonequilibrium quasiparticles in these double spin-filter tunnel junctions.
IV. CONCLUSIONS

In conclusion, we have fabricated double spin-filter tunnel junction in the configuration NbN-GdN1-Ti-GeN2-NbN. The conductance spectra in these double spin-filter tunnel junctions were found to be analogous to a highly asymmetric SINIS-type tunnel junction. We have demonstrated spin-polarized quasiparticle control in these double spin-filter tunnel junction with R-H measurements done at different bias voltage above and below gap voltage $eV = 2\Delta$. Hysteresis in the R-H loop was found to be absent for sub-gap bias current. Absence of hysteresis in R-H loop may be considered as an experimental signature of non-equilibrium spin-polarized quasiparticle accumulation. Although nonequilibrium effects cannot be inferred conclusively from these experiments, these preliminary experimental results are of fundamental importance and calls for further experimental and theoretical investigation. Magnetic manipulation of quasiparticles is pivotal for the advancement of quasiparticle spintronics [10].

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Supplementary Information

Spin-polarized quasiparticle control in a double spin-filter tunnel junction

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CONTENTS

Series of tunnel junctions with different configuration were fabricated from multilayer stacks NbN-GdN-NbN, NbN-Ti-GdN-NbN, and NbN-GdN1-Ti-GdN2-NbN. The thickness variation in different stacks was achieved by controlling the rotation speed of the sample stage during deposition. Below a detailed summary of all measurements done with these tunnel junctions are shown.

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NbN-GdN1-Ti(t)-GdN2-NbN tunnel junction

SFig. 1: SERIES-1 :I-V and normalized conductance spectra $G(V)/G_N$ of the NbN(50 nm)-GdN1(2 nm)-Ti(t)-GdN2 (2 nm)-NbN(50 nm) double spin-filter device with Ti thickness in the range 1-8 nm. The GdN1 and GdN2 layers were deposited with 8 % and 4 % N2 and Ar gas mixture. The conductance spectra at 4.2 K were measured with a standard lock-in technique.
Sfig. 2: SERIES-2: I-V and normalized conductance spectra $G(V)/G_N$ of the NbN(50 nm)-GdN1(2 nm)-Ti-GdN2 (2 nm)-NbN(50 nm) double spin-filter device with Ti thickness in the range 1-4 nm. Both the GdN1 and GdN2 layers were deposited with 8 % N$_2$ and Ar gas mixture. The conductance spectra at 4.2 K were measured with a standard lock-in technique. The junction with $>$1 nm Ti is equivalent to a NbN(50 nm)-GdN(4 nm)-NbN(50 nm) tunnel junction which is at the thickness limit of tunneling-type transport.
SFig. 3: The $R(T)$ of few representative double spin-filter tunnel junction with different thickness of Ti spacer and deposition gas mixture for GdN(1,2). The magnitude of resistance may not correspond to the thickness of GdN in these double junctions as all the $R(T)$ measurements were done with different bias current. Note that these tunnel junctions show nonlinear IV.
SFig. 4: (Color online) R-H loops of Jn1 (the same Jn as reported in the manuscript) measured at different temperatures with a bias current of $I = 200\mu\text{A}$. 
Reproducibility of spin-valve behavior in NbN-GdN1-Ti(t)-GdN2-NbN tunnel junction

SFig. 5: (Color online) R-H loops and I-V curve of Jn-2 (Note that each chip contain 8 identical junctions and measurements done on Jn-1 on the same chip is reported in the manuscript) measured at 300 mK with different bias current.

SFig. 6: (Color online) R-H loops and I-V curve of Jn-4 on the same chip measured at 300 mK with different bias current.
SFig. 7: (Color online) R-H loops and I-V curve of Jn-6 on the same chip measured at 300 mK with different bias current.

SFig. 8: (Color online) R-H loops and I-V curve of Jn-7 on the same chip measured at 300 mK with different bias current.

SFig. 9: (Color online) The R-H loop of the Jn-1 measured at 1.6 K with different bias current. The R-H loops measured below sub-gap current were found to be extremely sensitive to temperature stability during the measurement.

**Tunneling spectra of NbN-Ti-GdN-NbN tunnel junction:**

The NbN-Ti-GdN-NbN tunnel junction with thick ~9 nm Ti can be regarded as a N-I-S-type tunnel junction.
Normalized tunneling conductance of a NIS junction at a bias voltage $V$ can be written as:

$$\frac{G_s(V)}{G_N(V)} = \frac{d}{d(eV)} \int_{-\infty}^{\infty} N(E)[f(E) - f(E + eV)]dE,$$

(3)

where $f(E)$ is Fermi-Dirac distribution function and $N(E)$ is the normalized BCS density of state of the superconductor. Here $G_N(V)$ is the normal state conductance of the junction. Following Dynes approach, the quasiparticle density of states can be written as,

$$N(E) = N(0)\left|\frac{E/\Delta - i\gamma}{\sqrt{(E/\Delta - i\gamma)^2 - 1}}\right|.$$ Here the smearing parameter $\gamma$ is included to consider finite lifetime of quasiparticles. SFig. 9(b) shows fitting of Eq. (3) to conductance spectra of the NbN-Ti(9 nm)-GdN(2.6 nm)-NbN tunnel junction.

SFig. 10: (a) Temperature dependence of resistance of the NbN-Ti(9 nm)-GdN(2.6 nm)-NbN tunnel junction. measured with $I = 1 \mu A$. (b) I-V and normalized conductance spectra $G(V)/G_N$ of the tunnel junction measured at 4.2 K. The red solid line shows fitting to Eq. (3) with fitting parameter $\Delta_{\text{fit}} = 1.38$ meV and $\gamma = 0.325$. (c) Temperature evolution of conductance spectra from 4.2 to 9.1 K. (d) Temperature dependence of $\Delta_{\text{fit}}$ and $\gamma$. A BCS model fit (red solid line): $\Delta(T) = \Delta(0) \tanh(1.74\sqrt{(T_C - T)/T})$ gives $\Delta(0) = 1.416$ meV and $T_C = 10.91$ K. The smearing parameter $\gamma$ can be seen to increase exponentially (black solid line) with temperature.
Tunneling spectra of NbN-GdN (t)-NbN tunnel junction:

The quasiparticle tunneling conductivity \( G(V) = \frac{dI}{dV} \) in a S-I-S junction can be written as:

\[
\frac{G_s(V)}{G_N(V)} = \frac{d}{d(eV)} \int_{-\infty}^{\infty} N(E + eV)N(E)\left[f(E) - f(E + eV)\right]dE + \frac{V}{R_{sh}}
\]  

(4)

Where \( N(E) \) is density of states of the superconductor and \( f(E) \) is the Fermi distribution function. Here \( R_{sh} \) is a shunt resistance in series with the S-I-S junction. The modified density of states with Dynes parameter can be expressed as,

\[
N(E) = N(0) \left| \operatorname{Re} \left( \frac{E/\Delta - \imath\gamma}{\sqrt{(E/\Delta - \imath\gamma)^2 - 1}} \right) \right|
\]

Here \( \Delta \) is the superconducting gap and \( \gamma \) is the smearing parameter. SFig. 11 and SFig. 12 shows typical \( dI/dV - V \) spectra of NbN-GdN-NbN tunnel junction with two different barrier transparency. The temperature dependence of \( \Delta \) was fitted to the BCS-type temperature dependence, \( \Delta(T) = \Delta(0) \tanh(1.74 \sqrt{(T_C - T)/T}) \).
SFig. 11: (a) Temperature evolution of normalized conductance spectra \( G(V)/G_N \) of NbN-GdN(1.7 nm)-NbN tunnel junction. The conductance spectra were measured with a standard lock-in technique. (b) I-V and normalized conductance spectra \( G(V)/G_N \) of the same junction at 4.2 K. Red solid line is the fit to Eq. (4) with fitting parameter shown. Temperature dependence of the fitting parameter \( \Delta \) and \( \gamma \). A BCS model; \( \Delta(T) = \Delta(0) \tanh(1.74 \sqrt{(T_C - T)/T}) \) gave \( \Delta(0) = 1.44 \text{ meV} \) and \( T_C = 9.86 \text{ K} \). (d) Temperature dependence of resistance \( R(T) \) of the same junction.

SFig. 12: (a) Temperature evolution of normalized conductance spectra \( G(V)/G_N \) of NbN-GdN(2.3 nm)-NbN tunnel junction. The conductance spectra were measured with a standard lock-in technique. (b) I-V and normalized conductance spectra \( G(V)/G_N \) of the same junction at 4.2 K. Red solid line is the fit to Eq. (4) with fitting parameter shown. Temperature dependence of the fitting parameter \( \Delta \) and \( \gamma \). A BCS model; \( \Delta(T) = \Delta(0) \tanh(1.74 \sqrt{(T_C - T)/T}) \) gave \( \Delta(0) = 1.5 \text{ meV} \) and \( T_C = 9.54 \text{ K} \). (d) Temperature dependence of resistance \( R(T) \) of the same junction.
SFig. 13: (Color online) Superconducting gap $\Delta$ of 20 NbN(50 nm)-GdN($d$)-NbN(50 nm) tunnel junctions plotted against resistance of the junction measured at 4.2 K ($R_N(4mV)$) with bias voltage 4 mV ($>2\Delta$). Maximum error of 0.25 mV was assumed due to smeared gap edges. Conductance spectra of each junction is shown in figures SFig. 14-16. All the 20 NbN-GdN-NbN tunnel junctions were not prepared in the same deposition. Therefore, in this graph $R_N(4mV)$ is plotted against $\Delta$ instead of thickness of GdN layer $d$ vs $\Delta$. Note that in a tunnel junction $R \propto e^{-\kappa d}$ with $\kappa = -\frac{\hbar}{2m}\sqrt{2m\Phi}$; where $\Phi$ is the barrier height and $m$ is electron mass.
SFig. 14: SERIES-1 (23431): I-V and normalized conductance spectra $G(V)/G_N$ of the NbN(50 nm)-GdN(t)-NbN(50 nm) spin-filter device with GdN thickness in the range 0.8-2.3 nm. Conductance $G(V)$ measurement was not done in junctions with critical current due to divergence at the origin. All the tunnel junctions were prepared from the trilayer stack deposited at the same time.
Fig. 15: SERIES-2 (23432): I-V and normalized conductance spectra \( G(V)/G_N \) of the NbN(50 nm)-GdN(t)-NbN(50 nm) spin-filter device with GdN thickness in the range 1.1-3.3 nm. Conductance \( G(V) \) measurement was not done in junctions with critical current due to divergence at the origin. All the tunnel junctions were prepared from the trilayer stack deposited at the same time.
SFig. 16: SERIES-3 (23434): I-V and normalized conductance spectra $G(V)/G_N$ of the NbN(50 nm)-GdN(t)-NbN(50 nm) spin-filter device with GdN thickness in the range 1.0-2.9 nm. All the tunnel junctions were prepared from the trilayer stack deposited at the same time.