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Coexistence and tuning of spin-singlet and triplet transport in spin-filter Josephson junctions

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Reviewers' comments:

Reviewer #2 (Remarks to the Author):

This is important contribution to the physics of interplay of superconducting and magnetic correlations. The research reported in the manuscript focuses on mechanisms of generation of spin-polarized triplet Cooper pairs at magnetically inhomogeneous S/F interfaces. It is shown that singlet and triplet pairs can be detected in NbN-GdN-NbN Josephson junctions where GdN is a ferromagnetic insulator. This work combines experimental studies of the temperature behavior of the critical current a set of junctions with different barrier thicknesses and theoretical modeling within a tight-binding Bogolioubov de Gennes approach. Good correspondence between theory and experiment is found.

To my opinion, the results are new and previous work is properly cited.

I am confident that this work will have strong impact on the field and therefore I can recommend the manuscript for publication in its present form.

Reviewer #3 (Remarks to the Author):

This manuscript presents experimental measurements of the temperature dependence of the critical Josephson current, including the effect of an external magnetic field, for S-FI-S junctions where S are superconductors (here, NbN) and FI a ferromagnetic insulator (GdN). These measurements are then compared to numerical simulations based on a tight-binding Bogoliubov-de Gennes model. The numerics indicate that a Rashba spin-orbit interaction and a random impurity potential are required to reproduce the experiments. From the numerics, the authors present an analysis of the induced superconducting correlations and are able to quantify the amplitude and symmetry of the proximity-induced spin-triplet Cooper pairs. In particular, they find interesting connections between the measured behaviour of the critical current and the computed amplitude of the different pairing functions.

The study of hybrid junctions between superconductors and magnetic materials has become an active field of research with very promising potential applications. The comparison between theory and experiment presented in this manuscript and the analysis of the induced pairing correlations are thus, in my opinion, timely, interesting, and worthy of publication. However, I find that additional substantial improvement in the presentation and further clarification on the results is needed before I can recommend publication in Communications Physics. The main problems that I find are the following:

1. The measurements in figure 2 already appear on Ref. 33 [Phys. Rev. Lett. 122, 047002 (2019), figure 2(e,f,g)] by some of the authors, with the novelty here being the effect of an external magnetic field shown in figure 4. The numerical model was presented in Ref. 43 [Phys. Rev. B 100, 094501 (2019)], which states similar conclusions about the importance of the impurity potential and the spin-orbit coupling. While these previous works are cited, their overlap with the present results is not clearly described in the manuscript. In my opinion, the main novelty of the present work is the complementary combination of measurements and numerics for the analysis of singlet and triplet

correlations. Therefore, the authors could be more clear about how their results complement previous works.

2. Since the numerics are such an important part of these results (more than half of the figures show numerical results only), it is surprising that the theoretical model and the relevant parameters used are only described in the Supplementary Material. Maybe the main definitions could be included in the Methods section or even in the main text. For example, the reading of the first paragraphs of "section II Results" would benefit from a better description of the relevant parameters.

3. Is the external magnetic field included in the numerical calculations? Is it related to the small on-site fluctuations of the exchange field \delta_h? (see text after Equation 8 in the Supplementary Material). Is it possible to fit the measurements with a magnetic field in figure 4? The external magnetic field plays an important role in these results, but I miss a more detailed exploration of the dependence of the tight-binding calculations on the field.

4. Finally, I find the way the manuscript is written confusing. Partly because all the details of the numerical calculations, which are needed to understand most of the figures and results, are missing (relegated to the Supplementary Material). But also because, throughout the text but especially in the abstract, it is not clear when a concluding remark is due to an experimental observation or the analysis of the numerical results. The conclusions (last paragraph of "section III Discussion") are an exception, since there the main results are clearly listed. By contrast, the abstract and other parts of the text are filled with confusing statements about the main results of this manuscript.

For example, when the authors claim in the abstract that they "use the power of the Josephson effect for a quantitatively accurate proof of the coexistence and tunability of singlet and triplet transport" (lines 3-4). Or later (lines 37-38), after saying that the present work is an extension of the experiments in Ref. 33 with the novelty of an external magnetic field, they claim that the "magnetic field [is] an unambiguous knob to demonstrate coexistence and tuning of singlet and triplet components". This is very misleading because the pairing amplitudes are computed numerically using a model that does not explicitly include the external field and, moreover, the measurements with field are not directly compared to the numerics.

In summary, I recommend that the authors revise the manuscript so they can present the numerical calculations more clearly and describe better the effect of the external field. In doing so, they could make the exposition of their results more comprehensible, avoid misleading claims and try to write the text for a broader readership. Here are other, maybe minor, questions and comments:

1. The description of the characteristic shapes of the I_c curves in the introduction: plateau, cusp, or non-monotonic, seems to assume that the reader is familiar with the results of Ref. 33. In the introduction, without the support of a figure like figure 2 or 6, it is not obvious what these terms mean.

2. In figure 1 of the main text, the authors associate the separation between superconductors (dF) with the thickness L of the lattice model. However, when listing the simulation parameters for figure 2 in the Supplementary note 2, they keep L fixed (L=8) and change the width W: W=24 for panels (a,d) with dF=3nm, W=28 for panels (b,e) with dF=3.5nm, and W=32 for panels (c,f) with dF=4nm. What is thus the connection between L and dF?

3. Figure 2(c) seems to have a smaller critical temperature compared to (a) and (b). Could you explain?

4. Several times, the position inside the lattice is referred to as “the number of sites”, see figure 3 and its description on page 3 (eg., line 101). I think “the number of sites” could be more commonly interpreted as the *total* number of sites along one direction of the lattice, eg., L. The fact that L is fixed for the different thicknesses also adds to the confusion. Moreover, on first reading, it is not clear on page 3 if the lattice is being explored along the x or y direction.

5. The correlation functions in figure 3 are evaluated at phase \phi=0. Their behaviour at zero phase is then used to interpret the measurements of the critical current, which corresponds to a finite phase between 0 and pi. How do the different pairing functions change with the phase? Is their behaviour at zero phase the same that for the phase that gives the critical current?

6. Why is the case with dF=4nm not included in figure 4? Is the effect of the magnetic field for dF=4nm too small? The change is bigger for dF=3nm than for dF=3.5nm (line 134), however, it is not observed for dF=1.5nm (line 142). Do the authors have measured the effect of the field on the junctions with dF= 1.5, 1.75, 2, and 2.5 reported in Ref. 33? It would be interesting to know why the effect vanishes for both short (dF=1.5nm) and “long” (dF=4nm) junctions.

7. In the Supplementary note 2 the authors mention that “small changes in L provide large modifications in the system transport properties”. This seems to indicate a critical dependence on L, which is kept fixed throughout the manuscript (L=8). The results of Ref. 43 do not seem to show such a strong dependence, see figures 4 and 5 for L=50. Could you explain the choice of L=8?

8. Is it possible to “complete” figure 6 by showing the cases with strong SOC and impurity potential (that is, the missing top right panels)?

9. For completeness, the authors could provide the expressions for the T matrices in Equation 11 of the Supplementary Material, especially since they have a simple form, see Equation 13 in Ref. 43.

10. Ref. 53 could include the Erratum Phys. Rev. Lett. 11, 104 (1963), where the correct formula for the Josephson current is given.

Response to Referee #1

Josephson junctions underpinning a wide range of applications of superconductors are of everlasting great research interests. Needless to say, the authors of Manuscript COMMSPHYS-21-0458-T has reinvigorated this field by using a ferromagnetic insulator as the tunnelling barrier in their junction. As mentioned in the introduction of this manuscript and reported in other recent publications, e.g., ACS Nano 11, 5358 (2017) and Sci. Adv. 6, eaaz2536 (2020), SF hybrids host a large variety of emergent quantum phenomena of vast value in potential applications. Here, the authors use their SFS junction to unveil one of the most intriguing macroscopic quantum phenomena in physics, i.e., spin-triplet pairing. Their new findings not only provide critical insight into fundamental physics but also pave the path towards highly advanced applications in spintronics. Apart from the novelty and impact as mentioned above, this work also gains my support from the following aspects:
*The combined experimental and theoretical research provides rich and solid data to support their argument of the coexistence of spin-singlet and spin-triplet pairing, i.e., the non-monotonic behaviors of the Ic-T curves and especially the emergence of the bump in Ic-T.
*Ingredients such as disorder and spin-orbit coupling were taken into account for accurate simulations which, yet, still fit to the experimental data fairly well.
*The manuscript is well structured and easy to follow.

Thus, I recommend this work for a quick publication after the following revisions.

1. Despite the clear descriptions and cited references in the introduction, maybe the authors can use more words to clarify some terms, e.g., equal-spin triplets.

We thank the Referee for his/her positive comments and for the suggested references, which we included in the bibliography of the manuscript. In order to address the first point, in “Introduction”, we properly define “equal-spin triplets” and “opposite-spin triplets”.

JJs with multiple F-layer barriers have been theoretically and experimentally studied in connection to unconventional triplet superconductivity with equal-spin Cooper pairs, characterized by total spin-momentum S=1 and spin z-component S_z=±1 ($|11\rangle_{S,S_z} = |\uparrow\uparrow\rangle$ *and* $|1-1\rangle_{S,S_z} = |\downarrow\downarrow\rangle$)*, which can be artificially generated in these structures [5, 6, 8–16]. Compared to spin-singlet Cooper pairs and opposite-spin triplet Cooper pairs (total spin-momentum S=1 and spin z-component S_z=0,* $|10\rangle_{S,S_z} = 1/\sqrt{2}\left(\ |\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle\right)$*), the spin-triplet Cooper pairs are immune to the exchange field of the F-layer and can carry a non-dissipative spin current [5, 6, 8–16].*

In addition, we have included information on the theoretical model in the manuscript (Section “Spin-filter Josephson junctions and microscopic modelling”), thus explaining more carefully our jargon.

2. Fig. 1b is far more important than Fig. 1a, because it is richer in physics and highlights the schematic microscopic picture of the key findings. Thus, it would be better if the

authors increased the size of Fig. 1b. If necessary, Fig. 1 can be reconstructed by setting its Panel a as an inlet.

We appreciate the Referee comment, and we replace Fig.1 according to his/her suggestion. We also change every reference to figure 1 (a) or (b) in the manuscript, thus referring to main figure and inset in figure 1, respectively.

3. In contrast to spin-singlet superconductivity, spin-triplet pairs are good friends of applied magnetic fields. Although data obtained at different magnetic fields and temperatures are plotted in Fig. 4 as contours, it will be much better if the authors can include extra panels in Fig. 4 to show some vertical cutlines of the contours, i.e., field dependence of Ic at fixed temperatures.

We thank the Referee for his/her suggestion. Vertical cutlines in contour-plots reported in Fig.4 are standard Fraunhofer-like curves reported in the Supplementary Material (Supplementary Figure 2). We think that extra-panels in Fig. 4 do not add further information to the main aim of figure 4, which is to highlight the peculiar behavior of the Ic(T) curves in presence of magnetic field. Nevertheless, we understand the point raised by the Referee, and we add a deeper discussion on the dependence of the Ic on the magnetic field at fixed temperature in "Results" (line 139):

"The dependence of the Ic as a function of the normalized magnetic field H/H_0 is reported in Supplementary Material for the JJ with GdN thickness dF = 3.0nm at three selected temperatures:0.3K, 3K where a minimum of the $I_c(T)$ curve is measured for H/H_0 =75%, and 7K where a maximum of the $I_c(T)$ is observed for the same value of H/H_0. While a standard Fraunhofer-like $I_c(H)$ dependence is recovered at the three selected temperatures, for magnetic fields close to a quantum flux and at high temperature, e.g. 7K, I_c is larger than the value measured at low temperature, e.g. 0.3K. In order to stress this point, the IV curves measured at H/H_0 = 75% for the three selected temperatures are shown in Supplementary Figure 2 as a term of reference."

Response to Referee #2:

This is important contribution to the physics of interplay of superconducting and magnetic correlations. The research reported in the manuscript focuses on mechanisms of generation of spin-polarized triplet Cooper pairs at magnetically inhomogeneous S/F interfaces. It is shown that singlet and triplet pairs can be detected in NbN-GdN-NbN Josephson junctions where GdN is a ferromagnetic insulator. This work combines experimental studies of the temperature behavior of the critical current a set of junctions with different barrier thicknesses and theoretical modeling within a tight-binding Bogolioubov de Gennes approach. Good correspondence between theory and experiment is found.

To my opinion, the results are new and previous work is properly cited.

I am confident that this work will have strong impact on the field and therefore I can recommend the manuscript for publication in its present form.

We really appreciate the Referee opinion about our work. No question has been raised.

<u>Response to Referee#3:</u>

This manuscript presents experimental measurements of the temperature dependence of the critical Josephson current, including the effect of an external magnetic field, for S-FI-S junctions where S are superconductors (here, NbN) and FI a ferromagnetic insulator (GdN). These measurements are then compared to numerical simulations based on a tight-binding Bogoliubov-de Gennes model. The numerics indicate that a Rashba spin-orbit interaction and a random impurity potential are required to reproduce the experiments. From the numerics, the authors present an analysis of the induced superconducting correlations and are able to quantify the amplitude and symmetry of the proximity-induced spin-triplet Cooper pairs. In particular, they find interesting connections between the measured behaviour of the critical current and the computed amplitude of the different pairing functions.

The study of hybrid junctions between superconductors and magnetic materials has become an active field of research with very promising potential applications. The comparison between theory and experiment presented in this manuscript and the analysis of the induced pairing correlations are thus, in my opinion, timely, interesting, and worthy of publication. However, I find that additional substantial improvement in the presentation and further clarification on the results is needed before I can recommend publication in Communications Physics. The main problems that I find are the following:

1. The measurements in figure 2 already appear on Ref. 33 [Phys. Rev. Lett. 122, 047002 (2019), figure 2(e,f,g)] by some of the authors, with the novelty here being the effect of an external magnetic field shown in figure 4. The numerical model was presented in Ref. 43 [Phys. Rev. B 100, 094501 (2019)], which states similar conclusions about the importance of the impurity potential and the spin-orbit coupling. While these previous works are cited, their overlap with the present results is not clearly described in the manuscript. In my opinion, the main novelty of the present work is the complementary combination of measurements and numerics for the analysis of singlet and triplet correlations. Therefore, the authors could be more clear about how their results complement previous works.

We agree with the Referee that the main novelty of our work is the joint theoretical/experimental effort to relate the measurements of the unconventional temperature behaviour of the critical current to possible triplet pairing by using a microscopic modelling of the spin filter Josephson junctions. In this respect, we have emphasized this point as follows:

- In the Abstract, we remove the sentence:

"We use the power of the Josephson effect for a quantitatively accurate proof of the coexistence and tunability of singlet and triplet transport in ferromagnetic spin filter junctions."

and we add the sentence

“We build on previous achievements on spin-filter ferromagnetic Josephson junctions (JJs) and find unique correspondence between neat experimental benchmarks in the temperature behavior of the critical current and theoretical modeling based on microscopic calculations, which allow to determine a posteriori spin-singlet and triplet correlation functions. This kind of combined analysis provides an accurate proof of the coexistence and tunability of spin-singlet and triplet transport”

- In “Introduction”, we replace the sentence:

“Here we demonstrate alternative accurate methods to assess the spin-triplet transport…”

with

“By using a tight-binding Bogolioubov de Gennes approach, we model the $I_C(T)$ curves in the whole temperature range, along with the corresponding current-phase relation (CPR) as a function of the temperature T. It turns out that measurements of the temperature behavior of the critical current along with microscopic modeling approach provide an alternative accurate method to assess the spin-triplet transport, which can be extended to different types of JJs”

We also explicitly include in section “Experimental results and Theoretical Interpretation” the role played by the combined theoretical/experimental approach.

2. Since the numerics are such an important part of these results (more than half of the figures show numerical results only), it is surprising that the theoretical model and the relevant parameters used are only described in the Supplementary Material. Maybe the main definitions could be included in the Methods section or even in the main text. For example, the reading of the first paragraphs of “section II Results” would benefit from a better description of the relevant parameters.

We thank the Referee for raising this interesting point. In the revised version of the manuscript, we move the theoretical model and the relevant parameters from Supplementary Material to the main text (sections “Spin-filter Josephson junctions and microscopic modelling”, “Experimental results and theoretical interpretation” and “Discussion”).

3. Is the external magnetic field included in the numerical calculations? Is it related to the small on-site fluctuations of the exchange field \delta_h? (see text after Equation 8 in the Supplementary Material). Is it possible to fit the measurements with a magnetic field in figure 4? The external magnetic field plays an important role in these results, but I miss a more detailed exploration of the dependence of the tight-binding calculations on the field.

We do not include the Peierls phase in the hopping parameters due to the external magnetic field, nor we study the dynamics of the magnetization in barrier. It would include extra complications that are not justified in this manuscript. The main point here is that our samples have a ferromagnetic tunnel (FI) barrier. Qualitatively, the main role of the external field in these samples is to trigger the magnetic order in the GdN layer. This effect is qualitatively accounted for mimicking the role of the external magnetic field by reducing the impurity scattering in FI layer with SOC (see Fig.5 in the main text).

4. Finally, I find the way the manuscript is written confusing. Partly because all the details of the numerical calculations, which are needed to understand most of the figures and results, are missing (relegated to the Supplementary Material). But also because, throughout the text but especially in the abstract, it is not clear when a concluding remark is due to an experimental observation or the analysis of the numerical results. The conclusions (last paragraph of "section III Discussion") are an exception, since there the main results are clearly listed. By contrast, the abstract and other parts of the text are filled with confusing statements about the main results of this manuscript. For example, when the authors claim in the abstract that they "use the power of the Josephson effect for a quantitatively accurate proof of the coexistence and tunability of singlet and triplet transport" (lines 3-4). Or later (lines 37-38), after saying that the present work is an extension of the experiments in Ref. 33 with the novelty of an external magnetic field, they claim that the "magnetic field [is] an unambiguous knob to demonstrate coexistence and tuning of singlet and triplet components". This is very misleading because the pairing amplitudes are computed numerically using a model that does not explicitly include the external field and, moreover, the measurements with field are not directly compared to the numerics.

We thank the Referee for having point out this weakness in the manuscript. We accurately changed/dropped ambiguous sentences throughout the manuscript, as reported in the response to point 1 and in the list of changes.

In summary, I recommend that the authors revise the manuscript so they can present the numerical calculations more clearly and describe better the effect of the external field. In doing so, they could make the exposition of their results more comprehensible, avoid misleading claims and try to write the text for a broader readership. Here are other, maybe minor, questions and comments:

1. The description of the characteristic shapes of the I_c curves in the introduction: plateau, cusp, or non-monotonic, seems to assume that the reader is familiar with the results of Ref. 33. In the introduction, without the support of a figure like figure 2 or 6, it is not obvious what these terms mean.

We thank the Referee for his/her comment. We address his/her suggestion in "Introduction", by better discussing the differences between standard Ic(T) curves in literature and the ones reported in this work. The paragraph in line 43 is changed as follows:

"The large variety of materials and configurations employed in diffusive SFS JJs in literature allows to access to a wide range of behaviors for the thermal dependence of the I_c [1, 5]. Particularly relevant for our work, the possibility of generating oscillations in the superconducting order parameter by means of a finite exchange field in the F interlayer results in a π phase-shift in the CPR and a sudden drop of the I_c towards zero at temperatures $T_\pi < T_c$, followed by an increase of the I_c for $T > T_\pi$ [3, 5]. Therefore, the non-

monotonic behavior for the I_c (T) in systems in which a 0-π transition occurs is characterized by a peculiar cusp at the transition point T_π [3, 5]. In this work, we focus on the peculiar behavior of the I_c (T) in tunnel ferromagnetic spin-filter JJs, in which an unconventional 0-π transition occurs. Above a GdN thickness d_F =3.0nm, the I_c is not completely suppressed at T_π, thus suggesting that a 0-π transition broadened in a range of temperatures of the order of some K occurs [33]. Therefore, in the devices discussed here the I_c (T) curve shows a region in which the I_c is constant in a wide range of temperatures, i.e. it shows a plateau, or it shows a non-monotonic trend characterized by a non-zero local minimum, i.e. the I_c (T) exhibits an incipient 0-π transition [33]."

2. In figure 1 of the main text, the authors associate the separation between superconductors (dF) with the thickness L of the lattice model. However, when listing the simulation parameters for figure 2 in the Supplementary note 2, they keep L fixed (L=8) and change the width W: W=24 for panels (a,d) with dF=3nm, W=28 for panels (b,e) with dF=3.5nm, and W=32 for panels (c,f) with dF=4nm. What is thus the connection between L and dF?

We agree with the referee that we did not well clarify the role of the parameter L in the simulations and its connection with the experimental d_F. Here, we try to answer to his/her question, as well as we accordingly adjusted the main text to make this point clearer (please see "Experimental Results and Theoretical Interpretation"). In our description, we needed to consider the short-junction limit due to the tunnel nature of the barrier. Indeed, this leads to a sizeable critical current dropping with increasing the barrier thickness (as it is also noticeable from the experimental measurements in Ref.33). In our lattice model, this implies dealing with junctions of a few lattice sites, hence we choose L = 8 and keep it fixed in all the numerical simulations. For this reason, we are not able to establish a direct correspondence between the experimental barrier thickness d_F and the model parameter L, that we kept fixed in all the simulations. However, the main effect of increasing d_F (thus the magnetic area of the ferromagnet) consists in enhancing the magnetic activity of the junction, as reported in previous works in which a clear dependence of the spin-polarization and magnetic phenomena has been observed when increasing the thickness of the FI layer (see Pal, 2014, Nature Communications; Massarotti, 2015, Nature Communications; Caruso, 2019, Physical Review Letters; Ahmad, 2020, Journal of Superconductivity and Novel Ferromagnetism). Therefore, we assume to mimic this behavior by using the exchange field flux Φ=LWh and the on-site impurity potential strength V_{imp} as effective parameters, that we vary accordingly to fit the different experimental samples. In particular, by changing the width of the barrier W, we are able to increase the exchange field flux (modeling the enhanced magnetic activity of the JJ) without modifying the exchange field h of the FI (that we assume to be an intrinsic property of the GdN). In this picture, we model the shortest junction (with d_F=3.0nm) by choosing the lowest value of the exchange field flux Φ and the highest value of V_{imp}, whereas the viceversa holds for the d_F=4.0nm JJ, as it is explained in the following table.

Experimental barrier	Theoretical width of	Exchange field flux	Impurity potential

thickness d_F (nm)	the barrier W (in number of lattice sites)	Φ =hLW (in a.u.) (with h=0.25 in units of the hopping t and L=8 in number of lattice sites)	strength V_{imp} (in units of the hopping t)
3.0	24	48	0.30
3.5	28	56	0.37
4.0	32	64	0.23

3. Figure 2(c) seems to have a smaller critical temperature compared to (a) and (b). Could you explain?

The critical temperature of NbN/GdN/NbN Josephson junction in Fig. 2(c) differs from those in (a) and (b) of less than 10%, we think that the lower T_c in Fig. 2(c) has to be accounted to the larger thickness of the GdN barrier and to lower values of the critical current density.

4. Several times, the position inside the lattice is referred to as "the number of sites", see figure 3 and its description on page 3 (eg., line 101). I think "the number of sites" could be more commonly interpreted as the *total* number of sites along one direction of the lattice, eg., L. The fact that L is fixed for the different thicknesses also adds to the confusion. Moreover, on first reading, it is not clear on page 3 if the lattice is being explored along the x or y direction.

We understand the point raised by the Referee and we agree on his/her comment. By looking at figure 1 in the main text and in the Supplementary material, we indicate a given position in the lattice with the vector $\vec{r} = j\vec{x} + m\vec{y}$, which is thus identified by the couple of indices (j, m). In figure 3 we show the correlation functions at different values of the position along the x-direction (with index j), hence evaluating them at each fixed "transverse stripe" of the barrier (with j=1,…,L). In the manuscript (line 115, figures 3 and 5 and Table I), we now refer to the position in the lattice as "lattice position along x (index j)" instead of "number of sites" in order to clarify that the lattice is being explored along x and the total number of sites in the transport direction is L.

5. The correlation functions in figure 3 are evaluated at phase \phi=0. Their behaviour at zero phase is then used to interpret the measurements of the critical current, which corresponds to a finite phase between 0 and pi. How do the different pairing functions change with the phase? Is their behaviour at zero phase the same that for the phase that gives the critical current?

The correlation functions are related to the possible pairing mechanisms induced in the FI barrier by proximity effects. Thus, they must be evaluated taking into account the equilibrium state of the junction at fixed conditions (i.e. temperature T, geometric configuration and effective parameters). For these reasons, since all the junctions at temperature T=0.025 T_c (at which we calculate the correlation functions) are in the 0-equilibrium state (as it is observable from the corresponding current phase relations in figure 2), we choose ϕ=0. By increasing the temperature, modifications of the results compared to the lowest temperature are negligible.

6. Why is the case with dF=4nm not included in figure 4? Is the effect of the magnetic field for dF=4nm too small?

The JJ with d_F=4nm is characterized by currents of the order of few tens nanoamperes. Our analysis of the $I_c(T)$ curve in field requires to measure the Fraunhofer pattern curve up to the critical temperature of the device. When increasing the temperature, the Fraunhofer pattern curve is hardly measurable, since the critical current is suppressed around some nanoampere. For Josephson energy E_J comparable or smaller than the thermal energy ($E_J \leq k_B T$), in fact, fluctuations of I_c become a physical and intrinsic limitation, which does not allow to experimentally appreciate any variation in the $I_c(T)$ curves as a function of the magnetic field.

The change is bigger for dF=3nm than for dF=3.5nm (line 134), however, it is not observed for dF=1.5nm (line 142). Do the authors have measured the effect of the field on the junctions with dF= 1.5, 1.75, 2, and 2.5 reported in Ref. 33? It would be interesting to know why the effect vanishes for both short (dF=1.5nm) and "long" (dF=4nm) junctions.

We stress that the tuning of the $I_c(T)$ curve shape as a function of the magnetic field is expected to occur for all the JJs that show a plateau or a non-monotonic $I_c(T)$ behavior at zero field. The case with d_F=4nm is not included in figure 4 because of the considerations discussed in the previous point. Even though not reported in this work, devices with thicknesses 1.75nm, 2nm and 2.5 nm have shown a standard behavior in presence of a magnetic field, as it occurs for the junction with d_F=1.5nm. As reported in the Supplementary Material, we have demonstrated that the effect of the magnetic field on this junction does not affect the $I_c(T)$ curve shape, since at zero field the experimental data show small deviation from a standard Ambegaokar-Baratoff relation. Therefore, the dependence on an external magnetic field of the $I_c(T)$ curve has its onset in JJs that show unconventional $I_c(T)$ features, and the vanishing of the effect for the 4nm JJ is related to strongly reduced J_c in this specific device.

7. In the Supplementary note 2 the authors mention that "small changes in L provide large modifications in the system transport properties". This seems to indicate a critical dependence on L, which is kept fixed throughout the manuscript (L=8). The results of Ref. 43 do not seem to show such a strong dependence, see figures 4 and 5 for L=50. Could you explain the choice of L=8?

We thank the referee for raising this issue. We understand that our wording was somehow confusing, and we have modified that sentence in "Experimental Results and Theoretical Interpretation" and in the Supplementary material as:

"Tunnel junctions experience an exponential suppression of the critical current when increasing the barrier thickness [1]. In our model, this implies dealing with systems of few lattice sites, hence, we choose L=8 and keep it fixed in all the numerical simulations, in agreement with the short-junction limit".

Our results agree with those of Ref. 43. Our samples are tunnel junction where exponential suppression of the currents as a function of the length joins typical oscillations of ferromagnetic junctions. In order to be in the tunnel regime, we choose a small number of sites and optimized our parameter to match the experimental findings. While in the SNS regime we expect small modifications of the currents increasing the length of the N region (see fig. 2a of Ref. 43), in the case of SFIS junctions, the current is expected to be strongly oscillating as a function of L (see fig. 2b of Ref. 43). In that limit (say L from 4-20) small changes in the length of the junction may give rise to strong deviations in the current, in agreement with our findings.

8. Is it possible to “complete” figure 6 by showing the cases with strong SOC and impurity potential (that is, the missing top right panels)?

We thank the referee for raising this point. We have added the missing panels to improve Fig. 6 and we provide additional description on the added panels in “Discussions”.

9. For completeness, the authors could provide the expressions for the T matrices in Equation 11 of the Supplementary Material, especially since they have a simple form, see Equation 13 in Ref. 43.

We have added the explicit expression of the T matrices in the Supplementary Material.

10. Ref. 53 could include the Erratum Phys. Rev. Lett. 11, 104 (1963), where the correct formula for the Josephson current is given.

We thank the Referee for his/her suggestion, and we add in the bibliography the suggested reference.

LIST OF CHANGES (MANUSCRIPT)

- In "Abstract", we change the sentence in line 3 with:

  "We build on previous achievements on spin-filter ferromagnetic Josephson junctions (JJs) and find unique correspondence between neat experimental benchmarks in the temperature behavior of the critical current and theoretical modeling based on microscopic calculations, which allow to determine a posteriori spin-singlet and triplet correlation functions. This kind of combined analysis provides an accurate proof of the coexistence and tunability of spin-singlet and triplet transport"

- In "Introduction", we add references in line 14 of the new manuscript:

  [8] C. C.-B. Z. J.-C. PENG Lin, LIU Yong-Sheng, Influence of magnetic scattering and interface transparency on superconductivity based on a ferromagnet/superconductor heterostructure, Chinese Physics Letters 28, 087401 (2011).
  [9] G. Zhang, T. Samuely, N. Iwahara, J. Kaˇcmarˇcík, C.Wang, P.W. May, J. K. Jochum, O. Onufriienko, P. Szabó, S. Zhou, P. Samuely, V. V. Moshchalkov, L. F. Chibotaru, and H.-G. Rubahn, Yu-shiba-rusinov bands in ferromagnetic superconducting diamond, Science Advances

- In "Introduction", we change paragraph starting from line 14 in the old manuscript version with:

  "JJs with multiple F-layer barriers have been theoretically and experimentally studied in connection to unconventional triplet superconductivity with equal-spin Cooper pairs, characterized by total spin-momentum S=1 and spin z-component S_z=±1 ($|11\rangle_{\mathrm{S,S_z}} = |\uparrow\uparrow\rangle$ or $|1-1\rangle_{\mathrm{S,S_z}} = |\downarrow\downarrow\rangle$), which can be artificially generated in these structures [5, 6, 8–16]. Compared to spin-singlet Cooper pairs and opposite-spin triplet Cooper pairs (total spin-momentum S=1 and spin z-component S_z=0, $|10\rangle_{\mathrm{S,S_z}} = 1/\sqrt{2}(\;|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$), the spin-triplet Cooper pairs are immune to the exchange field of the F-layer and can carry a non-dissipative spin current [5, 6, 8–16]."

- In "Introduction", we change line 40 with:

  "By using a tight-binding Bogolioubov de Gennes (BdG) approach, we model the $I_c(T)$ curves in the whole temperature range, along with the corresponding current-phase relation (CPR) as a function of the temperature T. It turns out that measurements of the temperature behavior of the critical current along with microscopic modeling approach provide an alternative accurate method to assess the spin-triplet transport, which can be extended to different types of JJs:"

- In “Introduction”, we change paragraph starting from line 43 as follows:

  “The large variety of materials and configurations employed in diffusive SFS JJs in literature allows to access to a wide range of behaviors for the thermal dependence of the I_c [1, 5]. Particularly relevant for our work, the possibility of generating oscillations in the superconducting order parameter by means of a finite exchange field in the F interlayer results in a π phase-shift in the CPR and a sudden drop of the I_c towards zero at temperatures $T_\pi < Tc$, followed by an increase of the I_c for $T > T_\pi$ [3, 5]. Therefore, the non-monotonic behavior for the $I_c(T)$ in systems in which a 0-π transition occurs is characterized by a peculiar cusp at the transition point T_π [3, 5]. In this work, we focus on the peculiar behavior of the $I_c(T)$ in tunnel ferromagnetic spin-filter JJs, in which an unconventional 0-π transition occurs. Above a GdN thickness d_F =3.0nm, the I_c is not completely suppressed at T_π, thus suggesting that a 0-π transition broadened in a range of temperatures of the order of some K occurs [33]. Therefore, in the devices discussed here the $I_c(T)$ curve shows a region in which the I_c is constant in a wide range of temperatures, i.e. it shows a plateau, or it shows a non-monotonic trend characterized by a non-zero local minimum, i.e. the $I_c(T)$ exhibits an incipient 0-π transition [33].”

- We add the section “Spin-filter Josephson junctions and microscopic modelling” after the “Introduction” section.

- We move lines 59-62 from section “Results” (now: “Experimental results and theoretical interpretation”) to the beginning of the new section “Spin-filter Josephson junctions and microscopic modelling”.

- We replace “experimental results” in line 63 of Section “Results” of the old manuscript with “the S/FI/S junctions”, now in “Spin-filter Josephson junctions and microscopic modelling”.

- In “Spin-filter Josephson junctions and microscopic modelling”, we include the description of the microscopic model Hamiltonian (from line 82 to 102 of the new version of the manuscript), previously reported in Supplementary Material.

- In “Spin-filter Josephson junctions and microscopic modelling”, we include the recursive Green’s function (RGF) technique (from line 104 to 115 of the new version of the manuscript), previously reported in Supplementary Material.

- In “Spin-filter Josephson junctions and microscopic modelling”, we include the definition of s- and p-wave spin-pairing correlation functions (from line 116 to 124 of the new version of the manuscript), previously reported in Supplementary Material.

- We modify the title of “Results” section in “Experimental results and theoretical interpretation”.

- In “Experimental results and theoretical interpretation”, we add a section on the simulation parameters (from line 133 to 145 of the new version of the manuscript), previously reported in Supplementary Material.

- In “Experimental results and theoretical interpretation”, we add a deep discussion on the role of the parameter L in the model and the method used to mimic the variation in the FI barrier thickness with effective control parameters (from line 146 to 157 of the new version of the manuscript), previously reported in lines 81-86 of the old version of the manuscript and the Supplementary Material.

- In “Results” (now: “Experimental results and theoretical interpretation”), “Discussion” and captions in Fig.3 and Fig.5, we replace each reference to the “number of sites” with “position in the lattice along the x-direction, with index j=1,…,L”, or “lattice position along x (j)”. We also change the x-axis label in Fig.3 and Fig.5. Finally, we change “lattice site #4” with “lattice position j=4” all over the manuscript.

- In “Results” (now: “Experimental results and theoretical interpretation”), we change paragraph 131-139 as follows:

  “The dependence of the I_c as a function of the normalized magnetic field H/H_0 is reported in Supplementary Material for the JJ with GdN thickness d_F = 3.0nm at three selected temperatures: 0.3K, 3K where a minimum of the $I_c(T)$ curve is measured for H/H_0 =75%, and 7K where a maximum of the $I_c(T)$ is observed for the same value of H/H_0. While a standard Fraunhofer-like $I_c(H)$ dependence is recovered at the three selected temperatures, for magnetic fields close to a quantum flux and at high temperature, e.g. 7K, the I_c is larger than the value measured at low temperature, e.g. 0.3K. In order to stress this point, the IV curves measured at H/H_0 = 75% for the three selected temperatures are shown in Supplementary Figure 2 as a term of reference.”

- In “Results” (now: “Experimental results and theoretical interpretation”), line 143, we have added the following reference:

  “[54] V. Ambegaokar and A. Baratoff, Tunneling between superconductors, Phys. Rev. Lett. 11, 104 (1963).”

- In “Results” (now: “Experimental results and theoretical interpretation”), we remove from line 154-155:

“, as experimentally observed in Ref.[60] and predicted by full atomistic simulations in Refs.[61, 62].”

- In “Results” (now: “Experimental results and theoretical interpretation”), we add in line 163 information on the simulation parameters, previously reported in Supplementary Material.

- In “Discussion”, we include the simulation parameters used for the curves in Fig.6 (from line 251 to 253 of the new version of the manuscript), previously reported in Supplementary Material.

- We add the section “Data Availability” and “Code Availability” at the end of “Methods” section.

- Figure 1 (a) and (b) are replaced by a single figure 1, in which 1 (b) becomes an inset. Therefore, we also change the caption of figure 1 and every reference to Fig. 1 (a) and (b) thorough the manuscript with “Fig. 1” and “inset of Fig. 1”, respectively.

- Figure 3: we change the x-axis into “lattice position along x (j)” and each reference to lattice “site” in the caption of Figure 3.

- Figure 5: we change the x-axis into “lattice position along x (j)”.

- Figure 6: we include additional panels.

- We add the affiliation of Roberta Caruso:

  Brookhaven National Laboratory, Upton, New York 11973-5000, USA.

- We remove Francesco Tafuri’s affiliation “[3]CNR-SPIN; UOS Napoli; Monte S. Angelo; via Cinthia; I-80126 Napoli; Italy”.

- We add in Bibliography and in the main text references to:

  M. Minutillo, R. Capecelatro, and P. Lucignano, Realization of 0-π states in SFIS Josephson junctions. The role of spin-orbit interaction and lattice impurities, arXiv preprint arXiv:2108.04292 (2021).

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have made great efforts to thoroughly and carefully address the comments raised by the referees. In my opinion, given the impact of the research and the satisfying revisions, this manuscript deserves to be accepted to Commun. Phys. for publication.

Reviewer #3 (Remarks to the Author):

After carefully reading the rebuttal letter and the new manuscript, I thank the authors for the effort taken answering my questions and implementing my suggestions. They have satisfactorily addressed all of my concerns, and, in my opinion, also the most important issues raised by the other referees. Consequently, I find the new version of the manuscript improved, clearer and well structured.

As I mentioned in my previous report, the comparison between theory and experiment presented in this manuscript, together with the analysis of the induced pairing correlations, are convincing results which would undoubtedly be of interest for researchers in the community. Therefore, I recommend that the manuscript is accepted for publication in Communications Physics.

Dear Editor,

We would like to thank the Referees for their overall positive judgment of our work. We are very pleased by the fact that all of them assess that the manuscript contains results of high profile and of interest for a broad community. The Referees do not suggest additional modifications to the manuscript.

REVIEWERS' COMMENTS:

Referee #1

The authors have made great efforts to thoroughly and carefully address the comments raised by the referees. In my opinion, given the impact of the research and the satisfying revisions, this manuscript deserves to be accepted to Commun. Phys. for publication.

We thank the Referee#1 for his/her positive comments.

Referee #3

After carefully reading the rebuttal letter and the new manuscript, I thank the authors for the effort taken answering my questions and implementing my suggestions. They have satisfactorily addressed all of my concerns, and, in my opinion, also the most important issues raised by the other referees. Consequently, I find the new version of the manuscript improved, clearer and well structured.

As I mentioned in my previous report, the comparison between theory and experiment presented in this manuscript, together with the analysis of the induced pairing correlations, are convincing results which would undoubtedly be of interest for researchers in the community. Therefore, I recommend that the manuscript is accepted for publication in Communications Physics.

We thank Referee#3 for his/her positive comments.