Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Rogers and coworkers present a study of thin film structures coupling a conventional superconductor to a spin–active Cu-C60 interface. This is quite an impressive work, combining several elaborate experimental characterization tools together with inhomogeneous superconductivity calculations. The paper convincingly demonstrates i) the spin-activity of the Cu-C60 interface, ii) the superconducting proximity coupling between the Nb and the C60 layers, and iii) an enhanced magnetic penetration (weakened diamagnetism) in the superconductor, in structures displaying the spin-converter interface. The rest of the study aims at establishing that this last effect is due to a paramagnetic contribution, which is imputed to a superconducting spin-triplet component. The analysis supporting this last claim is scientifically sound, and I am really impressed by the amount and complexity of samples studied. My only criticism would be that while the manuscript text is conservatively stating merely that the observed reduction of magnetic screening is consistent with superconducting triplet correlations, this becomes a firm conclusion in the last few sentences (and in the title). I would thus recommend either circumstantiating this claim (and weakening the title), or discussing a few alternative scenarios and how to discard them. This is maybe a bit naïve, but how about for example paramagnetic contributions from conventional superconducting bound states, which might be present at the diverse interfaces (see for instance Phys. Rev. Lett. 80, 5782 (1998) and Phys. Rev. Lett. 82, 3336 (1999))?

Besides this point, I think this is a really nice work and could definitely deserve publication in Communications Physics.

Minor note:
- There is a typo at the end of Fig. 3 caption; the last “313 nm” should read “218 nm” I guess.

Reviewer #2 (Remarks to the Author):

The manuscript titled “Spin-singlet to triplet Cooper pair converter interface” written by Rogers et al. reports a careful study of hybrid superconducting/molecular/normal metal structures. The properties of these structures were characterized by a combination of electron transport, magnetometry and low-energy muon spin rotation measurements. Transport properties were studied as a function of the molecular film (here C⁶⁰ molecules) thickness, which show to influence the critical temperature. Spin-ordering in the heterostructures was investigate by low-energy muon spin rotation capable to directly probe the Meissner state across the sample depth. These measurements revealed that the integration of a Cu/C60 spin-converter interface in the heterostructure results in a paramagnetic screening contribution that the authors attribute to a conversion of spin-singlet Cooper pair states into odd-frequency triplet states. The manuscript reads smoothly, and results are clearly reported. Measurements discussion is fair as well as the literature discussion. I recommend publishing the manuscript without further modifications.

Reviewer #3 (Remarks to the Author):

The authors investigate the Meissner response of the superconducting junctions with Cu/C_{60} spin-converter interface utilising the electron transport, magnetometry, and low-energy muon spin rotation (muSR). Comparing the muSR results of the junctions with and without a spin-converter interface, they concluded the paramagnetic Meissner effect occurs in the junction with a spin-converter interface. The paramagnetic Meissner effect is induced by the odd-frequency spin-triplet s-wave pairs generated by the spin-rotational symmetry breaking by the spin-converter interface. In order to make their conclusion more sound, they prepared the so-called multi-probe samples with and without a spin-converter interface, where they measure the local magnetic field and find the magnetic-field enhancement by the spin-converter interface.

I think the manuscript is clearly written, and presents very interesting results, and hence I recommend publication. However, I do have some questions/comments I would like the authors to consider.

1. I would like the authors to clarify what is the most significant difference between an N/SAI/S junction and N/C_60/Cu/C_60/S one, where N, SAI, S means the normal-metal, spin-active interface such as thin film of a ferromagnet, and superconductor. I understood that Cu/C_60 interface acts as a sort of ferromagnet. Is there any phenomenon which is unique to Cu/C_60 but not to an SAI? Is there any advantage to use Cu/C_60 instead of an SAI?

2. The authors mention the long-range spin-triplet component (LRTC) in Introduction. This component can penetrate into the ferromagnetic metal in a longer length scale than the spin-singlet component. Is this relevant to this study? I think in this study the triplet pairs (whether Sz=\pm 1 or 0) penetrate into Au.

3. Regarding the theoretical part. The authors use the Usadel formalism with the Nazarov boundary condition. Is this formalism valid even in this complicated junctions? There are a spin-converter interface and fullerene layers. The untypical interface would require the extension of the boundary condition. In addition, the electronic structure of fullerene may influence the Usadel equation and the fullerene layers would not in the diffusive limit probably. It would be appreciated the authors clarify those points.

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Finally, I list my minor suggestions for improvements.

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Besides this point, I think this is a really nice work and could definitely deserve publication in Communications Physics.

We thank the reviewer on their positive assessment and recognising the effort to put together different complex techniques. We agree that the final conclusion would be strengthened by elaborating upon the alternative scenarios that could indeed result in the weakened diamagnetism of the SC layer and enhanced paramagnetism in the normal metal –please see the highlighted text in the manuscript.

The reviewer points out that they are impressed by the number and complexity of the number of samples we chose to study. It was indeed with consideration of the possible phenomena present at these diverse interfaces that we decided to do so. However, we acknowledge that this could have been better discussed in the previous version of the manuscript. To answer the reviewer's question directly: orbital paramagnetism (Phys. Rev. Lett. 80, 5782 (1998)) or interfacial trapped π-states (Phys. Rev. Lett. 82, 3336 (1999)) may be disregarded since we compare heterostructures where the N/S interface is consistently chosen to be a Nb/C60 interface. We should therefore maintain consistent changes of DOS over the interface in both the spin-converter and control samples. Furthermore, Phys. Rev. Lett. 80, 5782 (1998) suggests an upper field limit of 0.1 G for the paramagnetic instability originating from the orbital paramagnetism. We, therefore, ensured that we exceeded this field condition by measuring in a substantially larger field. Nonetheless, in the previous supplementary information, we did consider how such phenomena would manifest in the conducted muon measurements. If there were paramagnetic screening currents / trapped flux /vortex states highly localised to the N/S interface, we would have expected to see a *significantly larger depolarisation* rate of the muons that stop In this region for the sample where we observe the paramagnetic effect. By contrast, we see *similar or slightly smaller* values of the spin-converter sample's depolarisation rate, allowing us to disregard such phenomena.

We are pleased to add the suggested references in addition to a strengthened discussion in the main manuscript. We hope that the reviewer agrees that a reader would now benefit from greater clarity over the assignment of the paramagnetic screening effect to the presence of odd-frequency correlations.

Minor note:

- There is a typo at the end of Fig. 3 caption; the last "313 nm" should read "218 nm" I guess.

We also thank the reviewer for pointing out the typo. This has now been changed to read "218 nm".

<u>Reviewer #2 (Remarks to the Author):</u>

The manuscript titled "Spin-singlet to triplet Cooper pair converter interface" written by Rogers et al. reports a careful study of hybrid superconducting/molecular/normal metal structures. The properties of these structures were characterised by a combination of electron transport, magnetometry and low-energy muon spin rotation measurements. Transport properties were studied as a function of the molecular film (here C^{60} molecules) thickness, which show to influence the critical temperature. Spin-ordering in the heterostructures was investigate by low-energy muon spin rotation capable to directly probe the Meissner state across the sample depth. These measurements revealed that the integration of a Cu/C60 spin-converter interface in the heterostructure results in a paramagnetic screening contribution that the authors attribute to a conversion of spin-singlet Cooper pair states into odd-frequency triplet states. The manuscript reads smoothly, and results are clearly reported. Measurements discussion is fair as well as the literature discussion. I recommend publishing the manuscript without further modifications.

We are pleased that the reviewer is satisfied with the quality of the work, which we hope is further improved in the revised version.

Reviewer #3 (Remarks to the Author):

The authors investigate the Meissner response of the superconducting junctions with Cu/C_{60} spin-converter interface utilising the electron transport, magnetometry, and low-energy muon spin rotation (muSR). Comparing the muSR results of the junctions with and without a spin-converter interface, they concluded the paramagnetic Meissner effect occurs in the junction with a spin-converter interface. The paramagnetic Meissner effect is induced by the odd-frequency spin-triplet s-wave pairs generated by the spin-rotational symmetry breaking by the spin-converter interface. In order to make their conclusion more sound, they prepared the so-called multi-probe samples with and without a spin-converter interface, where they measure the local magnetic field and find the magnetic-field enhancement by the spin-converter interface.

I think the manuscript is clearly written, and presents very interesting results, and hence I recommend publication. However, I do have some questions/comments I would like the authors to consider.

We thank the reviewer for the positive and constructive comments which have addressed point by point below.

1. I would like the authors to clarify what is the most significant difference between an N/SAI/S junction and N/C_60/Cu/C_60/S one, where N, SAI, S means the normal-metal, spin-active interface such as thin film of a ferromagnet, and superconductor. I understood that Cu/C_60 interface acts as a sort of ferromagnet. Is there any phenomenon which is unique to Cu/C_60 but not to an SAI? Is there any advantage to use Cu/C_60 instead of an SAI?

We thank the reviewer for the interesting question. The Cu/C_{60} interface is essentially acting as a weak, thin ferromagnetic layer with localised spin ordering. However, we have drawn a distinction because this behaviour is not intrinsic, it only emerges when the molecule and metal are brought together, and the magnetic properties of this layer are not the same as a thin ferromagnet or even a superparamagnet (please see our previous work in Nature 524, 69–73(2015) and PNAS, 114(22), 5583 (2017)). We don't see suppression of the superconducting condensate (i.e. lower Tc) when interfacing with the spin converter structure, as we would expect for a superconducting/ ferromagnet interface. Therefore, we are cautious about assigning the `spin-active' term, given the usual inference of conventional ferromagnetic layers. However, we would be happy for the convention to be changed from spin-convertor to spin-active throughout the manuscript if the reviewer recommends that it is more appropriate to do so? In considering this comment, we noticed a break in our nomenclature in Fig. 3(e) where we label the Cu/C_{60} interface as $I_{S\text{-}A}$. We have changed this to $I_{S\text{-}C}$ as it is throughout the text.

In a broader context, we note that in terms of engineering interfaces for future technological applications, the ability to use simple, low-cost and widely available non-magnetic materials to engineer tuneable interfaces capable of spin manipulation could offer several advantages over conventional ferromagnetic elements. There is, therefore, perhaps some advantage in maintaining the distinction, even though we acknowledge that the interfaces are in this context fundamentally “spin-active”.

2. The authors mention the long-range spin-triplet component (LRTC) in Introduction. This component can penetrate into the ferromagnetic metal in a longer length scale than the spin-singlet component. Is this relevant to this study? I think in this study the triplet pairs (whether Sz=\pm 1 or 0) penetrate into Au.

The reviewer makes a good point. We feel it is worth mentioning the LRTC as it provides important technological context, especially for readers from molecular magnetism or spintronics backgrounds. As implied by the reviewer, there is no ferromagnetic layer in our structures that would necessarily generate S_z=1 pairs (though the possible presence of spin disorder at the interfaces does not rule out this possibility. Therefore, we have added the statement: "A possible future development would be the addition of a spin-selective ferromagnetic layer that could be used in conjunction with the spin converter interface to generate the Sz=1 LRTC. The triplet filtering can be used for super-spintronics or in coherence and interferometry applications." to the part of the conclusion where we relate the work to the technological application of the LRTC, whilst adding clarity to the fact that we aren't only probing the Sz=1 triplet state. Also, we have slightly amended the wording of the original statement in the introduction to: "Typical super-spintronic devices require the generation of an $S_z = \pm 1$ long-range spin-triplet component (LRTC) to preserve the superconducting correlations inside ferromagnetic layers."

We believe these additions now make it more clear that the LRTC is not as necessary in this system given that we don't include any ferromagnetic layers, as suggested by the referee.

3. Regarding the theoretical part. The authors use the Usadel formalism with the Nazarov boundary condition. Is this formalism valid even in this complicated junctions? There are a spin-converter interface and fullerene layers. The untypical interface would require the extension of the boundary condition. In addition, the electronic structure of fullerene may influence the Usadel equation and the fullerene layers would not in the diffusive limit probably. It would be appreciated the authors clarify those points.

We want to thank the reviewer for their insightful comment regarding this theory. Indeed, the Nb/C60 is a very untypical interface. Through our transport measurements, we observe that it effectively proximitises as if it were metallic, albeit with an effective Cooper pair propagation length that is likely limited by the small mean free path in the molecular layer. We accept that the chosen framework is limited in its accurate description of the flux screening in the full S/M/N system. Therefore it is highly possible that the calculated value of the penetration depth for the N/S model may not accurately represent what may be happening in the system. We would, however, like to emphasise that our use of the theoretical framework is really to demonstrate the significant difference between the temperature-dependent measured data in Fig. 4, which is dominated by the screening signal in the niobium layer only (due to the formation of muonium), and that which would be expected for a relatively straightforward model of the S that assumes no modification of the order parameter in the Nb layer. Despite the naivety of the N/S model, we see remarkable reproduction of the measured data in Fig. 4(d) whilst using input parameters derived from the transport measurements in the C60, which suggests that any modification of the theory needed to adapt to the molecular layer should at least reproduce the measured dependence of <B>(T) inside the Nb. Because of our awareness of these limitations, we are careful not to suggest that we can model the expulsion in the Au or C60 layers. This would certainly need more advanced theory.

To acknowledge the suggested limitations in the theoretical framework in this regard, we have amended some of the text. This includes the addition: "It is important to note that we cannot accurately describe the complex S/M/$I_{S\text{-}C}$/M/N structure. However, since the proximitised C_{60} behaves metallically, we can describe it as a simple S/N junction to simulate its effect on the S-layer. Using the experimentally observed mean free path of the C_{60} and interface parameters tuned to match the calculated Tc with that found experimentally, we expect that the calculated suppression of the order parameter inside the S-layer its response to a small externally applied field, are not unrealistic"

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- H. Walter et al. PRL (1998) [10.1103/PhysRevLett.80.3598].
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We thank the reviewer for the suggested texts, which we found very interesting and highly relevant. These have now been referenced within the main manuscript.

Finally, I list my minor suggestions for improvements.

5. The authors take the vertical axis of Fig.2(b) for "Tc suppression'. However, I think Tc/Tc_{control sample} vs C_{60} thickness is more intuitive with which we can see the recovery of Tc with increasing the C_{60} thickness.

We thank the reviewer for this suggestion. We are happy to change the normalisation to make it more intuitive to the reader. It has been modified as suggested. The main text now includes the phrase "The S/M/N transition temperature has been normalised with respect to the Nb control film and is shown in Fig. 2(b)."

6. At the bottom of page 12, there is a typo "consistent with consistent with".

We also thank the reviewer for pointing out the typo. The repeated phrase has been removed.

<u>Additional revisions</u>

We noticed a format error in the caption of Table1 where a misplaced $ sign has now been removed.

We noticed further deviations from the I_{S-C} labelling in supplementary information Fig. S3

The author S. Lee has been amended to S.L. Lee

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

All questions and comments were satisfactorily addressed. I recommend publication.

Reviewer #3 (Remarks to the Author):

The authors addressed all of my concerns. The manuscript is very clear to me and general readers. The contents are fascinating and useful. I would like to recommend the publication.