Evidence for spin-selectivity of triplet pairs in superconducting spin-valves

N. Banerjee[1], C. B. Smiet[1], R. G. J. Smits[1], A. Ozaeta[2], F. S. Bergeret[2], M. G. Blamire[1], J. W. A. Robinson[1]*

[1]Department of Materials Science and Metallurgy, University of Cambridge, 27 Charles Babbage Road, Cambridge CB3 0FS, United Kingdom

[2]Centro de Física de Materiales (CFM-MPC), Centro Mixto CSIC-UPV/EHU, Manuel de Lardizabal 4, E-20018 San Sebastián, Spain.

* jjr33@cam.ac.uk

Spin selectivity in a ferromagnet results from a difference in the density of up- and down-spin electrons at the Fermi energy as a consequence of which the scattering rates depend on the spin orientation of the electrons. This property is utilized in spintronics to control the flow of electrons by ferromagnets in a ferromagnet (F1)/normal metal (N)/ferromagnet (F2) spin valve, where F1 acts as the polarizer and F2 the analyzer. The feasibility of superconducting spintronics depends on the spin sensitivity of ferromagnets to the spin of the equal-spin triplet Cooper pairs which arise in superconductor(S)-ferromagnet(F) heterostructures with magnetic inhomogeneity at the S-F interface. Here, we report a critical temperature dependence on magnetic configuration in current-in-plane F-S-F spin-valves with a holmium spin mixer at the S-F interface providing evidence of a spin selectivity of the ferromagnets to the spin of the triplet Cooper pairs.

The dependence of the critical temperature (T_C) of ferromagnet-superconductor (F-S) bilayers has its origin in the spatially oscillating components in the wave function of the singlet Cooper pairs induced by the exchange field in the F layers.[1] The exchange field introduces a momentum mismatch between the spin-up and spin-down electrons that form a singlet pair and this results in a weak oscillatory dependence superimposed on the decrease of T_C with increasing thickness of the F layer.[2-4] In a F-S-F spin-valve the reversal of one F layer modifies the spatial properties of the decaying oscillation, resulting in a spin-switch effect in which the T_C is greater when the F layer moments are antiparallel (AP) than when they are parallel (P).[5,6,7] This (standard) spin-switch effect has been experimentally demonstrated[8,9].

This behavior should be substantially modified in S-F systems in which conversion between singlet pairs and odd-frequency spin-one triplet pairs is possible, for example by introducing magnetic non-collinearity (spin-mixer layers) at the S-F interface.[10,11] Since spin-one triplet pairs are immune to pair breaking by the exchange field in F, the proximity effect coupling between S and F layers is enhanced. As with the conventional superconductor – normal metal proximity effect, the increased “leakage" of pairs from the S layer should reduce the singlet pair amplitude within it and hence decrease the T_C of the structure.[12] A number experiments have been performed to test these predictions: for example in S-F'-F structures, Leksin *et al.*[13] and Zdravkov *et al.*[14] both recently demonstrated a minimum in T_C when the F and F' layers were orthogonal – the configuration which theoretically maximises singlet-triplet pair conversion.

More direct evidence for the generation of triplet pairing was obtained from S-F-S Josephson junctions in which, if interfacial spin-mixer layers were present, supercurrents could be measured through F-layer thicknesses much larger than the singlet coherence length.[15-20] Since spin-one triplet pairs, unlike singlet pairs, can carry spin these results mean that combining superconducting- and spin-electronics (superconducting spintronics) opens up real potential for practical low temperature applications.[21]

Conventional spintronics relies on the spin-selectivity of ferromagnets, which originates from the difference between the spin-up and spin-down density of states at the Fermi level. By extension, the realisation of superconducting spintronics requires a selectivity between spin up-up and spin down-down triplet pairs; however, the experiments performed to date are not spin-sensitive and the polarisation of a triplet supercurrent is so far unknown. In this Letter we report experimental results which demonstrate a T_C-dependence on the magnetic configuration in current-in-plane F-S-F spin-valves which incorporate a Ho spin mixer layer at the S-F interface, providing the first evidence for a sensitivity to triplet pair spin direction in a ferromagnet.

The experiments were performed on Py(8nm)/ Ho(d_{Ho})/ Nb(d_{Nb})/ Ho(d_{Ho})/ Py(5nm)/ FeMn(5nm) heterostructures (Fig. 1(a)). The bottom Py ($Ni_{80}Fe_{20}$) layer is pinned by exchange bias to FeMn while the orientation of the free (top) Py layer can be switched by applying an in-plane magnetic field that is greater than its coercive field so P- or AP- states can be achieved. The Nb is interfaced by Ho since this rare-earth helimaget has previously been shown by our group[18] and by Sosnin *et al.*[22] to be a spin mixer. The stacks were capped with 4 nm of (non-superconducting) Nb to prevent oxidation of the top Py layer. The central Nb layer thickness (d_{Nb}) was varied to achieve a superconducting transition in the 2 - 7 K range. The thicknesses of the Ho layers (d_{Ho}) were varied in the 0 - 7 nm range.

Results

In Figs. 1(b)-1(d) we have plotted the magnetization vs in-plane magnetic field for several spin-valves with different Ho layer thicknesses close to 20 K measurements using a cryogen-free vibrating sample magnetometer. At high positive fields the spin-valve is in a P-state and as the magnetic field direction is reversed, the free Py layer switches and a stable AP-state is obtained. Upon increasing the negative field the exchanged-biased Py layer eventually switches so the spin-valve returns to the P-state. The increase in coercivity of both Py layers (Fig. 1 inset) and a reduction in the field range of the AP state as a function of Ho layer thickness suggest an exchange coupling at the Ho/Py interfaces.

Magnetic field- and temperature-dependent resistance measurements of unpatterned samples were performed using a 4-point technique in a pulse-tube cryocooled measurement system at a constant current of 200 μA. The temperature stability of the system was better than 5 mK. We investigated the behavior of the spin-valves with several different Ho and Nb layer thicknesses by measuring the resistance (R) of the structures in both the P- and AP-states by sweeping the applied field (H) from positive to negative values and back again. For comparison we also measured R as a function of temperature T for both the P- and AP-states. However, because the effect of the magnetic state on T_C is in the mK range, R-H at constant temperature was found to be a more reliable probe of the superconducting state.

Figure 2 shows R-H data from two different samples with different Nb and Ho layer thicknesses. We first focus on the plots 2(a,b)(i) where each spin-valve contains Ho layers. The different lines represent measurements taken at different temperatures within the resistive transition of the sample and no offset has been added in plotting the data: each curve consists of a forward and a backward field sweep. The data shows a pronounced, but temperature dependent, peak in R at the fields corresponding to the AP-state meaning that, in all cases, the T_C is lower in the AP-state than in the P-state (i.e. the inverse of the standard spin-valve effect discussed in the introduction). The change in resistance for each curve (called 'magnetoresistance' here, MR) is plotted in Fig. 2(a,b)(ii), where the MR is given by (R(AP)-R(P))/R(P). The MR increases as the temperature decreases with no MR above $T_{C.}$ This implies, the MR is related to a change in T_C induced by the magnetic state of the valve and is not due to conventional magnetoresistance effects due to differential scattering of spin-up and spin-down electrons in the Py layers.

We have also directly measured the R-T dependence at zero-field for P and AP orientations as shown in Fig. 2(a)(ii). The P-state was set by applying a positive saturation field followed by a reduction to zero field, or in the AP-state by first applying a large positive field, ramping to -15 mT and then to zero field. In both cases T_C(P) is of the order of 10 mK larger than T_C(AP).

The behavior of these structures is the inverse of the standard behavior seen for simple F-S-F structures with no mixer layers.[8,9] In order to confirm this, we have also measured structures without any Ho (Fig. 2(c)). In order to achieve a measurable T_C, the thickness of the Nb layer was increased to 32 nm to compensate the additional suppression of T_C since the Py layers are in direct contact with Nb. The pink curve shows the R as the field is swept from positive to negative, and shows a clear dip from around -1 to -18 mT, and the reverse scan (blue curve) shows a dip of similar magnitude from 1 to 4 mT: thus a standard spin-switch effect is observed in this case. There is a prominent asymmetry in the width of the dips in the forward and reverse field directions which is also observed (less prominently) in the R(H) data with Ho. This is explained in the supplementary information where it is shown that the asymmetries are related to the magnetic state in the spin valves, which can be understood with reference to the hysteresis loops for the corresponding Ho thickness.

To summarise, the data in Fig. 2 shows two behaviors: in the absence of Ho a standard spin-valve effect is observed but the addition of Ho results in an inverse spin-valve effect in which the T_C is enhanced when the Py layers are P rather than AP.

Discussion

Before considering an explanation for this behavior involving the generation and diffusion of spin-triplet pairs we will first discuss possible effects arising from fringing fields[23-26,] and spin-imbalance[27] in Nb; factors which have been advanced as enhancing T_C in the P-state. In the fringing fields scenario T_C could be suppressed as a consequence of magnetic dipolar coupling between the two F (Py) layers which introduces flux into the superconductor (Nb). This suppression should be greatest close to the coercive field of the Py layers when the density of the fringing fields from Neel domain walls is maximized.[21] This explanation cannot, however, explain our results, as samples without Ho show a well-defined *decrease* in R at coercivity and in the AP state, which translates into an enhancement rather than a decrease of T_C (similar results are reported in Ref. 9). Even if we assume that exchange coupling with Ho introduces additional domain walls into the Py, given the high saturation field of Ho thin-films (~4T)[18,28] the domain configuration would hardly change in the field range investigated and so we would expect to see a monotonic increase in resistance with field rather than a sharp switching as observed in our data. Moreover we see an asymmetry in the resistance peaks in R(H) measurement for samples with Ho which clearly suggests that the R(H) measurements reflect the exchange-bias observed in the hysteresis loops and thus the magnetic state of the spin valve is the underlying factor responsible for the observed resistance peaks. This is discussed in detail in the supplementary information.

We now consider the spin-imbalance scenario in S of an F/S/F spin-valve[27] which assumes that spin-polarized quasiparticles flowing in the F layers scatter into the S. Since their transmission probability into the opposite F layer is smaller in the AP case, the spin-imbalance induced in the S is larger than in the P case. In other words, the spin-sensitivity to the relative F orientation is via quasiparticles. This scenario was originally introduced to explain inverse spin-valve behavior in Py/Nb/Py structure similar to our own, but more recent work on the same structures[26] suggest that a fringing field model is actually more likely. In any case spin-imbalance in the spin-valves structure with Ho is much less likely than in the simple F-S-F structures which show the standard spin-switch behavior. This is because since Ho is strongly spin-scattering, the up- and down-spin channels mix and the mean free paths for scatter is around a nm (estimates from resistivity measurements of Nb/Ho/Nb junctions[26]). Therefore, the net flow of scattered quasiparticles carries little or no net spin into the Nb. Even in the absence of Ho we see no evidence of spin-accumulation since in the transition region the resistance is lowest in the AP-state, which implies the short spin-diffusion length of Py is enough to nullify the net spin of the quasiparticles. This is consistent with Ref. 9, which reports Py/Nb/Py spin-valves. The combined scattering in Py and Ho means that we can rule out spin-accumulation as a possible explanation of our data.

This analysis eliminates both fringing field and quasiparticle scattering as a means of explaining the inverse spin-switch behavior seen in our Py/Ho/Nb/Ho/Py devices and so we consider instead the possibility of spin-one triplet pairs as the transmitters of spin from one F layer to another.

In the spin-valves with Ho we assume the presence of both singlet pairs and spin-one triplets pairs (which are generated by Ho) and so the observed T_c change with magnetic configuration of the spin valves must result from a combination of the standard spin-switch effect and the transmission of spin-one triplet pairs. The standard spin-switch effect, which involves singlet and triplet pairs with zero-spin projection, will always favor a higher T_c in the AP-state since the average exchange field acting on the pairs is lower. This necessarily implies that the increase in T_c observed in the P-state in samples with Ho must be linked to the transmission of triplet pairs and not to the singlet pairs. Below we bring forward the most likely explanation of how the triplets control T_c.

If up-up and down-down triplet pairs were equally involved in proximity coupling with the F layer then the T_C suppression would be independent of the magnetic orientation of the spin-valve. Instead we assume that at the interface those triplet pairs with spins parallel to the majority spin direction in a Py are more likely to enter it. Spin conservation at the interface therefore implies that opposite spin pairs are more likely to return to the Nb layer. On the Nb side these have a spatial range of at least the singlet coherence length[21] and so, for the spin valve systems considered here, can interact with the other S-F interface. If this F layer is AP to the other F layer then the pair has a higher probability of entering it. In other words, the proximity suppression of T_C due to the presence of spin-one triplet pairs, which is governed by the probability of pairs exiting the S, is enhanced in the AP-State in comparison with the P-State. This is consistent with our observations. The above interpretation is illustrated in Fig. 3. This explanation necessarily requires both the spin-triplet pairs to be able to cross Nb and that there is a spin-selectivity of a F to the spin of the triplet pairing. In view of recent theory[29] these assumptions are most likely to be fulfilled in our samples: firstly, spin-selectivity is an intrinsic property of ferromagnets with large spin-splitting at the Fermi level[30] and secondly all triplet components of the condensate induced at the S/F boundary can propagate into a superconductor a distance of the superconducting coherence length[29] $\xi_S = \sqrt{D/(2\pi * T_C)}$ which is 30 nm in Nb. Indeed this effect leads to a finite magnetic moment in the superconductor as predicted in Refs. [29,31] and observed in Ref. [32].

Since the magnitude change of MR (Fig. 2) for samples without Ho is smaller and of opposite sign to those samples with Ho, we can conclude that the spin-selectivity of triplet pairs is dominating the behavior of T_c over the singlet proximity effect. Exactly how these two opposing behaviours interact is hard to model since a microscopic theory involving the band structure of the F layers is required in order to account for the polarisation of the F layers.

As discussed in the introduction, spin-selectivity is an indispensible requirement for the future development of spin electronics and so this result represents an important milestone in this field.

Finally, it is worth noting the most dramatic and reproducible observations of the inverse spin-switch effect have occurred in manganite-$YBa_2Cu_3O_{7-d}$-manganite spin valves.[33,34] In both publications this was assumed to arise from the spin-imbalance effect discussed earlier. Although this scenario is more probable in high temperature superconducting (HTS) systems than in the Nb-based devices discussed here because of the availability of quasiparticle states at all energies (HTS), our results raise the intriguing possibility of the involvement of triplet pairing in this behaviour. There is increasing evidence that spin-disorder at manganite-HTS interfaces creates an intrinsic spin-mixer layer,[35-37] and the 100% spin polarisation of the manganites suggests that the dependence of the proximity effect on spin-valve configuration should be significantly higher than in systems involving half-metallic ferromagnets.

Methods

The films were grown by sputter deposition onto unheated oxidized silicon substrates in an ultra-high vacuum chamber. The chamber was cooled via a liquid nitrogen jacket to achieve a base pressure below 10^{-8} Pa. All targets were pre-sputtered for at least 15 minutes prior to film growth. The different layers were grown in 1.5 Pa of Ar in series by passing the substrates below stationary magnetrons. Growth rates were pre-calibrated by atomic force microscopy on step-edges created by partial lift-off of thin-films deposited on patterned substrates.

References

1. Buzdin, A. I. Proximity effects in superconductor-ferromagnet heterostructures. *Rev. Mod. Phys.* **77**, 935-976 (2005).

2. Jiang, J. S., Davidović, D., Reich, D. H., & Chien C. L. Oscillatory Superconducting Transition Temperature in Nb/Gd Multilayers. *Phys. Rev. Lett.* **74,** 314-317 (1995).
3. Garifullin, I. A. *et al.* H. Re-entrant superconductivity in the superconductor/ferromagnet V/Fe layered system. *Phys. Rev. B* **66,** 020505(R) (2002).
4. Zdravkov, V. I. *et al.* Reentrant superconductivity in superconductor/ferromagnetic-alloy bilayers *Phys. Rev. B*. **82,** 054517 (2010).
5. Tagirov, L. R. Low-Field Superconducting Spin Switch Based on a Superconductor/Ferromagnet Multilayer. *Phys. Rev. Lett.* **83,** 2058-2061 (1999).
6. Buzdin, A. I., Vedyaev, A. V., & Ryzhanova, N. N. Spin-orientation–dependent superconductivity in F/S/F structures Europhys. Lett. **48,** 686 (1999).
7. Baladié, I., Buzdin, Ryzhanova, A. N., & Vedyayev, A. Interplay of superconductivity and magnetism in superconductor/ferromagnet structures. *Phys. Rev. B* **63,** 054518 (2001).
8. Gu, J. Y., You, C.-Y., Jiang, J. S., Pearson, J., Bazaliy, Ya. B., & Bader, S. D. Magnetization-Orientation Dependence of the Superconducting Transition Temperature in the Ferromagnet-Superconductor-Ferromagnet System:CuNi/Nb/CuNi. *Phys. Rev. Lett.* **89,** 267001 (2002).
9. Moraru, I., Pratt Jr., W., & Birge, N. O. Observation of standard spin-switch effects in ferromagnet/superconductor/ferromagnet trilayers with a strong ferromagnet. *Phys. Rev. B* **74,** 220507(R) (2006).
10. Bergeret, F. S., Volkov, A. F., & Efetov, K. B. Long-Range Proximity Effects in Superconductor-Ferromagnet Structures *Phys. Rev. Lett.* **86,** 4096 (2001).
11. Fominov, Ya. V., Golubov, A., & Kupriyanov M. Triplet Proximity Effects in FSF Trilayers. *JETP Lett.* **77,** 510 (2003).
12. Fominov, Ya. V., Golubov, A., Karminskaya, T. Yu., Kupriyanov, M. Yu., Deminov, R. G., & Tagirov. L. R. Superconducting Triplet Spin Valve. *JETP Lett.* **91,** 308 (2010).
13. Leksin, P. V. *et al.* Evidence for Triplet Superconductivity in a Superconductor-Ferromagnet Spin Valve. *Phys. Rev. Lett.* **109,** 057005 (2012).
14. Zdravkov, V. I. *et al.* Experimental observation of the triplet spin-valve effect in a superconductor-ferromagnet heterostructure. *Phys. Rev. B* **87,** 144507 (2013).
15. Khaire, Trupti S., Khasawneh, Mazin A., Pratt, W. P., & Birge Norman O. Observation of spin-triplet superconductivity in co-based Josephson junctions. *Phys. Rev. Lett.* **104,** 137002 (2010).
16. Khasawneh, M. A., Khaire, T. S., Klose, C., Pratt, Jr., W. P., & Birge N. O. Spin-triplet supercurrent in Co-based Josephson junctions. *Supercond. Sci. Technol.* **24,** 024005 (2011).
17. Sprungmann, D., Westerholt, K., Zabel, H., Weides, M., Kohlstedt, H. Evidence for triplet superconductivity in Josephson junctions with barriers of the ferromagnetic Heusler alloy Cu_2MnAl. *Phys. Rev. B* **82,** 060505(R) (2010).
18. Robinson, J. W. A., Witt, J. D. S., & Blamire, M. G. Controlled Injection of Spin-Triplet Supercurrents into a Strong Ferromagnet**.** *Science* **329,** 59 (2010); Witt, J. D. S., Robinson, J. W. A., & Blamire, M. G Josephson junctions incorporating a conical magnetic holmium interlayer. *Phys. Rev. B.* ***85,*** 184526 (2012); Halász, Gábor B., Blamire, M. G., & Robinson, J. W. A. Magnetic-coupling-dependent spin-triplet supercurrents in helimagnet/ferromagnet Josephson junctions. *Phys. Rev. B* **84,** 024517 (2011); Usman, I. T. M. *et al.* Evidence for spin mixing in holmium thin film and crystal samples. *Phys. Rev. B* **83,** 144518 (2011).
19. Anwar, M. S., Veldhorst, M., Brinkman, A., & Aarts, J., Long range supercurrents in ferromagnetic CrO_2using a multilayer contact structure. *Appl. Phys. Lett.* **100,** 052602 (2012).
20. Klose, Carolin *et al.* Optimization of Spin-Triplet Supercurrent in Ferromagnetic Josephson Junctions. *Phys. Rev. Lett.* **108,** 127002 (2012).
21. Eschrig, Matthias Spin-polarized supercurrents for spintronics. *Phys. Today* **64(1),** 43-49 (2011).
22. Sosnin, I., Cho, H., Petrashov, V. T. & Volkov, A. F. Superconducting Phase Coherent Electron Transport in Proximity Conical Ferromagnets. *Phys. Rev. Lett.* **96,** 157002 (2006).
23. Stamopoulos, D., Manios, E., & Pissas, M. Stray-fields-based magnetoresistance mechanism in $Ni_{80}Fe_{20}$-Nb-$Ni_{80}Fe_{20}$ trilayered hybrids. *Phys. Rev. B* **75,** 184504 (2007).
24. Zhu, J., Cheng, X., Boone, C., & Krivorotov, I. N. Origin of the Inverse Spin Switch Effect in

Superconducting Spin Valves. *Phys. Rev. Lett.* **103,** 027004 (2009).

25. Flokstra, M., van der Knaap, J. M. & Aarts, J. Magnetic coupling in superconducting spin valves with strong ferromagnets. *Phys. Rev. B* **82,** 184523 (2010).
26. Steiner R. & Ziemann P. Magnetic switching of the superconducting transition temperature in layered ferromagnetic/superconducting hybrids: Spin switch versus stray field effects. Phys. Rev. B **74,** 094504 (2006).
27. Rusanov, A. Yu., Habraken, S., & Aarts, J. Inverse spin switch effects in ferromagnet-superconductor-ferromagnet trilayers with strong ferromagnets. *Phys. Rev. B* **73,** 060505(R) (2006).
28. Witt, J. D. S. *et al.* Strain dependent selection of spin-slip phases in sputter deposited thin-film epitaxial holmium. *J. Phys.: Condens. Matter* **23,** 416006 (2011).
29. Bergeret, F. S., Volkov, A. F., & Efetov, K. B. Induced ferromagnetism due to superconductivity in superconductor-ferromagnet structures. *Phys. Rev. B* **69,** 174504 (2004); *ibid*, Spin screening of magnetic moments in superconductors. *Euro Phys. Lett.* **66,** 111 (2004).
30. Grein, R., Eschrig, M., Metalidis, G., and Schön, Gerd Spin Dependent Cooper Pair Phase and Pure Spin Supercurrents in Strongly Polarized Ferromagnets. *Phys. Rev. Lett.* **102,** 227005 (2009).
31. Löfwander, T., Champel, T., Durst, J., & Eschrig, M. Interplay of Magnetic and Superconducting Proximity Effects in Ferromagnet-Superconductor-Ferromagnet Trilayers. *Phys. Rev. Lett.* **95,** 187003 (2005).
32. Xia Jing, Shelukhin, V., Karpovski, M., Kapitulnik, A., & Palevski, A., Inverse Proximity Effect in Superconductor-Ferromagnet Bilayer Structures. *Phys. Rev. Lett.* **102,** 087004 (2009).
33. Peña, V. *et al.* Giant Magnetoresistance in Ferromagnet/Superconductor Superlattices. *Phys. Rev. Lett.* **94**, 057002 (2005).
34. Pang, B. S. H., Bell, C., Tomov, R. I., Durrell, J. H., & Blamire, M. G. Pseudo spin-valve behavior in oxide ferromagnet/superconductor/ferromagnet trilayers. *Phys. Lett. A* **341,** 313–319 (2005).
35. Kalcheim, Y., Kirzhner, T., Koren, G. & Millo, O. Long-range proximity effect in $La_{2/3}Ca_{1/3}MnO_3$/(100)$YBa_2Cu_3O_{7-\delta}$ ferromagnet/superconductor bilayers: Evidence for induced triplet superconductivity in the ferromagnet. *Phys. Rev. B* **83**, 064510 (2011).
36. Kalcheim, Y., Millo, O., Egilmez, M., Robinson, J. W. A. & Blamire, M. G. Evidence for anisotropic triplet superconductor order parameter in half-metallic ferromagnetic $La_{0.7}Ca_{0.3}MnO_3$ proximity coupled to superconducting $Pr_{1.85}Ce_{0.15}CuO_4$. *Phys. Rev. B* **85**, 104504 (2012).
37. Visani, C. *et al.* Equal-spin Andreev reflection and long-range coherent transport in high-temperature superconductor/half-metallic ferromagnet junctions. *Nat. Phys.* **8**, 539-543 (2012).

Acknowledgements

This work was funded by the Royal Society through a University Research Fellowship “Superconducting Spintronics” held by J.W.A.R. M.G.B acknowledges funding from the UK EPSRC and the European Commission through an ERC Advanced Investigator Grant "Superspin". C.B.S. and R.G.J.S were supported by the Erasmus exchange programme and the Leiden Outbound Grant. C.B.S. acknowledges Prof. Jan Aarts’ for scientific input. The work of F.S.B and A. O. have been supported by the Spanish Ministry of Economy and Competitiveness under Project FIS2011-28851-C02-02. The work of A. O. have also been supported by the CSIC and the European Social Fund under JAE-Predoc program and the EU-FP 7 MICROKELVIN project (Grant No. 228464).

Author Contributions

J.J.W.A.R. had the idea of the experiment. N.B., and C.B. prepared the samples and performed the experiments. R.G.J.S. helped perform some of the transport measurements. A.O. and F.S.B. comments on the paper and performed detailed theoretical calculations in order to help us understand the experiment. J.W.A.R., N.B., and M.G.B. co-wrote the paper. All authors read the manuscript.

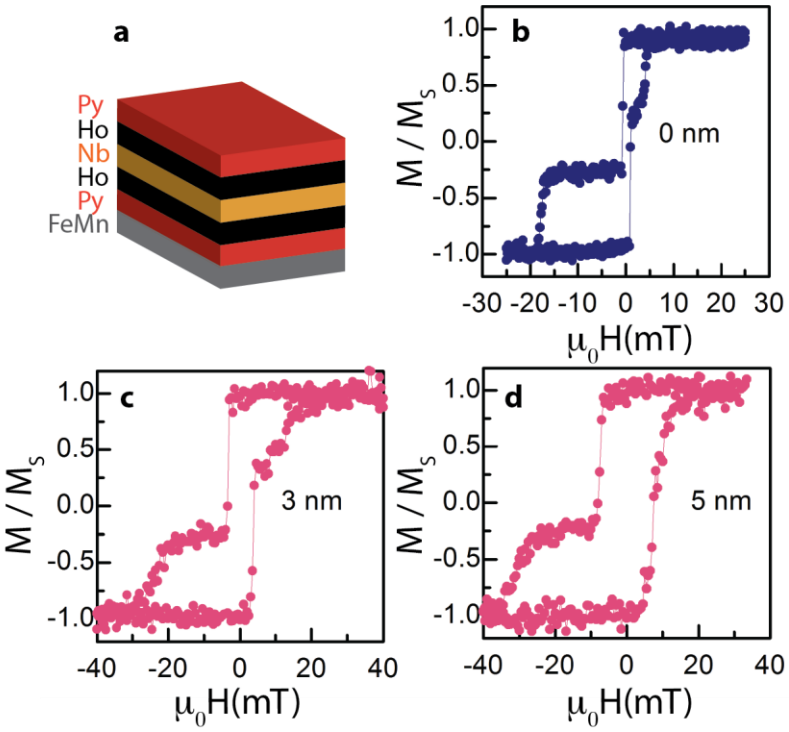


Figure 1 │Magnetic characterization of the spin valves. (a-d) The spin-valves were cooled in a positive field to 20 K to measure the M-H loops with the field applied in the plane of the samples. **(a)** An illustration of the exchange bias structures and magnetic properties of Nb(4nm)Py(8nm)/Ho/Nb/Ho/Py(5nm)/FeMn(5nm) spin-valves. **(b)** Sample without Ho layers. **(c)** Spin valve with 3-nm-thick layer of Ho and **(c)** spin valve with 5-nm-thick layer of Ho.

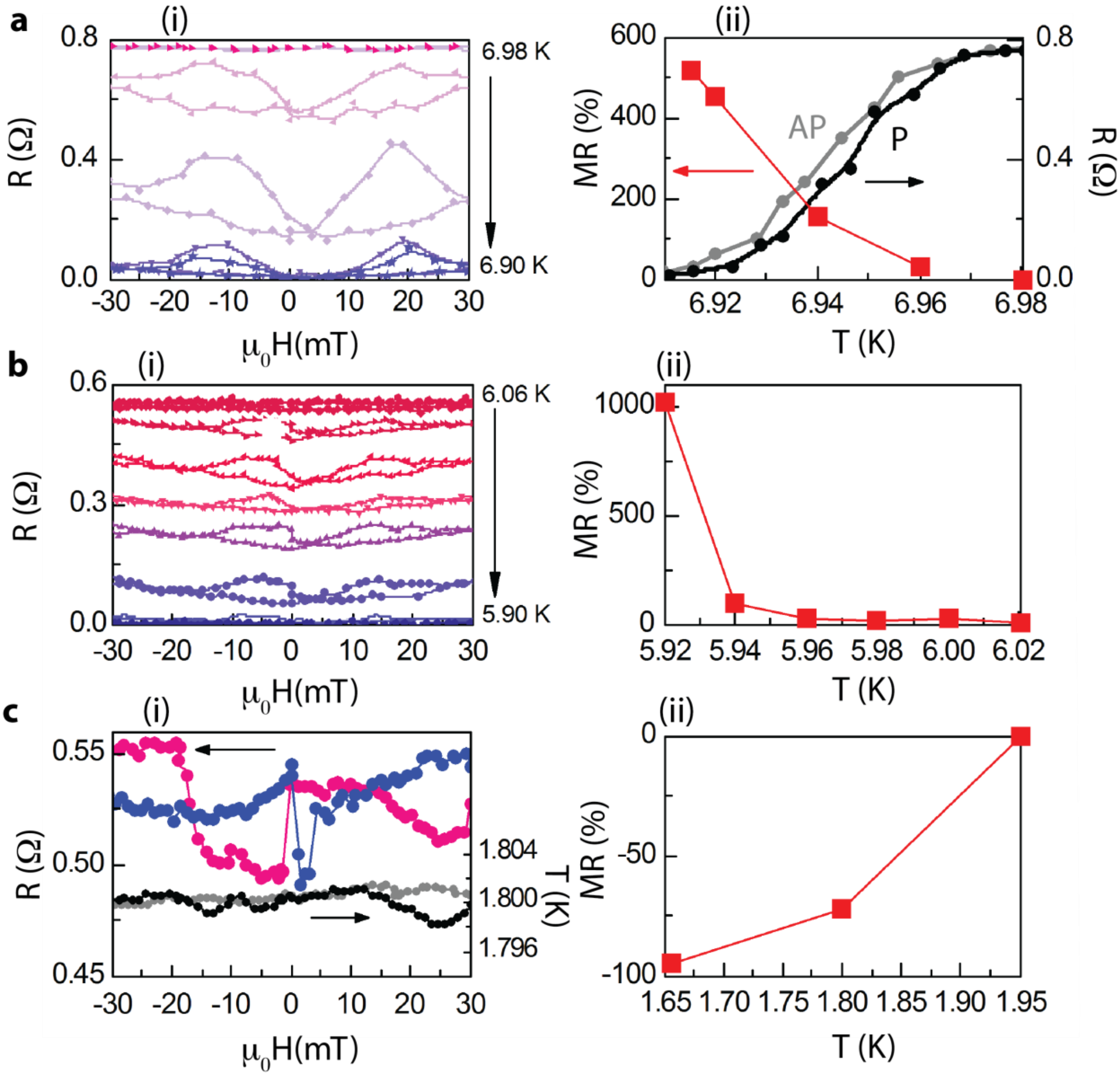


Figure 2 │ Transport properties of the spin valves. (a-c) Four point resistance measurements of Nb(4nm)/ Py(8nm)/ Ho(d_{Ho})/ Nb(d_{Nb})/ Ho(d_{Ho})/ Py(5nm)/ FeMn(5nm) spin-valves in the superconducting transition. **(a) (i)** R-H for 5-nm-thick top and bottom Ho layers and 26-nm-thick Nb layer and in **(ii)**, MR vs T and RT data for the same sample. **(b) (i)** R-H for 5-nm-thick Ho layers and 20-nm-thick Nb layer and in (b) (ii), MR vs T for the same sample. **(c) (i)** R-H for 32-nm-thick layer of Nb and no Ho (arrows indicate field sweep direction) and in **(ii)**, MR vs T for the same sample.

.

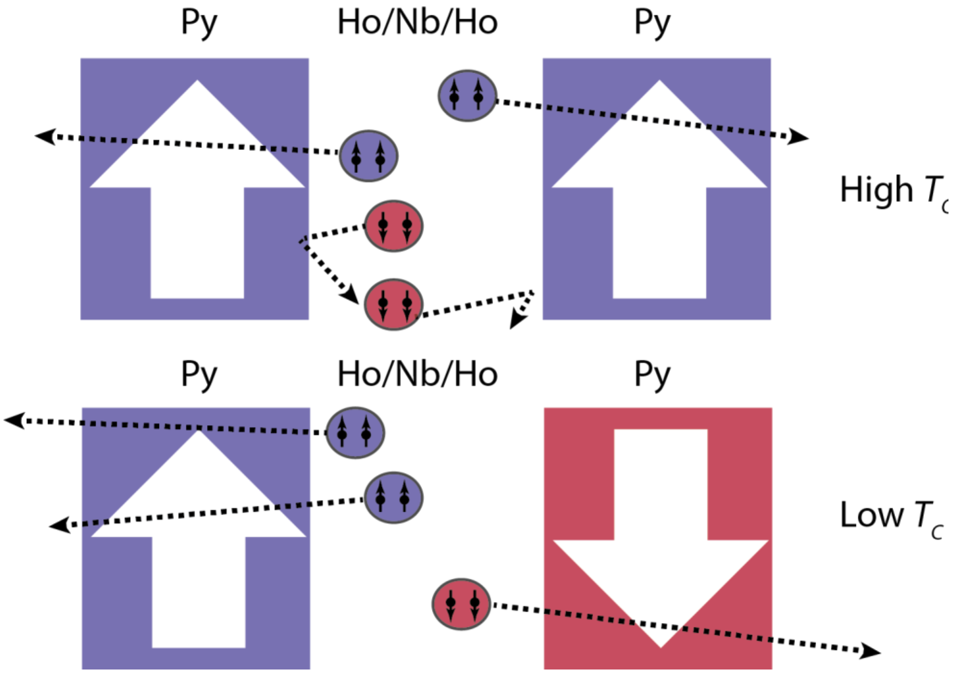
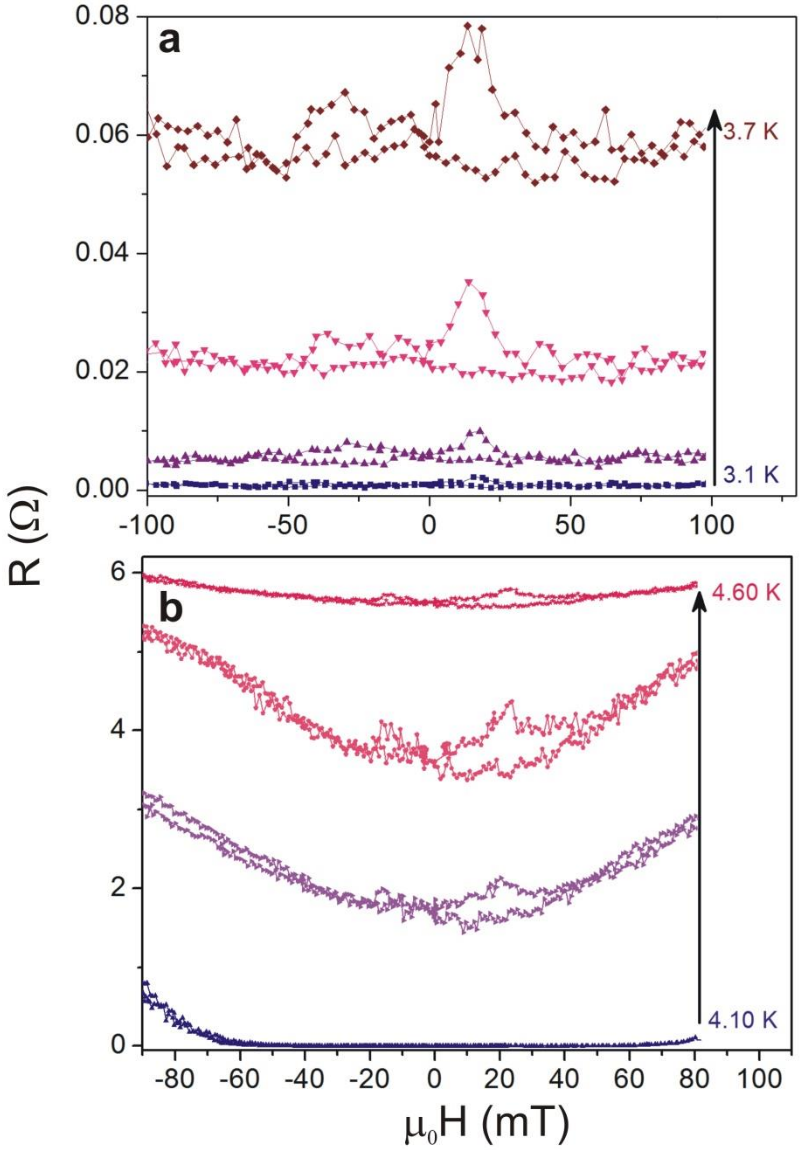
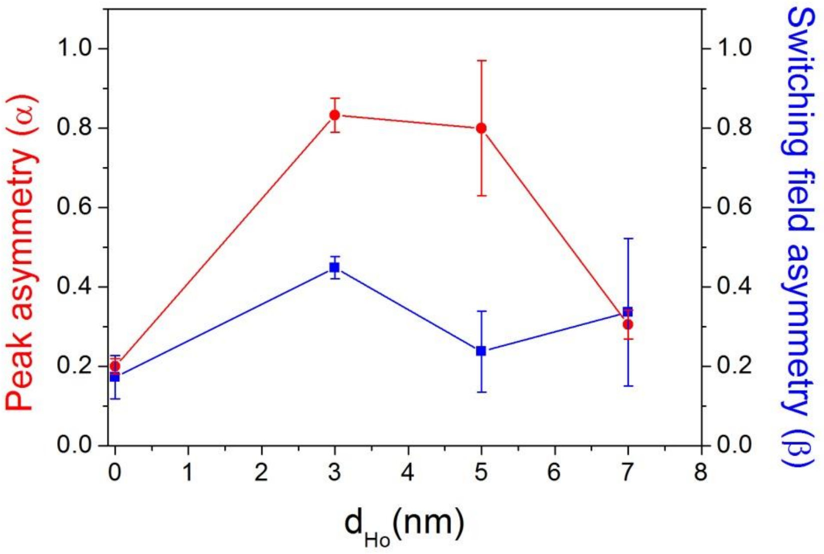


Figure 3 │ Illustration of the spin selectivity. A cartoon illustrating the possible behavior of spin-one triplet pairs in Py/Ho/Nb/Ho/Py/FeMn F-S-F spin-valves in the parallel (top) and antiparallel states (bottom).



Supplementary Figure S1: Four point resistance measurements of Nb(4nm)/ Py(8nm)/ Ho(d_{Ho})/ Nb(d_{Nb})/ Ho(d_{Ho})/ Py(5nm)/ FeMn(5nm) spin-valves in the superconducting transition. (a) R-H for 2-nm-thick top and bottom Ho layers and 26-nm-thick Nb layer. (b) R-H for 7-nm-thick Ho layers and 26-nm-thick Nb layer. The arrows show the direction of increasing temperature in equal steps with the minimum and maximum values indicated.



Supplementary Figure S2: Asymmetry in peak widths. The peak width asymmetries are calculated from hysteresis loops (α) and R(H) measurements (β) plotted as a function of Ho thickness. The error bars represent one standard deviation on each side.