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Extremely long range, high-temperature Josephson coupling across a half metallic ferromagnet

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ABSTRACT

The Josephson effect results from the coupling of two superconductors across a spacer such as an insulator, a normal metal or a ferromagnet to yield a phase coherent quantum state. However, in junctions with ferromagnetic spacers very long range Josephson effects have remained elusive. Here we demonstrate extremely long range (micrometric) high-temperature (tens of K) Josephson coupling across the half-metallic manganite $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ combined with the superconducting cuprate $\text{YBa}_2\text{Cu}_3\text{O}_7$. These planar junctions, in addition to large critical currents, display the hallmarks of Josephson physics, such as critical current oscillations driven by magnetic flux quantization and quantum phase locking effects under microwave excitation (Shapiro steps). The latter display an anomalous doubling of the Josephson frequency predicted by several theories. In addition to its fundamental interest, the marriage between high temperature, dissipationless quantum coherent transport and full spin polarization brings opportunities for the practical realization of superconducting spintronics, and opens new perspectives for quantum computing.

The antagonism between ferromagnetism, in which the exchange field tends to spin-polarize the conduction electrons, and singlet superconductivity, in which electrons form Cooper pairs with opposite spins, makes their coexistence challenging¹. In bulk samples, this has been

observed only recently in a P-doped EuFe_2As_2 compound with extremely weak exchange interaction between electrons and localized moments². Contrarily, equal-spin triplet superconductivity can survive in strong ferromagnets, although only rare bulk materials are considered triplet superconductors³.

Seminal theoretical studies⁴⁻¹⁰ showed that triplet correlations can be generated at the interface between a ferromagnet (F) and a superconductor (S). Triplets allowed explaining the observation of supercurrents across very thick (tens or even hundreds of nm) ferromagnets combined with conventional low-temperature (few K) superconductors¹¹⁻¹⁴. Despite many hints of triplet superconductivity, very long range Josephson effects have remained elusive, especially in the intriguing case of unconventional high-temperature superconductors combined with half-metal ferromagnets¹⁵⁻¹⁹. Much work has followed to identify various mechanisms that can lead to the opposite-spin singlet to equal-spin triplet conversion. Generally, these include spin-mixing, which leads to generation of opposite-spin ($S_z=0$) triplet component out of the singlet one, and spin-flip, which produces the equal-spin ($S_z=\pm 1$) triplet component. At the microscopic level, those processes result from spin dependent scattering at interfaces with strong ferromagnets⁵, and from the presence of an inhomogeneous magnetization^{4,6,8,9,20,21} or a momentum dependent exchange field due to the spin-orbit interaction^{22,23}. A number of experiments based on conventional (s-wave) low-temperature superconductors have found critical currents across S/F/S junctions for F thickness in the tens of nanometers range^{12,14,19,24,25}, which is much larger than expected for the singlet superconductor/ferromagnet proximity effect, and therefore supports the triplet scenario. In these vertical junctions, the triplet generation was engineered through the artificial design of a magnetic inhomogeneity at the interface with the superconductor, e.g. by intercalating various ferromagnetic layers with different magnetic anisotropy or spin texture^{12,14,19,24,25}. Experiments on lateral (planar) devices, particularly based on the half-metallic ferromagnet CrO_2 , found critical currents that decay over even longer distances –up to a few hundred of nm^{11,13}. Because in half-metals the conduction

electrons are fully spin-polarized, and consequently the penetration of opposite-spin singlet correlations is forbidden, that experimental observation was considered evidence for the generation of triplet superconductivity, which was explained¹⁰ by the presence of strong spin dependent scattering at S/F interface combined with intrinsic magnetic inhomogeneities²⁶.

Because of their fundamental and technological interest, heterostructures of unconventional (d-wave) high-temperature cuprate superconductors combined with half-metallic manganites have attracted much attention, and hints for triplet proximity effect have accumulated along the years: unexpected superconducting proximity effect¹⁵, an induced superconducting gap^{16,18}, Andreev reflection and coherent transport^{17,27}, as well as supercurrents¹⁹ over length scales of the order few tens-of-nm have been experimentally observed in vertical junctions (although at relatively low temperatures ~ 10 K). Notwithstanding, the demonstration of long-range Josephson effects has remained elusive. Proving Josephson coupling requires evidencing the macroscopic phase coherent state. A first signature is the observation of flux quantization effects in the critical current: in principle, the Josephson current must vanish for integer number of flux quanta, giving rise to the well-known Fraunhofer diffraction pattern. While realized in triplet Josephson junctions based on low-temperature superconductors^{25,28}, flux quantization effects have been never observed in cuprate/manganite junctions. They are clearly demonstrated in the present experiments. The second signature are the so called Shapiro steps in the $I(V)$ characteristics measured under microwave radiation²⁹. These originate from the current phase relation $I = I_0 \sin \varphi$, where the phase φ difference between the electrode evolves in time under application dc voltage bias V according to $\frac{\partial \varphi}{\partial t} = \frac{2\pi}{\Phi_0} V$, with $\Phi_0 = \frac{h}{2e}$ being the flux quantum, yielding an ac current with the Josephson frequency $f_J = \frac{V}{\Phi_0}$. Resonant absorption of microwave radiation occurs when the Josephson frequency is a multiple of the microwave's frequency, yielding steps in the $I(V)$ curve measured under microwave illumination at voltages given in the conventional case by $V = n\Phi_0 f$, with $n = 0, 1, 2, \dots$. To the best of our

knowledge, these phase locking effects have never been reported for triplet Josephson junctions. They are very clear in the present experiments, and they present the anomalous periodicity expected in the context of triplet Josephson effects^{23,30}.

We have fabricated planar S/F/S Josephson junctions [Figure 1 (a)] in which two YBCO electrodes are separated a micron apart by a LSMO wire. These microstructures were fabricated from oxide films grown epitaxially on SrTiO₃ by high pressure pure oxygen sputtering technique. LSMO layers (30 nm thick) were first patterned into a wire (20 or 25 microns wide) using electron beam lithography (see the green layout in the optical image of Figure 1 (a)). Amorphous alumina (a-Al₂O₃, hereafter ALO) templates fabricated with electron beam lithography were used to define the YBCO contacts [yellow pattern in Figure 1 (a)] separated by micron size gaps. The YBCO (50 nm thick) was grown on top of the alumina templates using the same high-pressure sputtering technique as for the manganite. Holes in the ALO template allow epitaxial growth of YBCO in the contacts, while the barrier area was protected by the thick ALO on top of which the YBCO is known to grow amorphous and highly insulating³¹ (see below). The spacing between YBCO contacts was ~1μm [see Figure 1 (b)]. An enlarged view of the portion of LSMO wire protected by the ALO stripe separating the YBCO contacts and thus defining the width of the Josephson barrier is shown in Figure 1 (c).

The ex-situ growth of the YBCO on top of the manganite wires did not degrade the structure nor the chemistry of the interfaces as compared to vertical structures grown in situ. A demonstration of the high quality of the YBCO/LSMO interface grown ex-situ is shown in the scanning transmission electron microscopy (STEM) images displayed in Figures 1 (d-f). A low magnification high angle annular dark field (HAADF) image in Figure 1 (d) shows that the sample grows flat and continuous over long lateral distances. In Figure 1 (d and e) the high-resolution image displays the YBCO/LSMO interface, which appears epitaxial, atomically smooth and free of disorder with the same crystalline quality as the interfaces grown in situ. As it turns out, the growth at 900 °C

in a pure oxygen plasma has the effect of (re)conditioning the surface after exposure to atmosphere or processing. Figure 1 (f) displays an atomic resolution Electron Energy Loss Spectroscopy (EELS) chemical map of the interface alongside a high resolution HAADF image. While the HAADF image indicates which atom corresponds to each atomic column, the colored map presents a color mixing of Ba $M_{4,5}$ (red), La $M_{4,5}$ (yellow) and Mn $L_{2,3}$ (blue) elemental EELS maps, where the interface is observed to be chemically sharp with no intermixing detected. The interfacial termination planes between YBCO and LSMO are also evidenced and, as in the in-situ samples, correspond to BaO facing MnO_2 planes with missing CuO chains at the interface^{32,33}. The Cu-O-Mn superexchange path across the interface induces a magnetic state in the Cu which may play a central role in the triplet generation^{17,27}.

To ascertain the magnetic properties of the LSMO nanowire, and particularly within the gap in between the two YBCO electrodes, we examine the magnetic domain structure of LSMO/ YBCO hybrids with the same geometry of our devices via Spatially resolved Photoemission Electron Microscopy (SPEEM) using X-ray magnetic circular dichroism (XMCD) as magnetic contrast mechanism. XMCD was measured at the Mn L_3 -edge (640.3 eV) as the normalized difference in absorption of circular polarized radiation with left and right helicity. Color code in the image of Figure 1 (g) is set by the projection of the sample magnetization along the direction of propagation of the beam. Colored arrows in the figure mark the direction of the magnetization. PEEM images of Figure 1 (g) show that the LSMO bridge can be homogeneously magnetized along [110] directions corresponding to the biaxial easy axes of the manganite, although in some situations states with coexisting micron size domains were observed at remanence. In all cases the domain geometry ensures that there are wide regions with homogeneous magnetization connecting the YBCO contacts where the triplet pairs can propagate freely.

The main panel of Figure 2 (a) shows resistance curves of three different samples with YBCO contacts of similar size separated 1 micron on LSMO wires of different widths (20 or 25 microns).

Transport was measured by injecting current between the two neighboring YBCO contacts while voltage was measured along the LSMO wire with contacts at its ends (as shown in the schematic displayed in Figure 2(b)). Between room temperature and down to the transition temperature of the YBCO (90 K), the (4-probe) resistance is dominated by the LSMO wire. This is illustrated by Fig. 2 (c), which shows that the temperature dependence of the resistance of the devices (the blue curve is in example taken from Fig. 2a) is qualitatively similar to that of a single LSMO film of the same thickness (black curve in Figure 2c). Control devices consisting of YBCO deposited on the ALO mask and without the LSMO micro wire had resistances at least four orders of magnitude higher, with the resistance exceeding in most cases the measurement limit (10 M Ω) at the temperatures of the experiment. These large resistance values correspond to the YBCO grown on top of amorphous ALO. Similarly to other oxide perovskites grown on amorphous oxides³⁴, YBCO grown on the amorphous ALO is amorphous and strongly insulating³¹. See orange curve in Figure 2 c corresponding to a device with no LSMO wire and YBCO electrodes separated 1 μm . This insulating behavior, found in each of the numerous control devices we fabricated, rules out the presence of conducting paths across ALO that may yield a short between YBCO contacts.

The YBCO/LSMO devices (Fig. 2a) display a broad resistive transition where different steps can be observed (see inset in Figure 2 (a)) when temperature is decreased below ~ 90 K. This is because proximity effect is established gradually and, given the planar geometry of the device (see sketches in Figure 2 (b) and 3 (c)), contributions from different sample regions are concurrently measured. First, the transition of the YBCO electrodes is detected. Thus, a first step in the R(T) is observed around 90 K. Due to the very short YBCO coherence length (~ 1 nm along the a-axis), the onset of superconductivity can occur at ~ 90 K far from the interface with the LCMO while, within a few unit cells from it, the proximity effect depresses T_c , and the transition is completed at a much lower temperature ~ 60 -65 K. Below this temperature, the resistance of the LSMO lying directly underneath the YBCO electrodes gradually decreases with decreasing

temperature, due to the proximity effect. However, in order to observe zero resistance and a faster drop to zero resistance, Josephson coupling need to be established between the “proximitized LSMO banks” across the $\sim 1 \mu\text{m}$ long LSMO wire that is not covered by superconducting YBCO. In the $R(T)$, the onset of Josephson coupling shows as a shoulder at ~ 40 K.

To determine the critical current of the devices, $I(V)$ curves were measured as a function of temperature (Figure 2(d)). Figure 2(d) display selected IV curves, while a full set is presented in Supplementary Figure 1. Linear scale I-V plots show considerable rounding as recently observed in other Josephson junctions with ferromagnetic barriers³⁵. Double logarithmic scale plots of the $I(V)$, see Fig. 2d, show that the critical current is indeed much lower than anticipated from the linear scale plots, and that a rather restrictive voltage criterion (\sim hundred of nanovolts) is necessary to reach the critical current regime. As discussed in the Supplementary Information Section 2, fits of the low-voltage portion of $I(V)$ curves to the Ivachenko - Zil'berman model³⁶ showed that the critical current is limited by thermal fluctuations, and that the higher voltage regime of the $I(V)$ is dominated by the contribution of parts of the planar device (YBCO electrodes and proximitized LSMO lying directly underneath YBCO) having higher resistance and critical current than the bare LSMO wire that bridges the YBCO electrodes and plays de role of Josephson barrier. The dominance of those elements' contribution, which as discussed above is evident also in $R(T)$ at sufficiently high temperatures, is unavoidable in this type of planar geometries as discussed earlier³⁵. Yet the analysis of the lower voltage range (below a few μV) allows us to estimate the critical current (see Supplementary Information Section 2), which is shown Figure 2 (e). Values in the range of $10^2 - 10^3 \text{ A/cm}^2$ are found, i.e., much lower than the critical current (a few 10^7 A/cm^2) typically found in YBCO wires at the same temperatures. The temperature dependence of the critical current was analyzed in the framework of the theory of mesoscopic diffusive SNS junctions³⁷ to obtain the Thouless energy E_{Th} , the energy scale for the mesoscopic proximity effect. Details can be followed in Supplementary Information sections 2

and 3. The Thouless energy was found to be $E_{Th} = 95 \pm 10 \mu\text{eV}$, not far from the $E_{Th} = 54 \mu\text{eV}$ reported in proximity effect experiments with CrO_2 ^{13,38}. Since the CrO_2 sample had a 700-nm gap, if we scale the 54 μeV CrO_2 Thouless energy to a sample length of 1 micron, it would be reduced by half, so we should compare 95 μeV for LSMO with 27 μeV for CrO_2 , values which are still reasonably close. This similarity is not surprising since both materials are half metal oxides with rather similar electronic properties. Measurements on devices with shorter ($L=750 \text{ nm}$) and longer ($L=2 \mu\text{m}$) LSMO spacing were in agreement with the expected $1/L^2$ scaling the Thouless energy (See Supplementary Section 3), further demonstrating the robustness of this analysis.

Measurements of the junction resistance R as a function of the magnetic field H applied in the plane of the LSMO microwire allowed for the observation of quantum interference effects demonstrative of Josephson coupling. At low temperatures (below the critical temperature of the proximitized LSMO) and sufficiently high injected current I , the junction's resistance periodically oscillates as a function of the applied field. This is shown in the example displayed in Figure 3 (a), which corresponds to a $R(H)$ measured at $T=30 \text{ K}$ with $I=25 \mu\text{A}$ and different angles θ between the applied in-plane magnetic field and the LSMO wire [geometry shown in Figure 3 (b)]. As it can be seen, the oscillations' period depends on the angle between the in-plane magnetic field and the LSMO wire. This is further evidenced in Figure 3 (c), which displays a contour plot of the resistance (color scale) as a function of the magnetic field magnitude and angle θ with respect to the LSMO wire [geometry shown in Figure 3 (b)]. The contour plot is obtained from a large set of $R(H)$, measured for varying θ (every 2° for $0^\circ \leq \theta \leq 360^\circ$) at constant $T=30\text{K}$ and $I=25 \mu\text{A}$. In this plot, the magnetoresistance oscillations show as a "wavy pattern" with mirror symmetry around $\theta=0^\circ$, 90° , 180° and $\theta=270^\circ$. The pattern results from the oscillations' period being the shortest when magnetic field is perpendicular to the LSMO wire (around $\theta=90^\circ$ and $\theta=270^\circ$) and gradually increasing as the magnetic field is rotated towards the direction of the LSMO wire ($\theta=0^\circ$ and $\theta=180^\circ$). The periodic magnetoresistance oscillations [Fig.

3 (a)] result from the Fraunhofer oscillation of the critical current as a function of the magnetic flux Φ threading the junction. The angular (θ) dependence of the oscillations [Fig. 3 (c)] results from the junction's geometry, more specifically from the angular dependence of the magnetic flux across the junction, $\Phi(H, \theta) = \mu_0 H A_{eff} |\sin(\theta)|$, with A_{eff} an effective junction area as shown in Fig. 3 (b). The resistance oscillations pattern comes from the critical current one when a magnetic flux is applied (Fraunhofer pattern). We show in figure 3(d) the calculated critical current $I_c(H, \theta) = I_{c0} \left| \frac{\sin \pi \frac{\Phi}{\Phi_0}}{\pi \frac{\Phi}{\Phi_0}} \right|$, with I_{c0} being the maximum critical current, Φ_0 the flux quantum ($2.07 \cdot 10^{-15}$ Wb), and $\Phi(H, \theta)$ as described above, for a rectangular Josephson junction, which qualitatively reproduces the experimental pattern (figure 3(c)). The period of the oscillations [~ 0.036 T for $\theta=270^\circ$] corresponds to an effective area $A_{eff} \sim 0.101 \mu\text{m}^2$. Considering the vertical dimension of the wire is limited by the LSMO thickness ($w = 30$ nm) and that the effective junction area is given by $A_{eff} = L_{eff} \times w$, we extract an effective junction width $L_{eff} \sim 3.4 \mu\text{m}$, which is in excess of the distance between the YBCO electrodes ($1 \mu\text{m}$). The origin of this L_{eff} will be discussed below.

Confirmation of the phase-locked response of the Josephson coupling is obtained from the irradiation of the sample with microwaves at different powers while the I(V) curves were recorded at a temperature close to the onset of the supercurrent in LSMO. The resonant absorption of the microwave signal by the AC Josephson current produced characteristic interference patterns evidencing phase coherence. This is clearly seen in Figure 4(a), which displays the differential resistance $dR \equiv dV/dI$ (color scale) as a function of the bias current I and microwave power, with $f=9.997$ GHz and measured at $T=37$ K. The same data can be plotted as a function of the voltage across the junction V [Figure 4 (b)], which reveals the presence of Shapiro steps at constant voltages that appear as vertical lines in the interference pattern. This resonant response constitutes direct evidence of the sinusoidal current phase relationship of the Josephson junction. Strikingly, though, steps appear at half-integer factors of the Josephson

voltage $V_{Josephson} = \Phi_0 f$ evidencing a doubling of the Josephson frequency and thus the preponderance of the second harmonic term in the current phase relation. Half-integer Shapiro steps have been predicted^{23,30,39,40} and experimentally observed in S/F/S vertical (tunnel) junctions with weak ferromagnets at the verge of the 0- π transition^{41,42}. The half-integer Shapiro steps observed in our junctions constitute a fingerprint of proximity effect governed by a slowly decaying 2nd harmonic, which is one of the expected scenarios for the long-range propagation of triplet superconductivity in ferromagnets³⁰. Interestingly, from the observation of 2nd harmonic dominance in the Shapiro steps one expects a halving of the period of Fraunhofer oscillations, that is, $\mu_0 \Delta H = \phi_0 / 2A_{eff}$. Considering this, the period of the Fraunhofer pattern (Fig. 3) yields accordingly an effective junction length of $L_{eff} \sim 1.7$ micron (instead of the $L_{eff} \sim 3.4$ micron obtained with an analysis in terms of the 1st harmonic of the current phase relation). This effective length of the Josephson junction, $L_{eff} = L + 2\lambda$, is in much better agreement with actual length of the junction, $L = 1 \mu\text{m}$, and yields a value ~ 350 nm for the YBCO penetration depth. This value, somewhat larger than the 140 nm of YBCO single crystals, is expected for a thin film in proximity with LSMO. A more accurate quantitative description of the above observations will require future theoretical studies of proximity effect between unconventional superconductors and half metals in a planar geometry.

The demonstration of extremely long-range (micrometric) triplet Josephson coupling between d-wave, high-temperature superconducting electrodes across a half-metallic ferromagnets is important at various levels. Fundamentally, it brings up various questions. For example, which is the mechanism governing the singlet to triplet conversion (which seems to be very efficient in view of the quantitative agreement obtained with the predictions of theories of SNS junctions³⁷), especially considering the planar geometry, or what is the induced pairing symmetry in the LSMO, i.e. whether nodal pairing is preserved or a conversion into s-wave occurs⁴³. In addition, the present findings have much relevance in the field of superconducting spintronics^{44,45} thanks to the gathering of i) the very high temperature (tens of K) for which triplet Josephson effects

are observed and ii) the very long (micrometric) distance over which phase coherence is preserved in the half-metal in a planar device. Moreover, the fact that triplet supercurrents are necessarily fully spin polarized in a half metal, and that both ac and dc triplet Josephson effects are demonstrated for the first time, opens unprecedented opportunities as they pave the way to novel logic gates⁴⁶, full superconducting switches, nonvolatile random access memories⁴⁷ and quantum computing^{48,49}. Furthermore, the half-metallic Josephson junctions should reveal an anomalous Josephson effect with a non-zero phase difference φ_0 at the ground state, which is determined by the mutual orientation of the magnetization in the half-metal and interface magnetizations^{10,50}. Such unusual φ_0 junction could serve as an important building block of “quiet qubit”⁴⁸, and may provide a unique mechanism of direct coupling between magnetism and phase dynamics in Josephson junctions⁴⁹.

Methods

LSMO was grown on (001)-oriented SrTiO₃ single crystals in a high O₂ pressure (3.2 mbar) d.c. sputtering system at 900°C. In situ annealing was done at 800°C with 900mbar O₂ pressure for 1 hour. Electron beam lithography was performed in a Raith50 module mounted on a SEM Zeiss EVO50 to obtain LSMO microwires and to define amorphous alumina patterns. Amorphous alumina was grown in a d.c. sputtering at $7.3 \cdot 10^{-3}$ mbar atmosphere (Argon/Oxygen 2:1) at room temperature. YBCO was grown on top of LSMO and a-ALO template in a high O₂ pressure (3.4 mbar) d.c. sputtering system at 900°C. In situ annealing was done at 800°C with 900mbar O₂ pressure for 1 hour. R(T), R(H) and I(V) were performed in a Helium close cycle cryostat down to 15K applying a maximum current of 800uA and magnetic field up to 4000 Oe while measuring the voltage. Voltage contacts were placed at the ends of the LSMO wire see sketch in Figure 1 (b), although we used control experiments with all 4 contact on the YBCO wires to check that normal state resistance R_n is not limited by interface resistance. See Supplementary Figure S1.

The magnetic domain structure of the ferromagnet superconducting hybrids was examined by means of photoemission electron microscopy (PEEM) using X-ray magnetic circular dichroism (XMCD) as magnetic contrast mechanism. Experiments have been done at the SPEEM station at the UE49/PGMa beam line of the synchrotron radiation source BESSY II at Helmholtz-Zentrum Berlin. The angle of incidence of the incoming radiation with respect to the sample surface is 16° . The sample was mounted on a sample holder which allows application of in plane magnetic field pulses up to ± 1000 Oe. Magnetic imaging was done in remanence after applying the desired magnetic field value. Images with a $10 \mu\text{m}$ field of view were collected at the Mn L_3 -edge (640.3 eV) for circularly polarized radiation with clockwise ($\sigma+$) and counterclockwise ($\sigma-$) helicities. The data has been normalized to a background image and drift corrected before their averaging. The XMCD images were calculated as $(\sigma--\sigma+)/(\sigma+\sigma+)$. Measurements were conducted at 50 K.

Four-probe $R(H)$ measurements were carried out in a closed cycle refrigerator, equipped with an electromagnet and a sample rotating stage. The dc resistance $R=V/I$ was measured by injecting an electrical current I with a dc source and measuring the voltage V with a nanovoltmeter. The voltage offsets were removed by measuring both current polarities. The magnetic field was applied in the plane of the sample while the latter was rotated every 2° .

For the measurement of the Shapiro steps the microwave signal was delivered through a semi-rigid coaxial cable terminated by a wide band spiral antenna placed in front of the sample (~ 1 cm above it) and connected to a generator in a continuous wave mode at frequency f and power P . Differential resistance curves were obtained by numerically differentiating the $I(V)$ characteristic after applying a moving average window to smooth the data.

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Data availability statement. The data used in this paper are available from the authors upon reasonable request

Authors contributions. DS-M and FC grew the samples and performed resistance and critical current measurements. DS-M and SM measured angle dependent transport with contributions from AS, XP, and AB. DS-M, LM and SV measured x-ray absorption. D S-M, SM measured Shapiro steps with the guidance and analysis of CF-P, NB and JL. AIB contributed to the theoretical understanding and modelling. GO, VR, JG-B, MR, GF, JT, AR, FM, MG-H worked in sample growth and characterization in different stages of the project. MC and JM G-C did the microscopy. JS designed the overall experiment and J-EV contributed with the design of the Josephson characterization. JS and J-EV wrote the manuscript with inputs and help of JL, AIB, SM, D S-M, CL. All authors discussed results and revised the manuscript.

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References

1. Buzdin, A. I. Proximity effects in superconductor-ferromagnet heterostructures. *Rev. Mod. Phys.* **77**, 935–976 (2005).
2. Stolyarov, V. S. *et al.* Domain Meissner state and spontaneous vortex-antivortex generation in the ferromagnetic superconductor $\text{EuFe}_2(\text{As}_{0.79}\text{P}_{0.21})_2$. *Sci. Adv.* **4**, eaat1061 (2018).
3. Aoki, D. & Jaques Flouquet. Ferromagnetism and Superconductivity in Uranium Compounds. *J. Appl. Phys.* **81**, 1–11 (2012).
4. Bergeret, F. S., Volkov, A. F. & Efetov, K. B. Long-range proximity effects in superconductor-ferromagnet structures. *Phys. Rev. Lett.* **86**, 4096–4099 (2001).
5. Eschrig, M., Kopu, J., Cuevas, J. C. & Schön, G. Theory of Half-Metal/Superconductor Heterostructures. *Phys. Rev. Lett.* **90**, 4 (2003).
6. Houzet, M. & Buzdin, A. Long range triplet Josephson effect through a ferromagnetic trilayer. *Phys. Rev. B* **76**, 060504 (2007).
7. Asano, Y., Sawa, Y., Tanaka, Y. & Golubov, A. A. Odd-frequency pairs and Josephson current through a strong ferromagnet. *Phys. Rev. B* **76**, 224525 (2007).
8. Sperstad, I. B., Linder, J. & Sudbø, A. Josephson current in diffusive multilayer superconductor/ferromagnet/ superconductor junctions. *Phys. Rev. B - Condens. Matter Mater. Phys.* **78**, 104509 (2008).
9. Halterman, K., Valls, O. T. & Barsic, P. H. Induced triplet pairing in clean s -wave superconductor/ferromagnet layered structures. *Phys. Rev. B* **77**, 174511 (2008).
10. Eschrig, M. & Löfwander, T. Triplet supercurrents in clean and disordered half-metallic ferromagnets. *Nat. Phys.* **4**, 138–143 (2008).
11. Keizer, R. S. *et al.* A spin triplet supercurrent through the half-metallic ferromagnet CrO_2 . *Nature* **439**, 825–7 (2006).
12. Khaire, T. S., Khasawneh, M. a., Pratt, W. P. & Birge, N. O. Observation of Spin-Triplet Superconductivity in Co-Based Josephson Junctions. *Phys. Rev. Lett.* **104**, 137002 (2010).
13. Anwar, M. S., Czeschka, F., Hesselberth, M., Porcu, M. & Aarts, J. Long-range supercurrents through half-metallic ferromagnetic CrO_2 . *Phys. Rev. B* **82**, 100501 (2010).
14. Robinson, J. W. a, Witt, J. D. S. & Blamire, M. G. Controlled injection of spin-triplet supercurrents into a strong ferromagnet. *Science (80-)*. **329**, 59–61 (2010).
15. Sefrioui, Z. *et al.* Ferromagnetic/superconducting proximity effect in $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ superlattices. *Phys. Rev. B* **67**, 214511 (2003).
16. Dybko, K. *et al.* Possible spin-triplet superconducting phase in the $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{YBa}_2\text{Cu}_3\text{O}_7/\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ trilayer. *Phys. Rev. B* **80**, 144504 (2009).
17. 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).

18. Kalcheim, Y., Millo, O., Egilmez, M., Robinson, J. W. A. & Blamire, M. G. Evidence for anisotropic triplet superconductor order parameter in half-metallic ferromagnetic $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_3\text{O}$ proximity coupled to superconducting $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$. *Phys. Rev. B* **85**, 104504 (2012).
19. Egilmez, M. *et al.* Supercurrents in half-metallic ferromagnetic $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$. *EPL (Europhysics Lett.)* **106**, 37003 (2014).
20. Crouzy, B., Tollis, S. & Ivanov, D. A. Josephson current in a superconductor-ferromagnet-superconductor junction with in-plane ferromagnetic domains. *Phys. Rev. B - Condens. Matter Mater. Phys.* **76**, 134502 (2007).
21. Buzdin, A. I., Mel'nikov, A. S. & Pugach, N. G. Domain walls and long-range triplet correlations in SFS Josephson junctions. *Phys. Rev. B - Condens. Matter Mater. Phys.* **83**, 144515 (2011).
22. Bergeret, F. S. & Tokatly, I. V. Singlet-triplet conversion and the long-range proximity effect in superconductor-ferromagnet structures with generic spin dependent fields. *Phys. Rev. Lett.* **110**, 1–6 (2013).
23. Mel'nikov, A. S., Samokhvalov, A. V., Kuznetsova, S. M. & Buzdin, A. I. Interference Phenomena and Long-Range Proximity Effect in Clean Superconductor-Ferromagnet Systems. *Phys. Rev. Lett.* **109**, 237006 (2012).
24. Klose, C. *et al.* Optimization of spin-triplet supercurrent in ferromagnetic Josephson junctions. *Phys. Rev. Lett.* **108**, 127002 (2012).
25. Glick, J. A. *et al.* Phase control in a spin-triplet SQUID. *Sci. Adv.* **4**, eaat9457 (2018).
26. Voltan, S., Singh, A. & Aarts, J. Triplet generation and upper critical field in superconducting spin valves based on CrO_2 . *Phys. Rev. B* **94**, 054503 (2016).
27. Visani, C. *et al.* Magnetic field influence on the proximity effect at $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ superconductor/half-metal interfaces. *Phys. Rev. B* **92**, 014519 (2015).
28. Lahabi, K. *et al.* Controlling supercurrents and their spatial distribution in ferromagnets. *Nat. Commun.* **8**, 1–7 (2017).
29. Shapiro, S. Josephson currents in Superconducting tunneling: The effect of microwaves and other observations. *Phys. Rev. Lett.* **11**, 80–82 (1963).
30. Richard, C., Houzet, M. & Meyer, J. S. Superharmonic long-range triplet current in a diffusive Josephson junction. *Phys. Rev. Lett.* **110**, 1–5 (2013).
31. Perez-Muñoz, A. M. *et al.* In operando evidence of deoxygenation in ionic liquid gating of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$. *Proc. Natl. Acad. Sci. U. S. A.* **114**, 215–220 (2017).
32. Hoffmann, A. *et al.* Suppressed magnetization in $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ superlattices. *Phys. Rev. B - Condens. Matter Mater. Phys.* **72**, 140407 (2005).
33. Salafranca, J. & Okamoto, S. Unconventional Proximity Effect and Inverse Spin-Switch Behavior in a Model Manganite-Cuprate-Manganite Trilayer System. *Phys. Rev. Lett.* **105**, 256804 (2010).
34. Schneider, C. W., Thiel, S., Hammerl, G., Richter, C. & Mannhart, J. Microlithography of electron gases formed at interfaces in oxide heterostructures. *Appl. Phys. Lett.* **89**, 12–14 (2006).

35. Bhatia, E. *et al.* Nanoscale Domain Wall Engineered Spin-Triplet Josephson Junctions and SQUID. *Nano Lett.* **21**, 3092–3097 (2021).
36. M., I. Y. & Zil'berman, L. A. The Josephson effect in small tunnel contacts. *Sov. J. Exp. Theor. Physics*, **28**, 1272 (1969).
37. Dubos, P. *et al.* Josephson critical current in a long mesoscopic S-N-S junction. *Phys. Rev. B* **63**, 064502 (2001).
38. Anwar, M. S., Veldhorst, M., Brinkman, A. & Aarts, J. Long range supercurrents in ferromagnetic CrO₂ using a multilayer contact structure. *Appl. Phys. Lett.* **100**, 052602 (2012).
39. Buzdin, A. Peculiar properties of the Josephson junction at the transition from 0 to π state. *Phys. Rev. B - Condens. Matter Mater. Phys.* **72**, 1–4 (2005).
40. Houzet, M., Vinokur, V. & Pistoiesi, F. Superharmonic Josephson relation at 0- π -junction transition. *Phys. Rev. B - Condens. Matter Mater. Phys.* **72**, 1–4 (2005).
41. Sellier, H., Baraduc, C., Lefloch, F. & Calemczuk, R. Half-integer shapiro steps at the 0- π crossover of a ferromagnetic josephson junction. *Phys. Rev. Lett.* **92**, 1–4 (2004).
42. Ryazanov, V. V. *et al.* Coupling of two superconductors through a ferromagnet: Evidence for a π junction. *Phys. Rev. Lett.* **86**, 2427–2430 (2001).
43. Volkov, A. F. & Efetov, K. Proximity Effect and Its Enhancement by Ferromagnetism in High-Temperature Superconductor-Ferromagnet Structures. *Phys. Rev. Lett.* **102**, 077002 (2009).
44. Linder, J. & Robinson, J. W. A. Superconducting spintronics. *Nature Physics* vol. 11 307–315 (2015).
45. Eschrig, M. Spin-polarized supercurrents for spintronics: a review of current progress. *Rep. Prog. Phys.* **78**, 104501 (2015).
46. Villegas, J. & Crete, D. G. . Logic circuit based on spin valves of the spin-polarized supercurrent type and circuit integrating such logic gates. *US20170085269A1* (2017).
47. Vernik, I. V. *et al.* Magnetic josephson junctions with superconducting interlayer for cryogenic memory. *IEEE Trans. Appl. Supercond.* **23**, 1701208 (2013).
48. Ioffe, L. B., Geshkenbein, V. B., Feigel'man, M. V, Fauchère, A. L. & Blatter, G. Environmentally decoupled sds -wave Josephson junctions for quantum computing. *Nature* **398**, 679–681 (1999).
49. Mironov, S., Meng, H. & Buzdin, A. Magnetic flux pumping in superconducting loop containing a Josephson ψ junction. *Appl. Phys. Lett.* **116**, 162601 (2020).
50. Mironov, S. & Buzdin, A. Triplet proximity effect in superconducting heterostructures with a half-metallic layer. *Phys. Rev. B - Condens. Matter Mater. Phys.* **92**, 184506 (2015).

FIGURE CAPTIONS

Figure 1: Devices' structure. (a) Optical image of the device. Amorphous Al₂O₃ and YBCO are

on top of the LSMO micro wire. (b) AFM image of the device before YBCO deposition. The Al_2O_3 stripe between YBCO electrodes is continuous all along the LSMO. (c) AFM image of the Al_2O_3 stripe separating the YBCO electrodes and defining the width of the barrier (before YBCO deposition). (d) Low magnification STEM-HAADF image of a LSMO/YBCO bilayer grown ex-situ on (001) STO substrate (e) High resolution STEM-HAADF image of the same interface. (f) Left: Atomic resolution HAADF image where atomic positions of each element are indicated by arrows in the different atomic columns. Right: Ba $M_{4,5}$ (red), Mn $L_{2,3}$ (blue) and La $M_{4,5}$ (yellow) elemental EELS map in a color mix of a spectrum image of a region of interest from the same HAADF image. (g) XMCD image of the LSMO nanowire between the two YBCO contacts measured at the Mn L_3 -edge (640.3 eV) at 50 K. Black line separates two XMCD images of uniformly magnetized LSMO taken at remanence.

Figure 2: Superconducting characterization. (a) Resistance vs. temperature of three different devices. Separation between YBCO contacts is 1 micron. Width of the LSMO wire electrodes is 20um (red) 25um (blue and green). Current level is 1 μA (red and green) and 20 μA (blue) (b) Schematic of the planar devices and wiring used. (c) Resistance vs. temperature of a 30 nm thick LSMO single layer (black) to be compared with that of a junction device (blue). The orange curve corresponds to a control sample with no LSMO wire and YBCO electrodes separated 1 μm . (d) I(V) curves at different temperatures between 18K and 40K. Inset: Critical temperature as a function of temperature as extracted from the I(V) with the criterion shown in panel (e). (e) IV curves in double logarithmic scale. Notice that the critical current is attained at very low voltages shown by the vertical line.

Figure 3: Flux quantization effects. a) Resistance oscillations as a function of magnetic field ($-0.4\text{T} < H < 0.4\text{T}$) for different directions with LSMO wire. Notice the disappearance of the oscillations when the field is applied parallel to the junction. Curves are shifted in resistance for clarity (b) Schematic illustrating the orientation of magnetic field. Blue shaded areas in the LSMO wire are used to sketch proximitized regions. (c) Contour plot of the resistance as a function of magnetic field ($-0.4\text{T} < B < 0.4\text{T}$) with different orientations relative to the LSMO wire. Angle θ is varied in steps of 2° (d) Simulation of the oscillation pattern of the I_c/I_0 assuming a Fraunhofer dependence of the critical current of the magnetic flux.

Figure 4: ac Josephson effect. (a) Shapiro steps pattern of the differential resistance as a function of current for different microwave power levels for a frequency of 9.997GHz. (b) Shapiro steps as a function of voltage normalized to the Josephson voltage ($V_{\text{Josephson}} = \frac{hf}{2e}$) for microwave radiation of 9.997 GHz. Solid (dashed) lines correspond to Integer (half-integer) Shapiro steps.