Superconductivity-induced change in magnetic anisotropy in epitaxial ferromagnet-superconductor hybrids with spin-orbit interaction

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Abstract

The interaction between superconductivity and ferromagnetism in thin film superconductor/ferromagnet heterostructures is usually reflected by a change in superconductivity of the S layer set by the magnetic state of the F layers. Here we report the converse effect: transformation of the magnetocrystalline anisotropy of a single Fe(001) layer, and thus its preferred magnetization orientation, driven by the superconductivity of an underlying V layer through a spin-orbit coupled MgO interface. We attribute this to an additional contribution to the free energy of the ferromagnet arising from the controlled generation of triplet Cooper pairs, which depends on the relative angle between the exchange field of the ferromagnet and the spin-orbit field. This is fundamentally different from the commonly observed magnetic domain modification by Meissner screening or domain wall-vortex interaction and offers the ability to fundamentally tune magnetic anisotropies using superconductivity - a key step in designing future cryogenic magnetic memories.

Superconductivity (S) is usually suppressed in the presence of ferromagnetism (F) [1,5]. For example, in F/S/F spin-valves the transition temperature $T_C$ of the S layer is different for a parallel alignment of the F layer moments compared to an anti-parallel alignment [6,9]. Interestingly, for non-collinear alignment of the F layer moments in spin-valves [10,12] or Josephson junctions [13,22], an enhancement in the proximity effect is found due to the generation of long-range triplet Cooper pairs, which depend on the pair-breaking exchange field in the F layers. So far, the reciprocal modification of the static properties of the ferromagnet by superconductivity has been limited to restructuring [23] and pinning of magnetic domains walls (DWs) by Meissner screening and vortex-mediated pinning of DWs [24,27].

Modification of the magnetization dynamics in the presence of superconductivity has been studied in [28-30]. Recently, theoretical and experimental results have indicated an underlying role of Rashba spin-orbit coupling (SOC), resulting in an enhancement of the proximity effect and a reduction of the superconducting $T_C$, along with enhanced spin pumping and Josephson current in systems with a single F layer coupled to Nb through a heavy-metal (Pt) [37,43]. In this context, V/MgO/Fe [44] has been shown to be an effective system to study the effect of SOC in S/F structures with fully epitaxial layers.

At first glance, altering the magnetic order in S/F heterostructures leading to a change in the direction of magnetization appears non-trivial due to the difference in the energy scales associated with the order parameters. The exchange splitting of the spin-bands and the superconducting gap are about $10^3$ K and $10^1$ K, respectively. However, this fundamentally changes if one considers the possibility of controlling the magneto crystalline anisotropy (MCA) by manipulating the competing anisotropy landscape with superconductivity, since the MCA energy scales are comparable to the superconducting gap energy. Interestingly, emergent triplet superconducting phases in S/SOC/F heterostructures offer the possibility to observe MCA modification of a F layer coupled to a superconductor through a spin-orbit coupled interface, triggered by the superconducting phase [45].

In this communication, we present evidence that cubic in-plane MCA in V/MgO/Fe(001) system is modified by the superconductivity of V through SOC at the MgO/Fe interface [46]. Our detailed characterization of the coercive fields of the rotated soft Fe(001) and sensing hard (Fe/Co) ferromagnetic layers by tunnelling magnetoresistance effect (TMR) [47] along with numerical simulations dismisses the Meissner screening and DW-vortex interactions as a source of the observed effects.

The magnetic tunnel junction (MTJ) multilayer stacks have been grown by molecular beam epitaxy (MBE) in a chamber with a base pressure of $5 \times 10^{-11}$ mbar following the procedure described in [48]. The samples were grown on [001] MgO substrates. Then a 10 nm thick seed of anti-diffusion MgO underlayer is grown on the substrate
FIG. 1: (a) Sketch of the junctions under study. Fe(10 nm)/Co(20 nm) is the hard (sensing) layer while Fe(10 nm) is the soft ferromagnet where spin reorientation transitions are investigated. (b) Sketch showing the top view without the hard Fe/Co layer, with the 4-fold in-plane magnetic energy anisotropy expected for the Fe(001) atomic plane of the magnetically free layer, for temperatures above $T_C$ (yellow line) and well below $T_C$ (dashed cyan). Note that during the epitaxial growth, the Fe lattice is rotated by 45 degrees with respect to MgO. Parts (c) and (d) show in-plane spin reorientation transitions between parallel (P), perpendicular in-plane (PIP) and antiparallel (AP) relative magnetization alignments of the soft and hard F layers for a 30×30 μm² junction at $T=10$ K (above $T_C$). Indices above the inset sketches indicate the direction of the soft layer. The in-plane rotation has been carried out with the angle $\Phi_H$ of the magnetic field relative to the Fe[100] axis going from $-30$ to 390 degrees.

To trap the C from it before the deposition of the Fe (or V). Then the MgO insulating layer is epitaxially grown by e-beam evaporation, the thickness approximately $\sim 2$ nm and so on with the rest of the layers. Each layer is annealed at 450 C for 20 mins for flattening. After the MBE growth, all the MTJ multilayer stacks are patterned in 10-40 micrometre-sized square junctions (with diagonal along [100]) by UV lithography and Ar ion etching, controlled step-by-step in situ by Auger spectroscopy. The measurements are performed inside a JANIS® He³ cryostat. The magnetic field is varied using a 3D vector magnet. For the in-plane rotations, the magnetic field magnitude was kept at 70-120 Oe, far away from the soft Fe(001) and hard Fe/Co layers switching fields obtained from in-plane TMRs (see Supplemental Material S1,S2 [49]). This way, only the soft layer is rotated and the difference in resistance can be attributed to the angle between the soft and hard layers.

Figure 1 shows the device structure with the Fe/Co hard layer sensing the magnetization alignment of the 10-nm thick Fe(001) soft layer. A typical TMR plot above $T_C$ is shown in Figure 1c. The resistance switching shows a standard TMR between the P and AP states. However, the epitaxial Fe(001) has a four-fold in-plane anisotropy with two orthogonal easy axes - [100] and [010] - (Figure 1b). These MCA states could be accessed by an in-plane rotation of the Fe(001) layer with respect to the Fe/Co layer using field greater than the coercive field of the Fe(001) layer without disrupting the Fe/Co magnetization (see also Supplemental material S1 [49] for the magnetic characterization of the Fe/Co layer). This is shown in Figure 1d, where TMR is plotted as a function of the in-plane field angle with respect to the [100] direction angle $\Phi_H$. This gives rise to four distinct magnetization states with P, perpendicular in-plane (PIP) and AP states reflected by the TMR values. Supplemental Material S3 [49] discusses the weak magnetoelastic coupling between the two FM layers (detected through resistances in-between the P and AP states in the virgin state of different samples), showing that it does not affect the capability to reorient the soft layer independently of the hard one. It also demonstrates that the soft layer retains different magnetic directions at zero field.

Figure 2 analyzes the most probable in-plane magnetization orientations of the Fe(001) layer through magnetic field rotations at fixed temperatures from above to below $T_C$. Typically, no qualitative changes in TMR are observed above and below $T_C$ in the $0$-$\pi$ field rotation angle ($\Phi_H$) span (Figure 2a). However, in the $\pi$-$2\pi$ range, the TMR qualitatively changes below $T_C$/2, possibly indicating new stable magnetization states along different directions to the ones stabilized by the principal crystallographic axes (Figure 2a).

To ascertain the exact angle $\Phi_{FM}$ between the two F layers, we have calibrated the magnetization direction of the soft layer with respect to the hard Fe/Co using the Slonczewski formula (Supplemental Material S4 [49]). The applicability of the macrospin approach to describe TMRs and magnetization reorientation resides in the high effective spin polarization obtained ($P = 0.7$ [47], approaching to the values typically reported for Fe/MgO in a fully saturated state [50, 51]. Figure 2c is a histogram representing the probability of obtaining a specific $\Phi_{FM}$ as temperature is lowered from above to below $T_C$. We observe that the most probable Fe(001) directions are oriented along the [100] and [010] principal axes above $T_C$/2, while below $T_C$/2 it splits in three branches roughly oriented along $\pi/4$ angles. The split of the [001] state into three branches is also visualized in Fig2b, with a plot of the counts vs. temperature around the [110], [010] and [101] magnetization directions.

Interestingly, once the rotation is initiated in the AP configuration, the magnetization apparently locks in the ($\pi + \pi/4$) or [110] state (Figures 2a,d,f). This probably arises due to the improved initial macrospin alignment,
which is not fully achieved in the AP state with a preceding P-AP rotation. We believe that with the full 2π field rotation, magnetization inhomogeneities or local DWs created during the P-AP state rotation help to overcome MCA energy barriers more easily. The suggested suppression of the local DWs with the magnetization rotation initiated from the AP state can be indirectly inferred from the broadening of the [T00] to [0T0] transition in the normal state detected as a small (extrinsic) number of counts around [T10] (Figure 2). For a more systematic analysis, we performed a series of in-plane TMR measurements along different directions relative to the symmetry axes. The first experiment (i) was performed with an initial saturation field of ±1 kOe in the [100] direction, followed by a TMR in the [210] direction (between [100] and [110]). The second (ii) initially saturates both the hard and soft layers along the [100] direction. Then, a minor loop is performed starting from zero field and going up to 150 Oe along the [110] axis.

Both experiments further suggest the possibility of superconductivity-induced changes of MCA. The inset of Figure 3a shows the full field sweep range in the first (i) configuration, and Figure 3a zooms in close to the AP configuration. When we sweep the field in the [210] direction, we detect a weak but robust resistance upturn configuration. When we sweep the field in the [210] direction, we detect a weak but robust resistance upturn (Figure 3a) roughly corresponds to an 8-10 degree rotation, we detect a weak but robust resistance upturn (Figure 3a) roughly corresponds to an 8-10 degree rotation.
field, $H_{\text{switch}}$, required to reorient the soft layer from [110] towards [210] when $T$ decreases below $T_C/2$, could reflect the superconductivity-induced MCA energy minimum along the [110] direction.

The minor TMR loops along [110] (Figure 3c) realized after saturation along [100] point on a thermally induced magnetization reorientation from [100] towards [110] even at zero field, in a temperature range below $T_C$ where the barrier between adjacent energy minima is comparable to $k_B T$. The zero-field reorientation becomes less probable when the thermal energy is insufficient to overcome the barrier (Figure 3d). An estimation of the in-plane normal-state MCA energy barrier done through magnetization saturation along [100] and [110] provides a value of only a few µeV/atom (Supplemental Material S5). However, the real barrier is determined by the nucleation volume, which depends on the exchange length in the material. With a DW width of about 3 nm for Fe(001) we estimate the MCA barrier to be at least $10^3 - 10^4$ mV.

Before describing our explanation of the MCA modification of Fe(001) in the superconducting state of V(40 nm)/MgO(2 nm)/Fe(10 nm) system, we discard alternative interpretations of the observed effects. Meissner screening, if present, would introduce a 10% correction to the actual magnetic field independently of the external field direction (see Supplemental Material S2). The reason for the weak in-plane field screening could be the small superconductor thickness (40 nm), only slightly exceeding the estimated coherence length (26 nm). On the other hand, intermediate multidomain states are expected to be absent in when magnetization is directed along [110] (Supplemental material S6). Indeed, our experiments show that magnetization, when locked below $T_C$ in the $(\pi + \pi/4)$ angle, hardly depends on the absolute value of the external field along [110] varied between 0 and 100 Oe. Moreover, simulations of the vortex-DW interaction using MuMax3 and TDGL codes discard the vortex mediated DW pinning scenario including when interfacial magnetic defects created by misfit dislocations are considered (see Supplemental material S2). The vortex pinning mechanism also contradicts that only the $(0 - \pi)$ field rotation span (Figure 2a) gets affected below $T_C/2$. The observed irrelevance of the junction area (Supplemental material S7) contradicts the importance of the vortex-edge DWs interaction. The shape and vortex-DW interactions, if relevant, would strengthen magnetization pinning along [110], but not [110] (Supplemental material S6, S7). Finally, we also indicate that the MCA modification from singlet superconductivity would not enable any zero field rotation to non-collinear misalignment angles, in contrast to our data (Fig. 3d).

To explain our results, we consider the possibility in which the invariance of the superconducting proximity effect to magnetization rotation is broken in the presence of SOC. It has been predicted that triplet-superconductivity is effectively generated even for weakly spin-polarized ferromagnets with a small spin-orbit field. In addition to generating triplet pairs, the SOC also introduces an angle-dependent anisotropic depairing field for the triplets. In V/MgO/Fe, the Rashba field is caused by a structural broken inversion symmetry at the

![FIG. 4: Numerical modelling. (a), Free energy $F$ vs in-plane magnetization angle $\Phi_{FM}$ for temperatures below the superconducting critical temperature and just above the critical temperature ($T_{C1}^+$). The free energy is plotted relative to the free energy in the AP configuration $F_{AP}$ and has been normalized to the hopping parameter $t$ used in the tight-binding model. (b), Illustration of the physical origin of the change in magnetic anisotropy induced by the superconducting layer. Above $T_C$, $V$ is a normal metal and the soft Fe layer has a 4-fold in-plane magnetic energy anisotropy (yellow line). Below $T_C$, $V$ is superconducting and influences the soft Fe layer via the proximity effect: a leakage of Cooper pairs into the ferromagnet. Due to the SOC at the interface, a magnetization-orientation dependent generation of triplet Cooper pairs occurs. The generation of triplets is at its weakest for a magnetization pointing in the [010] direction, giving a maximum for the superconducting condensation energy gain. This modifies the magnetic anisotropy of the soft Fe layer (cyan line), enabling magnetization switching to the [010] direction (blue arrow). The magnetic anisotropy does not show the weak AP coupling between the two Fe layers, causing an absolute minimum in $\Phi_{FM} = \pi$ (a).](image-url)
effects in magnetic Josephson junction technology and simulation superspintronic technologies. Specifically, they could establish superconductors as tunable sources of magnetic anisotropies and active ingredients for future low dissipation screening and vortex induced domain wall pinning, even though the spin-triplet mechanism and performed simulations require many assumptions. Our results even the capability to reorient the soft layer independently of the hard one) described by an additional contribution $f_{AF} \cos(\Phi_{FM})$ with a constant parameter $f_{AF} > 0$.

Figure 7 shows the total free energy of the system as a function of the IP magnetization angle $\Phi_{FM}$ for decreasing temperatures. Due to the increase in the superconducting proximity effect, additional local minima appear at $\Phi_{FM} = n\pi/2 + \pi/4$, where $n = 0, 1, 2, \ldots$ (i.e., [110], [110], [110], and [110], respectively). This is a clear signature for the proximity-induced triplet correlations. These are most efficiently generated at angles $\Phi_{FM} = n\pi/2$ (i.e., [100], [010], [100], and [010]) for a heterostructure with a magnetic layer that has a cubic crystal structure like Fe [45]. As a result, the decrease in the free energy is stronger at angles $\Phi_{FM} = n\pi/2 + \pi/4$ where more singlet Cooper pairs survive. Our numerical results thus confirm that the experimentally observed modification of the anisotropy can be explained by the presence of SOC in the S/F structure alone, without including superconducting proximity effects from misalignment between the Fe(100) and Fe/Co layers. Moreover, Figure 4 illustrates why the $\Phi_{FM} = n\pi/2 + \pi/4$ states only appear experimentally when the external field is rotated from an AP to P alignment (Figure 2). Because of the weak AP coupling between the ferromagnetic layers, the energy thresholds for reorienting the magnetization from one local minimum to the next are higher under a rotation from AP to P alignment.

In conclusion, we present experimental evidence for superconductivity-induced change in magnetic anisotropy in epitaxial ferromagnet-superconductor hybrids with spin-orbit interaction. We believe this mechanism is fundamentally different from the previous reports of magnetisation modification arising from Meissner screening and vortex induced domain wall pinning, even though the spin-triplet mechanism and performed simulations require many assumptions. Our results establish superconductors as tunable sources of magnetic anisotropies and active ingredients for future low dissipation superspintronic technologies. Specifically, they could provide an opportunity to employ spin-orbit proximity effects in magnetic Josephson junction technology and approach it to Fe/MgO-based junctions that are widely used in commercial spintronic applications.

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SUPPLEMENTARY MATERIAL

In the supplementary material, the section S1 presents a magnetic characterization of the hard Fe/Co layer of the junctions under study. Section S2 presents a magnetic characterization of the soft Fe(001) layer and studies the possible influence of the Meissner screening on the coercive fields of the soft and hard layers. Section S3 estimates the strength of the weak antiferromagnetic coupling between magnetically soft and hard electrodes. Section S4 provides details about the calibration of the angle between the soft and hard layers using the Slonczewski formula, as well as discussing the possible sources of error for this calibration and their magnitude. Section S5 provides an estimation for the magnetano-anisotropic energy barrier between the [110] and [100] magnetization directions, normalized per volume or per atom. Section S6 numerically evaluates the possible domain walls pinning by superconducting vortices. Section S7 discusses the contribution of the shape to the magnetic anisotropy. Finally, section S8 provides details on the theoretical modelling of the observed effects.
S1. Magnetic characterization of the hard Fe/Co layer

Figure S5 shows the magnetic characterization of the hard Fe/Co bilayer, determined from a typical spin-valve M-H loop on a standard Fe/MgO/Fe/Co single crystal MTJ system (continuous layers, unpatterned). The nominal thickness of the layers on this sample, MgO(100)/Fe(30 nm)/MgO(2 nm)/Fe(10 nm)/Co(20 nm), has been chosen to optimize the magnetic properties of the MTJ stack [48]. The TMR measurements of the coercive fields of the hard ($H_{C,Hard}$) and soft ($H_{C,Soft}$) layers in MTJs under study show that they are well separated from the external field values used to rotate the soft layer. Figure S6 also shows that the hard layer switching fields obtained from TMRs along [100], [010] and [110] measured in our junctions remain far above the typical range of 70-120 Oe which is used to rotate the soft layer. Moreover, Figure S7 also shows the typical temperature dependence of $H_{C,Hard}$, demonstrating its independence with temperature from well above to well below $T_C$.

FIG. 5: Magnetic characterization of a Fe(30 nm)/MgO/Fe(10 nm)Co(20 nm) structure, at room temperature, along the [100] direction.

S2. Magnetic characterization of the soft Fe(001) layer and estimation of the Meissner screening

The magnetostatic Meissner screening has been discussed mainly in studies with perpendicular magnetization [24]. In the case of the experiments with in-plane field rotation which we carry out, such field expulsion could induce some screening of the external magnetic field applied to invert or rotate the magnetization of the soft Fe(001) layer (which is the closest to the superconductor), and with less probability affect the switching of the more distant hard Fe/Co layer. Figure S8 shows that the hard layer switching fields obtained from TMRs along [100], [010] and [110] measured in our junctions remain far above the typical range of 70-120 Oe which is used to rotate the soft layer. Moreover, Figure S9 also shows the typical temperature dependence of $H_{C,Hard}$, demonstrating its independence with temperature from well above to well below $T_C$.

FIG. 6: Coercive field of the hard Fe/Co layer for magnetic field oriented along different crystallographic directions [100], [110] and [210], above the superconducting critical temperature ($T = 5$ K). The grey band shows the typical field range used to manipulate the magnetization of the soft Fe(100) layer in the rotation experiments.

S3. Estimation of the weak antiferromagnetic coupling of the two ferromagnetic layers.

In order to quantify the unavoidable weak antiferromagnetic magnetostatic coupling between the rotated soft Fe(001) and the practically fixed hard FeCo layer, we show low field TMR measurements where the AP state is achieved and then maintained at zero field (Figure S9a). One clearly observes that the AP and P states can be...
obtained as two different non-volatile states, and therefore the antiferromagnetic coupling is not sufficient to antiferromagnetically couple the two layers at zero field. The stability of the P state against the antiferromagnetic coupling is confirmed by the temperature dependence of the resistance in the P and AP states. The P state shows stable resistance values at least below 15 K (Figure S9b). This means that the antiferromagnetic coupling energy is well below 2 mV.

S4. Calibration of the angle between the two ferromagnetic layers

In order to estimate the angle between the two ferromagnets for the TMR measurements and rotations, we used the Slonczewski model [56]. By using values of the resistance in the AP, P and PIP states established above $T_C$, we can calculate the desired angle $\theta$ with the follow-
ing expression:

\[ G^{-1} = G_1^{-1} + \left[ G_2 \left(1 + p^2 \cos \theta \right) \right]^{-1}. \]  

(1)

Here, \( G \) is the total conductance of the sample, \( G_1 \) and \( G_2 \) are the conductances of each of the two tunnel barriers, and \( p \) is the spin polarization in the ferromagnets, for which we obtain values between 0.7 and 0.8 depending on the sample (the value being robust for each individual one).

In order to ascertain the precision of this calibration method, an analysis of the different errors has been performed. First, an standard error propagation calculation was done to estimate the uncertainty in the resistance values, taking typical values for the current and voltage of 100 nA and 5 mV, respectively, which gives us a typical resistance value of 50 kΩ. The current is applied using a Keithley 220 Current Source, which has an error of 0.3% in the operating range according to the user manual. The voltage is measured using a DMM-522 PCI multimeter card. In the specifications, the voltage precision is said to be 5 1/2 digits. With all this, the resistance error obtained is \( \Delta R = 75.08 \Omega \) or a 0.15% of relative error. Using this value, the error bars in the measurements shown in the main text would be well within the experimental points.

For the calculated angle, the error propagation method is not adequate. It gives errors bigger than 360 degrees for some angles, and in general over 30 degrees. This is clearly not what it is observed in reality: the performed fits are quite robust, showing little variance in the estimated angle when changing the input parameters all that is reasonable. Instead, we have used a typical rotation performed on a 30 \( \times \) 30 \( \mu \)m\(^2\) sample. The fitting to the Slonczewski formula needs three input values: the resistance in the P state (\( R_P \)), the resistance in the AP state (\( R_{AP} \)) and the resistance in the PIP state (\( R_{PIP} \)). Using these, a numerical algorithm calculates the spin polarization (\( p \)), the resistance of the F/F barrier (\( R_{FF} \)), and the resistance of the F/S (F/N) barrier (\( R_{NF} \)). These give us the total resistance of the sample as a function of the angle \( \Phi_{FM} \) between the two ferromagnets or, reciprocally, the angle as a function of resistance. For our estimation, we have varied the value of the \( R_{PIP} \) input parameter from the lowest to the highest possible in the PIP state of the rotation, as well as taking an intermediate value which would be used in a normal analysis (the P and AP resistance values are always taken as the minimum and maximum resistance values in the rotation respectively). The calculated parameters for the resistance of each barrier and the polarization may slightly vary from one fitting to another, but the overall fitting remains remarkably stable, as shown in Figure 10.

As expected, the difference is higher for the PIP state, and minimum in the P and AP state that are “fixed”.

![Figure 10](image_url)

**FIG. 10:** (a) \( \Phi_{FM} \) as a function of resistance for the fittings with maximum, usual, and minimum \( R_{PIP} \) used, in the P-AP resistance range. (b) difference of calculated angle vs resistance (in the P-AP resistance range) for the fittings with maximum and minimum \( R_{PIP} \) used.

The difference doesn’t exceed 7 degrees, and it keeps below 2 degrees near the P and AP states.

**S5. Saturation magnetization for thin Fe(001) films in [100] and [110] directions**

Different M vs H measurements were performed at room temperature on a 10 nm thick Fe films, both for the easy [100] and hard [110] crystallographic axes, in order to estimate the magnetocrystalline anisotropy (MCA) energy. The results are depicted in Fig. S11. Using the saturation field for the two directions, the anisotropy energy can be estimated as \( K_{Fe} = M_{Fe} H_{Sat}/2 = 5.1 \times 10^6 \text{ erg} \cdot \text{cm}^{-3} \), where \( M_{Fe} = 1714 \text{ emu/cm}^3 \) is used. The anisotropy energy per unit cell is therefore \( MAE = 6.764 \mu \text{eV} \), or 3.337 \( \mu \text{eV} \) per atom. The obtained energy barrier is similar to the one measured using ferromagnetic resonance [57].

As shown in Fig. S12, the experimental MCA energy
values have been theoretically confronted with theoretical/numerical calculations of the angular in-plane variation of magnetic anisotropy, using the ab-initio Wien2k FP-LAPW code \[58\]. The calculations were based on a supercell model for a V/MgO/Fe/MgO slab similar to the experimental samples. To insure the requested extreme accuracy in MCA energy values (\(\mu\)eV energy range), a thoroughly well-converged \(k\) grid with significantly large number of \(k\)-points has been involved. Within these circumstances, our theoretical results for the Fe(001) thin films show standard fourfold anisotropy features and reasonable agreement with the experimentally estimated figures with a maximum theoretical MAE of 4.9 \(\mu\)eV per atom (expected theoretical under-estimation of the magnetocrystalline energy within the GGA approach). Note that the superconducting-V induced MCA modulation features cannot be described within the ab-initio FP-LAPW approach, describing the V in its normal metallic state. Therefore, the below \(T_C\) experimentally observed MCA energy modulations have to be clearly related to the proximity effect in the superconducting V/MgO/Fe(001) system and not to any specific MCA feature of Fe(001) in the V/MgO/Fe(001)/MgO complex stacking sequence.

FIG. 11: M vs H measurements on a 10 nm thick Fe film for the easy \([100]\) (a) and hard \([110]\) (b) crystallographic axis. The saturation field (\(H_{sat}\)) for the easy axis is around 10 Oe, while for the hard direction it reaches up to 600 Oe.

FIG. 12: Ab-initio calculation of magnetocrystalline anisotropy energy (MAE) as a function of the in-plane orientation angle \(\theta\), defined in the inset. Solid line is a phenomenological fit to a \(\sin^2(2\theta)\) function.

S6. Evaluation of the vortex induced pinning of domain walls

Using MuMax3 \([52]\), we have compared numerically the DWs formation along the \([100]\) and \([110]\) magnetization directions. The simulations took place in samples with \(3 \times 3 \mu\)m\(^2\) lateral dimensions (100 nm rounded corners were used as the devices have been fabricated by optical lithography), with \(512 \times 512 \times 16\) cells, at \(T = 0\). The rest of the parameters used were \(A_{ex} = 2.1 \times 10^{-11}\) J/m for the exchange energy, \(M_{sat} = 1.7 \times 10^6\) A/m for the saturation magnetization, a damping parameter \(\alpha = 0.02\), and crystalline anisotropy parameters \(K_{C1} = 4.8 \times 10^4\) J/m\(^3\) and \(K_{C3} = -4.32 \times 10^5\) J/m\(^3\). The goal of the simulations was to evaluate the DW formation and their interaction with the superconducting vortices induced by the vertical component of the stray fields at a 2-3 nm from the Fe(001) surface. We observed that, depending on the external field, in the range of 70-1000 Oe both edge-type and inner-type DWs are formed when the field is directed along \([100]\), and mainly edge type DWs are formed with field along \([110]\) (Figure S13a). We have also calculated the interaction \(I\) between the DW related excess exchange energy \(E_{ex}\) and the vertical component of the stray fields, \(B_{eff}\) (Figure S13b):

\[
I = \int_0^{N_x} \int_0^{N_y} |B_{eff}| E_{ex} F_{xy} dxdy
\]

Where \(N_x\) and \(N_y\) are the total number of cells in each dimension of the simulation, and \(F\) is a filter “Vortex generation function” that takes into account the simulated dependence of the number of vortices on the vertically applied field (Figure S13c). The vortices were simulated using the Time Dependent Ginzburg Landau code develop.
FIG. 13: (a) Typical DW formation mapped by MuMax3 simulations for the [110] and [100] applied field directions in the non-saturated (70 Oe) and saturated (1000 Oe) field regimes. The color map represents the out of plane component of the magnetization, while the red arrows indicate the in plane direction. (b) Values of the 2D integral $I$ between the local exchange energy (DWs) and the perpendicular component of the stray fields at a distance of 2-3 nm from the ferromagnet, taking into account the vortex generation function $F$. (c) Vortex generation function $F$, represented as number of vortices formed in a 5 × 5 µm$^2$ square 40 nm thick superconducting Vanadium film as a function of the applied perpendicular field (normalized by the second critical field $H_{c2}$), simulated at $T = 2$ K by using the TDGL code described in [59]. The insert shows a typical image of the vortices at $H=0.15H_{c2}$.

The TDGL simulations took place in $5 \times 5$ µm$^2$ Vanadium samples with 200 × 200 cells, at $T = 2$ K, with a coherence length $\xi_0 = 2.6 \times 10^{-8}$ based on our experimental estimations for the studied devices, $\kappa = 3$ and $T_C = 4$ K. A uniform field was applied in the perpendicular direction, its magnitude varying from $0.1H_{c2}$ to $0.6H_{c2}$, and the number of vortices generated in the relaxed state were counted.

The second critical field in the vertical direction ($H_{c2} = 3$ kOe) was determined experimentally. The estimated interaction shows that in the weakly saturated regime, when the inner DWs could emerge and the DW-vortex interaction increases, such interaction should pin the magnetization along the [100] direction, corresponding to the MCA already present in the normal state, therefore blocking any magnetization rotation towards the [110] direction, contrary to our experimental observations. The possible reason for the irrelevance of the DW-vortex interaction in our system is that inner DWs are expected to be of Neel-type for the thickness considered [60]. Finally we mention that our numerical evaluations show that, if present, the vortex-DW interaction should remain dominant for the magnetization directed along [100] respect [110] and for the magnetic field range 70-1000 Oe also without $K_{C3}$ parameter providing the MCA energy minima along [110]. In these simulations the Fe(001) layer has been considered to be smooth. In order to further approach simulations to the experiment, we have also verified that the conclusions above are not affected by the introduction of interfacial magnetic disorder due to mismatch defects (every 30 lattice periods) with 25% excess of Fe moment at the Fe/MgO interface [61]. More detailed simulations involving also interface roughness could be needed to further approach the real experimental situation.

**S7. Magnetization alignment along [110] and irrelevance of the junction area for the superconductivity induced MCA modification**

As we mentioned in the main text, our experiments point that Fe(001) layers are close to a highly saturated state when the magnetization is directed along [100] or equivalent axes. On the other hand, micromagnetic simulations (Figure S13) show that the magnetization alignment is more robust in the [110] direction (or equivalent) rather than in the [100] direction (or equivalent). So, if we indeed reach a highly saturated state in the [100] direction, this should also be the case for the [110] direction. Therefore, the emergent stable tunneling magnetoresistance states we observe experimentally below $T_c$, cannot be explained in terms of the intermediate multi-domain states but rather correspond to the dominant [110] magnetization alignment of the Fe(001) layer.

As shown in Figure S14, our experiments shows that the observed effects remain qualitatively unchanged when the junction area is varied about an order of magnitude.
S8. Modelling

We describe the V/MgO/Fe structure by the Hamiltonian [45]

\[
H = -t \sum_{(i,j), \sigma} c^\dagger_{i,\sigma} c_{j,\sigma} - \sum_{i,\sigma} (\mu_i - V_i) c^\dagger_{i,\sigma} c_{i,\sigma} - \sum_i U_i n_{i,\uparrow} n_{i,\downarrow} + \sum_{i,\alpha,\beta} c^\dagger_{i,\alpha} (h_i \cdot \sigma) c_{i,\beta} - \frac{i}{2} \sum_{(i,j), \alpha, \beta} \lambda_i c^\dagger_{i,\alpha} \tilde{n} \cdot (\sigma \times d_{i,j})_{\alpha,\beta} c_{j,\beta}
\]  (3)

deﬁned on a cubic lattice. The ﬁrst term describes nearest-neighbor hopping. The second term includes the chemical potential and the potential barrier at the insulating MgO layers. The remaining terms describes superconducting attractive on-site interaction, ferromagnetic exchange interaction, and Rashba spin-orbit interaction, respectively. These are only nonzero in their respective regions. In the above, \(t\) is the hopping integral, \(\mu_i\) is the chemical potential, \(V_i\) is the potential barrier that is nonzero only for the MgO layer, \(U > 0\) is the attractive on-site interaction giving rise to superconductivity, \(\lambda_i\) is the local spin-orbit coupling magnitude, \(\tilde{n}\) is a unit vector normal to the interface, \(\sigma\) is the vector of Pauli matrices, \(d_{i,j}\) is a vector from site \(i\) to site \(j\), and \(h_i\) is the local magnetic exchange ﬁeld. The number operator used above is deﬁned as \(n_{i,\sigma} \equiv c^\dagger_{i,\sigma} c_{i,\sigma}\), and \(c^\dagger_{i,\sigma}\) and \(c_{i,\sigma}\) are the second-quantization electron creation and annihilation operators at site \(i\) with spin \(\sigma\). The superconducting term in the Hamiltonian is treated by a mean-ﬁeld approach, where we assume \(c_{i,\uparrow} c_{i,\downarrow} = (c_{i,\uparrow} c_{i,\downarrow}) + \delta\) and neglect terms of second order in the ﬂuctuations \(\delta\).

We consider a system of size \(N_x \times N_y \times N_z\) setting the interface normals parallel to the \(x\) axis and assuming periodic boundary conditions in the \(y\) and \(z\) directions. To simplify notation in the following, we deﬁne \(i \equiv i_x, j \equiv j_x, \| \equiv (i_x, i_y)\) and \(k \equiv (k_y, k_z)\). We apply the Fourier transform

\[
c_{i,\sigma} = \frac{1}{\sqrt{N_y N_z}} \sum_k c_{i,k,\sigma} e^{i(k \cdot i \|)}
\]  (4)

to the above Hamiltonian and use that

\[
\frac{1}{N_y N_z} \sum_{i \|} c^{\dagger}_{\|,k'} i \| = \delta_{k,k'}
\]  (5)

FIG. 14: In-plane ﬁeld rotation experiments with \(H = 70\) Oe below (blue) and above (red) \(T_C\) for \(10 \times 10\) (a), \(20 \times 20\) (b) and \(30 \times 30\) (c) \(\mu m^2\) junctions. (d) Histograms of the calculated angle between the two FM layers \(\phi_{FM}\) for these same rotations, above and below \(T_C\), showing the \([110]\) states for low temperatures. (e) and (f) show the evolution of the \([110]\), \([010]\) and \([110]\) (PIP) states with temperature for the same \(10 \times 10\) and \(30 \times 30\) \(\mu m^2\) junctions (qualitatively similar evolution is shown in Figure 2f in the main text for the \(20 \times 20\) \(\mu m^2\) junction).
We choose a new basis
\[ B_{i,k}^\dagger = [c_{i,k,\uparrow}^\dagger, c_{i,k,\downarrow}^\dagger, \bar{c}_{i,-k,\uparrow}, \bar{c}_{i,-k,\downarrow}] \] (6)
spanning Nambu×spin space, and rewrite the Hamiltonian as
\[ H = H_0 + \frac{1}{2} \sum_{i,j,k} B_{i,k}^\dagger H_{i,j,k} B_{i,k}. \] (7)

Above, the Hamiltonian matrix is given by
\[ H_{i,j,k} = \epsilon_{i,j,k} \hat{\tau}_i \hat{\sigma}_0 + \delta_{ij} \left[ \Delta_i \hat{\tau}_x \hat{\sigma}_y - i \Delta_i \hat{\tau}_y \hat{\sigma}_x \right] \\
+ h_i^x \hat{\tau}_i \hat{\sigma}_0 + h_i^y \hat{\tau}_i \hat{\sigma}_0 + h_i^z \hat{\tau}_i \hat{\sigma}_0 \\
- \lambda_i \sin(k_z) \hat{\tau}_i \hat{\sigma}_x + \lambda_i \sin(k_z) \hat{\tau}_i \hat{\sigma}_y, \] (8)

where \( \Delta_i \) is the superconducting gap which we solve for self-consistently, \( \hat{\tau}_i \hat{\sigma}_\delta \equiv \hat{\tau}_i \otimes \hat{\sigma}_\delta \) is the Kronecker product of the Pauli matrices spanning Nambu and spin space, \( \hat{\tau}^\pm \equiv (\hat{\tau}_1 \pm i \hat{\tau}_2)/2 \), and
\[ \epsilon_{i,j,k} = -2t \left[ \cos(k_y) + \cos(k_z) \right] \delta_{ij} - t \left( \delta_{i,j+1} + \delta_{i,j-1} \right) - (\mu_i - V_i) \delta_{i,j}. \] (9)

The constant term in Eq. (7) is given by
\[ H_0 = - \sum_{i,k} \left\{ 2t \left[ \cos(k_y) + \cos(k_z) \right] + \mu_i - V_i \right\} \]
\[ + N_y N_z \sum_{i} \frac{\Delta_i^2}{U_i}. \] (10)

We absorb the sum over lattice sites in Eq. (7) into the matrix product by defining a new basis
\[ W_k = [B_{1,k}^\dagger, \ldots, B_{N_x,k}^\dagger, B_{N_y,k}^\dagger]. \] (11)

Eq. (7) can then be rewritten as
\[ H = H_0 + \frac{1}{2} \sum_k W_k^\dagger H_k W_k, \] (12)

where
\[ H_k = \begin{bmatrix} H_{1,1,k} & \cdots & H_{1,N_y,k} \\ \vdots & \ddots & \vdots \\ H_{N_x,1,k} & \cdots & H_{N_x,N_y,k} \end{bmatrix} \] (13)
is Hermitian and can be diagonalized numerically. We obtain eigenvalues \( E_{n,k} \) and eigenvectors \( \Phi_{n,k} \) given by
\[ \Phi_{n,k}^\dagger = [\phi_{n,k}^\dagger, \phi_{n,N_y,k}^\dagger, \bar{\phi}_{n,N_x,k}, \bar{\phi}_{n,k}^\dagger], \]
\[ \phi_{n,k}^\dagger = [\psi_{n,k}^\dagger, \psi_{n,N_y,k}^\dagger, \bar{\psi}_{n,N_x,k}, \bar{\psi}_{n,k}^\dagger]. \] (14)

The diagonalized Hamiltonian can be written on the form
\[ H = H_0 + \frac{1}{2} \sum_{n,k} E_{n,k} \gamma_{n,k}^\dagger \gamma_{n,k}, \] (15)

where the new quasi-particle operators are related to the old operators by
\[ c_{i,k,\uparrow} = \sum_n w_{i,n,k} \gamma_{n,k}, \]
\[ \bar{c}_{i,-k,\uparrow} = \sum_n \bar{w}_{i,n,k} \gamma_{n,k}, \]
\[ c_{i,k,\downarrow} = \sum_n \bar{w}^*_{i,n,k} \gamma_{n,k}, \]
\[ \bar{c}_{i,-k,\downarrow} = \sum_n w^*_{i,n,k} \gamma_{n,k}. \] (16)

The superconducting gap is given by \( \Delta_i = U_i \langle c_{i,\uparrow}^\dagger c_{i,\downarrow} \rangle \). We apply the Fourier transform in Eq. (4) and use Eq. (16) in order to rewrite the expression in terms of the new quasi-particle operators. Also using that \( \langle \gamma_{n,k}^\dagger \gamma_{m,k} \rangle = f(E_{n,k}/2) \delta_{n,m} \), we obtain the expression
\[ \Delta_i = - \frac{U_i}{N_y N_z} \sum_{n,k} v_{i,n,k} w_{i,n,k}^* \left[ 1 - f(E_{n,k}/2) \right] \] (17)

for the gap, that we use in computing the eigenenergies iteratively. Above, \( f(E_{n,k}/2) \) is the Fermi-Dirac distribution.

Using the obtained eigenenergies, we compute the free energy,
\[ F = H_0 - \frac{1}{\beta} \sum_{n,k} \ln(1 + e^{-\beta E_{n,k}/2}), \] (18)

where \( \beta = (k_B T)^{-1} \). The preferred magnetization directions are described by the local minima of the free energy. In the main body of the paper, we use this to explain the possible magnetization directions of the soft ferromagnet when rotating an IP external magnetic field over a 2\( \pi \) angle starting at a parallel alignment with the hard ferromagnet.

Other relevant quantities to consider in modelling the experimental system is the superconducting coherence length and the superconducting critical temperature. In the ballistic limit, the coherence length is given by \( \xi = \hbar v_F/\pi \Delta_0 \), where \( v_F = \frac{1}{2} \frac{dE_k}{dk} \) is the Fermi velocity related to the normal-state eigenenergy \( E_k = -2t[\cos(k_y) + \cos(k_y) + \cos(k_z)] - \mu \), and \( \Delta_0 \) is the zero-temperature superconducting gap \( \Delta \). We choose an initial guess with a magnitude very close to zero and with a lattice site dependence similar to that of the gap just below \( T_c \).

In the main plot showing the free energy under IP rotations of the magnetization, we have chosen parameters
$t = 1$, $\mu_S = \mu_{SO} = \mu_F = 0.9$, $V = 2.1$, $U = 1.35$, $\lambda = 0.4$, $h = 0.8$, $N_0^2 = 30$, $N_{SO}^2 = 3$, $N_F^2 = 8$, and $N_y = N_z = 60$. All length scales are scaled by the lattice constant $a$, all energy scales are scaled by the hopping parameter $t$, and the magnitude of the spin-orbit coupling $\lambda$ is scaled by $ta$. In order to make the system computationally manageable, the lattice size is scaled down compared to the experimental system, however the results should give qualitatively similar results as long as the ratios between the coherence length and the layer thicknesses are reasonable compared to the experimental system. For this set of parameters, the superconducting coherence length is approximately $0.6 N_y^2$. Since the coherence length is inversely proportional to the superconducting gap, $U$ has been chosen to be large in order to allow for a coherence length smaller than the thickness of the superconducting layer. Although this results in a large superconducting gap, the modelling will qualitatively fit the experimental results as long as the other parameters are adjusted accordingly. We therefore choose the local magnetic exchange field so that $h \gg \Delta$, as in the experiment. For this parameter set, $h \approx 20\Delta$. The order of magnitude of $\lambda$ is $1 \text{ eV}\AA$, given that $t \sim 1 \text{ eV}$ and $a \approx 4 \text{ Å}$. This is realistic considering Rashba parameters measured in several materials [33]. The Rashba spin-orbit field at the interfaces of $V$/MgO/Fe is caused by a structural inversion asymmetry across the MgO layer, and breaks the inversion symmetry at the MgO interfaces [44]. This causes generation of triplet-superconductivity even for weakly spin-polarized ferromagnets with a small spin-orbit field [44]. We are therefore not dependent upon a strong magnetic exchange field and a strong spin-orbit field for realizing the observed effects. For the AF coupling contribution to the free energy, we set $f_{AF} = 0.01$ in order to fit the anisotropy of the experimental system just above $T_C$.

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