Observation of the dominant spin-triplet supercurrent in Josephson spin valves with strong Ni ferromagnets

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We study experimentally nanoscale Josephson junctions and Josephson spin-valves containing strong Ni ferromagnets. We observe that in contrast to junctions, spin valves with the same geometry exhibit anomalous $I_\phi(H)$ patterns with two peaks separated by a dip. We develop several techniques for in-situ characterization of micromagnetic states in our nano-devices, including magnetoresistance, absolute Josephson fluxometry and First-Order-Reversal-Curves analysis. They reveal a clear correlation of the dip in supercurrent with the antiparallel state of a spin-valve and the peaks with two noncollinear magnetic states, thus providing evidence for generation of spin-triplet superconductivity. A quantitative analysis brings us to a conclusion that the triplet current in out Ni-based spin-valves is approximately three times larger than the conventional singlet supercurrent.

I. INTRODUCTION

Spin-polarized ferromagnetism is antipathetic to spin-singlet superconductivity. However, destruction of singlet Cooper pairs in a ferromagnet is not an instant process. Pairing correlations survive over a certain time/distance, during which precession of spins in exchange field may create a correlated triplet pair. The corresponding odd-frequency spin-triplet order parameter has been predicted theoretically using various approaches [1][10]. This inspired intense experimental search for this exotic state in Superconductor/Ferromagnet (S/F) heterostructures [17][28]. Although supercurrents through F were reported many times [17][23], it is difficult to prove their triplet nature. First, even singlet-singlet current can flow over long ranges in clean or weak ferromagnets [1][15][10][30]. The singlet current is reduced in strong F, which should be material of choice for a critical test. Second, the supercurrent strongly depends on usually unknown domain structure in F [20][27][31], flux quantization in S [32][33], both influenced by size and geometry. This uncertainty can be obviated in nanoscale devices with mono- (or few) domain F-layers and with the flux-quantization field larger than the coercive field [32]. Finally, the long-range triplet current should appear only in the noncollinear magnetic state [2][6][10]. Therefore, unambiguous identification of the pairing order is only possible if the micromagnetic state of the actual device is known. It is not sufficient to analyze similar large-area heterostructures because their magnetic properties (coercive fields, domain structure, shape anisotropy) would be different from a nano-device. In order to prove/disprove the triplet nature of supercurrent it is necessary to demonstrate its correlation/anticorrelation with the non-collinear state [19][21][24][28]. In the end, it is all about having an in-situ control over the micromagnetic state of the studied nano-device. This is our main motivation.

The noncollinear magnetic state can be controllably created in mono-domain spin valves. The simplest is the pseudo spin valve F$_1$NF$_2$ with two F$_{1,2}$ layers separated by a normal metal (N) spacer. Triplet current in this case is second-harmonic with respect to the phase difference and is proportional to the difference between $F_1$ and $F_2$ [8][10][13][14][16] (see the Appendix for more details). Therefore, an asymmetric spin-valve F$_1 \neq F_2$ is needed for generation of the triplet current. The asymmetry (different coercive fields) is also needed for controllable tuning of the relative magnetization angle $\alpha$ between F$_{1,2}$ layers.

Here we study experimentally nano-scale SFS Josephson junctions (JJ’s) and SF$_{1}$NF$_{2}$S Josephson spin-valves (JSV’s). We use strong F (Ni) to suppress singlet currents and to make triplet currents dominant. We focus on development of various methods for in-situ characterization of micromagnetic states in our nano-devices, including magnetoresistance (MR), absolute Josephson fluxometry (AJF) and First-Order-Reversal-Curves (FORC) analysis. We observe that JSV’s behave qualitatively differently compared to JJ’s with similar geometry: they exhibit non-Fraunhofer $I_\phi(H)$ modulation with two distinct peaks separated by a dip. In-situ characterization reveals a clear correlation of the supercurrent dip with the antiparallel (AP) state of the JSV and the peaks with two noncollinear states around it. This provides an in-situ evidence for generation of spin-triplet superconductivity.

II. EXPERIMENTAL

We study nanoscale Nb(200nm)/Ni(5nm)-Cu(10nm)-
Ni(7nm)/Nb(200nm) JSV’s and Nb(200nm)/Ni(5-10nm)/Nb(200nm) JJ’s. Thin film multilayers are deposited by dc-magnetron sputtering and patterned into µm size bridges by photolithography and reactive ion etching. Subsequently they are transferred into a dual-beam Scanning Electron Microscope (SEM) / Focused Ion Beam (FIB) and nanoscale devices are made by 3D FIB nanosculpturing [20][34]. Both JJ’s...
III. PROPERTIES OF SFS JUNCTIONS

Figures 2(a,b) represent measured $I_c(H)$ patterns in the easy axis orientation for JJ’s (a) Ni (5 nm) with area $164 \times 896 \text{ nm}^2$, and (b) Ni (7 nm) with areas $200 \times 1000 \text{ nm}^2$. Fig. 2(c) shows $I_c(H_{\perp})$ along the hard axis for the same Ni (5 nm) JJ. Up and down field sweeps are shown. They exhibit hysteresis due to finite coercivity. From Figs. 2(a,b) it can be seen that the hysteresis starts/ends at about $\pm 1.5 \text{ kOe}$, which represents the saturation field. In all cases SFS JJ’s exhibit regular Fraunhoffer-type $I_c(H)$, indicating good uniformity of Ni-interlayers.

A. First-Order-Reversal-Curves analysis

FORC is a powerful tool for in-situ characterization of magnetic states in complex ferromagnetic structures. The analysis starts at the same saturated state. Then field is swept to a reversal field $H_{max}$ and measurements are carried out on the way back to the saturated state. Figs. 2(d,e) represent $I_c(H_{\perp})$ FORC’s for a JJ with Ni (10 nm). Thin black lines represent the upward sweep from the saturated $\downarrow \downarrow$ state. Red, green and blue lines are FORCs with different $H_{\perp max}$. FORC’s show very little hysteresis up to $H_{\perp max} \approx 1.1 \text{ kOe}$ and then rapidly jump to the saturated $\uparrow \uparrow$ state. This reflects an abrupt remagnetization of the Ni-nanoparticle within the JJ. Note, that the curves for different $H_{\perp max}$ in Figs. 2(d,e) have the same Fraunhofer-type shape, which just shifts upon remagnetization of the Ni-interlayer.

B. Absolute Josephson fluxometry

AJF is based on flux quantization in Josephson devices, due to which minima and maxima of $I_c$ occur at integer and half-integer flux quanta $\Phi_0$ within a device. Magnetization is related to flux via: $M = (\Phi/Ld^* - H)/4\pi$, where $L$ is the size and $d^*$ the magnetic thickness of the device. Thus absolute values of $M$ can be obtained at discrete fields determined by the flux quantization field $\Delta H = \Phi_0/Ld^*$.

C. Combined AJF+FORC

For nano-devices with large $\Delta H$ the discreetness of AJF is a limitation. To obviate this problem we combine AJF with FORC, which allows continuous determination of $M(H_{\perp max})$ for arbitrary small devices. For example, the central maxima of FORC’s in Figs. 2(d,e) correspond to $\Phi = 0$. Therefore, fields at which they occur, $H(I_{c0})$, represent absolute values of remnant magnetization $M_{rem} = -H(I_{c0})/4\pi$. Since we can vary $H_{\perp max}$ with arbitrary small step, we can get a continuous $M_{rem}(H_{\perp max})$ curve from such AJF+FORC analysis even for very small devices. This is demonstrated in Fig. 2(f) where red circles represent $-H(I_{c0}) = M_{rem}/4\pi$ as a function of $H_{\perp max}$ for FORC’s from Fig. 2(d,e). It is seen that $M_{rem}$ switches rapidly at $H_{\perp max} \gtrsim 1.1 \text{ kOe}$, which represents the coercive field. Blue squares represent $I_{c0}$, which apparently stays constant. Thus, hysteresis in SFS JJ’s is trivial: remagnetization of the F-layer changes the internal flux, which just shifts $I_c(H)$ patterns without changing their shapes.
FIG. 2. (color online). Characteristics of SFS (Nb/Ni/Nb) Josephson junctions with different Ni thicknesses. (a) and (b) $I_c(H_{∥})$ patterns for up and down field sweeps for junctions with (a) 5 nm and (b) 7 nm Ni thickness in field parallel to the long side. (c) $I_c(H_{⊥})$ modulation for the same Ni (5 nm) junction in the field perpendicular to the long side. Note that $I_c(H)$ has Fraunhofer-type modulation at both field orientations. (d-f) First-order-reversal-curves analysis of $I_c(H_{⊥})$ patterns for a junction with Ni (10 nm). Black curves in (d,e) represent the upwards sweep from the saturated negative state. Red, green and blue curves represent reversal curves with different $H_{max}$. (f) The summary of FORC analysis from (d,e). Red circles (right axis) show the position of the central maximum, representing the remnant magnetization in the junction. Blue squares (left axis) show the amplitude of the central peak. It is seen that remagnetization of the SFS junction leads to a trivial hysteresis: the $I_c(H)$ patterns maintain their shapes and merely shift due to changing magnetization of the F-interlayer.

IV. PROPERTIES OF JOSEPHSON SPIN VALVES

Figure 3 shows $I_c(H)$ patterns for three JSV’s with different sizes from the same chip in easy (a,c,e) and hard (b,d,f) axis orientations. In a stark contrast to SFS JJ’s, Figs. 2(a,b), JSV’s with a similar geometry exhibit a profound distortion of the central $I_c(H)$ maximum in the easy axis orientation. The distortion depends on the size. For the narrowest JSV’s, (a) $L = 160$ nm, the central maximum splits into two peaks, separated by a dip. With increasing JSV size, (e) $L = 300$ nm, the splitting decreases. For the hard axis orientation, corresponding to larger sizes: (d) 510 nm, (b) 860 nm and (f) 900 nm, the distortion seemingly disappears and $I_c(H)$ patterns acquire Fraunhofer-type shapes. The latter indicates good uniformity of the barrier [36]. Therefore, the double-peak distortion in the easy axis orientation for the same devices can not be ascribed to non-uniformity or defects. This is our central observation that we will analyze below.

A. Hard-axis orientation

We start with the hard-axis orientation because in this case $I_c(H_{∥})$ patterns have Fraunhofer-type shapes facilitating similar analysis as for SFS junctions. Figure 3(a) represents $I_c(H_{⊥})$ FORC’s for a JSV $300 \times 900$ nm$^2$. Thin white lines represent the upward sweep and thick color lines the FORC’s with different $H_{max}$. The lower curve indicates that up to the end of the central peak, $H_{⊥max} \lesssim 274$ Oe, FORC’s are fully reversible. Above it a hysteresis appears. However, in contrast to SFS JJ’s, see Fig. 2(e), the hysteresis is non-trivial: Remagnetization of JSV’s leads both to the shift and distortion of $I_c(H)$ patterns. In particular, it leads to a significant reduction of the central maximum, $I_{c0}$, which reaches minimum at $H_{⊥max} \approx 718$ Oe. With further increase of $H_{⊥max}$, $I_{c0}$ grows back and recovers to the original value when $H_{⊥max}$ exceed the saturation field, see Fig. 3(b). Figs. 3(c,d) represent AJF+FORC analysis: (c) $M_{rem}(H_{max})$ and (d) $I_{c0}(H_{max})$. It is seen that $I_{c0}$ is reduced by up to a factor two within the hysteresis region, marked by vertical lines, demonstrating the non-trivial type of hysteresis, compared to SFS junctions, see Fig. 2(f).

B. Easy-axis orientation

In Figure 4 we analyze behavior of the $160 \times 860$ nm$^2$ JSV’s in the easy axis orientation. Fig. 4(a) represents FORC analysis. FORC’s are reversible until $H_{∥max}$ passes the first $I_c(H_{∥})$ peak in the upward sweep (thin white lines). At higher fields hysteresis appears, accom-
FIG. 3. (color online). \(I_c(H)\) modulation patterns for three Josephson spin valves with different sizes, made at the same chip. (a,c,e) in fields parallel and (b,d,f) perpendicular to the long side. (a,b) JSV 160 × 860 nm\(^2\) at \(T \simeq 2\) K, (c,d) JSV 165 × 510 nm\(^2\) at \(T \simeq 2\) K. (e,f) JSV 300 × 900 nm\(^2\) at \(T \simeq 0.6\) K. Note non-Fraunhofer double-peak patterns in parallel field orientation.

Panied by the reduction of \(I_c\). The \(I_c\) reaches a minimum when \(H_{\parallel max}\) passes the second maximum at 816 Oe. At \(H_{\parallel max} = 916 − 1473\) Oe a state with one dominant peak is observed. With further increase of \(H_{\parallel max} \geq 1475\) Oe, the second peak reemerges. Finally, for \(H_{\parallel max}\) larger than the saturation field, \(\simeq 2\) kOe, the reversal curve becomes mirror symmetric with respect to the upward curve. Thus, hysteresis in JSV’s is non-trivial for both field orientations: the appearance of hysteresis is always accompanied by the reduction of supercurrent, as indicated in Figs. 4 (d) and 5 (c).

C. Difference between SFS junctions and JSV’s

To understand the difference in behavior of JJ’s and JSV’s, we first note that the conventional Fraunhofer \(I_c(H)\) modulation in JJ’s occurs due to flux quantization with field independent critical current density, \(J_c(H) = \text{const}\). The observed trivial hysteresis in SFS junctions suggests that upon remagnetization of a single F-layer only the total flux changes, but \(J_c\) remains unchanged. Conversely, the non-trivial hysteresis in JSV’s indicates that \(J_c\) is not constant, but depends on the relative orientation \(\alpha\) of the two F-layers. It is anticipated [8][10][32] that the triplet component should vanish in the collinear \(\alpha = 0, 180, 360^\circ\) states and should have maxima in the noncollinear \(\alpha = 90, 270^\circ\) states, see numerical analysis in the Appendix.

The origins of magnetic hysteresis in JJ’s and JSV’s are also different. For JJ’s with a single F-layer it is caused predominantly by the shape anisotropy. Presence of the second F-layer in JSV’s leads to another mechanism caused by magnetostatic interaction between F\(_1,2\)-layers, which favors the AP state. In a mono-domain case remagnetization of a JSV starts by a scissor-like rotation of \(M_{1,2}\) [32]. Such rotation is reversible and non-hysteretic. It continues until the softer F\(_1\)-layer flips and JSV switches into the AP state. Magnetostatic stability of the AP state leads to the appearance of hysteresis: the field is reversed, the spin valve will remain in the AP state. With increasing field the harder F\(_2\)-layer also flips and JSV enters into the second scissor-like non-collinear state, which gradually turns into the parallel \(\uparrow\uparrow\) state [32]. Micromagnetic simulations for our JSV’s, presented in the Appendix, confirm such a behavior but also demonstrate that remagnetization may involve formation of two domains. Few domains do not change the overall picture, but lead to additional hysteresis associated with the disappearance of each domain wall.

To summarize the above discussion, the principle difference between JJ’s and JSV’s is in \(J_c(H)\) dependence, which is constant for JJs and depends on magnetization orientation, \(J_c[\alpha(H)]\), for JSV’s. During remagnetization \(\alpha(H)\) varies from 0 to 360\(^\circ\) passing two times through noncollinear states \(\alpha = 90^\circ\) and \(270^\circ\). Therefore, the triplet component should have two peaks at \(\alpha = 90^\circ\) and \(270^\circ\), surrounding a dip in the AP state \(\alpha = 180^\circ\), while the singlet component should have one sharp maximum in the AP state, see the Appendix. This provides a robust qualitative test for the nature of the dominant supercurrent: Since the appearance of hysteresis in JSV is caused by the switching from the noncollinear scissor-state to the collinear AP state, the associated change
in $I_c$ should unambiguously reveal the dominant type of supercurrent. If $I_c$ increases than it is singlet and if decreases - triplet. The later is qualitatively consistent with our observations, see Figs. 4 (d) and 5 (c).

**D. In-situ characterization of JSV state**

Unambiguous confirmation of the triplet nature of supercurrent requires detailed knowledge of the micromagnetic state. Figs. 5 (b-e) represent the in-situ analysis of the magnetic state evolution for the easy axis orientation of the JSV. Fig. 5 (b) represents hysteresis, i.e., area between upward $I_c(H)$ and FORC’s from Fig. 4 (a). Fig. 5 (c) shows amplitudes of the first (left) main peak $I_{c1}$ in FORC’s. Fig. 5 (c) shows high-bias spin-valve magnetoresistance measured at the same device [35]. Parallel and AP states of JSV correspond to minima and maxima of MR, respectively [20]. Such the analysis provides a self-consistent understanding of the magnetic state evolution in the JSV. The saturation field, at which FORC’s stop changing, see Fig. 4 (b), and MR reaches minimum, see Fig. 5 (e), is $\sim \pm 2$ kOe. At $H < -2$ kOe the JSV is in the $\downarrow\downarrow$ parallel state $\alpha \simeq 0$. In a broad range $-2$ kOe < $H_{\text{max}}$ < 26 Oe, there is no hysteresis. Consequently, the JSV is in a mono-domain noncollinear scissor state with reversible rotation $0 < \alpha < 180^\circ$. Hysteresis appears at $H_{\text{max}} \gtrsim 26$ Oe, indicating switching into the magnetostatically stable AP state $\alpha \simeq 180^\circ$, as confirmed by the large value of MR. At $H_{\text{max}} > 816$ Oe a sudden change occurs both in hysteresis, Fig. 5 (b),

![FIG. 4](image1.png)

**FIG. 4.** (color online). Experimental FORC analysis for a JSV’s 300 × 900 nm² in the hard axis orientation at $T = 1.0$ K. (a) Thin white lines represent $I_c(H)$ for the upward field sweep starting from the saturated $\downarrow\downarrow$ state. Thick color lines are reversal curves starting from different $H_{\text{max}}$. The curves are shifted vertically for clarity. $H_{\text{max}}$ are indicated by circles and/or text. (b) Mirror-symmetric $I_c(H)$ curves for upward (blue) and downward (red) field sweeps from saturated $\downarrow\downarrow$ and $\uparrow\uparrow$ states. (c) Position of the central maximum of $I_c$ FORC’s as a function of the reversal field $H_{\text{max}}$. It represents absolute values of remnant magnetization of the JSV. (d) Amplitudes of the central maximum of downward FORC’s $I_{c0}(\text{down})$, normalized by that for the upward sweep $I_{c0}(\text{up})$ as a function of the reversal field $H_{\text{max}}$. Note that in contrast to SFS junctions, Fig. 2 (f), remagnetization of the JSV leads to a non-trivial hysteresis, which is accompanied by a significant reduction of the supercurrent.

![FIG. 5](image2.png)

**FIG. 5.** (color online). In-situ characterization of a JSV 160 × 860 nm² in the easy axis orientation at $T = 1.2$ K. (a) Experimental FORC’s (thick color lines) for different $H_{\text{max}}$, indicated by circles. The curves are shifted vertically for clarity. (b) Hysteresis of $I_c(H)$, equal to the area between upward and reversal curves in (a). (c) Amplitude of the first supercurrent peak $I_{c1}(H \sim 0)$. Arrows in the top of panel (b) depict the magnetic state of the JSV, as described in the text. (d) $I_c(H)$ patterns for upward (blue) and downward (red) field sweeps from saturated $\downarrow\downarrow$ and $\uparrow\uparrow$ states. (e) Spin-valve magnetoresistance, measured at bias current much larger than $I_c$, Dashed vertical arrows in (d,e) indicate a clear correlation between the dip in $I_c$ and the maximum of MR, corresponding to the AP state of the spin-valve.
and $I_{c1}$, Fig. 5(c). It indicates a switching out of the AP state into a second noncollinear state $180^{\circ} < \alpha < 360^{\circ}$. At $H_{|| max} \approx 1473$ Oe there is yet another jump in both hysteresis, and $I_{c1}$, before reaching the saturated $\uparrow \uparrow$ parallel state, $\alpha = 360^{\circ}$, at $\sim 2$ kOe. Such a two-step switching from AP to $\uparrow \uparrow$ parallel state is fully consistent with micromagnetic simulations presented in the Appendix and is due to formation of two domains in both layers. At $H_{|| max} \approx 1473$ Oe the thinner F-layer switches into the monodomain state, followed by the thicker F-layer close to the positive saturation field. Arrows in the top part of Fig. 5(b) sketch the evolution of magnetic states during the remagnetization.

E. Correlation between the supercurrent and the magnetic state in JSV

Now we can look at correlations between the supercurrent and the magnetic state. In Fig. 5(d) we show $I_c(H)$ patterns for this JSV. Let’s focus on the upward field sweep (blue line). It has a double-peak structure. Solid vertical lines going through Figs. 5(b–e) emphasize that the dip with the AP state. Most straightforwardly this is seen from comparison with the MR. Dashed arrows in Figs. 5(d,e) indicate that the dip in $I_c$ corresponds to the maximum of MR. Furthermore, FORC analysis, Figs. 5(b,c), indicates that the field range of the primary hysteresis, $26$ Oe $< H_{|| max} < 816$ Oe, associated with magnetostatic stability of the AP state, coincides with the field range between the two peaks and that the appearance of this hysteresis is accompanied by the reduction of the $I_{c1}$ peak. Consequently, entrance into the AP state leads to a significant reduction of $I_{c1}$ through the JSV. However, the supercurrent recovers upon leaving the collinear AP state in both direction, resulting in the observed double-peak $I_c(H)$ pattern. We emphasize that such behavior is opposite to expectations for the singlet current, which should be at maximum in the AP state and is consistent with the predictions for the odd-frequency spin-triplet supercurrent, see the Appendix.

We note that such an unusual behavior has not been reported in an earlier similar work [20] on JSV’s containing diluted CuNi ferromagnets because in that case the dominant supercurrent ($\sim 80\%$) had a singlet nature. An estimation based on numerical fitting of our data, presented in the Appendix, indicates that in our Ni-based JSV’s the triplet current amplitude is approximately three times larger than the singlet. This helps to uncover the characteristic double-peak modulation, which provides an unambiguous evidence for generation of the spin-triplet order parameter. Yet, even in Ni-based JSV’s the dip in the AP state does not go to zero, indicating that there is still a significant subdominant ($\sim 30\%$) singlet supercurrent even through a relatively thick Ni.

V. CONCLUSION

To conclude, both singlet and triplet supercurrents can flow through S/F heterostructures [11][15]. The unique feature of our work that adds to further understanding of the triplet state, along with earlier experimental works [17][28], was development of in-situ characterization techniques for an accurate assessment of micromagnetic states in actual nano-devices. In particular, we developed a new AJF+FORC technique, a powerful tool allowing absolute magnetometry of nano-devices and accurate identification of micromagnetic states. We fabricated and studied nano-scale Josephson junctions and (pseudo) spin valves with Ni-interlayers. Small sizes enabled mono-(or few) domain configurations, which could be unambiguous identified. A strong F (Ni) was employed for reduction of the singlet current, enabling the dominant triplet component. This was instrumental for observation of an extraordinary behavior of JSV’s, qualitatively different from similar-size SFS JJ’s. Namely, $I_c(H)$ modulation of studied JSV’s had two main peaks separated by a dip and exhibited a non-trivial hysteresis, accompanied by reduction of $I_c$. The in-situ characterization showed a clear correlation of the $I_c$ dip with the antiparallel state of the spin valve and the two peaks to the two noncollinear states aside of it, thus providing unambiguous evidence for generation of the spin-triplet order parameter.

Appendix: Numerical modeling of Josephson spin-valve

To clarify the behavior of JSV we perform numerical analysis. Josephson current in JSV’s has three main components [10]: the short-range spin-singlet $I_{ss}$, the long-range spin singlet $I_{sl}$ and the long-range spin-triplet $I_{tr}$. Their local values depend on relative angles, $\alpha(x,y)$, between magnetizations, $M_{1,2}$, of the two F-layers and the Josephson phase difference between S-electrodes $\varphi(x,y)$:

$$I_{ss}(x,y) = I_{s0} \cos^2[\alpha(x,y)/2] \sin[\varphi(x,y)], \quad (1)$$

$$I_{sl}(x,y) = I_{s0} \sin^2[\alpha(x,y)/2] \sin[\varphi(x,y)], \quad (2)$$

$$I_{tr}(x,y) = I_{t0} \sin[\alpha(x,y)] \sin[2\varphi(x,y)]. \quad (3)$$

To calculate $I_c(H)$ we follow the procedure from Ref. [32]. First we perform a micromagnetic simulation in OOMMF, which provides the two-dimensional distribution of components $M_{x1,2}(x,y)$ and $M_{y1,2}(x,y)$. Next, we calculate $\varphi$ by direct integration of:

$$\frac{\partial \varphi(x,y)}{\partial y} = \frac{2\pi d_x}{\Phi_0} H_x + \frac{2\pi d_1}{\Phi_0} 4\pi M_{x1} + \frac{2\pi d_2}{\Phi_0} 4\pi M_{x2}, \quad (4)$$

$$\frac{\partial \varphi(x,y)}{\partial x} = \frac{2\pi d_y}{\Phi_0} 4\pi M_{y1} + \frac{2\pi d_2}{\Phi_0} 4\pi M_{y2}. \quad (5)$$

Here $H_x$ is the applied magnetic field in the $x$-direction and $d_{1,2}$ are the thicknesses of $F_{1,2}$ layers. The total su-
percurrent \( I_s = I_{ss} + I_{sl} + I_{tr} \), Eq. (1-3), is integrated over the JSV area using the obtained values \( \alpha(x,y) \) and \( \varphi(x,y) \). To find the critical current we maximized the supercurrent with respect to the integration constant. For more details of the simulation procedure see Ref. \[32]\.

In Figures 6 and 7 we show results corresponding to one of the studied JSV's Ni(5nm)/Cu(10nm)/Ni(7.5nm) with sizes 160×860 nm\(^2\). Simulations are shown for the following set of supercurrent amplitudes: \( I_{ss0} = 0.1, I_{sl0} = 1.0, I_{tr0} = 3.0 \), which fits well the experimental data. From this we conclude that the triplet current amplitude in our JSV's is approximately three times larger than the singlet, \( I_{tr0}/(I_{sl0} + I_{ss0}) \approx 3 \).

Fig. 6 (a) shows the magnetization curve along the easy axis (see the inset). Black lines represent the major hysteresis loop and color lines - FORC's with \( H_{\parallel \text{max}} \) indicated by circles. The spin valve appears to be at the border between the mono- and the two-domain states. Upon sweeping of the field upwards from the saturated \( \downarrow \downarrow \) state, magnetization in F-layers first curves into a C-shape (state-A), which is reversible without hysteresis (see the green line). Then the \( F_1 \) and \( F_2 \) layers switch sequentially into the state with two domains (states B and C) simultaneously flipping the \( x \)-component of magnetization. Hysteresis appears in the state B (red line), which corresponds to the AP state.

Fig. 6 (b) shows amplitudes of the long-range singlet (blue) and triplet (red) supercurrents for an upward field sweep. In the AP state-B the singlet amplitude is large and the triplet is small. On both sides of it, there are two noncollinear states A, C with large triplet and small singlet amplitudes. At large positive/negative fields the JSV is in the parallel state with vanishing of both singlet and triplet long-range components. The shape of \( I_c(H) \)
pattern of the JSV depends on relative amplitudes of singlet and triplet components. Fig. 6(c) shows the case with the dominant triplet current ($I_{c\parallel} = 0.1$, $I_{c\perp} = 1.0$, $I_{tr\parallel} = 3.0$) for the total (black), singlet (blue) and triplet (red) currents. Since in this case the total current is dominated by the triplet current, $I_c(H)$ has two peaks corresponding to the noncollinear states A and C, separated by a dip, corresponding to the AP state, similar to the experimental data in Fig. 5(d).

In Figs. 6 (d-i) we analyze $I_c(H)$ FORC's, corresponding to $M(H)$ curves with the same color in Fig. 6 (a). Panel (d) represents the case when $H_{c1max}$ is within the first $I_c(H)$ peak. Here the spin valve is in the reversible noncollinear state-A. In (e) $H_{c2max}$ is within the dip in the AP state-B. As emphasized in the main text, the fingerprint of the AP state is the appearance of hysteresis, see red line in Fig. 6 (a). Panel (f) demonstrates that within this hysteresis the singlet current is increased (top panel) and the triplet decreased (bottom panel). Thus, the change of the current upon appearance of hysteresis tells us about the nature of the dominant supercurrent. Since in our simulations the triplet current is dominant, there is an overall drop of $I_c$ at the hysteretic branch, as seen in Fig. 6 (e). Panels (g) and (h) show FORC's after switching out of the AP state B into the noncollinear state C with domains. Note that along with some metastability associated with domains, in Fig. 6 (h) we observe a net enhancement of the central noncollinear peak at the expense of the second peak. Finally, panel (i) shows $I_c(H)$ starting from fully saturated states. Overall, presented simulations are in a good agreement with experimental data for JSVs' from Fig. 5 (a). Simulations reproduce both the double-peak structure of $I_c(H)$ for points A) the first non-collinear state upon coherent rotation from the negative parallel state, B) antiparallel state and C) the second non-collinear state upon switching from the AP state.

Fig. 7(c) shows sample-averaged values of normalized triplet, $\propto \sin^2(\alpha)$ (red) and singlet, $\propto \sin^2(\alpha/2)$, (blue) current amplitudes, see Eqs. (3) and (2). The behavior of both components is similar to the easy axis case, Fig. 6 (b). I.e., in this respect the field orientation does not make a principle difference. However, the $I_c(H)$ pattern is strongly affected in this orientation. Figs 7 (d-f) show magnetic field modulation of (d) triplet, (e) singlet, and (f) total currents. It can be seen that although triplet current amplitude in panel (c) has two clear peaks (A, C), however, the large length $L$ of the JSV at this field orientation makes $\Delta H$ much smaller than the coercive field. Therefore at points A and C with the largest triplet amplitudes there are already many flux quanta inside the JSV, suppressing the triplet critical current by more than an order of magnitude. As a result, the characteristic double-maxima feature becomes unrecognizable in the total $I_c(H)$ modulation. Thus, the difference between easy and hard axis orientations is entirely due to the flux quantization effect. Nevertheless, both numerical simulations, see Figs. 6(b) and 7(c) and experimental analysis in Figs. 4 and 5 demonstrate that the essential physics remains independent of the field orientation.

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FIG. 7. Simulations for the hard axis orientation. (a) Magnetization curves. (b) Configurations of magnetization at points A, B, C. (c) Sample-averaged values of normalized triplet \(\alpha \sin^2(\alpha)\), red and singlet \(\alpha \sin^2(\alpha/2)\), blue current amplitudes for the upward field sweep. Panels (d-f) show magnetic field modulation patterns for (d) triplet, (e) singlet, and (f) total critical currents. It can be seen that although triplet current amplitude in (c) has two clear peaks in noncollinear states A, C, however, small flux quantization field at this field orientation leads to rapid damped oscillations, which suppresses the triplet supercurrent (see the vertical scale). Therefore, the characteristic double-maxima become unrecognizable in the total \(I_c(H)\) modulation (f). This explains how flux quantization effect changes \(I_c(H)\) patterns for JSV’s from a double- to a single-peak for easy- and hard-axis orientations, respectively.

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