Half-Metallic Superconducting Triplet Spin MultiValves

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We study spin switching effects in finite-size superconducting multivalve structures. We examine F\(_1\)F\(_2\)SF\(_3\) and F\(_1\)F\(_2\)SF\(_3\)F\(_4\) hybrids where a singlet superconductor (S) layer is sandwiched among ferromagnet (F) layers with differing thicknesses and magnetization orientations. Our results reveal a considerable number of experimentally viable spin valve configurations that lead to on-off switching of the superconducting state. For S widths on the order of the superconducting coherence length \(\xi\), non-collinear magnetization orientations in adjacent F layers with multiple spin-axes leads to a rich variety of triplet spin-valve effects. Motivated by recent experiments, we focus on samples where magnetizations in the F\(_1\) and F\(_2\) layers exist in a fully spin-polarized half metallic phase, and calculate the superconducting transition temperature, spatially and energy resolved density of states, and the spin-singlet and spin-triplet superconducting correlations. Our findings demonstrate that superconductivity in these devices can be completely switched on or off over a wide range of magnetization misalignment angles due to the generation of equal-spin and opposite-spin triplet pairings.

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I. INTRODUCTION

Over the last two decades, the interplay of superconductivity and ferromagnetism has fueled interest in exploring ferromagnet (F) and superconductor (S) hybrid structures for low temperature spintronic applications\(^1\)–\(^7\) One intriguing consequence of this interplay is the creation of spin-triplet Cooper pairs that was predicted theoretically\(^5\),\(^6\),\(^7\). To confirm the generation of these unconventional pairs, much progress has been made so far, both theoretically and experimentally\(^8\)–\(^20\). One of the first signatures of the existence of spin-polarized superconducting correlations was observed in a planar half-metallic Josephson junction\(^30\). Since a half-metal supports only one spin direction, it was concluded that the supercurrent should be carried by an equal-spin triplet channel.

The two kinds of basic spin valves that have been mainly studied both experimentally and theoretically are F\(_1\)SF\(_2\) and SF\(_1\)F\(_2\) based structures\(^3\),\(^31\)–\(^45\). These systems offer simple and controllable platforms that can reveal signatures of spin triplet superconducting correlations. If differing ferromagnetic materials, constituting the left and right F layers are chosen properly, they respond to an external magnetic field in different ways, providing active control of the magnetization misalignment angles through variations in the intensity and direction of an external magnetic field. It was shown that the superconducting transition temperature\(^46\)–\(^50\) and density of states\(^5\),\(^5\),\(^5\)–\(^5\) reveal prominent signatures of the long-ranged spin-triplet superconductivity\(^5\) as a function of magnetization misalignment. Nevertheless, a direct and clear observation of the equal-spin triplet pairings in superconducting hybrids is still elusive.

In a recent experiment\(^48\) involving a SF\(_1\)F\(_2\) spin valve, it was observed that the superconducting critical temperature \(T_c\) in MoGe-Ni-Cu-CrO\(_2\), where F\(_2\) is the half-metallic compound, CrO\(_2\), can have a variation as high as \(\Delta T_c \sim 800\) mK when varying the magnetization misalignment angle. This order of \(T_c\) variation is much larger compared to when standard ferromagnets are used [i.e., CuNi-Nb-CuNi\(^39\), CoO\(_x\)-Fe\(_1\)-Cu-Fe\(_1\)-Pb\(^39\), Co-Cu-Py-Cu-Nb\(^40\), and Co-Nb-Co-Nb\(^45\)], albeit using a relatively strong out-of-plane magnetic field of \(H \sim 2T\). This was consistent with theory that demonstrated the largest variations in \(T_c\) of a SF\(_1\)F\(_2\) spin valve\(^50\) occurred when F\(_2\) is a half-metal, rather than a ferromagnet with a smaller exchange energy\(^47\). A very recent experiment\(^47\) involving the half metal La\(_{0.6}\)Ca\(_{0.4}\)MnO\(_3\), consisted of a LCMO-Au-Py-Cu-Nb stack under the influence of a much weaker in-plane magnetic field of \(H \sim 3.3mT\) to rotate the magnetization. This configuration demonstrated a slight improvement with \(\Delta T_c \sim 150mK\) compared to experiments involving standard ferromagnets that yielded \(\Delta T_c \sim 50mK\)\(^39\), and \(\Delta T_c \sim 120mK\(^40\). Since strong variations in \(T_c\) can be representative of singlet superconductivity weakening and its conversion to the triplet channel, it is of fundamental interest to create spin valve structures with the largest variations in \(T_c\) by a weak external magnetic field. On the one hand, it can provide unambiguous signatures of the presence of equal-spin triplet correlations under magnetization rotation. On the other hand, by restricting the magnetization variations to reside in-plane, the overall magnetization state in the ferromagnet can be manipulated with much weaker fields. This low-field magnetization control considerably increases device reliability and provides an effective spin switch for technological purposes.

In this paper, we introduce the F\(_1\)F\(_2\)SF\(_3\) and F\(_1\)F\(_2\)SF\(_3\)F\(_4\) multivalves (depicted in Fig. 1), where the two outer F layers are half-metallic. The presence of the additional ferromagnetic layers amplifies the singlet-triplet conversion process, leading to a larger spin valve effect compared to standard spin valves described above. We show this by considering a wide range of layer thicknesses and magnetizations to achieve a broad critical temperature variation as the magnetization in one of the F layers rotates. As mentioned earlier, the transition temperature of a spin valve system\(^46\)–\(^48\),\(^50\) and the density of states\(^5\)–\(^5\) are two experimental quantities that can determine the degree in which triplet pair conversion takes place for a given device.
Our investigation is within the ballistic regime using a microscopic self-consistent formalism that can accommodate the large variation in energy scales present in the problem. In particular, the multivalve structures considered here contain conventional ferromagnets with relatively weak exchange fields \( (h/E_F \ll 1) \) in addition to the surrounding half metals \( (h/E_F = 1) \). Our results demonstrate that, stemming from nontrivial interlayer spin-valve effects, superconductivity can be effectively switched on and off over a wide range of relative magnetization misalignment angles. We note that this feature is absent or occurs to a limited extent in the previously studied\(^{30,55} \) individual F\(_1\)F\(_2\)SF\(_2\) and SF\(_1\)F\(_2\) spin-valves. We complement the \( T_c \) studies by investigating various pair correlations and the corresponding local density of states, where the emergence of a zero-energy peak is associated with the presence of equal-spin triplet correlations.

The rest of the paper is organized as follows. We first present an overview of the theoretical framework used. In Sec. II A we study the superconducting transition temperature for two multivalve configurations. In Sec. II B we present the corresponding local density of states, paying particular attention to the DOS at zero energy. Lastly, we present the singlet and triplet superconducting correlations generated and discuss their behavior in Sec. II C. Finally, we give concluding remarks in Sec. III.

II. THEORY AND RESULTS

To accurately describe the ballistic multivalve configurations displayed in Fig. 1, we solve the spin-dependent Bogoliubov-de Gennes (BdG) equations in a fully self-consistent manner. In this formalism we denote the quasiparticle's energy and their associated probability amplitudes by \( \epsilon_n, u_{n\sigma}, \) and \( v_{n\sigma} \) \((\sigma = \uparrow, \downarrow)\), respectively:

\[
\begin{pmatrix}
\mathcal{H}_0 - h_z & -h_x + i h_y & 0 & \Delta(x) \\
-h_x - i h_y & \mathcal{H}_0 + h_z & \Delta(x) & 0 \\
0 & \Delta'(x) & -\mathcal{H}_0 - h_z & -h_x - i h_y \\
\Delta'(x) & 0 & -h_x + i h_y & -\mathcal{H}_0 + h_z \\
\end{pmatrix} \times 
\begin{pmatrix}
u_{n\uparrow} \\
u_{n\downarrow} \\
u'_{n\uparrow} \\
u'_{n\downarrow} \\
\end{pmatrix} = 
\begin{pmatrix}
u_{n\uparrow} \\
u_{n\downarrow} \\
u'_{n\uparrow} \\
u'_{n\downarrow} \\
\end{pmatrix},
\]

in which \( \Delta(x) \) represents the pair potential, calculated self-consistently as shown below [Eq. (3)]. For the in-plane magnetization rotations considered here, the components of the exchange field \( h = (h_x(x), h_y(x), h_z(x)) \), so that the exchange field which vanishes in the \( S \) layer, can in general vary between the ferromagnet layers. The free-particle Hamiltonian \( \mathcal{H}_0(x) \) is defined as:

\[
\mathcal{H}_0(x) \equiv \frac{1}{2m} \left( -\frac{d^2}{dx^2} + \frac{k^2_x + k^2_y}{\pi} \right) - E_F + U(x),
\]

where \( E_F \) denotes the Fermi energy, and \( U(x) \) is a spin-independent scattering potential. The multivalve layers in Fig. 1, are translationally invariant in the \( yz \) plane and hence quasiparticle motion in this plane is appropriately described by the good quantum numbers \( k_x \) and \( k_y \). For this reason, all spatial variations take place in the \( x \) direction, and the system is considered quasi-one dimensional.

Ferromagnetism and superconductivity are two competing types of ordering and their simultaneous existence in space results in nontrivial spatial profiles for the pair potential \( \Delta(x) \). Therefore, to account properly for proximity effects, it is necessary to obtain the pair potential in a self-consistent manner via:

\[
\Delta(x) = \frac{g(x)}{2} \sum_n \left[ u_{n\uparrow}(x) v_{n\downarrow}^*(x) + u_{n\downarrow}(x) v_{n\uparrow}^*(x) \right] \tanh \left( \frac{\epsilon_n}{2T} \right),
\]

where \( g(x) \) is the pair coupling constant that is nonzero solely inside the superconductor layer and the sum above is restricted to the quantum states with positive energies below the Debye energy cutoff \( \omega_D \).

To compute the transition temperature, we use the fact that \( \Delta(x)/\Delta_0 \ll 1 \) close to the critical temperature, where \( \Delta_0 \) is the bulk pair potential at zero temperature, \( T = 0 \). In this regime, the self-consistency equation [Eq. (3)] can be linearized near the transition through a perturbative expansion of the quasiparticle amplitudes and energies and retaining terms to first order fixer. As part of the linearization process, the pair potential \( \Delta(x) \) and quasiparticle amplitudes are Fourier expanded in a sine-wave basis. For example, the zeroth-order wavefunctions are written, \( u^0_{n\sigma} = \sqrt{2/d} \sum_p u^0_{n\sigma}(k_p) \sin(k_p x) \) and \( v^0_{n\sigma} = \sqrt{2/d} \sum_p v^0_{n\sigma}(k_p) \sin(k_p x) \), where \( k_p = p\pi/d \). Using standard perturbation theory techniques described elsewhere,\(^{34,57} \) we arrive at the following matrix eigenvalue problem:

\[
\Delta_i = \sum_q J_{iq}\Delta_q,
\]
FIG. 2. The normalized transition temperature as a function of magnetization rotation in F$_3$ layer of a F$_3$F$_3$SF$_3$ spin multivale. In panel (a) the thickness of F$_2$ layer changes while in (b) the thickness of F$_3$ varies.

where the $\Delta_q$ are the first-order expansion coefficients for $\Delta(x)$, and $J_{iq}$ are the matrix elements written entirely in terms of zeroth-order quantities:

$$J_{iq} = \frac{gN_0}{8\pi k_F d} \int d\epsilon \sum_{m,n} \left( T_{qmn} F_{inn} \tanh \left( \frac{\epsilon_n}{2T} \right) \right)$$

where $\epsilon_{\perp} = \sqrt{(2m)(k_F^2 + k_z^2)}$ is the quasiparticle kinetic energy in the transverse direction, $N_0$ is the density of states at the Fermi energy, and $F_{qmn}$ are the unperturbed zeroth-order energies. The zeroth-order quasiparticle amplitudes ($u^\dagger_{mn}$) and corresponding energies ($\epsilon_n$) are found by solving Eq. (1) with $\Delta(x) = 0$. We also define, $T_{qmn} = \pi \sqrt{2d} \sum_{p,r} K_{qpr} (u_{mp}^\dagger v_{nr} + u_{mp} v_{nr}^\dagger)$, where

$$gK_{qpr} = (2/d)^{3/2} \int_0^d dx g(x) \sin(k_x x) \sin(k_p x) \sin(k_r x).$$

Experimentally accessible information regarding the quasiparticle spectra is contained in the local density of single-particle excitations in the system. This includes zero-energy signatures in the form of peaks in the density of states (DOS), which can reveal the emergence of equal-spin triplet pairings in either the ferromagnet or superconductor regions. The total local density of states, $N(x, \epsilon)$, includes contributions from both the spin-up and spin-down quasiparticle states: $N(x, \epsilon) = N_1(x, \epsilon) + N_2(x, \epsilon)$, where

$$N_q(x, \epsilon) = -\sum_n \left[ |u_{mr}(x)|^2 f'(\epsilon - \epsilon_n) + |v_{nr}(x)|^2 f'(\epsilon + \epsilon_n) \right]$$

in which $f'(\epsilon) = \partial f(\epsilon) / \partial \epsilon$ is the derivative of the Fermi function.

In order to study the various superconducting correlations that can arise, we define the triplet pair amplitudes in terms of the field operators:

$$f_0(t) = \frac{1}{2} \left[ \langle \psi_1(t) \psi_2(0) \rangle - \langle \psi_2(t) \psi_1(0) \rangle \right],$$

$$f_1(t) = \frac{1}{2} \left[ \langle \psi_1(t) \psi_2(0) \rangle + \langle \psi_2(t) \psi_1(0) \rangle \right],$$

$$f_2(t) = \frac{1}{2} \left[ \langle \psi_1(t) \psi_2(0) \rangle + \langle \psi_2(t) \psi_1(0) \rangle \right].$$

where $t$ is the relative time in the Heisenberg picture. If we consider the quantization axis fixed along the $z$ axis, the triplet amplitudes $(f_0(t), f_1(t), f_2(t))$ can be written in terms of the quasiparticle amplitudes:

$$f_0(t) = \frac{1}{2} \sum_n \left[ u_{n1}(t) v_{n2}^*(t) - u_{n2}(t) v_{n1}^*(t) \right] \zeta_n(t),$$

$$f_1(t) = -\frac{1}{2} \sum_n \left[ u_{n1}(t) v_{n2}^*(t) + u_{n2}(t) v_{n1}^*(t) \right] \zeta_n(t),$$

$$f_2(t) = -\frac{1}{2} \sum_n \left[ u_{n1}(t) v_{n2}^*(t) - u_{n2}(t) v_{n1}^*(t) \right] \zeta_n(t),$$

where $\zeta_n(t)$ is given by,

$$\zeta_n(t) = \cos(\epsilon_n t) - i \sin(\epsilon_n t) \tanh \left( \frac{\epsilon_n}{2T} \right).$$

We consider an in-plane Stoner-type exchange field for each magnetic layer,

$$h_i = h(0, \sin \theta_i, \cos \theta_i),$$

where $h_i$ are the magnitudes of the exchange field in each layer denoted by $i$, and $\theta_i$ are the angles that $h_i$ make with the $z$-axis (see Fig. 1). In situations where it is more convenient to align the quantization axis with the local exchange field direction, we perform a spin rotation using the transformations found in the Appendix. The result is

$$f'_0 = f_0 \cos \theta_i + i \sin \theta_i f_2,$$

$$f'_1 = f_1,$$

$$f'_2 = \cos \theta_i f_1 + i \sin \theta_i f_0.$$

When presenting results, we normalize all lengths by the Fermi wavevector $k_F$, e.g., $D_i = k_F d_i$, $X = k_F x$. We set the superconducting coherence length to the normalized value of $k_F d_0 = 100$. The half-metallic layers have fixed widths $D_{1,4} = 100$. The quasiparticle energies are scaled by the bulk superconducting gap $\Delta_0$, and the critical temperature of a sample by $T_0$, the transition temperature of its bulk counterpart. The local DOS is normalized by the DOS of a normal metal.
The procedure for identifying the transition temperature $T_c$ versus the magnetization strength of $F_3$ is shown (see Fig. 1(a)), where $F_3$ is a half-metallic with two minima at $T_3 = 0.8\pi$ and $D_2 \approx 10$. The critical temperature shows a nonmonotonic behavior vs both $\theta_3$ and $D_2$ with two minima at $D_2 \sim 10$ and $D_2 \sim 20$. Similar trends are obtained when $D_3$ varies. Nonetheless, Fig. 2(b) shows an effectively stronger spin valve effect with $T_c$ spanning the range $0 < T_c/T_0 < 0.8$, compared to Fig. 2(a) which has $0 < T_c/T_0 < 0.5$. In addition, at $D_3 \sim 20$, the critical temperature is vanishingly small for all values of $\theta_3$. These results demonstrate an effective spin switch that can turn superconductivity on or off using a multivalve $F_1F_2SF_3$ configuration with experimentally accessible parameters.

Next, we incorporate an additional half-metallic layer, and consider the superconducting critical temperature of the $F_1F_2SF_3F_4$ multivalve in Fig. 3. Having the outer $F_1$ and $F_4$ layers half-metallic with $h_{1,4} = E_F$ (and thus only one spin band at the Fermi level) maximizes the generation of equal-spin triplet pairs. In Figs. 3(a), 3(b), and 3(c) the normalized critical temperature $T_c/T_0$ is plotted as a function of the magnetization orientation angles in the $F_{2,3}$ layers $\theta_2$ and $\theta_3$. Here, the weaker inner ferromagnets have the exchange fields $h_2 = h_3 = 0.05E_F$ and widths $D_2 = D_3 = 10$. The superconductor has $D_2 = 250$, corresponding to a relative thickness $d_2/\xi_0 = 2.5$. In Figs. 3(a), 3(b), and 3(c) the magnetization in the right half-metal $F_4$ is set along (a) $z$, (b) $y$, and (c) $-z$, while while the orientation of the left half-metal $F_1$ is fixed along the $z$ direction. As seen, in Fig. 3(a), the critical temperature is zero at $\theta_{2,3} \sim \pi/2$. Thus, at this magnetization configuration, the system transitions to a normal resistive state for all temperatures. By changing the magnetization
alignment in $F_3$, panels (b) and (c) demonstrate that regions of very low $T_c$ (blue regions) shift to larger $\theta_2$ and $\theta_3$. The critical temperature mappings in (a)-(c) suggest that a large number of combinations of magnetization alignments leads to effective spin switches with large critical temperature variations $\Delta T_c(\theta_3) = T_{c,\text{max}}(\theta_3) - T_{c,\text{min}}(\theta_3)$. In Fig. 3(b), as seen, the minimum of $T_c$ exceeds zero and also regions of very small $T_c$ occur over narrower angular ranges of $\theta_2$ and $\theta_3$.

Next in Fig. 3 we investigate how the critical temperature is modified when varying $\theta_3$ (we set $\theta_2 = \pi/2$) and (d) the dimensionless thickness $D_2$ ($D_3 = 10$) or (e) the thickness $D_3$ ($D_2 = 10$). For certain ferromagnet widths $D_{2,3} \sim 10$ and 20, the critical temperature is severely diminished for a broad range of $\theta_3$. Indeed, Fig. 3(d) shows that if the system is in a superconducting state at $T = 0$, superconductivity is completely switched off for orientations with $\theta_3 \sim \pi/2$ and $\sim 1.5\pi$. This type of switching effect occurs over an extended angular range $0.5\pi \leq \theta_3 \leq 1.5\pi$ for $D_3 \sim 20$, indicative of a strong spin valve effect in this regime. The same feature appears in Fig. 3(e) except now the on-off superconductivity switching regime occurs at $D_3 = 10$ and broadens to $0.2\pi \leq \theta_3 \leq 1.5\pi$.

The choice of relatively weak ferromagnets for $F_2$ and $F_3$ generates opposite-spin triplet correlations in those regions and a subsequent conversion into spin-triplet pairs that can propagate within the half-metallic regions, modifying $T_c$. To identify the optimal ferromagnetic strengths, we show in Fig. 3(f) the critical temperature vs the normalized magnetization strength in both $F_2$ and $F_3$. For simplicity, we set $h_2 = h_3 = h$. It is evident that there is a broad region spanned by $h$ and $\theta_3$ in which $T_c \approx 0$, creating an effective superconductivity on-off switch. In particular, superconductivity is shown to disappear at any temperature when $0.2\pi \leq \theta_3 \leq \pi$ and $0.03 \leq h/E_F \leq 0.06$. Nonetheless, $\theta_3 = 0.5\pi$ and $1.5\pi$, overall, provides smaller critical temperatures within $0.03 \leq h/E_F \leq 0.22$. These results demonstrate that the use of ferromagnets $F_{2,3}$ with $h/E_F \ll 1$ causes the greatest decline in the superconducting state. Below, this will be discussed in terms of the population of both the equal-spin and opposite-spin triplet correlations in each relevant region of the spin valve. Lastly, in a similar fashion we show the importance of using half-metallic outer layers to achieve enhanced spin valve effects. In Fig. 3(g), we exhibit the critical temperature vs the normalized exchange energy $h_1/E_F$. The ferromagnets again have $D_{2,3} = 10$ and $h_{2,3}/E_F = 0.05$. The magnetization in $F_2$ is aligned according to $\theta_3 = 0.5\pi$ (along the y-direction) and the first outer layer has $h_1 = E_F$. The exchange field strength in $h_1$ varies from 0 to $E_F$, or equivalently, from a nonmagnetic normal metal to a fully spin polarized half-metal. Consistent with previous studies, the results shown here demonstrate that when the layer adjacent to the weak ferromagnet has one spin state present at the Fermi energy, the greatest variations in $T_c$ as a function of magnetization rotation can occur. Switching between superconducting and normal resistive states has previously been found in $F_1$FeF$_2$ and $F_1$FeS$_2$ structures. By incorporating multiple half-metallic layers, the variations in $T_c$ found here, with a fairly thick S layer, are considerably larger than what has been previously reported for the simpler Fe$_1$Fe$_2$ and Fe$_1$FeS$_2$ counterparts.

The conversion of opposite-spin triplet pairs into equal-spin triplet pairs is enhanced by coupling a weaker ferromagnet with the half-metal due in part to the preservation of phase coherence that would otherwise be destroyed by the single-spin half-metal. To exemplify this, and to compare the relative strengths of the two types of spin valves, we present in Fig. 4, the normalized $T_c$ as a function of magnetization orientation $\theta_3$ for both the (a) $F_1$FeS$_2$ and (b) $F_1$FeS$_2$Fe$_4$ types of structures. The thickness of the superconducting layer is fixed at $D_3 = 250$, which serves to effectively illustrate which device configuration leads to the largest $\Delta T_c$ variations. The ferromagnetic layers $F_1$ and $F_4$ are half metallic, while the other layers are much weaker, standard ferromagnets. The remaining parameters are set identical to the cases previously shown. We examine three magnetization orientations in the $F_2$ layer: $\theta_2 = 0, \pi/2, \pi$, while rotating $\theta_3$ continuously from 0 to $2\pi$. In the top panel, the angle $\theta_2 = 0$ corresponds to a FSF configuration, while for the lower panel it coincides with a $F_1$FeS$_2$F$_3$ structure. Thus, by appropriately varying the magnetizations, the multivalves can be reduced to their simpler $F_1$FeS$_2$ and $F_1$FeS$_2$ spin-valve counterparts. As seen, $\theta_3 = 0$ induces the weakest variations in $T_c$ for both devices. The case with $\theta_2 = \pi$ introduces moderate variations in $T_c$, while the largest changes arise when $\theta_2 = \pi/2$, corresponding to the spin multivalve configurations introduced in this paper. It is evident that
FIG. 5. The normalized spatial ($X = k_F x$) and energy ($\epsilon/\Delta_0$) resolved density of states of a ballistic $F_1 F_2 S F_3 F_4$ structure. The $F_{1,4}$ layers are in the half-metallic phase, and the magnetization in the $F_{1,2,4}$ layers is fixed along the $z$ axis. Each panel (a)-(e) corresponds to a different magnetization orientation, with $\theta_1 = 0, \pi/3.6, \pi/2, \pi/1.65, \text{and} \pi$, respectively.

The critical temperature for the $F_1 F_2 S F_3 F_4$ multivalve reaches zero when $\theta_2 = \pi/2$ and $0.48\pi \leq \theta_3 \leq 0.52\pi$, which is equivalent to a normal resistive state. Thus, it is apparent that the $F_1 F_2 S F_3 F_4$ device can provide stronger variations in the superconducting critical temperature compared to $F_1 F_2 S F_3$ and simpler spin valves.

B. Density of States

The study of single-particle excitations in these systems can reveal important signatures in the proximity induced singlet and triplet pair correlations. A useful experimental tool that probes these single-particle states is tunneling spectroscopy, in which the local DOS, $N(x, \epsilon)$, can be measured as a function of position $x$ and energy $\epsilon$. In Fig. 5, the local DOS is shown as a function of the normalized quasiparticle energy $\epsilon/\Delta_0$ and normalized location $X$ within the $F_2 S F_3$ region of a $F_1 F_2 S F_3 F_4$ multivalve. All plots are normalized to the corresponding DOS in a bulk sample of $S$ material in its normal state. To be consistent, we use the same layer thicknesses and exchange field magnitudes found in Fig. 3(a). In Figs. 5(a)-5(e) the magnetization in the $F_1$ layer is rotated incrementally according to $\theta_1 = 0, \pi/3.6, \pi/2, \pi/1.65, \text{and} \pi$. The $F_{1,2,4}$ layers have their magnetizations oriented along $z$, i.e., $\theta_{1,2,4} = 0$.

We note that, as seen in Fig. 3, the variation of both $\theta_2$ and $\theta_3$ results in a fairly wide range of magnetization directions where $T_c \sim 0$, and thus in those cases the system is not superconducting at any temperature. Hence, $\theta_2 = 0$ is chosen so that $T_c$ is nonzero over a wider range in parameter space. In (a), the magnetizations in each layer are collinear and directed along $z$, allowing only the opposite-pair correlations to exist. Although there is no gap in the energy spectrum, near the center of the superconductor ($X \sim 225$), traces of the BCS-like

![FIG. 6. The opposite-spin ($f_0$) and equal-spin ($f_1$) pairings vs $\theta_3$, averaged over each region in a $F_1 F_2 S F_3 F_4$ structure where $F_{1,4}$ are half-metallic. In the top row, (a) and (b), we set $\theta_3 = 0$, in the middle row, (c) and (d), $\theta_2 = \pi/2$, while in the bottom row, (e) and (f), $\theta_2$ is equal to $\pi$.](image-url)
energy structure are seen. The self-consistent proximity effects within the vicinity of the interfaces (found near the endpoints of the X range) however result in an increase of subgap states $|e/\Delta_0| < 1$. We see in Fig. 3(b) that the deviation of the magnetization orientation in $F_3$ from 0 to $\pi/3.6$ results in a peak at zero energy near the $S/F_3$ interface ($x \sim 360$), and within the S region. As Fig. 5(c) and 5(d) depict, as the magnetization rotates closer to the $\pi/2$ orientation, this zero energy peak region becomes more localized and pronounced in energy, extending deeper into the S layer. Finally, in Fig. 3(e), the relative magnetizations are again collinear, with the magnetization in $F_3$ antiparallel to the $z$-axis ($\theta_3 = \pi$), resulting in the disappearance of the zero energy mode and subsequent splitting into separate Andreev bound states. Note that the microscopic method used here accounts for the significant band curvature near the Fermi energy arising from the strong spin-splitting effects of the half-metallic layers, as evidenced by the particle-hole asymmetry in the DOS.50,59,60

C. Superconducting Correlations

To correlate the spin-triplet superconducting correlations to the nonmonotonic behavior of $T_c$, we plot the opposite-spin ($f_0$) and equal-spin ($f_1$) pair correlations in Fig. 6. We have averaged $f_0$ and $f_1$ over each region separately, identified by $F_1$, $F_2$, $S$, $F_3$, and $F_4$. In this fashion, one can readily evaluate the spatial distribution of the different pairings, denoted by $|f_0|$ and $|f_1|$. We consider the $F_1 F_2 S F_3 F_4$ configuration with the same parameter values used in Fig. 3(a). In Figs. 6(a) and 6(b) we set $\theta_2 = 0$, in 6(c) and 6(d) $\theta_2 = \pi/2$, and in 6(e) and 6(f) $\theta_2 = \pi$. The outer half-metals also have $\theta_1 = \theta_4 = 0$. For the top and bottom rows when $\theta_2 = 0$, $\pi$, respectively, all of the magnetic layers have collinear relative magnetizations (except of course for $F_3$, which has $\theta_3$ varying). In these configurations, we see that $f_1$ is zero at $\theta_1 = 0$ and $\theta_3 = \pi$, since then, all layers are collinear with a single quantization axis which prohibits the generation of equal-spin triplet pairs. We see that $f_0$ exhibits the same behavior as $T_c$, at $\theta_2 = 0, \pi/2, \pi$, i.e., decreases as $\theta_1$ approaches $\sim \pi/2$. The $f_1$ triplet pair-
ing however increases simultaneously, demonstrating a direct link between the appearance of equal-spin correlations and a decrease in $T_c$. In cases where $\theta_1 = 0$ and $\pi$, we see that the $f_1$ correlation has a large amplitude in the right half-metal $F_4$. It is also evident that $f_0$ is negligible in the half-metallic regions $F_1$ and $F_4$, since this opposite spin triplet channel is energetically unstable in the regions with only one spin band present. The magnetization direction in $F_2$ results in drastic changes: As can be seen in Figs. 6(c) and 6(d), $f_1$ correlations penetrate all regions throughout the multivalue and both $f_0$ and $f_1$ vanish within $0.35\pi \leq \theta_1 \leq 0.75\pi$ which is consistent with the $T_c$ results where superconductivity disappears in the middle $S$ layer.

The superconductor width plays a key role in the range of magnetization angles that result in strong spin valve effects. In the top row of Fig. 7, three larger $S$ widths corresponding to $D_S = 260, 270,$ and $D_S = 400$ are presented. Previously in Fig. 3(a) we found that for $D_S = 250$ and $\theta_2 \approx \pi/2$, there was a sizable range of angles $\theta_1$ in which the singlet correlations vanished at $T = 0$. In these regions of parameter space, the triplet correlations must also vanish as demonstrated in Fig. 6(c) and (d). If the thickness of superconductor layer increases, the pair breaking effects of the surrounding magnets become less detrimental to superconductivity, and the angular window that results in the system transitioning to a normal resistive metal narrows and eventually disappears altogether. For instance, in Fig. 7(a), although the overall trends are the same, the interval in which $f_1 = 0$ has been reduced to $0.4\pi \leq \theta_2 \leq 0.65\pi$ compared to the $D_S = 250$ case in Fig. 6(d). For $D_S = 270$ in Fig. 7(b), the triplet correlations $f_1$ now pervade every region of the spin valve. Instead of vanishing over a certain range of magnetization orientation angles, $f_1$ dips to a minimum at $\theta_2 \approx \pi/2$ in every layer, including the superconductor. Finally, if the $S$ layer is increased to $D_S = 400$ as shown in (c), the behavior of $f_1$ changes drastically throughout the system. The three layers consisting of $S$, $F_3$ and the half-metal $F_4$, which previously had dips in $f_1$ at $\theta_2 \approx \pi/2$, now have their situations reversed so that the $f_1$ triplet population is enhanced in those regions. Thus for example, for thin $S$ widths the $f_1$ correlations in the half-metal $F_4$ are constrained to vanish over a range of magnetization misalignment angles, including at $\theta_1 = \pi/2$, and as the $S$ width increases, the constraint is lifted and what was once an absence or minimum of $f_1$, now peaks for the magnetically inhomogeneous situation $\theta_2 \approx \pi/2$.

Further information can be gathered from the spatial dependence of the pair correlations. In Fig. 7(d), the singlet pair correlations are shown as a function of dimensionless position $X$ for several magnetization orientations $\theta_1$. The parameter values used here are identical to those implemented in Figs. 6(c) and (d), where $D_S = 250$ and $\theta_2 = \pi/2$. Each curve represents a different magnetization orientation described by the angle $\theta_2$ shown in the legend. As observed in Fig. 3, $T_c$ exhibits considerable variations when rotating from $\theta_1 = 0$, where all magnetizations are aligned along the $z$ direction (except in $F_2$, which is directed along $y$), to $\theta_1 = \pi/2$, where the magnetizations between the ferromagnets and half-metals are orthogonal. Indeed, the transition temperature rapidly diminishes until eventually the system transitions to the normal state for all temperatures. This is reflected in the self consistent singlet pair correlations at $T = 0$ (Fig. 7(d)), where the rapid decline in the $S$ region as a function of $\theta_1$ is clearly evident. The singlet correlations of course vanish in the half-metallic regions where only one spin state is permitted. Moreover, due to the asymmetric magnetization profile, the singlet profile is not symmetric, exhibiting a greater presence of singlet correlations in $F_3$ compared to $F_2$. Due to the singlet-triplet conversion that takes place, we see a corresponding increase in the $f_0$ triplets in $F_2$ and decrease in $F_3$ (panels (e) and (f)). Although the opposite-spin triplet and singlet correlations cannot reside in the half-metallic regions, in Fig. 7(e), we see the presence of the equal-spin triplet components $f_1$ and $f_2$ in the half-metal ($F_1$). Since these triplet pairs so not suffer from the energetically unfavorable Zeeman splitting, they can subsist in the half-metal. Although $f_1$ and $f_2$ propagate throughout the entire $F_1$ region with a slow spatial variation, these correlations are absent in the other half-metal $F_3$ which has its magnetization collinear with the adjacent ferromagnet. By rotating the magnetization in $F_3$ however, Fig. 6(b) shows that $f_{1,2}$ triplet pairs can be created in $F_4$, peaking when $\theta_3 \approx \pi/6$. This is shown in detail in Fig. 7(f) where the equal-spin triplet pairs have also been amplified in the superconductor.

### III. Conclusions

In summary, motivated by recent theoretical progress and experimental advancements in superconducting spin valves, we have proposed $F_1F_2SF_3$ and $F_1F_2SF_3F_4$ superconducting triplet spin multivalves that host multiple spin valve effects among adjacent $F$ layers. We calculated the superconducting transition temperature, and the spatially-resolved density of states vs the magnetization orientations and layer thicknesses. Our results reveal that due to proximity effects and spin-valve effects involving singlet and triplet conversion and creation, these structures offer stronger superconducting spin-switching and spin-triplet generation compared to the basic single $SF_1F_2$ and $F_1SF_2$ spin-valve counterparts. In order to provide insight into these switching effects and accurate details of the behavior of the pair correlations in both the singlet and triplet channels, we performed our calculations using a microscopic self-consistent theory that is capable of handling the broad range of length and energy scales involved. This method also allows for multiple Andreev reflections and corresponding resonances in the ballistic regime where the mean free path is much larger than the system thickness. Using this formalism, coupling between layers and quantum effects arising from interfering quasiparticle trajectories within adjacent layers is accounted for. The results shown here demonstrated that the proposed hybrid structures can provide unambiguous signatures of the presence of equal-spin triplet correlations that can arise under relatively weak in-plane external magnetic fields, thus increasing device reliability.
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Appendix A: Spin Rotation

Here we outline the spin rotations involving the triplet components \( (f_0, f_1, f_2) \), affording a clearer physical interpretation of the results. The central quantity that we use to perform the desired rotations is the spin transformation matrix \( \mathcal{T} \) in particle-hole space. The quasiparticle amplitudes transform as,

\[
\Psi'_n(x) = \mathcal{T}\Psi_n(x),
\]

where \( \Psi_n(x) = (u_n(x), u_n^*(x), v_n(x), v_n^*(x)) \), and the prime denotes quantities in the rotated system. The matrix \( \mathcal{T} \) can be written solely in terms of the angles that describe the local magnetization orientation. In particular, when the orientation of the exchange fields in a given layer is expressed in terms of the angles given in Eq. (11), we can write:

\[
\mathcal{T} = \begin{bmatrix}
\cos(\theta_i/2) & -i\sin(\theta_i/2) & 0 & 0 \\
-i\sin(\theta_i/2) & \cos(\theta_i/2) & 0 & 0 \\
0 & 0 & \cos(\theta_j/2) & -i\sin(\theta_j/2) \\
0 & 0 & -i\sin(\theta_j/2) & \cos(\theta_j/2)
\end{bmatrix}
\]

Using the spin rotation matrix \( \mathcal{T} \), it is also possible to transform the original BdG equations \( \mathcal{H}\Psi_n = e_n\Psi_n \) (Eq. (1)) by performing the unitary transformation: \( \mathcal{H}' = \mathcal{T}\mathcal{H}\mathcal{T}^{-1} \), with \( \mathcal{T}^{\dagger}\mathcal{T} = 1 \). As is the case under all unitary transformations, the eigenvalues here are preserved, but the eigenvectors are modified in general according to Eq. (A1). Thus we can write,

\[
u'_n = \cos(\theta_i/2)u_n - i\sin(\theta_i/2)v_n,
\]

\[
u'_n = \cos(\theta_i/2)u_n - i\sin(\theta_i/2)v_n,
\]

\[
u'_n = \cos(\theta_i/2)v_n - i\sin(\theta_i/2)u_n,
\]

\[
u'_n = \cos(\theta_i/2)v_n - i\sin(\theta_i/2)u_n.
\]

Thus for example, the terms involved in calculating the singlet pair correlations (Eq. (3)), obey the following relation between the transformed (primed) and untransformed quantities:

\[
u'_n = \nu_n + \nu_n^* = \nu_n + \nu_n^*.
\]

Thus, the terms that dictate the singlet pairing are invariant for any choice of quantization axis, transforming as scalars under spin rotations.

The terms governing the triplet amplitudes on the other hand are not invariant under spin-rotation. The relevant particle-hole products in Eq. (9a) that determine \( f_0 \), upon the spin transformations obey the following relationships:

\[
u'_n = \nu_n + \nu_n^* = \nu_n + \nu_n^*.
\]

For \( f_2 \) however, it is straightforward to show that

\[
u'_n = \nu_n + \nu_n^* = \cos\theta f_2 + \sin\theta f_0.
\]

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