Meissner effect probing of odd-frequency triplet pairing in superconducting spin valves

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Superconducting correlations which are long ranged in magnetic systems have attracted much attention due to their spin-polarization properties and potential use in spintronic devices. Whereas experiments have demonstrated the slow decay of such correlations, it has proven more difficult to obtain a smoking gun signature of their odd-frequency character which is responsible, e.g., for their gapless behavior. Here we demonstrate that the magnetic susceptibility response of a normal metal in contact with a superconducting spin valve provides precisely this signature, namely, in the form of an anomalous *positive* Meissner effect, which may be tuned back to a conventional *negative* Meissner response simply by altering the magnetization configuration of the spin valve.

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I. INTRODUCTION

Superconducting spin valves with double ferromagnet layers [1] (i.e., F_1/F_2) have very recently attracted interest due to the appearance of long-range spin and triplet supercurrents when the two F layers have misaligned magnetizations [2–4]. These structures are of considerable interest, both experimentally for being feasible to fabricate, and also because such long-range Josephson currents were thought to occur only in systems with more complicated magnetic inhomogeneity [5–12]. Most experimental efforts aimed at revealing the long-range triplet correlations have measured the supercurrent [13–15], which conveyed the long-range nature of the triplet correlations but not their intrinsic odd-frequency nature. We consider here a $S/F_1/F_2/N$ spin-valve configuration, where S represents the superconductor and N a normal nonmagnetic metal. For this type of spin valve, we show that it is possible to utilize the Meissner response to unambiguously determine the existence of long-ranged triplet correlations *and* their odd-frequency symmetry. This originates from proximity effects [16], in which Cooper pairs in S populate the adjacent normal metal or ferromagnet regions [17]. These induced superconducting correlations respond to an applied magnetic field H_a by setting up Meissner screening currents, which lead to observable changes in the magnetic susceptibility χ.

The Meissner response was previously investigated [18–20] as a way to elucidate proximity effects in hybrid nonmagnetic S/N systems. More recently these types of structures revealed peculiar proximity-induced reentrance effects in χ with changing H_a or temperature T [21–23]. These effects were attributed to an enhancement of the paramagnetic contribution to the susceptibility [24–26], which in the diffusive regime can depend on the existence of electron-electron interactions in the N layer [27]. Meissner effects in monodomain ferromagnets in contact with superconductors have also been considered [28,29]. If there are spin-dependent interactions, including those arising from sequences of F layers with misaligned magnetizations, a richer variety of proximity effects can emerge, including the well-known induced equal-spin odd-frequency triplet correlations with long-range penetration into a ferromagnet. Spin-dependent scattering at an S/N interface can also generate odd-frequency correlations that modify the Meissner response, turning it paramagnetic [30], resulting in χ oscillations as T is varied. A vanishing Meissner response may be indicative of the onset of the familiar damped oscillations that a Cooper pair undergoes when entering a ferromagnet [31]. It is therefore of importance to understand not only how the odd triplet pairing extends throughout the system, as described by the anomalous Green's function, but also to know their symmetry. There have been considerable efforts to find ways to experimentally measure the symmetries or long-range nature of triplets, including surface impedance measurements [32] and local signatures in the density of states (DOS) [33–39].

Recent experiments [40] involving double-magnet superconducting spin valves have shown that by varying the relative in-plane magnetization angle θ, the critical temperature T_c can be lowest when the magnetizations are nearly orthogonal, reflecting the increased presence of equal-spin triplet pairs [41], in agreement also with theoretical works [42–45]. The long-range triplet pair correlations always vanish when the magnetizations are collinear but can, due to phase shifts at the F_1/F_2 interface, oscillate and vanish at an intermediate θ [43]. By considering an $S/F_1/F_2/N$ spin valve, manipulating θ controls the long-range odd-frequency triplets that propagate in the N region. Thus when the long-range triplet correlations are absent or very weak, we show that the magnetic susceptibility is negative, corresponding to the conventional Meissner response. However, when there is a strong misalignment of the mutual magnetizations and the long-ranged triplets are enhanced, we find that the magnetic susceptibility is positive, corresponding to an anomalous Meissner response. Therefore by measuring χ, it is possible to determine from its magnitude the presence of the long-ranged correlations, while its sign indicates their odd-frequency character. A main advantage of this prediction compared to previous works is that the odd-frequency symmetry is revealed by an overall sign change, which is easily experimentally observable, in contrast to more

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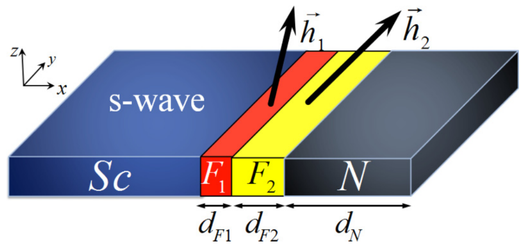

FIG. 1. (Color online) Schematic of the proposed metallic $S/F_1/F_2/N$ junction. The x axis is perpendicular to the interfaces separating the different layers. Two ferromagnetic layers (F_1, F_2) with thicknesses d_{F1} and d_{F2} are sandwiched between a superconductor (S) and normal metal (N) with thickness d_N. The exchange field in the ferromagnetic layers, $\vec{h}_1$ and $\vec{h}_2$, may be misaligned as shown in the figure. An external magnetic field $\vec{H}_a$ (not shown) is applied in the z direction, resulting in a spatially dependent Meissner supercurrent $\vec{J}(x)$ flowing in the y direction.

subtle signatures such as scaling behavior and a combination of multiple spectroscopic fingerprints [36,37].

II. THEORY AND FORMALISM

To model a realistic experimental system, we will consider the diffusive limit for a metallic $S/F_1/F_2/N$ junction as shown in Fig. 1. The superconducting proximity effect in such a setup may be described by using a Green's function approach, where the superconducting correlations are quantified via the so-called anomalous Green's function $\hat{f}$. In the presence of a magnetic exchange field, it is necessary to consider carefully the spin structure of the Green's function, which in general will consist of a spin-singlet and spin-triplet part. In order to compute the Meissner response of the normal metal, we first need to obtain the anomalous Green's function in the normal part of the junction. This is done by solving the quasi-classical Usadel equation with proper boundary conditions at the interface region of the superconductor ($x = 0$) and vacuum ($x = d_{F1} + d_{F2} + d_N$). In typical experiments, the interface transparency is rather low (tunneling limit) such that the linearization of the Usadel equation is a very good approximation, corresponding to an anomalous Green's function which satisfies $|\hat{f}| \ll 1$. We decompose the anomalous Green's function into singlet even- ($\mathbb{S}$) and triplet odd-frequency ($\vec{\mathbb{T}}$) components as [45]

$$\hat{f}(\varepsilon) = i[\mathbb{S}(\varepsilon) + \vec{\mathbb{T}}(\varepsilon).\vec{\tau}]\tau_y. \tag{1}$$

Above, $\vec{\tau}$ is a vector composed of Pauli matrices. The linearized Usadel equations then have the following general form (with $\sigma = \pm 1$):

$$\begin{aligned}
&-\sigma\partial_x^2\mathbb{T}_{x-} + i\partial_x^2\mathbb{T}_{y-} + 2i[-\varepsilon(-\sigma\mathbb{T}_{x-} + i\mathbb{T}_{y-})\\
&\quad -\sigma\mathbb{S}_-(h_x - \sigma i h_y)] = 0,\\
&-\sigma\partial_x^2\mathbb{S}_- + \partial_x^2\mathbb{T}_{z-} + 2i[-\sigma h_x\mathbb{T}_{x-} - \sigma h_y\mathbb{T}_{y-}\\
&\quad -(-\sigma\mathbb{S}_- + \mathbb{T}_{z-})(\varepsilon + \sigma h_z)] = 0.
\end{aligned} \tag{2}$$

Here, D is the diffusion constant, $\vec{h} = (h_x, h_y, h_z)$ is the magnetic exchange field, which is nonzero inside the ferromagnetic layers and absent in the normal metal layer. The above equations are to be supplemented with the Kupriyanov-Lukichev boundary conditions at the S/F_1 interface and are valid in the tunneling limit [46]. The transparency of the interface to quasiparticle tunneling is determined by the parameter ζ, which depends on the the ratio between the resistance of the interface and the resistance in the diffusive normal region. We obtain the following boundary conditions at the S/F_1 interface:

$$(\zeta\partial_x - c^*(\varepsilon))(-\sigma\mathbb{T}_{x-} + i\mathbb{T}_{y-}) = 0, \tag{3}$$

$$(\zeta\partial_x - c^*(\varepsilon))(-\sigma\mathbb{S}_- + \mathbb{T}_{z-}) + \sigma s^*(\varepsilon) = 0. \tag{4}$$

Here $\mathbb{T}_{x,y,z\mp} \equiv \mathbb{T}_{x,y,z}(\mp\varepsilon)$, whereas $s(\varepsilon)$, $c(\varepsilon)$ are the off-diagonal and normal components of the bulk Green's function for the superconducting region. The general expression for the supercurrent density in the system reads $\vec{J}(\vec{r}) = \vec{J}_0 \int d\varepsilon \mathrm{Tr}\{\rho_3(\hat{G}[\hat{\partial},\hat{G}])^K\}$, in which K represents the Keldysh component of the matrix. Here J_0 is a normalization constant. When the normal part of the system is subject to an external magnetic field H_a oriented in the z direction (see Fig. 1), the supercurrent flowing parallel to the interfaces (along y) can be expressed in terms of the short-range spin-0 correlations and the long-range spin-1 triplet correlations as

$$\begin{aligned}
J(x) = -J_0 8ieA(x)\sum_\sigma \int_0^\infty d\varepsilon[\sigma\mathbb{S}(\sigma\varepsilon,x)\mathbb{S}^*(-\sigma\varepsilon,x)\\
-\sum_{j=\{x,y,z\}} \sigma\mathbb{T}_j(\sigma\varepsilon,x)\mathbb{T}_j^*(-\sigma\varepsilon,x)]\tanh(\varepsilon\beta/2).
\end{aligned} \tag{5}$$

Since the magnetic field $\vec{B}$ and the vector potential $\vec{A}$ are related by $\vec{\nabla} \times \vec{A} = \vec{B}$, Maxwell's equation must be taken into account together with the above equation for the supercurrent. If we use the Coulomb gauge, $\vec{\nabla} \cdot \vec{A} = 0$, Maxwell's equation reduces to

$$d^2A(x)/dx^2 = -4\pi J(x)/c. \tag{6}$$

Similar to previous works considering the proximity-induced Meissner effect [30], we assume that the external magnetic field is expelled entirely from the superconductor and thus $A(0) = 0$. The normal metal is taken to be sufficiently wide so that the external magnetic field fully penetrates the rightmost end of it. The system susceptibility is expressed by

$$\chi = \frac{1}{H_a d_N}\int M(x)dx = \frac{1}{4\pi H_a d_N}\int (B - H_a)dx, \tag{7}$$

where $\vec{B} = \vec{H}_a + 4\pi\vec{M}$, and $\vec{M}$ is the magnetization.

III. RESULTS AND DISCUSSION

We have solved numerically the Usadel equations with their boundary conditions, computed the supercurrent density, and used the latter to solve Maxwell's equation in order to obtain the magnetic susceptibility. This corresponds to a linear-response theory, since $J(x) \propto A(x)$, which nevertheless compares very well to experiments [20]. To enhance the level of induced triplet correlations in our system, the thickness of one magnet should be much larger than the other one [47]: the thin F layer generates the triplet components, whereas the thick F layer filters out the short-range components. We thus set $d_{F1} = 0.15\xi_S$, $d_{F2} = 1.95\xi_S$, and $d_N = 2.5\xi_S$, where

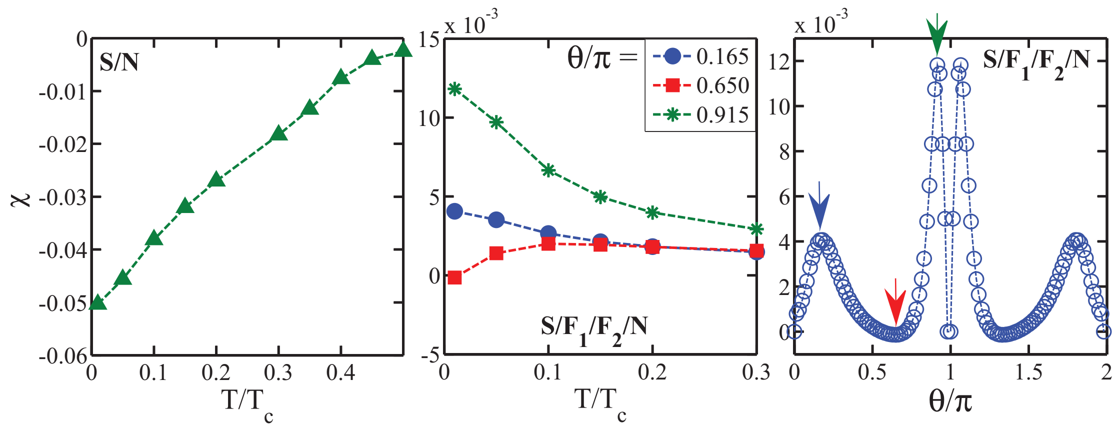

FIG. 2. (Color online) Meissner effect in S/N (left panel) and $S/F_1/F_2/N$ (middle and right panels) structures as a function of temperature T and magnetic misalignment angle θ. In the S/N structure the singlet superconducting correlations give rise to a conventional Meissner effect ($\chi < 0$), while in the $S/F_1/F_2/N$ structure an anomalous Meissner effect ($\chi > 0$) appears which may be controlled via the relative magnetization directions in the double F layers.

ξ_S is the superconducting coherence length, which is typically of the order of tens of nanometers in diffusive metals. For simplicity, the exchange field is taken to reside solely in the y-z plane, so that the angle θ describes the in-plane misalignment between the two magnets. To ensure that the linearized Usadel equations are valid, we consider the tunneling limit and set $\zeta = 6$, whereas the exchange field is taken to be that of a weak ferromagnetic alloy, $|\vec{h}_1| = |\vec{h}_2| = 10\Delta_0$. All lengths in the system are normalized by the superconducting coherence length ξ_S and correspondingly, all energies by the superconducting gap at zero temperature Δ_0. Throughout the calculations we set that $\hbar = k_B = 1$, and unless otherwise noted, $T = 0.05T_c$.

To make contact with previous works considering nonmagnetic S/N bilayers, we plot in the left panel of Fig. 2 the proximity-induced susceptibility χ integrated over the entire N region with $|\vec{h}_1| = |\vec{h}_2| = 0$. As seen, the temperature dependence of χ displays a diamagnetic behavior, as expected for the conventional Meissner effect, and vanishes as $T \to T_c$ [20]. We now investigate what happens when the spin valve is present and denote the misalignment angle between the F layers as θ. The middle panel of Fig. 2 gives the temperature dependence of χ for an $S/F_1/F_2/N$ junction when considering different values of θ. It is clear that the susceptibility changes sign compared to the nonmagnetic case, indicating a paramagnetic Meissner effect. As we shall demonstrate below, this anomalous magnetic response is entirely due to the odd-frequency nature of the triplet correlations.

We have also computed how χ changes as a function of the misalignment angle θ as it makes a full cycle, starting from a parallel magnetization configuration (P, $\theta = 0$) to antiparallel (AP, $\theta = \pi$) and then back to parallel ($\theta = 2\pi$). This is shown in the right panel of Fig. 2, which displays several noteworthy features. First, it is seen that χ practically vanishes at $\theta = 0$ and $\theta = \pi$. This is consistent with the fact that in these cases a single quantization axis can be defined for the whole system and hence there are no long-ranged odd-frequency triplets for these configurations, reflecting the absence of proximity effects. Second, two peaks develop as soon as one moves slightly away from the P and AP configuration, whereas χ has a minimum and nearly vanishes for an intermediate value of θ near $\pi/2$. This might seem odd at first glance. Indeed, one might expect the triplet correlations to be the largest in magnitude when the misalignment angle deviates the most from the P and AP configuration, namely, at $\pi/2$.

Before resolving this issue, we demonstrate that the anomalous Meissner effect shown in Fig. 2 is due to the odd-frequency correlations induced in the normal metal. By directly computing the spin-0 singlet and $S_z = 0$ triplet contribution $\mathbb{T}_z$ to the current in Eq. (5), we find that these are several orders of magnitude smaller (and nearly zero) compared to the contribution from $\mathbb{T}_y$. The latter is exactly the long-ranged triplet component, which must have an odd-frequency symmetry since our system is diffusive (only s-wave symmetry survives). Now, one may show analytically [30] that in the scenario where purely odd-frequency correlations are present, the Meissner response will be paramagnetic such that $\chi > 0$. Therefore the behavior of χ in our system provides a clear signature, not only for the long-ranged proximity effect, but *simultaneously* for its odd-frequency character. Another advantage with the proposed setup is that one may explicitly tune the Meissner response by changing the misalignment angle θ, since the latter turns on and off the long-ranged odd-frequency correlations. We note that the magnitude of the proximity-induced susceptibility χ ($\sim$0.01) is due to the assumption of a weak proximity effect and is well within experimental reach [20,48].

Figure 3 illustrates the crossover between a fully and partially anomalous Meissner effect, $\chi > 0$, as a function of magnetization misalignment θ for several choices of the thickness of $F2$. The thickness of $F1$ is kept fixed at $d_{F1} = 0.15\xi_S$, whereas the thickness of second ferromagnetic layer varies from $d_{F2} = 0.85\xi_S$ to $d_{F2} = 1.95$, where $d_{F1} \ll d_{F2}$. As seen, the anomalous Meissner effect is most pronounced ($\chi > 0$ for all θ) when $d_{F1} \ll d_{F2}$. When d_{F2} is reduced, the contribution to the Meissner effect from the short-ranged correlations, including the conventional even-frequency ones that yield a standard Meissner effect, is no longer negligible and thus χ becomes negative for an increased regime of misalignment angles θ.

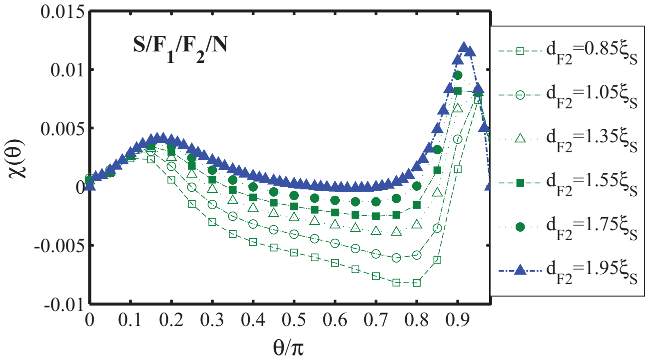

FIG. 3. (Color online) Meissner effect (χ) in $S/F_1/F_2/N$ connections against the noncollinearity angle θ for various values of $F2$ layer thicknesses: $d_{F2}/\xi_S = 0.85,1.05,1.35,1.55,1.75,1.95$. The thickness of $F1$ and normal metal layers are fixed at $d_{F1} = 0.15\xi_S$ and $d_N = 2.5\xi_S$, respectively. The anomalous Meissner effect ($\chi > 0$) also depends on the magnetic layer thicknesses and is strongest where $d_{F2} \gg d_{F1}$, for instance.

Having established this, we now turn to the peculiar behavior near $\pi/2$ for the susceptibility. The key thing to note here is that contrary to what one might expect initially, the generation of triplet correlations is *not* necessarily the strongest for a maximum misalignment angle near $\pi/2$. In fact, the proximity-induced equal-spin triplets can actually vanish all together for θ near $\pi/2$. This was recently shown in Ref. [43] and explained in terms of phase slips of the singlet and triplet correlation functions due to the finite S/F_1 interface transparency, causing them to vanish at a critical misalignment angle θ_c, which in the tunneling limit and for $h \gg \Delta_0$ was close to $\pi/2$. To investigate if a similar situation is present in our setup, we plot in Fig. 4 the imaginary and real part of the long-ranged spin-triplet Green's function $\mathbb{T}_y(\varepsilon,x)$. We consider quasiparticle energies close to the Fermi level, $\varepsilon/\Delta_0 = 0.1$. This is a representative choice, since the main contribution to the current in Eq. (5) comes precisely from low-energy excitations. Figure 4 shows how the anomalous Green's function depends on the misalignment angle for several positions in the N metal: $x/\xi_S = \{2.15,3.00,3.50,4.50\}$. As seen, the anomalous Green's function displays qualitatively a similar dependence on θ as the susceptibility, with peaks occurring close to 0 and π and a local minimum near $\pi/2$. This is then consistent with the phase-slip phenomenon in Ref. [43] and explains the angular dependence of χ. The misalignment angle θ then controls the long-ranged proximity effect in the N part, which in turn alters the magnitude of the Meissner response, the *sign* of which is nevertheless always positive due to the odd-frequency character of the correlations.

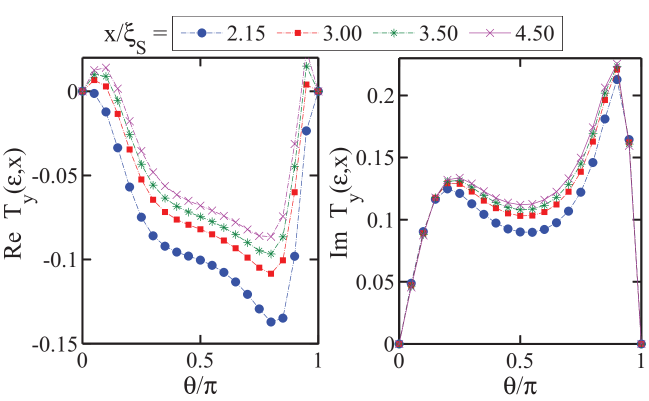

FIG. 4. (Color online) Real and imaginary parts of the y component of the decomposed spin Green's function ($\mathbb{T}_y(\varepsilon,x)$) versus magnetization misalignment angle θ at several locations inside the N region of the $S/F_1/F_2/N$ configuration considered here. Since energies close to the Fermi level comprise the main contribution to the proximity effect, we have set $\varepsilon = 0.1\Delta_0$ as a representative choice.

What is the physical origin of the paramagnetic Meissner effect encountered here which serves as a signature for long-ranged odd-frequency pairing? To explain this, it is useful to recall that a nonlinear Meissner effect also is known to occur in d-wave superconductors [49]. In that case, the temperature dependence of the magnetic penetration depth has been experimentally observed to be nonmonotonic, with a minimum at an intermediate temperature between $T = 0$ and $T = T_c$. This is in contrast to what one sees for a conventional Meissner effect, which simply reduces the penetration depth as the temperature decreases, consistent with the left panel of our Fig. 2. The explanation behind this phenomenon for d-wave superconductors is the existence of low-energy Andreev bound states which have a paramagnetic contribution to the Meissner effect, thus competing with the shielding supercurrent and producing the nonlinear behavior [50]. The key observation is now that Andreev-bound states and odd-frequency pairing are intimately related, as shown in Ref. [51]. In fact, the appearance of zero-energy bound states in unconventional superconductors may be reinterpreted as a manifestation of odd-frequency pairing correlations, such that the latter should also contribute paramagnetically to the Meissner effect. We nevertheless emphasize that in our case, the anomalous Meissner effect is produced from garden-variety s-wave superconductivity combined with a spin-valve structure, i.e., without the need of any unconventional superconducting materials.

One experimental challenge regarding measurements of the predicted paramagnetic Meissner effect occurring in superconducting spin valves comes from the influence of stray fields from the ferromagnetic regions. However, we note that the positive χ appears with a similar dependence on the misalignment angle θ even if one considers the susceptibility as obtained from integration over the entire nonsuperconducting region, i.e., $F_1/F_2/N$. The response of the bulk superconducting region is diamagnetic and could be expected to dominate the total susceptibility if one were to measure the susceptibility response of the entire heterostructure. In order to prove the combined odd-frequency and long-ranged nature of the superconducting proximity effect, it would therefore be necessary to conduct a local measurement of the susceptibility. As mentioned earlier, a measurement of the susceptibility response of the normal part would provide the most clear signature of the anomalous Meissner effect, although it would also be seen even if one were to include the ferromagnetic parts in the measurement. Major experimental advances in achieving such local probing of spin susceptibility has recently been reported, demonstrating detection capability at the single-spin level on nano- and atomic scales by using so-called nitrogen-vacancy magnetometers [52]. The observation of a paramagnetic Meissner effect in the spin-valve setup of Fig. 1 would provide clear evidence of not only the long-ranged nature of the triplet correlations, but importantly, their odd-frequency characteristic, the latter not being observable in supercurrent measurements done so far in half-metals and strong ferromagnets [53].

IV. CONCLUSIONS

In conclusion, we have proposed a spin valve made of two layers of uniform ferromagnetic layers with unequal thicknesses attached to a superconducting lead from one side and a normal metal layer from other side. Our results demonstrate an anomalous positive Meissner effect which can be experimentally probed in the connected normal metal layer. We have shown that the anomalous Meissner effect appears due to the dominance of proximity odd-frequency triplet superconducting correlations. The theoretical proposed structure here may open new and feasible venues in experiment to study and investigate the proximity triplet superconducting correlations.

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