Spin Splitting Induced in a Superconductor by an Antiferromagnetic Insulator

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Inspired by recent feats in exchange coupling antiferromagnets to an adjacent material, we demonstrate the possibility of employing them for inducing spin splitting in a superconductor, thereby avoiding the detrimental, parasitic effects of ferromagnets employed to this end. We derive the Gor'kov equation for the matrix Green's function in the superconducting layer, considering a microscopic model for its disordered interface with a two-sublattice magnetic insulator. We find that an antiferromagnetic insulator with effectively uncompensated interface induces a large, disorder-resistant spin splitting in the adjacent superconductor. In addition, we find contributions to the self-energy stemming from the interfacial disorder. Within our model, these mimic impurity and spin-flip scattering, while another breaks the symmetries in particle-hole and spin spaces. The latter contribution, however, drops out in the quasiclassical approximation and thus, does not significantly affect the superconducting state.

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Introduction.—Conventional Bardeen-Cooper-Schrieffer (BCS) superconductors [1] are incompatible with magnetic interactions as the latter tend to break the Cooper pairing [2] between the opposite-spin electrons. Nevertheless, the so-called Pauli contribution, associated with energy splitting of the two spin states, leads to interesting new phenomena when the spin-splitting is comparable to the "unperturbed" superconducting gap [3]. These include spatially inhomogeneous order parameter in an otherwise homogeneous superconductor [4,5], gapless superconductivity [6,7], and a first-order phase transition between superconducting and normal states [8,9], all of which have been experimentally observed [10,11]. Furthermore, hybrids incorporating such spin-split superconductors were recently predicted [12–14], and found [15,16], to exhibit large thermoelectric effects. The spin splitting in the superconducting layer may be induced by a magnetic field or via exchange coupling to a magnetic layer [16,17] and leads to intriguing transport properties reviewed in Ref. [18].

The success of "exchange biasing" a ferromagnet (FM) layer via its coupling to an adjacent antiferromagnet (AFM) has been instrumental in the contemporary memory technology [19–21]. A simplified picture of exchange biasing in FM-AFM bilayers requires the AFM interface to be uncompensated, i.e., possess finite surface magnetization [19,21,22]. Several theoretical models [22], most of which assume the AFM surface to be uncompensated, have been employed to understand the experiments. Recent progress in surface characterization methods [23] and epitaxial sample growth [24] has enabled to resolve [24,25] several previously open questions [19]. Numerous experiments [26–34] have succeeded in direct observation and quantification of uncompensated spins at interfaces thereby improving the understanding of their role in exchange bias and the control of the effect.

Recently, the presence of surface magnetization, stemming from broken translational symmetry at interfaces, in magnetoelectric AFMs has been predicted [35]. This has also been observed experimentally and exploited in achieving electrically switchable exchange bias [36] and magnetic memory [37] using α-Cr_2O_3. Furthermore, uncompensated AFM interfaces have been theoretically predicted to amplify transfer of magnonic spin from a magnetic insulator to an adjacent non-magnetic conductor [38,39].

In this Letter, we suggest employing insulating AFMs, with their uncompensated surfaces, to induce an effective exchange field in an adjacent superconducting layer. To the best of our knowledge, only FMs have been employed to this end so far. AFMs offer several advantages over FMs in this regard [40–42]. These include minimization of stray magnetic fields, the possibility of electrical tunability [36,37,43], avoiding parasitic negative effects of low-energy magnon excitations [44,45] and so on. The proximity effect due to metallic antiferromagnets has been investigated experimentally [46] and theoretically [47]. Antiferromagnetically ordered impurity chains may also give rise to the Majorana state [48].

Considering a two-sublattice magnetic insulator (MI)-superconductor (S) bilayer structure, we derive the Gor'kov equation for the matrix Green's function in S from a microscopic Hamiltonian including the interface [49]. Our model for MI encompasses the full range of single-domain magnets from ferro- to antiferro- via ferrimagnets

[38,45]. We explicitly include interfacial disorder in our model and find that the induced exchange field is resistant to it, within the Born approximation. We find that the effect of the MI layer is captured by a self-energy which includes interfacial disorder-mediated terms, in addition to the spin-splitting term. The latter is found to be large for an uncompensated interface with an AFM. For the system considered here, with the Hamiltonian diagonal in spin space [50], the interfacial disorder-mediated terms take a form identical to spin-independent impurity and spin-flip scattering. A third disorder contribution breaks the particle-hole and spin symmetries, but predominantly renormalizes the normal state properties leaving the superconducting state essentially unaffected.

Model and Hamiltonian.—We consider a MI-S bilayer (Fig. 1) with the S thickness d_S much smaller than the superconducting coherence length. MI is comprised by a single-domain two-sublattice magnetic insulator where sublattice magnetizations are considered static and collinear to the z axis. We consider S to be a BCS superconductor in the weak coupling regime such that the Hamiltonian in the grand canonical ensemble reads [49]:

$$\tilde{\mathcal{H}} = \int d^3r \left(\sum_{\alpha} \tilde{\psi}^{\dagger}_{\alpha}(\boldsymbol{r})[-\partial^2 + V_s(\boldsymbol{r})(\delta_{\alpha\uparrow} - \delta_{\alpha\downarrow})]\tilde{\psi}_{\alpha}(\boldsymbol{r}) + \sum_{\alpha,\beta} \frac{g}{2} \tilde{\psi}^{\dagger}_{\beta}(\boldsymbol{r})\tilde{\psi}^{\dagger}_{\alpha}(\boldsymbol{r})\tilde{\psi}_{\alpha}(\boldsymbol{r})\tilde{\psi}_{\beta}(\boldsymbol{r}) \right). \tag{1}$$

Here, $\tilde{\psi}_{\alpha}(\boldsymbol{r})$ is the electron annihilation operator for z-projected spin α at position $\boldsymbol{r}$, $\partial^2 \equiv \nabla^2/2m + \mu - V_i(\boldsymbol{r})$, μ is the chemical potential, m is electron effective mass, $V_i(\boldsymbol{r})[V_s(\boldsymbol{r})]$ represents the spin-independent (dependent) potential energy, $g(< 0)$ parametrizes the electron-electron attraction, and we have set $\hbar$ to 1. All operators are in the Heisenberg picture and are decorated by a ˜ above.

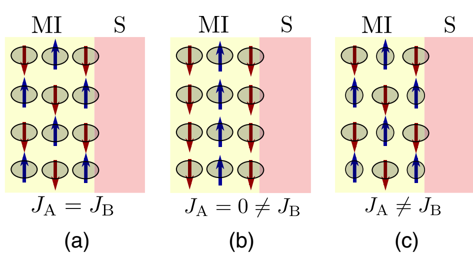

FIG. 1. Possible interface microstructures for MI-S bilayers. Sublattices A and B are depicted in blue and red, respectively. Cases (a) and (b), respectively, represent antiferromagnets with compensated and fully uncompensated interfaces with S. Case (c) depicts a ferrimagnet with a compensated interface. In this case, the symmetry of interfacial coupling between S and the two sublattices is broken [51,52] by, for example, different wave function clouds associated with the localized moments that comprise the sublattice. Interfacial disorder, accounted for in our model, is not depicted explicitly here.

The interface with MI results in the potential energy terms $V_{i,s}(\boldsymbol{r})$. For simplicity, we do not explicitly include bulk contributions to the potential energy here.

The MI-conductor interface is typically modeled as an effective exchange interaction between the spin densities on the two sides [38]:

$$\tilde{\mathcal{H}}_{\text{int}} = -\int d^2s \sum_{\mathcal{G}=\text{A},B} [J_{\mathcal{G}} \tilde{\boldsymbol{S}}_{\mathcal{G}}(\boldsymbol{s}) \cdot \tilde{\boldsymbol{S}}(\boldsymbol{s})]. \tag{2}$$

Here, $\boldsymbol{s}$ is the two-dimensional position vector in the interfacial plane defined by $y = 0$, $\tilde{\boldsymbol{S}}$ is the electronic spin density operator in S, and $\tilde{\boldsymbol{S}}_{A(B)}$ is MI sublattice $A(B)$ spin density operator. $J_{A(B)}$ parametrizes the exchange strength between the MI sublattice $A(B)$ and the S electrons, and depends upon the details of the interface such as its microstructure (Fig. 1). The magnetic spin densities are related to the corresponding magnetizations via the sublattice gyromagnetic ratios $\gamma_{A,B}$, assumed negative, $\tilde{\boldsymbol{M}}_{A,B} = -|\gamma_{A,B}|\tilde{\boldsymbol{S}}_{A,B}$. We consider sublattice $A(B)$ to be saturated along positive (negative) z direction with saturation magnetization $M_{A0(B0)}$.

Augmenting the interfacial interaction above [Eq. (2)] with a spin-independent contribution and disorder, the net interfacial Hamiltonian may be expressed as

$$\tilde{\mathcal{H}}_{\text{int}} = \int d^3r \sum_{\alpha} \tilde{\psi}^{\dagger}_{\alpha}(\boldsymbol{r}) U(\boldsymbol{s}) \delta(y)[a + b(\delta_{\alpha\uparrow} - \delta_{\alpha\downarrow})]\tilde{\psi}_{\alpha}(\boldsymbol{r}), \tag{3}$$

where a parametrizes the spin-independent contribution of the interfacial interaction and $b = J_A M_{A0}/2|\gamma_A| - J_B M_{B0}/2|\gamma_B|$. $U(\boldsymbol{s})$ accounts for the interfacial disorder which is modeled in a manner analogous to the treatment of impurities-mediated disorder in a bulk conductor [49,53]:

$$U(\boldsymbol{s}) = 1 + \sum_{\boldsymbol{s}_i} u(\boldsymbol{s} - \boldsymbol{s}_i), \tag{4}$$

with $u(\boldsymbol{s} - \boldsymbol{s}_i)$ representing the fluctuation in potential energy associated with a "disorder center" located at $\boldsymbol{s}_i$, and we assume $\int d^2s u(\boldsymbol{s}) = 0$. Employing Eq. (3), the potential energy contribution to the total Hamiltonian [Eq. (1)] corresponds to $V_{i[s]}(\boldsymbol{r}) = U(\boldsymbol{s})\delta(y)a[b]$.

Gor'kov equation.—We now formulate the problem at hand in terms of imaginary-time Green's functions in Nambu-spin space. Decorating four-dimensional entities (vectors and matrices) by a ˇ and two-dimensional by a ˆ above, we define $\check{\Psi}^{\dagger} \equiv [\tilde{\psi}^{\dagger}_{\uparrow}, \tilde{\psi}^{\dagger}_{\downarrow}, \tilde{\psi}_{\downarrow}, \tilde{\psi}_{\uparrow}]$. We further define the matrix, imaginary-time Green's function as [49]

$$\check{G}(x_1,x_2) \equiv -\hat{\tau}_z \otimes \hat{\sigma}_0 \langle \mathrm{T}_\tau \check{\Psi}(x_1)\check{\Psi}^\dagger(x_2)\rangle,$$
$$= \begin{bmatrix} G_{\uparrow\uparrow} & G_{\uparrow\downarrow} & F_{\uparrow\downarrow} & F_{\uparrow\uparrow} \\ G_{\downarrow\uparrow} & G_{\downarrow\downarrow} & F_{\downarrow\downarrow} & F_{\downarrow\uparrow} \\ -\bar{F}_{\downarrow\uparrow} & -\bar{F}_{\downarrow\downarrow} & \bar{G}_{\downarrow\downarrow} & \bar{G}_{\downarrow\uparrow} \\ -\bar{F}_{\uparrow\uparrow} & -\bar{F}_{\uparrow\downarrow} & \bar{G}_{\uparrow\downarrow} & \bar{G}_{\uparrow\uparrow} \end{bmatrix}, \quad (5)$$

where $\tau = it$ is the imaginary time, $x_1 \equiv (\boldsymbol{r}_1, \tau_1)$, $\hat{\tau}_{0,x,y,z}$ and $\hat{\sigma}_{0,x,y,z}$ are the identity and Pauli matrices in, respectively, the Nambu and spin spaces, and the outer product is expanded as

$$\hat{\tau}_z \otimes \hat{\sigma}_0 = \begin{bmatrix} \hat{\sigma}_0 & 0 \\ 0 & -\hat{\sigma}_0 \end{bmatrix}.$$

Employing the Heisenberg equation of motion for $\tilde{\psi}_\alpha(x_1)$ with the Hamiltonian given by Eq. (1), we obtain the dynamical equation for $G_{\alpha\beta}(x_1,x_2)$:

$$\frac{\partial G_{\alpha\beta}(x_1,x_2)}{\partial \tau_1} = -\delta_{\alpha\beta}\delta(x_1-x_2) + [\partial_1^2 - V_s(\boldsymbol{r}_1)(\delta_{\alpha\uparrow}-\delta_{\alpha\downarrow})] \times G_{\alpha\beta}(x_1,x_2) - i\sum_\gamma \Delta_{\alpha\gamma}(x_1)\bar{F}_{\gamma\beta}(x_1,x_2), \quad (6)$$

where $\Delta_{\alpha\beta}(x) \equiv i|g|F_{\alpha\beta}(x,x)$. In simplifying the four-point correlator above, we have employed Wick's theorem [54] and disregarded terms that lead to a mere renormalization of the chemical potential [49]. Dynamical equations for the other components of the matrix Green's function can be derived in an analogous manner [49]. All these equations may be expressed as a single Gor'kov equation for the matrix Green's function:

$$\check{\mathcal{G}}^{-1}(x_1)\check{G}(x_1,x_2) = \delta(x_1-x_2)\hat{\tau}_0 \otimes \hat{\sigma}_0, \quad (7)$$

where

$$\check{\mathcal{G}}^{-1}(x_1) = -\frac{\partial}{\partial \tau_1}\hat{\tau}_z \otimes \hat{\sigma}_0 + \partial_1^2 \hat{\tau}_0 \otimes \hat{\sigma}_0 - V_s(\boldsymbol{r}_1)\hat{\tau}_z \otimes \hat{\sigma}_z - \check{\Delta}(\boldsymbol{r}_1). \quad (8)$$

For a homogeneous superconducting state, the pair potential matrix may be chosen as $\check{\Delta}(\boldsymbol{r}) = -i\Delta\hat{\tau}_y \otimes \hat{\sigma}_z$ with real Δ [49,55].

Interfacial self energy.—Since the Gor'kov equation can rarely be solved exactly, we resort to perturbation theory within the Green's function method [53] and obtain the self-energy arising from the interfacial contribution to the Hamiltonian [Eq. (3)]. To this end, we express $\check{\mathcal{G}}^{-1}(x_1) = \check{\mathcal{G}}_0^{-1}(x_1) - \check{\mathcal{H}}_{\mathrm{int}}(x_1)$ as the sum of the clean superconducting layer plus the interfacial contribution, which assumes the form [using Eqs. (3) and (8)]:

$$\check{\mathcal{H}}_{\mathrm{int}}(x_1) = U(\boldsymbol{s}_1)\delta(y_1)[a\hat{\tau}_0 \otimes \hat{\sigma}_0 + b\hat{\tau}_z \otimes \hat{\sigma}_z] \equiv U(\boldsymbol{s}_1)\delta(y_1)\check{t}. \quad (9)$$

The evaluation of the corresponding self-energy follows the method analogous to the case of impurities-mediated disorder in a bulk conductor [49,53] and is detailed in the Supplemental Material [56]. Within this method, the so-called cross-diagram technique [49,53], the following assumptions are made. (i) The perturbation is assumed small thus making the Born approximation. (ii) We average over the positions $\boldsymbol{s}_i$ of the disorder centers. (iii) All diagrams with intersecting impurity scattering lines may be disregarded. (iv) We further neglect diagrams with more than two scattering events. In addition, we employ the quasiclassical approximation in treating the homogeneous superconducting state. With these assumptions, diagrams of all orders can be summed [49,53] and we obtain the main result of this Letter:

$$\check{\Sigma}_{\mathrm{int}}(\omega_n,\boldsymbol{p}) = \frac{1}{d_S}\left[\check{t} + N_{\mathrm{dis}}\int \frac{d^3p_1}{(2\pi)^3}|u(\boldsymbol{\kappa}-\boldsymbol{\kappa}_1)|^2\check{t}\check{G}(\omega_n,\boldsymbol{p}_1)\check{t}\right], \quad (10)$$

where the result is expressed concisely in the frequency ω_n and momentum $\boldsymbol{p}$ representation [56]. Here, $u(\boldsymbol{\kappa}) \equiv \int d^2s u(\boldsymbol{s})\exp(-i\boldsymbol{\kappa}\cdot\boldsymbol{s})$, N_{dis} is the areal density of disorder centers, $\boldsymbol{\kappa}$ is the in-plane component of the momentum $\boldsymbol{p}$, and d_S is the thickness of the S layer assumed to be much smaller than the superconducting coherence length. The Green's function for the proximity-coupled superconducting layer is given by $\check{G}^{-1}(\omega_n,\boldsymbol{p}) = \check{G}_0^{-1}(\omega_n,\boldsymbol{p}) - \check{\Sigma}_{\mathrm{int}}(\omega_n,\boldsymbol{p})$, in terms of the unperturbed Green's function $\check{G}_0(\omega_n,\boldsymbol{p})$ and the self-energy evaluated above.

Discussion.—The self-energy [Eq. (10)], stemming from the interface with MI, comprises a contribution independent of, and thus resistant to, interfacial disorder and a term proportional to the areal density of disorder centers N_{dis}. Apart from a small renormalization of the chemical potential, the former contribution is simply the effective exchange field, $\propto b = J_A M_{A0}/2|\gamma_A| - J_B M_{B0}/2|\gamma_B|$, induced in S. Thus, an AFM with uncompensated surface, for which $J_A \neq J_B$, $M_{A0} = M_{B0}$, and $\gamma_A = \gamma_B$, induces spin splitting in the adjacent S layer.

The interfacial disorder-mediated contribution to the self energy can be further divided into three terms with the integrands in Eq. (10), respectively, proportional to (i) $a^2(\hat{\tau}_0 \otimes \hat{\sigma}_0\check{G}\hat{\tau}_0 \otimes \hat{\sigma}_0)$, (ii) $b^2(\hat{\tau}_z \otimes \hat{\sigma}_z\check{G}\hat{\tau}_z \otimes \hat{\sigma}_z)$, and (iii) $ab(\hat{\tau}_0 \otimes \hat{\sigma}_0\check{G}\hat{\tau}_z \otimes \hat{\sigma}_z + \hat{\tau}_z \otimes \hat{\sigma}_z\check{G}\hat{\tau}_0 \otimes \hat{\sigma}_0)$. The term (i) looks like the self-energy due to nonmagnetic impurities [49]. Assuming isotropic scattering, this contribution drops out of the superconducting gap as well as the Eilenberger equations for s-wave superconductors, consistent with the Anderson theorem [57]. Assuming that $\check{\mathcal{G}}^{-1}(x_1)$ is diagonal in spin space, which is the case here [Eq. (8)], the total matrix Green's function is also diagonal in spin space. Taking this into consideration, term (ii) may be rewritten as $\propto \hat{\tau}_z \otimes \hat{\sigma}_0 \check{G} \hat{\tau}_z \otimes \hat{\sigma}_0$, which has the same form as the self-energy contribution due to spin-flip scattering via magnetic impurities [49]. The effect of such a term has been studied and is known to result in phenomena such as gapless superconductivity [7]. It also has consequences for the density of states [58–60] and leads to an enhancement of the Seebeck effect in magnet or superconductor heterostructures [61].

Again, accounting for the diagonal in spin space structure of the total Green's function, the contribution to the self-energy corresponding to the term (iii) assumes the matrix structure $\propto \hat{\tau}_0 \otimes \hat{\sigma}_z$, thereby breaking the symmetries in both Nambu and spin spaces. An explicit evaluation of the quasiclassical Green's function matrix shows that this term drops out on integrating over the excitation energy. Thus, this term renormalizes the normal-state properties of the S layer while dropping out in the quasiclassical description of the superconducting state. The analogous term in the self-energy evaluated beyond the Born approximation for magnetic impurities in a bulk superconductor, which does not lead to any spin splitting, was found to break the particle-hole symmetry [62]. Its key manifestation was asymmetric scattering with the Yu-Shiba-Rusinov states [63–65] resulting in a large thermoelectric effect [62].

In general, the Hamiltonian, and thus the total matrix Green's function, may be nondiagonal in spin space when, for example, the magnetization is spatially inhomogeneous or the superconductor exhibits unconventional same-spin electron pairing. Under those circumstances, terms (ii) and (iii) may not be interpreted as discussed above.

Here, we have considered a superconducting layer much thinner than the coherence length. For a thick superconductor, the evaluated self-energy may be incorporated in the boundary conditions for the Gor'kov equation in the bulk. Thus, our theory also provides a microscopic derivation of the boundary conditions describing the interface of a superconductor with a magnetic insulator, complementary to the corresponding evaluations within a scattering theory approach [66–68]. Furthermore, we have considered a single-domain magnet leaving possible generalizations to textured and multidomain interfaces for future work [69]. Reference [25] reviews exchange bias and the magnetic proximity effect together thereby delineating the connection between the two phenomena further and providing directions for generalizing our results.

From the experimental point of view, it is considered difficult to grow metals on insulators due to lattice mismatch. Such interfaces are inevitably disordered. Nevertheless, a strong interfacial exchange coupling has been observed in a wide range of such structures [51,52,70–75]. This is consistent with our result demonstrating that interfacial disorder does not lead to any qualitative changes in physics and the induced exchange field is resistant to this disorder. It, however, leads to additional spin-flip scatteringlike contributions which, in some cases [61,62,76,77], may be desirable.

As elaborated in the Supplemental Material [56], the existing literature on exchange bias [19] and spin-mixing conductance [70,75,78,79] provides valuable guidance regarding materials and corresponding expected spin splittings. Several AFMs, such as CoO, FeF_2, and FeS, may induce fields greater than 100 mT in a 10 nm thick superconducting layer [19,56]. Furthermore, multilayers incorporating one or more ferromagnetic seed layers are expected to be particularly effective [19,56], while still circumventing the disadvantages of spin splitting induced via a ferromagnetic layer.

Summary.—We have derived and solved the Gor'kov equation for two-sublattice magnetic insulator-thin superconductor bilayer structures. Starting with a microscopic description of the interface, we have evaluated the interfacial self-energy for the matrix (Nambu-spin space) Green's function in the superconducting layer. Our findings show that an antiferromagnet with an uncompensated surface, in addition to ferrimagnets, induces interfacial disorder-resistant spin splitting in the adjacent superconductor. Additional contributions mimicking nonmagnetic impurities and spin-flip scattering result due to the interfacial disorder. Our findings, in conjunction with related experiments [19,36,37,75], pave the way for employing antiferromagnetic insulators in inducing exchange field in an adjacent superconductor, thereby addressing the feasibility of a wide range of concepts and devices involving spin-split superconductors.

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