Enhancing triplet superconductivity by the proximity to a single superconductor in oxide heterostructures

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We show how in principle a coherent coupling between two superconductors of opposite parity can be realized in a three-layer oxide heterostructure. Due to strong intraionic spin-orbit coupling in the middle layer, singlet Cooper pairs are converted into triplet ones and vice versa. This results in a large enhancement of the triplet superconductivity, persisting well above the native triplet critical temperature.

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The prospect of realizing Majorana bound states that can be used for quantum information processing has led to a large interest in odd-parity superconductivity. Native triplet superconductivity, believed to be realized in, e.g., Sr2RuO4, is fragile and only present at very low temperatures [1]. It is known that a singlet superconductor (SC) can induce triplet pairing correlations in systems with Rashba spin-orbit coupling and/or ferromagnetism due to the proximity effect [2–10], and it has been suggested to induce triplet superconductivity in hybrid structures with such properties [11–14]. Rashba spin-orbit coupling and ferromagnetism can also give rise to a Josephson coupling between s- and p-wave SCs [15,16].

Here, we suggest an alternative way to improve the robustness of an odd-parity SC by tunnel coupling it to an even-parity singlet SC in all-oxide-based heterostructures. This mechanism is neither due to Rashba coupling nor ferromagnetism, but by virtue of a strong intraionic spin-orbit coupling inherent to late transition-metal compounds such as iridium oxide Sr2IrO4 [17]. To have a coherent coupling between two SCs of opposite parity, the tunneling has to “rotate” the Cooper pairs, since the wave functions of the odd-parity spin-triplet “ruthenate” SC (A layer) is separated from the odd-parity spin-triplet A layer by an insulating layer, the “iridate convertor” (see the inset of Fig. 1). The two SCs will show that a strong intraionic spin-orbit coupling in the middle layer, and in the Ru orbitals of the B layer, the “iridate convertor” (see the inset of Fig. 1). The Cooper pairs are converted into triplet ones and vice versa. This results in a large enhancement of the triplet superconductivity, persisting well above the native triplet critical temperature.

Model. The Hamiltonian of the system \( H = H_A + H_B + H_{AB} \) comprises the Hamiltonians \( H_A/B \) for the odd-parity spin-triplet A layer and the even-parity spin-singlet B layer, respectively, and the effective tunneling term \( H_{AB} \) due to the iridate convertor between these layers. The Hamiltonian for the triplet SC reads as

\[
H_A = \sum_{\mathbf{k}, \sigma} \epsilon_{\mathbf{k}}(\sigma) \mathbf{c}_{\mathbf{k}}^\dagger \mathbf{c}_{\mathbf{k} \sigma} + \text{H.c.},
\]

where \( \mathbf{c}_{\mathbf{k}} \) is the electron field in the Nambu form, \( \mathbf{c}_{\mathbf{k}} = (\mathbf{a}_{\mathbf{k}}^\dagger, \mathbf{a}_{\mathbf{k}}, \mathbf{a}_{-\mathbf{k}}^\dagger, \mathbf{a}_{-\mathbf{k}}) \), and

\[
\hat{H}_A(\mathbf{k}) = \begin{pmatrix} \hat{\xi}_A(\mathbf{k}) & \hat{\Delta}_A(\mathbf{k}) \\ \hat{\Delta}_A^*(\mathbf{k}) & -\hat{\xi}_A(\mathbf{k}) \end{pmatrix}.
\]

It is assumed that the dispersion \( \xi_A(\mathbf{k}) \) is independent of spin. The order parameter matrix \( \hat{\Delta}_A(\mathbf{k}) = i\mathbf{d}_{\mathbf{k}} \cdot \sigma \mathbf{S} \), where \( \sigma_j \) are the Pauli matrices.

The singlet-layer Hamiltonian \( H_B \) takes a similar form to \( H_A \), except for the replacements \( \hat{\xi}_B(\mathbf{k}) \rightarrow \hat{\xi}_A(\mathbf{k}) \), \( \hat{\Delta}_A(\mathbf{k}) \rightarrow \hat{\Delta}_B(\mathbf{k}) \) with \( \hat{\Delta}_B(\mathbf{k}) = i\sigma \mathbf{S} \Delta_{\mathbf{k}} \), and \( \hat{\Delta}_A \rightarrow \hat{\Delta}_B = (\mathbf{b}_{\mathbf{k}}, \mathbf{b}_{\mathbf{k}}^\dagger, \mathbf{b}_{-\mathbf{k}}, \mathbf{b}_{-\mathbf{k}}^\dagger) \).

A general tunneling term between the A and B layers can be written as

\[
T(\mathbf{k}) = \begin{pmatrix} \hat{T}(\mathbf{k}) & 0 \\ 0 & -\hat{T}^*(\mathbf{k}) \end{pmatrix}, \quad \hat{T}(\mathbf{k}) = \begin{pmatrix} P_k & R_k \\ S_k & Q_k \end{pmatrix}.
\]

Time-reversal invariance of the Hamiltonian gives the following restriction on the elements of the tunneling matrix: \( P_k = Q_{-k}^* \) and \( R_k = -S_{-k}^* \). The system under consideration is invariant under reflections about the \( xz \) plane, \( M_z \), where the position and spin transform as \( (x, y) \rightarrow (x, -y) \) and \( (S_x, S_y, S_z) \rightarrow (-S_x, S_y, -S_z) \). There is a similar symmetry under reflection about the \( yz \) plane, \( M_y \), so that in the spin sector \( M_x \) and \( M_y \) correspond to \( i\sigma_x \) and \( i\sigma_z \), respectively.
Since a spin-orbit coupling $L \cdot S$ is invariant under these symmetries, the tunnel Hamiltonian $\hat{T}$ is invariant under the combined operation $\mathcal{M}_s \mathcal{M}_t$ and obeys $\hat{T}(-\mathbf{k})\mathcal{M}_s = \hat{T}(\mathbf{k})$.

The free energy for the system can be calculated by integrating out the fermionic degrees of freedom $[20]$. It takes the form $F = F_A + F_B + F_{AB}$, where $F_{AB}(B)$ is the free energy for the $A(B)$ layer and $F_{AB}$ is the coupling between the two layers. Our focus is on the coupling between the two SCs. Assuming time-reversal invariance of the system and unitary $p$-wave superconductivity in the $A$ layer, the coupling to second order in $H_{AB}$ is $[2]$

$$F_{AB} \approx \frac{1}{2} \sum_{\mathbf{k}} W_{\mathbf{k}} \Delta^{s\tau}_{\mathbf{k}}(d^{\tau \dagger}_{\mathbf{k}}(\mathbf{P}_{-\mathbf{k}})^2 + |\mathbf{R}_{-\mathbf{k}}|^2)$$

$$+ (d^{\tau \dagger}_{\mathbf{k}} + i d^{\tau}_{\mathbf{k}}) \mathbf{P}_{-\mathbf{k}} R_{-\mathbf{k}} + (d^{\tau \dagger}_{\mathbf{k}} - i d^{\tau}_{\mathbf{k}}) \mathbf{P}_{-\mathbf{k}} R^{\tau \dagger}_{-\mathbf{k}} + \text{H.c.},$$

where only the terms sensitive to the phase difference between the two layers have been kept $[21]$. Here, $W_{\mathbf{k}} = (\Phi^{d}_{\mathbf{k}} - \Phi^{d}_{-\mathbf{k}}) / (E_{\mathbf{k}} - E_{-\mathbf{k}})$, a function $\Phi^{A/B} = \frac{1}{E_{A/B}} \tanh \frac{E_{A/B}}{2T}$, and $\beta$ is the inverse temperature.

The quasiparticle energies are given by $E_{A/B}(\mathbf{k}) = \sqrt{\frac{\Delta^{s\tau}_{\mathbf{k}}}{\mathcal{M}_s \mathcal{M}_t}} + |\Delta^{s\tau}_{\mathbf{k}}|^{2}$. The symmetry $\mathcal{M}_s \mathcal{M}_t$ corresponds to $\mathbf{k} \rightarrow -\mathbf{k}$ and a $\pi$ rotation of spins around the $z$ axis, such that the invariance of the $\mathbf{d}_{\mathbf{k}}$ vector implies $d_{i,j,k} = -d_{j,i,k}$ and $d_{i,j,k} = d_{j,i,k}$, and as a consequence, $d^{\tau \dagger}_{\mathbf{k}} \equiv 0$. In Eq. (3), this is reflected by the fact that $d_{i,j,k} (d_{i,j,k}^{\dagger})$ is multiplied by an odd (even) function $\mathbf{d}_{\mathbf{k}}$ of $\mathbf{k}$, so only $d_{i,j,k}$ terms may couple to a $\mathbf{k}$-even $\Delta^{s\tau}_{\mathbf{k}}$.

To illustrate the effect with a simple toy model, we assume that the elements of the tunneling matrix take the simple form $P_R = i P$ and $R_{\mathbf{k}} = R(\sin k_x - i \sin k_y)$, with $P$ and $R$ real. To get an idea which combinations of order parameters in the singlet and triplet layers give a nonvanishing coupling, we consider the following order parameters for the triplet layer $[2]$

$$\Gamma_{1/3}^{\tau} : \mathbf{d}_{\mathbf{k}} = \eta e^{i \theta} (\mathbf{e}_x \sin k_x \pm \mathbf{e}_y \sin k_y),$$

$$\Gamma_{2/4}^{\tau} : \mathbf{d}_{\mathbf{k}} = \eta e^{i \theta} (\mathbf{e}_x \sin k_y \mp \mathbf{e}_y \sin k_x),$$

and that the singlet-layer pairing has either s- or d-wave symmetry, $\Delta^{s\tau}_{\mathbf{k}} = \Delta_0 (\cos k_x + \cos k_y)$ and $\Delta^{d\tau}_{\mathbf{k}} = \Delta_0 (\cos k_x - \cos k_y)$, respectively. Here $\eta, \Delta_0 > 0$, and $\theta$ is the phase difference between the $p$- and $s/d$-wave order parameters. For this model, except for the cases $\Gamma_{1/3}$ and s wave or $\Gamma_{2/4}$ and d wave, the integrand in Eq. (3) is either exactly vanishing, antisymmetric under mirroring about the $y$ axis, or antisymmetric under a $90^\circ$ rotation about the origin.

Therefore only the combinations $(\Delta^{s\tau}_{\mathbf{k}}, \Gamma_{1/3}^{\tau})$ and $(\Delta^{d\tau}_{\mathbf{k}}, \Gamma_{2/4}^{\tau})$ give a nonvanishing $F_{AB}$. The first combination constitutes a fully gapped helical topologically nontrivial SC $[23–25]$.

**Proximity enhancement.** The coupling between the two SCs leads to a dramatic enhancement of the triplet order parameter, as we will see now. Close to the native critical temperature $T_{C0}$ of the A layer, the triplet order parameter is small and the free energy can be expanded in $\eta$. Assuming that the singlet pairing in the $B$ layer is robust, $\Delta_0 > T_{C0}$, and its variation near $T_{C0}$ is negligible, we can ignore the $F_B$ term since it only contributes a constant to the energy. To fourth order in $\eta$, the free energy can then be written as

$$F = (r - 1) a \eta^2 + \frac{1}{2} b \eta^4 - r a \eta \eta \cos \theta,$$

where $a, b > 0$ are constants describing the native A layer $[26,27]$, and $r = T / T_{C0}$. The enhancement factor $r$ is defined by $F(a, b, \theta) = -\eta \cos \theta$. Here, $\eta_0$ is the zero temperature gap at vanishing $F_{AB}$. For fixed $\eta$, it is clear that $F$ is minimized when $\cos \theta = \text{sgn}(r)$. The value attained by the p-wave order parameter is found by minimizing $F$ with respect to $\eta$. Figure 1 shows $\eta$ as a function of temperature for representative values of $r$ (see the discussion below). We see that the coupling to the $B$ layer can give a large enhancement of the triplet superconductivity persisting well above $T_{C0}$.

Due to the amplification of the triplet order parameter, the anomalous pair-tunneling current will persist also above the “native” critical temperature $T_{C0}$ in the form of a zero-bias peak in the differential conductance $[28]$. More directly, the proximity-enhanced triplet gap (proportional to $a \eta$) and its temperature dependence (see Fig. 1) can be probed by scanning tunneling microscopy (STM) or angle-resolved photoemission spectroscopy (ARPES) experiments.

**Iridate convertor.** We now discuss a possible realization of the layered structure that gives rise to a tunneling matrix of the form (2), where the diagonal elements have even parity and the off-diagonal ones have odd parity, providing a coherent coupling between the $A$ and $B$ SCs.

We assume that all three layers have a square lattice geometry with similar lattice constants. A possible candidate, which can be designed by a modern layer-by-layer growth technique $[29]$, could be the oxide heterostructure $\text{Sr}_2\text{RuO}_4/\text{Sr}_2\text{IrO}_4/\text{La}_2\text{CuO}_4$. The pairing in the $B$ (cuprate) layer takes place in the Cu $d_{x^2-y^2}$ orbitals designated by the annihilation operator $b_{\mathbf{\sigma} r}$, where $r$ is the (two-dimensional) lattice position and $\sigma$ the spin, while the pairing in the A (ruthenate) layer is assumed to take place in the Ru $d_{xy}$ orbitals $[30]$, labeled by $a_{\mathbf{\sigma} r}$. The relevant orbitals in the middle layer
odd-parity elements in the tunneling matrix. Due to the relative sign difference between hopping in the positive and negative $x$ directions. A similar argument applies to hopping between a $d_{x^2-y^2}$ orbital in the middle layer and a $d_{xy}$ orbital located at $r \pm e_x \pm e_z$; we denote this hopping by $t'$. Similar arguments apply to the hopping $t''$ between $d_{x^2-y^2}$ orbitals and $d_{xy}$ orbitals. There is also a hopping between the $d_{x^2-y^2}$ orbitals of the middle and A layer. In this case there is no relative minus sign for hopping in the opposite $x(y)$ direction and hopping to all next-nearest neighbors have the same magnitude, which gives a tunneling of the form $t''Y_{\alpha \beta}(\alpha_{dxz}, + \alpha_{dxy}) + H.c.$ Projecting the above $t'$ and $t''$ hopping processes onto the $d_{eff} = 1/2$ band, we find the tunneling Hamiltonian between the middle layer and the A layer:

$$H_{MA} = \frac{2}{\sqrt{3}} \sum_{k \alpha} f_{\alpha}^{\dagger} \left[ -i t' \sigma (\sin k_x - i \sigma \sin k_y) d_{\alpha}^{\dagger} k - \sigma + H.c. \right].$$

Introducing an effective charge transfer energy $\Delta E$ required to move an electron into the middle layer, we can calculate the effective tunneling Hamiltonian between the Cu $d_{x^2-y^2}$ orbitals and the Ru $d_{xy}$ orbitals to second order in $H_{BM}$ and $H_{MA}$. We then find that the elements of the tunneling matrix (2) are given by

$$P_k = i g_e (\sin^2 k_x + \sin^2 k_y),$$

$$R_k = -g_o (\cos k_x + \cos k_y) (\sin k_x - i \sin k_y),$$

with amplitudes $g_e = \frac{4}{\Delta E} t' t''$ and $g_o = \frac{4}{\Delta E} t'' t'''$ in even- and odd-parity channels, correspondingly. While it is difficult to quantify these constants, the above orbital-symmetry considerations confirm that the desired topology of the tunneling matrix, with an opposite parity of the diagonal and off-diagonal elements, is indeed realistic in perovskite-type oxide heterostructures.

Inserting now the tunneling coefficients (11) into Eq. (3), we find a nonvanishing $F_{AB}$ in the $(\Delta^{f}_{Bk}, \Gamma_2^d)$ and $(\Delta^{d}_{Bk}, \Gamma_4^d)$ channels (as it was observed above), and evaluate the corresponding coupling constants $r$ using circular Fermi surfaces for simplicity. The main contribution to $F_{AB}$ (3) stems from the region close to the Fermi surface in the A layer. Away from nesting of the Fermi circles in the A and B layers, the enhancement factor $r$ will be suppressed by $\delta^2 = |\xi_B(k_F^d) - \xi_A(k_F^d)|^2$, where $k_F^d$ is the Fermi-circle radius in the A layer. An estimate of the enhancement factor $r_1$ (due to

Projection of the Ir $t_{2g}$ states onto the $f_\sigma$ band gives the following correspondence:

$$(\alpha_{r,\sigma}; \beta_{r,\sigma}; \gamma_{r,\sigma}) \rightarrow \frac{1}{\sqrt{3}} ( -\sigma f_{r,-\sigma}; i f_{r,-\sigma}; f_{r,\sigma} ).$$

With this substitution in Eq. (7) and after a Fourier transformation, we arrive at the tunneling between the B layer and the middle ($M$) layer:

$$H_{BM} = \sum_{k \sigma} \sum_{k' \sigma'} t(k) (\sin k_x - i \sigma \sin k_y) d_{\sigma}^{\dagger} k + H.c.$$
for the cases of functions \[\text{Eqs. (12) and (13)}\] on the Fermi circle radius. The results to single-particle tunneling considered so far gives

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Figure 3 shows where \(f_0\) is a function that only depends on the Fermi-circle radius. Figure 3 shows another process that contributes to \(\delta_1\), and gives the following contribution to the enhancement factor:

\[r_2 \approx -\frac{g_{e,g}}{\Delta_0} N_0 V \ln \left( \frac{2\omega_c \Omega}{\Delta_0} \right) \left( \frac{\omega_c}{T_C} \right) f_2(k_F^B, k_F^B), \tag{13}\]

where \(N_0\) is the density of states and \(\omega_c\) is an upper frequency cutoff. The function \(f_2\) depends on the Fermi-circle radii in the \(A\) and \(B\) layers, and is plotted in Fig. 3 as a function of \(k_F^B\) (at \(k_F^B = \kappa_0 + \frac{\pi}{\Omega}\)). Assuming \(g_{e,g} \sim 0.1 \Delta E, N_0|V| \sim 0.5,\) and \(\omega_c/\Delta_0 \sim 10\), we find \(r_2 \approx \pm 2.5 f_2\). Depending on microscopics, \(r_2\) is positive for an attractive potential (e.g., for a phonon and/or magnetically mediated interaction \(V < 0\)), and negative for a repulsive one. From the above estimates, it seems plausible that the single-particle and pair-tunneling processes can give a sizable enhancement factor of the order of \(|r| \sim 1\), as used in Fig. 1. In addition, antiferromagnetic (AF) correlations are expected to arise in the iridate layer \([17,38]\). Since pseudospins are spin-orbit composite objects, their AF correlation is in fact a coherent mixture of real-spin singlets and triplets, implying that the iridate AF correlations will further facilitate a coherent singlet-triplet conversion.

In conclusion, we have shown how a coherent coupling between a triplet and a singlet SC can be achieved by means of a time-reversal invariant conversion layer that effectively rotates singlet Cooper pairs into triplets. The conversion is due to tunneling via the strong intragrain spin-orbit coupled states in the middle layer; a possible candidate for such a “pair convertor” might be the iridium oxide \(\text{Sr}_2\text{IrO}_4\). The coherent coupling leads to a dramatic enhancement of the triplet superconductivity, existing well above its “native” critical temperature \(T_C\). Experimentally, the enhanced triplet gap in the quasiparticle spectrum and its temperature dependence as shown in Fig. 1 can be verified using ARPES and STM techniques. The proximity mechanism considered here may also enable the stabilization of topologically nontrivial \(p\)-wave SCs.

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[9] A triplet order parameter can also be induced in a different type of system, such as in the vortex phase of high-\(T_c\) SCs [10].
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[18] Due to breaking of mirror symmetry in the $z$ direction, a Rashba-type spin-orbit coupling can be induced at the $AB$ interface. Except for very special and rare systems [19], this effect is small and therefore ignored here since it will not change the results qualitatively.
[21] Higher-order terms in $H_{AB}$ can give rise to a phase-dependent coupling between the two SCs even in the absence of a spin active interface, but these terms will in general be smaller and have a different dependence on the phase difference [22].
[30] The pairing in ruthenates is in reality of multiorbital nature and not yet fully understood (see, e.g., the recent works [31–34]); however, our assumption of the $xy$ orbital is not of a principal importance for illustration of the basic idea of the present Rapid Communication.
[36] Due to spin-orbit coupling, the Ir $d_{x^2-y^2}$ and $d_{xy}$ orbitals mix, and will give an effective hopping between Ir $d_{x^2-y^2}$ orbitals in the $B$ layer and the Ir $d_{xy}$ orbitals. There is also a term corresponding to hopping in the straight $e_z$ direction. This effective term will not change the physics and is therefore ignored to keep the model as simple as possible.