Enhancing triplet superconductivity by the proximity to a singlet 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- and even-parity superconducting condensates are orthogonal to each other. We consider a heterostructure consisting of three quasi-two-dimensional layers: The even-parity spin-singlet A layer and the odd-parity spin-triplet B layer, respectively, and the effective tunneling term HAB due to the iridate convertor between these layers. The Hamiltonian for the triplet SC reads as $H_A = \frac{1}{2} \sum_k \vec{\hat{A}}_k H_A(k) \vec{A}_k$, where the electron fields have been written in Nambu form, $\vec{\hat{A}}_k = (\hat{a}_{k\uparrow}, \hat{a}_{k\downarrow}, a_{\cdots k\uparrow}, a_{\cdots k\downarrow})$, and

$$\hat{H}_A(k) = \begin{pmatrix} \xi_A(k) & \hat{\Delta}_A(k) \\ \hat{\Delta}_A^\dagger(k) & -\xi_A(k) \end{pmatrix}. \tag{1}$$

It is assumed that the dispersion $\xi_A(k)$ is independent of spin. The order parameter matrix $\hat{\Delta}_A(k) = i\vec{d}_A \cdot \vec{\sigma} \sigma_j$, where $\sigma_j (j = x,y,z)$ are the Pauli matrices.

The single-layer Hamiltonian $H_B$ takes a form similar to $H_A$, except for the replacements $\xi_B(k) \rightarrow \xi_A(k)$, $\Delta_B(k) \rightarrow \Delta_A(k)$ with $\Delta_B(k) = i\sigma_j \Delta_B k$, and $\hat{\Delta}_B \rightarrow \hat{\Delta}_A$.

A general tunneling term between the A and B layers can be written as $H_{AB} = \frac{1}{2} \sum_k \hat{A}_k T(k) \hat{B}_k + H.c.$, where

$$T(k) = \begin{pmatrix} \hat{T}(k) & 0 \\ 0 & -\hat{T}^\dagger(-k) \end{pmatrix}, \quad \hat{T}(k) = \begin{pmatrix} P_k & R_k \\ S_k & Q_k \end{pmatrix}. \tag{2}$$

Time-reversal invariance of the Hamiltonian gives the following restriction on the elements of the tunneling matrix:

$$P_k = Q^*_{-k} \text{ and } R_k = -S^*_{-k}.$$ The system under consideration is invariant under reflections about the $yz$ plane, $\mathcal{M}_y$, where the position and spin transform as $(x,y) \rightarrow (x,-y)$ and $(\vec{S}_x, \vec{S}_y, \vec{S}_z) \rightarrow (-\vec{S}_x, -\vec{S}_y, -\vec{S}_z)$. There is a similar symmetry under reflection about the $yz$ plane, $\mathcal{M}_z$, so that in the spin sector $\mathcal{M}_y$ and $\mathcal{M}_z$ correspond to $i\sigma_y$ and $i\sigma_z$, respectively.
Since a spin-orbit coupling \( \mathbf{L} \cdot \mathbf{S} \) is invariant under these symmetries, the tunnel Hamiltonian \( \mathcal{T} \) is invariant under the combined operation \( \mathcal{M}_z \mathcal{M}_y \) and obeys \( \sigma_y \mathcal{T}(-\mathbf{k}) \sigma_z = \mathcal{T}(\mathbf{k}) \).

\( \Phi_1 \) and \( \Phi_2 \) are even functions of 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]

\[
F_{AB} \simeq \frac{1}{2} \sum_{\mathbf{k}} W_{\mathbf{k}} \Delta^*_A |d_{\mathbf{k}}(\mathbf{p}_{\mathbf{k}})|^2 + |R_{\mathbf{k}}|^2 + (d_{\mathbf{k}} + i d_{\mathbf{k}}^*) P_{\mathbf{k}} R_{\mathbf{k}} - (d_{\mathbf{k}} - i d_{\mathbf{k}}^*) P_{\mathbf{k}}^* R_{\mathbf{k}}^* + \text{H.c.},
\]

where only the terms sensitive to the phase difference and the other terms vanish. The quasiparticle energies are given by \( E_{AB}(\mathbf{k}) = \sqrt{E_{\mathbf{k}}^2 + \Delta^2_{AB} |\mathbf{k}|^2} \).

The symmetry \( \mathcal{M}_z \mathcal{M}_y \) corresponds to \( \mathbf{k} \to -\mathbf{k} \) and a \( \pi \) rotation of spins around the \( z \) axis, such that the invariance of the \( d_{\mathbf{k}} \) vector implies \( d_{\mathbf{k}/\mathbf{k}} = -d_{\mathbf{k}/\mathbf{k}}^* \) and \( d_{\mathbf{k}/\mathbf{k}} = d_{\mathbf{k}/\mathbf{k}}^* \), and as a consequence, \( d_{\mathbf{k}} \equiv 0 \). Eq. (3) is valid for the fact that \( d_{\mathbf{k}/\mathbf{k}}^* (d_{\mathbf{k}/\mathbf{k}}) \) is multiplied by an odd (even) function of \( d_{\mathbf{k}} \), so only \( d_{\mathbf{k}/\mathbf{k}}^* d_{\mathbf{k}/\mathbf{k}} \) terms may couple to a \( d_{\mathbf{k}} \)-even \( \Delta_{\mathbf{k}/\mathbf{k}} \).

To illustrate the effect with a simple toy model, we assume that the elements of the tunneling matrix take the simple form \( P_{\mathbf{k}} = i P \) 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],

\[
\begin{align*}
\Gamma_{1/3}^- &: \quad d_{\mathbf{k}} = \eta e^{i\theta} (\mathbf{e}_x \sin k_y \pm \mathbf{e}_y \sin k_x), \\
\Gamma_{2/4}^- &: \quad d_{\mathbf{k}} = \eta e^{i\theta} (\mathbf{e}_x \sin k_y \mp \mathbf{e}_y \sin k_x),
\end{align*}
\]

and that the singlet-layer pairing has either \( s \)- or \( d \)-wave symmetry. \( \Delta^s_{\mathbf{k}} = \Delta_0 (\cos k_x + \cos k_y) \) and \( \Delta^d_{\mathbf{k}} = \Delta_0 (\cos k_x - \cos k_y) \), 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 \( (\Delta_{\mathbf{k}/\mathbf{k}}^s, \Gamma_{1/3}^-) \) and \( (\Delta_{\mathbf{k}/\mathbf{k}}^d, \Gamma_{2/4}^-) \) 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, \( \Delta_0 \gg T_{C0} \), and its variation near \( T_{C0} \) is negligible, we can ignore the \( F_{ab} \) 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 layer [26,27], and \( r = T/T_{C0} \). The enhancement factor \( r \) is defined by \( \frac{F_{ab}}{\hbar_0} = -r \eta \cos \theta \). Here, \( \eta_0 \) is the zero temperature gap at \( \eta_0 \). 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 \( \eta \)) and its temperature dependence (see Fig. 1) can be probed by scanning tunneling microscopy (STM) or angle-resolved photoemission spectroscopy (ARPES) experiments.

**Iridate converter.** 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 \( \text{Cu}_d \) \( \hat{d}_{\mathbf{x},\mathbf{y}} \) orbitals designated by the annihilation operator \( b_{\mathbf{x},\mathbf{y}} \), with \( \mathbf{r} \) 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 \( \text{Ru}_d \) \( \hat{d}_{\mathbf{x},\mathbf{y}} \) orbitals [30], labeled by \( a_{\mathbf{x},\mathbf{y}} \). The relevant orbitals in the middle layer
are the Ir $t_{2g}$ orbitals $d_{xz}, d_{yz}, d_{xy}$ denoted below by $\alpha_{\sigma\sigma}, \beta_{\sigma\sigma}, \gamma_{\sigma\sigma}$, respectively. Figure 2 shows the orientation of the above orbitals.

We consider first the tunneling between the $B$ layer and the middle layer. Figure 2 shows the possible hopping paths between the two layers. There can be no hopping between a $d_{x\text{-}y^2}$ orbital located at $r$ and a $d_{xy}$ orbital located at $r \pm e_y + e_z$ for symmetry reasons [35]. On the other hand, hopping from a $d_{x\text{-}y^2}$ orbital located at $r$ to a $d_{xz}$ orbital located at $r \pm e_x + e_z$ is symmetry allowed. However, there will be a relative sign difference between hopping in the positive and negative $x$ directions. A similar argument applies to hopping between a $d_{x\text{-}y^2}$ orbital in the $B$ layer and a $d_{xy}$ orbital in the middle layer. The tunneling between the $B$-layer $d_{x\text{-}y^2}$ orbitals and the $d_{xz}$ and $d_{yz}$ orbitals in the middle layer is then

$$ t^{\hbar}_{\text{re}} [(\alpha_{r-e_x} - \alpha_{r+e_x}) - (\beta_{r-e_x} - \beta_{r+e_x})]_\sigma + \text{H.c.}, \quad (6) $$

where $t$ is the hopping strength. The relative sign difference for hopping in the opposite $x(y)$ direction will give rise to the odd-parity elements in the tunneling matrix. Due to the relative 45° rotation of the Cu $d_{x\text{-}y^2}$ and Ir $d_{xy}$ orbitals, there will always be equal contributions of opposite sign in an overlap integral [35]. The same argument gives a vanishing element for hopping in the straight $e_z$ direction [36].

We recall now that the spin and orbital states of the Ir ion are strongly entangled via intratomic spin-orbit coupling, $H_{SO} = \lambda \mathbf{L} \cdot \mathbf{S}$, which results in a completely filled $J_{\text{eff}} = 3/2$ quartet well below the Fermi level, and a half-filled $J_{\text{eff}} = 1/2$ doublet [17]. This implies that the tunneling process is mostly contributed by half-filled $J_{\text{eff}} = 1/2$ states, with the following wave functions [17,37]:

$$ |f_{\sigma} \rangle = \frac{1}{\sqrt{3}} [\sigma |yz, -\sigma \rangle + i |xz, -\sigma \rangle + |xy, \sigma \rangle]. \quad (7) $$

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

$$ (\alpha_{r,\sigma}; \beta_{r,\sigma}; \gamma_{r,\sigma}) \rightarrow \frac{1}{\sqrt{3}} (-\sigma f_{r,\sigma} - if_{r,-\sigma}; f_{r,\sigma}). \quad (8) $$

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:

$$ H_{BM} = \frac{2}{\sqrt{3}} \sum_{k\sigma} t^{\hbar}_{\text{re}} (\sin k_x - i \sigma \sin k_z) b^{\dagger}_{k\sigma} f_{k\sigma} + \text{H.c.}. \quad (9) $$

We now consider the tunneling between $t_{2g}$ orbitals in the middle layer and the $d_{xy}$ orbital in the A layer. By arguing as above, we find that there will only be hopping between $d_{xz}$ orbitals at $r$ and $d_{xy}$ orbitals at $r \pm e_y + e_z$; we denote this hopping by $t'$. Similar arguments apply to the hopping $t''$ between $d_{xz}$ orbitals and $d_{xy}$ orbitals. There is also a hopping between the $d_{xy}$ 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 $t' t'' I_{\text{re}} (\alpha_{r,\sigma}, \gamma_{r,\sigma}) + \alpha_{r,\sigma} + \gamma_{r,\sigma} + \text{H.c.}$ Projecting the above $t'$ and $t''$ hopping processes onto the $J_{\text{eff}} = 1/2$ band, we find the tunneling Hamiltonian between the middle layer and the A layer:

$$ H_{MA} = \frac{2}{\sqrt{3}} \sum_{k\sigma} f^{\dagger}_{k\sigma} [-i t' \sigma (\sin k_x - i \sigma \sin k_z) a_{k,-\sigma}$$

$$ + t'' (\cos k_x + \cos k_y) a_{k\sigma}] + \text{H.c.}. \quad (10) $$

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 $d_{x\text{-}y^2}$ 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

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

$$ R_k = -g_0 (\cos k_x + \cos k_y) (\sin k_x - i \sin k_y), \quad (11) $$

with amplitudes $g_e = \frac{4}{\Delta E} t^{\hbar} t'$ and $g_0 = \frac{4}{\Delta E} t^{\hbar} t''$ 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 $(\Delta^{r}_{k\sigma}, \Gamma_{-3})$ and $(\Delta^{d}_{k\sigma}, \Gamma_{-3})$ channels (as it was observed above), and evaluate the corresponding coupling constants $r$ using circular Fermi surfaces for simplicity. The main contribution to $F_{AB}$ 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 $\delta^2 = |\xi_B(k^B_F) - \xi_A(k^A_F)|^2$, where $k^B_F$ is the Fermi-circle radius in the A layer. An estimate of the enhancement factor $r_1$ (due
for the cases of results to single-particle tunneling considered so far) gives

$$f_1 \approx \frac{g_\varepsilon g_o}{\Delta_0} \frac{\Delta_0}{T_{C0}} \ln \left( \frac{\delta}{T_{C0}} \right) f_1(k^A_f),$$

(12)

where $f_1$ is a function that only depends on the Fermi-circle radius. Figure 3 shows $f_1(k^A_f)$ for the two combinations $(\Delta_{s1}, \Gamma_{s1})$ and $(\Delta_{d1}, \Gamma_{d1})$. For representative values of $g_{\varepsilon/o} \sim 0.18$, $\Delta_0/T_{C0} \sim 30$, and $\Delta_0/\delta \sim 0.2$, we find $r_1 \approx 1.5 f_1$.

Another process that contributes to $F_{AB}$ in addition to (3) is the scattering of a Cooper pair of relative moment $2p$ in the $B$ layer to a pair of relative momentum $2k$ in the $A$ layer, due to an electron-electron scattering potential of strength $V$ in the middle layer. This pair-tunneling process, sketched in the lower panel of Fig. 3, is not suppressed by the energy difference $\delta$ and gives the following contribution to the enhancement factor:

$$r_2 \approx -\frac{g_\varepsilon g_o}{(\Delta_E)^2 T_{C0}} N_0 V \ln \left( \frac{2\omega_c}{\Delta_0} \right) \ln \left( \frac{\omega_c}{T_{C0}} \right) f_2(k^A_f, k^B_f),$$

(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 radius in the $A$ and $B$ layers, and is plotted in Fig. 3 as a function of $k^A_f$ (at $k^B_f = k^A_f + \pi/B$). Assuming $g_{\varepsilon/o} \sim 0.1 \Delta_E$, $N_0|V| \sim 0.5$, 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-singlet 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 intrionic spin-orbit coupled states in the middle layer; a possible candidate for such a “pair convertor” might be the iridium oxide Sr$_2$IrO$_4$. The coherent coupling leads to a dramatic enhancement of the triplet superconductivity, existing well above its “native” critical temperature $T_{C0}$. 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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