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Spontaneous magnetization in unitary superconductors with time reversal symmetry breaking

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We report the study of spontaneous magnetization (i.e., spin-polarization) for time-reversal symmetry (TRS)-breaking superconductors with unitary pairing potentials, in the absence of external magnetic fields or Zeeman fields. Spin-singlet ($\Delta_s$) and spin-triplet ($\Delta_t$) pairings can coexist in superconductors whose crystal structure lacks inversion symmetry. The TRS can be spontaneously broken once a relative phase of $\pm\pi/2$ is developed, forming a TRS-breaking unitary pairing state ($\Delta_s \pm i\Delta_t$). We demonstrate that such unitary pairing could give rise to spontaneous spin-polarization with the help of spin-orbit coupling. Our result provides an alternative explanation to the TRS breaking, beyond the current understanding of such phenomena in the noncentrosymmetric superconductors. The experimental results of Zr$_3$Ir and CaPtAs are also discussed in the view of our theory.

Introduction—In condensed matter physics, superconductivity and magnetism are generally antagonistic to each other [1–3] and the interplay between them brings us intriguing phenomena. One of them is the Fulde-Ferrell-Larkin–Ovchinnikov (FFLO) state [4, 5] in which superconducting Cooper pairs carry a finite momentum induced by an external Zeeman field. Recent theoretical efforts have been made to realize chiral Majorana modes in the topological FF phase [6, 7] and the Majorana mode chain in the topological LO phase [8]. Another fascinating phenomenon is the spontaneous magnetization or spin-polarization (SP) in a time-reversal symmetric (TRS)-breaking superconductor (SC) [9–11], which continuously promotes extensive experimental and theoretical research [12, 13]. The TRS-breaking candidate SCs include Sr$_2$RuO$_4$ [14–16], ReT ($T =$ transition metal) [17–22], UPr$_3$ [23–25], UGe$_2$ [26, 27], URhGe [28], UCoGe [29, 30], PrOs$_3$Sb$_2$ [31], URu$_2$Si$_2$ [32, 33], SrPt$_4$As [34], Ru$_2$B$_2$ [35, 36], LaNiC$_3$ [37], LaNiGa$_3$ [38, 39], Bi/Ni bilayers [40], CaPtAs [41], Zr$_3$Ir [42], and others summarized in a recent paper [13]. More recently, iron-based SCs also exhibit TRS-breaking signatures [43, 44]. These exciting experimental discoveries arouse considerable attentions, and a great deal of theoretical progress has recently been made on TRS-breaking SCs with mixed pairing states [45–55], non-unitary pairing states [56–67], and Bogliubov Fermi surface [68–71].

There are mainly two direct ways to probe spontaneously TRS-breaking pairing states, including the zero-field muon-spin relaxation (µSR) [72–74], and the polar Kerr effect (PKE) [75, 76]. Firstly, the µSR is especially very sensitive to a small change of internal fields (with a resolution down to 10 µT) [77]. The enhancement of the zero-field muon-spin polarization rate in the superconducting state provides direct evidence for TRS breaking pairing states. Besides, the PKE measures the optical phase difference between two opposite circular-polarized lights reflected on a sample surface, thus it gives information about the TRS of a system. A finite PKE unambiguously points to TRS-breaking states. Theoretically, non-unitary spin-triplet pairing potentials, such as the $A_1$ phase in He$^3$ superfluid characterized by spin-triplet $a_t(k) = k_z(1, -i, 0)$ [78], could spontaneously induce SP in a homogeneous SC and thus naturally explaining experimental observations by the µSR and the PKE [1, 2]. However, one may still wonder if there is any mechanism other than the non-unitary spin-triplet pairing states to induce the SP in the TRS-breaking superconductors.

In this work, we report the discovery of SP in TRS-breaking unitary SCs. The spin-singlet pairing ($\Delta_s$) coexists with the spin-triplet pairing ($\Delta_t$) in both noncentrosymmetric SCs and superconducting thin films. Once a relative phase of $\pm\pi/2$ is developed ($\Delta_s \pm i\Delta_t$), TRS is spontaneously broken. By combing the symmetry analysis and the Ginzburg-Landau (GL) theory, we find that the interplay between spin-orbit coupling (SOC) and the $\Delta_s \pm i\Delta_t$ unitary pairing potential could give rise to SP in a homogeneous SC. The direction of the induced SP is perpendicular to both the SOC $g$-vector and the spin-triplet $d$-vector, even though both $g$ and $d$ are real vectors. Our result provides an alternative explanation for the TRS breaking phenomenon. The potential applications of our theory to recently discovered noncentrosymmetric SCs (e.g., Zr$_3$Ir and CaPtAs) are also discussed.

TRS-breaking unitary pairings—In the absence of external magnetic fields or Zeeman fields, the non-vanishing magnetism or SP in the superconducting states generally causes the spontaneous breaking of TRS. The SP can be generated by non-unitary pairing states with a complex spin-triplet $d$-vector, whose direction is parallel to $d \times d^*$. Alternatively, in this work, we explore the spontaneous SP induced by TRS-breaking unitary pair-
ing states in SCs whose crystal structure lacks inversion symmetry. For this propose, we start with a single-band model Hamiltonian [79],

$$\mathcal{H}_0 = \xi_k \sigma_0 + \alpha \vec{g} \cdot \vec{\sigma},$$

(1)

where $\xi_k = k^2 / 2m - \mu$ is the electron band energy measured from the Fermi energy $\mu$, $\vec{\sigma}$ denotes the Pauli matrices of electron spin, and $\alpha$ is the strength of SOC. The inversion symmetry is broken due to $\vec{g}(k) = -\vec{g}(-k)$. In this work, we mainly focus on the Rashba-type SOC which is given by $\vec{g} = (-k_y, k_x)$, allowed by the $C_{4v}$ point group. There are two Fermi surfaces with opposite chirality, and the two bands are $\epsilon_k = \epsilon_k \mp \alpha |\vec{g}|$, and the Fermi momentum is $k_F = \sqrt{2m\mu} / \alpha \rightarrow 0$.

Then we consider the superconducting pairing Hamiltonian with attractive interactions,

$$\mathcal{H}_{\text{int}} = \sum_{k,k',s_1,s_2} V_{k,k'} c_{k,s_1}^\dagger c_{k',s_2} c_{-k',s_2} c_{-k,s_1}$$

(2)

here $s_1, s_2$ are spin indexes. Applying the mean-field decompositions, we define the gap functions as $\Delta_{s_1, s_2}(k) = \sum_{k'} V_{k,k'} \langle c_{-k', s_2} c_{k', s_1} \rangle$. Here $\langle \cdots \rangle$ represents averaging over the thermal equilibrium states. After ignoring fluctuations, the mean-field pairing Hamiltonian becomes,

$$\mathcal{H}_\Delta = \sum_k \Delta_{s_1, s_2}(k) c_{k,s_1}^\dagger c_{-k,s_2} + \text{h.c.}$$

(3)

Due to the breaking of inversion symmetry, the even-parity pairing coexists with the odd-parity pairing [79]. The pairing potential is generally given by $\Delta(k) = \Delta_s \psi(k) + \Delta_t \vec{d}(k) \cdot \vec{\sigma} \sigma_g$, with $\psi(k) = \psi(-k)$ and the real spin-triplet $\vec{d}$-vector $\vec{d}(k) = -\vec{d}(-k)$ required by the Fermi statistic. And the pairing strengths $\Delta_{s,t} = |\Delta_{s,t}| \exp(i \theta_{s,t})$ are generally complex for the spin-singlet (triplet) pairing states. To break TRS, the mean-field pairing Hamiltonian becomes,

$$\mathcal{H}_{\text{int}} = \sum_{k,k'} V_{k,k'} c_{k,s_1}^\dagger c_{k',s_2} c_{-k',s_2} c_{-k,s_1} + \text{h.c.}$$

(2)

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$$\mathcal{H}_{\text{int}} = \sum_{k,k'} V_{k,k'} c_{k,s_1}^\dagger c_{k',s_2} c_{-k',s_2} c_{-k,s_1} + \text{h.c.}$$

(2)

(5)

$$\mathcal{F} = F_2 + F_4 + \gamma_1 (\Delta_s^* \Delta_t)^2 + (\gamma_2 \cdot \vec{M}) \Delta_s^* \Delta_t + \text{c.c.},$$

(5)

where $F_2 = \alpha_s |\Delta_s|^2 + \alpha_t |\Delta_t|^2 + \alpha_M |\vec{M}|^2$ and $F_4 = \beta_s |\Delta_s|^4 + \beta_t |\Delta_t|^4 + \beta_{st} |\Delta_s|^2 |\Delta_t|^2$. Here $\gamma_1 \neq 0$ indicates the pairing breaks TRS since the relative phase difference between singlet and triplet pairings is developed as $\pm \pi / 2$ [52]. The bilinear coupling term $\Delta_s^* \Delta_t$ pines the phase difference to an arbitrary non-zero value [82] in the low temperature. Hereafter, we consider both $\Delta_s$ and $\Delta_t$ belong to different representations of the lattice symmetry group thus the bilinear coupling is forbidden. Here $\alpha_M > 0$ indicates the lacking of intrinsic ferromagnetic ordering, and the $\gamma_2$-terms couple the SP $\vec{M}$ with the TRS-breaking unitary pairing $\Delta_s \pm i \Delta_t$, which satisfy both the global $U(1)$ symmetry ($\theta_{s,t} \rightarrow \theta_{s,t} + 2\pi$) and TRS. Once $\gamma_2 \neq 0$, $\vec{M}$ is spontaneously induced by unitary $\Delta_s \pm i \Delta_t$. By minimizing $\mathcal{F}$, we find

$$\vec{M} = -\frac{1}{\alpha_M} \text{Im}(\gamma_2 \Delta_s^* \Delta_t),$$

(6)

which will not alter the relative phase difference $\theta_{s,t}$ between $\Delta_s$ and $\Delta_t$ (see Sec. A in the Supplementary Materials [83]). It indicates that the unitary pairing states $\Delta_s \pm i \Delta_t$ coexist with the induced SP $\vec{M}$. The bulk magnetization vanishes in a purely clean system due to two reasons: the translational symmetry and the Meissner effect [84, 85]. Nevertheless, the boundary SP (see Fig. 1(b)) persists as well as the SP around impurities in

![FIG. 1. Illustrations of the p-wave pairings and the boundary SP. (a) The four p-wave pairings with in-plane $\vec{d}$ vectors, labeled by the black arrows. (b) The real space viewpoint of the boundary SP of a two-dimensional rectangular sample, marked by the red arrows. (c) The momentum space viewpoint of the $\vec{M}_{A_2}$-type SP that is parallel with $\vec{g} \times \vec{d}$, because of the product rule of the point group: $A_2 = A_2 \times A_1(B_1 \times B_2)$.](image)
the bulk, both of which can be detected by the μSR and the PKE.

Below we first use symmetry analysis to classify the \( \vec{\gamma}_2 \)-vector. The free energy in Eq. (5) preserves all the crystalline symmetries, which imposes strict constraints on the direction of the SP \( \vec{M} \). We take the \( C_{4v} \) point group as an example to identify the ferromagnetic-type SP induced by the TRS-breaking unitary pairing potential. The character table is shown in the Sec. B in the Supplementary Materials [83]. A full classification for the \( \vec{\gamma}_2 \)-terms by different symmetry groups is left for future work. The \( C_{4v} \) point group is generated by three independent symmetry operators \( (\sigma_x, \sigma_d \text{ and } R_{4z}) \): \( \sigma_x \) are the vertical reflection planes along \( x \) and \( y \); \( \sigma_d \) are the diagonal reflection planes along the \( x \pm y \) lines; \( R_{4z} \) is the four-fold rotation along \( z \) axis. To break both \( \sigma_d \) and \( \sigma_x \) simultaneously, only the \( z \)-component of \( \vec{\gamma}_2 \) is nonzero, namely, \( \vec{\gamma}_2 = (0, 0, \gamma_2^z) \), illustrated in Fig. (1(b)). It belongs to \( A_2 \) representation of \( C_{4v} \), shown in Table (S1) in the Supplementary Materials [83]. According to the product rule of the point group, we conclude that the SP could be induced by the interplay of the \( s \)-wave (\( d \)-wave) singlet pairing and the \( d_{xy} \) \( d_{xz, yz} \) \( s \)-wave \( \alpha \) \( \beta \)-triplet pairing, respectively.

\[
\begin{align*}
\hat{\Delta}_{s+i\sigma} & = (\Delta_s \Psi_s(k) + i \Delta_t \vec{d}_{A_2}(k) \cdot \vec{\sigma}) i \sigma_y, \\
\hat{\Delta}_{d+i\sigma} & = (\Delta_d \Psi_d(k) + i \Delta_t \vec{d}_{B_2}(k) \cdot \vec{\sigma}) i \sigma_y.
\end{align*}
\]  
(7)

Both the \( s+i\sigma \)- and \( d+i\sigma \)-pairing states are fully gapped in 2D SCs, while gap nodes can exist in 3D SCs.

Next, we investigate the important role of Rashba SOC for the establishment of the \( \vec{\gamma}_2 \)-term in Eq. (5). In this work, we study a single-band Hamiltonian with SOC in Eq. (1), and the results could also be generalized to multiband systems. The coupling coefficients of \( \vec{\gamma}_2 \) are calculated for the Hamiltonian \( \mathcal{H}_0 + \mathcal{H}_\Delta \),

\[
\gamma_2^i = \frac{1}{\beta} \sum_{k, \omega_n} \text{Tr} \left[ G_h(\vec{d} \cdot \vec{\sigma}) G_c \sigma_i G_c \sigma_0 \right],
\]  
(8)

with \( i = \{x, y, z\} \) and \( \beta = 1/(k_B T) \) the inverse of temperature. And the Matsubara Green’s function is \( G_c(k, \omega_n) = [\omega_n - \mathcal{H}_0(k)]^{-1} \) with \( \omega_n = (2n + 1)\pi/\beta \) and \( G_h(k, \omega_n) = G_c^*(-k, -\omega_n) \). We find that the direction of \( \vec{M} \) is perpendicular to both \( \vec{d} \) vector and the SOC \( \vec{g} \) vector, namely,

\[
\vec{\gamma}_2^z \propto i \sum_k \left\langle (\vec{g}(k) \times \vec{d}(k)) \cdot \psi_s(k) \right\rangle_{FS},
\]  
(9)

where \( \langle \cdots \rangle_{FS} \) denotes the average over the entire Fermi surfaces. Here we take the \( \Delta_s + i\Delta_t \) with \( s \)-wave pairing and \( \vec{d}_{A_2}(k) \) as an example [see Fig. 1(c)], where \( \vec{g} \times \vec{d}_{A_2} \approx \vec{c}^z_s(\sin^2 k_x + \sin^2 k_y) \). Moreover, we find the nonzero \( z \)-component of \( \vec{\gamma}_2^z \) to the leading order of \( \alpha k_F/k_B T_c \) as,

\[
\gamma_2^z = \frac{7\zeta(3)}{8\pi^3} \frac{\alpha k_F}{k_B T_c},
\]  
(10)

where \( \zeta(z) \) is the Riemann zeta function. The crucial role of SOC to the SP is manifest in Eq. (10), representing the key result of this work. Only when \( \alpha = 0 \), we can have \( \gamma_2^z \neq 0 \) as well as \( \vec{M}_z \neq 0 \). Therefore, SOC is indispensable to induce SP by a TRS-breaking unitary pairing \( \Delta_s + i\Delta_t \). Moreover, the sign of the SP is determined by \( \text{sign}(M_z) = \text{sign}(\alpha \Delta_s \Delta_t) \).

FIG. 2. The spontaneous SP induced by the \( d + i\sigma \) unitary pairing potential given by Eq. (7). The averaged SP on the boundary is calculated as a function of SOC strength for three cases, \( \Delta_t < \Delta_s \) (red circle), \( \Delta_s = \Delta_t \) (blue circle) and \( \Delta_t > \Delta_s \) (orange circle). Parameters: \( m_0 = 1, \mu = 1 \), and \( \Delta_t, \Delta_s = \{(1, 2), (1, 1), (2, 1)\} \) are used for the three cases.

To be more explicitly, we perform a numerical calculation for the averaged SP based on the solution of the Bogoliubov–de Gennes (BdG) Hamiltonian,

\[
\mathcal{H}_{BdG} = \begin{pmatrix} \mathcal{H}_0(k) & \mathcal{H}_\Delta \\ \mathcal{H}_\Delta^* & -\mathcal{H}_0^*(-k) \end{pmatrix},
\]  
(11)

where the Nambu basis \( \{c_{k, \uparrow}, c_{-k, \uparrow}, c_{-k, \downarrow}, c_{k, \downarrow}\}^T \) is used. Here \( \mathcal{H}_0(k) \) is given by Eq. (1) and \( \mathcal{H}_\Delta \) in Eq. (3). And the averaged boundary SP is defined as,

\[
\vec{M}_z = \frac{1}{N_t} \sum_{\vec{l}} \sum_{E_n} \left\langle E_n(\vec{l}) \right| \hat{P}_c \sigma_z(\vec{l}) \hat{P}_c \left| E_n(\vec{l}) \right\rangle_{BdG},
\]  
(12)

where \( \vec{l} \) is the “edge coordinate” on a \( N_x \times N_y \) rectangular lattice and \( \vec{l} \in \{(1, i_y), (i_x, N_y), (N_x, i_y), (i_x, 1)\} \) with total sites \( N_t = 2N_x + 2N_y - 4 \). \( \hat{P}_c \) is the projection operator into the particle subspace, and \( \left| E_n \right\rangle \) is solution of the BdG equation \( \mathcal{H}_{BdG} \left| E_n \right\rangle = E_n \left| E_n \right\rangle \).

Next, we take the \( d + i\sigma \) pairing states in Eq. (7) for an example. And the numerical result is shown in Fig. 2. It confirms that the averaged spontaneous SP \( \vec{M}_z = 0 \) corresponding to SOC strength \( \alpha = 0 \) and it increases as increasing the \( \alpha \), consistent with the analytical analysis.
als [83]). If $\Delta_s \ll \Delta_t$, triplet is dominated, the pairing function $\Delta_{s+ip} = \Delta_{d+ip} \sim \Delta_0 \sin \theta$ exhibits gap nodes. $\Delta_0$ being the superconducting gap at zero temperature whose value is also related to the magnitude of $\Delta_s$. Indeed, as shown by the fun 2 in Fig. 3(a), the calculated superfluid density $\rho_s$ shows a clear temperature dependence at low temperatures (see details of the calculations in Ref. 86).

While for $\Delta_s \sim \Delta_t$, as shown in Fig. 3(a), all fun 5 and fun 7 for $s + ip$ and fun 9 for $d + ip$ exhibit temperature-dependent superfluid density, similar to the case of fun 2. As for fun 3 ($\Delta_{s+ip} \sim \Delta_0 \sqrt{1 + \sin^2 \theta}$), the calculated $\rho_s$ is temperature-independent at low-$T$, an indication of fully-gapped SC, and shows remarkably good agreement with the experimental data. Therefore, the $s + ip$-pairing with $\Delta_s \sim \Delta_t$ might be applied to the noncentrosymmetric Zr$_3$Ir SC with weak SOC.

While for the strong SOC case (i.e., CaPtAs), the mixed pairings in Eq. (7) lead to 12 gap functions in total for the three different limits ($\Delta_s \ll \Delta_t$, $\Delta_s \sim \Delta_t$ and $\Delta_s \gg \Delta_t$) (see Sec. C in the Supplementary Materials [83]). Considering the presence of both broken TRS and superconducting gap nodes in CaPtAs, we exclude the pairing functions with $\Delta_s \gg \Delta_t$. In the case of $\Delta_s \sim \Delta_t$, for $s + ip$ pairing, all fun 3, fun 5, and fun 7 show a poor agreement with the experimental data (see Fig. S1 in Supplementary Materials [83]). Then, we focus on the gap functions with $\Delta_s \ll \Delta_t$. Both fun 2 (see Fig. 3(b) for $s + ip$) and fun 10 (see Fig. S1 for $d + ip$) exhibit a strong temperature-dependent superfluid density, and deviate significantly from the experimental data. However, for fun 8 ($\Delta_{s+ip} \sim \Delta_0 |\cos \phi \sin \phi \sin \theta|$), the calculated $\rho_s$ shows a good agreement with the experimental data over the entire temperature range. Furthermore, the $\rho_s$ calculated from fun 11 and fun 12 for $d + ip$ are also highly consistent with the experimental data, and the fitting result of fun 11 ($\Delta_{d+ip} \sim \Delta_0 |\cos \phi \sin \phi \sin \theta - \sin \theta + \sin \phi \cos \phi \sin \phi \cos \phi |$) is illustrated in Fig. 3(b). Therefore, both $s + ip$- and $d + ip$-pairings with $\Delta_s \ll \Delta_t$ might be applied to the noncentrosymmetric CaPtAs SC with strong SOC. By comparing with the experimental data, we demonstrate that our theory might be practicable to both nodal and nodeless superconductivity with broken TRS in the noncentrosymmetric SCs.

**Conclusion and discussion**. We briefly discuss the effects of the boundary SP on first-order (second-order) 2D topological SCs, which host topological Majorana edge (corner) states (see Sec. D in the Supplementary Materials [83]). A purely helical $p$-wave SC supports a pair of helical Majorana edge modes protected by TRS [87–89], which become fully gapped on each boundary for the $s + ip$ pairing states. Furthermore, a second-order topological SC is achieved [90–109] for the $d + ip$ case, which supports topological Majorana corner states (MCS). They are kind of Jackiw-Rebbi zero modes [110].

![FIG. 3. The superfluid density $\rho_s$ versus the reduced temperature $T/T_c$ for the noncentrosymmetric SCs.](image)

(a) Zr$_3$Ir with a weak SOC and (b) CaPtAs with a strong SOC. The experimental data determined by the transverse-field muon-spin rotation measurements were taken from Ref. 41 and 42. The solid lines represent the $\rho_s$ fitted by different pairing functions. For CaPtAs, it might be also fitted by pairing fun 5, 7, 10, and 12 (see Fig. S1 in the Supplementary Materials [83]).
sitting on each corner, protected by the combined $\sigma_4 T$ symmetry. Interestingly, we find that the boundary SP enlarges the edge gap to protect the MCSs.

In sum, we find that the TRS-breaking unitary pairing states could induce the spontaneous SP with the help of SOC, in the absence of external magnetic fields or Zeeman fields. We propose that both $s + ip$ and $d + ip$ spontaneously break TRS and give rise to SP, which is induced to be perpendicular to both the real spin-triplet $d$-vector and the SOC $g$-vector. The averaged boundary SP is also estimated $\sim 0.02$ meV for noncentrosymmetric SC, $\text{Zr}_3\text{Ir}$, which should be able to be detected in experiments. Moreover, our theory can quantitatively describe the superfluid density of $\text{Zr}_3\text{Ir}$ and $\text{CaPtAs}$ noncentrosymmetric SCs. Our result provides an alternative explanation to the TRS breaking, beyond the current understanding of such phenomena in the noncentrosymmetric superconductors. We also notice a recent theoretical work demonstrating that the pairing symmetry might be $d + ip$ for $\text{Sr}_2\text{RuO}_4$ [111]. Our theory may be also valid near an interface where spin-orbit coupling appears to explain the observations of broken TRS [14, 15].

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