$\pi/2$-Josephson junction as a topological superconductor

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Superconducting states with broken time-reversal symmetry are rarely found in nature. Here we predict that it is inevitable that the time-reversal symmetry is broken spontaneously in a superconducting Josephson junction formed by two superconductors with different pairing symmetries dubbed as $\pi/2$-Josephson junction. While the leading conventional Josephson coupling vanishes in such an $\pi/2$-Josephson junction, the second-order coupling from tunneling always generates chiral superconductivity orders with broken time-reversal symmetry. Josephson frequency in the $\pi/2$ junction is doubled, namely $\omega = 4eV/h$. The result can not only provide a way to engineer topologically trivial or nontrivial time-reversal breaking superconducting states, but also be used to determine the pairing symmetry of unconventional superconductors.

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I. INTRODUCTION

The van der Waals (vdW) Josephson junction [1], which is contacted by two close-by superconducting (SC) layers by vdW forces, has been realized in layered dichalcogenide superconductors recently [1,2]. This provides a platform to investigate the properties of two SC layers with different pairing symmetries forming in the junction. In general, the physics of the junction is controlled by the relative phase between the two SC order parameters, $\Delta\theta$. In a conventional Josephson junction which is formed by two s-wave SC layers, $\Delta\theta$ is typically zero in the absence of external or internal magnetic fields. It can be turned to nonzero by magnetic fields that break the time-reversal symmetry of the system explicitly. However, in the unconventional Josephson junction, $\Delta\theta$ can be nonzero in the ground state without external or internal magnetic fields [3–5]. A special case $\Delta\theta = \pm\pi/2$, which breaks time-reversal symmetry, is called chiral SC in the literature [6].

Superconductors with spontaneously time-reversal symmetry breaking (TRB) pairing states [7–15] have been widely sought. The most intriguing property of a TRB SC is the nontrivial topology, namely, a TRB SC can be a topological superconductor (TSC) [16–19], e.g., topological $p + ip$ [20–22] and $d + id$ [23–28] superconductors. The former $p + ip$ TSC can be realized in many spin-orbital coupling systems [22,29–37], while the latter $d + id$ TSC has only been proposed in honeycomb lattice systems [38], such as doped graphene [24,28,39–41], single $TiSe_2$ layer [42], and bilayer silicene [25]. Although the $d + id$ TSC exhibits many interesting phenomena, such as quantized boundary current [23,28], spontaneous magnetization [23,43], quantized spin and thermal Hall conductance [28,43], and geometric effects [44], there is no strong experimental evidence to support the presence of this chiral SC state.

Here, we ask whether a TRB SC can be spontaneously formed in a vdW Josephson junction. We show that the TRB takes place spontaneously in this Josephson junction formed by two SC layers with different pairing symmetries as illustrated in Fig. 1. For example, a $d + id$ TRB SC can be engineered in a junction with a $d_{x^2-y^2}$ SC layer close to a d_{xy} one. Furthermore, we prove that this $d + id$ SC constructed in this way is also a TSC. The junction has a distinct Josephson frequency, $\omega = 4eV/h$, which is twice the conventional Josephson frequency $2eV/h$. We discuss possible experimental realizations for this type of junction. The results can not only help to realize different SC states and design SC qubit devices [45,46], but also be used to determine the pairing symmetry of an unknown SC by the unique feature of the Josephson frequency.

Before we discuss specific models, we first present a general argument. Considering a general Bogoliubov–de Gennes (BdG) Hamiltonian of two SC layers connected through tunneling and expanding the free energy up to the fourth order of the tunneling, the free energy can be generally written as [3–5,47]

$$\mathcal{F} = \mathcal{F}_0 - J\cos\Delta\theta + g\cos^2\Delta\theta, \tag{1}$$

where the first term is the relative phase independent term, the second term is the conventional Josephson coupling term, and the last term can lead to spontaneous TRB. In a conventional Josephson junction, J is positive and much larger than g so that the third term can be ignored. Here the main finding is when two SC layers in a vdW Josephson junction have different pairing symmetries, J vanishes and g becomes the leading coupling from tunneling. Remarkably, g is always positive [47]. Thus, to minimize the free energy, $\Delta\theta = \pm\pi/2$, which breaks TRS spontaneously. Such a vdW Josephson junction is called $\pi/2$-Josephson junction in this paper.

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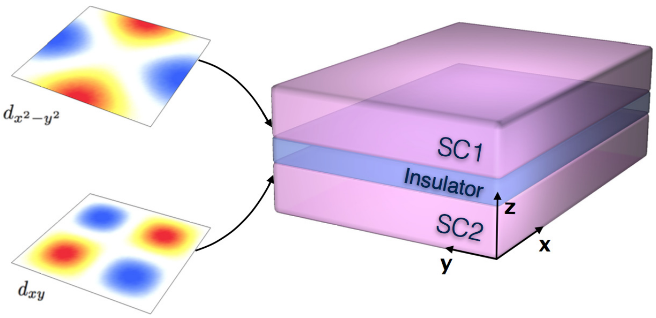

FIG. 1. The sketch of a vdW Josephson junction with two SC layers having two different pairing symmetries, e.g., $d_{x^2-y^2}$ and d_{xy}.

II. MODEL

More specifically, the above analysis can be modeled by a two-band superconductor in which the two bands have different SC orders $\Delta_\alpha(\mathbf{k})e^{i\theta_\alpha}$ and $\Delta_\beta(\mathbf{k})e^{i\theta_\beta}$ where θ_α and θ_β are the SC phases. Their relative phase, $\Delta\theta = \theta_\beta - \theta_\alpha$, is a physical quantity when the tunneling between two bands is induced. The general BdG Hamiltonian can be written as

$$H = \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger \mathcal{H}(\mathbf{k}) \Psi_{\mathbf{k}}, \tag{2}$$

where $\Psi_{\mathbf{k}} = (d_{\mathbf{k},\alpha,\uparrow}, d^\dagger_{-\mathbf{k},\alpha,\downarrow}, d_{\mathbf{k},\beta,\uparrow}, d^\dagger_{-\mathbf{k},\beta,\downarrow})^T$ and $\mathcal{H}(\mathbf{k})$ is defined as

$$\mathcal{H}(\mathbf{k}) = \begin{pmatrix} \epsilon_\alpha(\mathbf{k}) & \Delta_\alpha(\mathbf{k})e^{i\theta_\alpha} & t(\mathbf{k}) & 0 \\ \Delta_\alpha(\mathbf{k})e^{-i\theta_\alpha} & -\epsilon_\alpha(\mathbf{k}) & 0 & -t(\mathbf{k}) \\ t(\mathbf{k}) & 0 & \epsilon_\beta(\mathbf{k}) & \Delta_\beta(\mathbf{k})e^{i\theta_\beta} \\ 0 & -t(\mathbf{k}) & \Delta_\beta(\mathbf{k})e^{-i\theta_\beta} & -\epsilon_\beta(\mathbf{k}) \end{pmatrix}. \tag{3}$$

The tunneling $t(\mathbf{k})$ is taken to be real, and the eigenvalues of this Hamiltonian for each $\mathbf{k}$ are

$$E^{\pm\pm} = \pm\frac{1}{\sqrt{2}}\sqrt{\epsilon_\alpha^2 + \epsilon_\beta^2 + \Delta_\alpha^2 + \Delta_\beta^2 + 2t^2 \pm \sqrt{\left(\epsilon_\alpha^2 - \epsilon_\beta^2 + \Delta_\alpha^2 - \Delta_\beta^2\right)^2 + 4t^2[(\epsilon_\alpha + \epsilon_\beta)^2 + |\Delta_\alpha e^{i\theta_\alpha} - \Delta_\beta e^{i\theta_\beta}|^2]}}. \tag{4}$$

At zero temperature, the free energy is $\mathcal{F} = \sum_{\mathbf{k}}[E^{-+}(\mathbf{k}) + E^{--}(\mathbf{k})]$. We can expand the free energy up to the fourth order of $t(\mathbf{k})$. The free energy is given by Eq. (1), in which the parameters can be specified as

$$J = \sum_{\mathbf{k}}[t^2(\mathbf{k})g_1(\mathbf{k}) + t^4(\mathbf{k})g_3(\mathbf{k})]\Delta_\alpha(\mathbf{k})\Delta_\beta(\mathbf{k}), \tag{5}$$

$$g = \sum_{\mathbf{k}} t^4(\mathbf{k})g_2(\mathbf{k})\Delta_\alpha^2(\mathbf{k})\Delta_\beta^2(\mathbf{k}). \tag{6}$$

The explicit form of $g_i(\mathbf{k})$ is shown in the Supplemental Material [47]. While the functions of $g_i(\mathbf{k})$ are very lengthy, we can analyze their symmetry characters. For convenience, we consider a square lattice symmetry classified by the C_{4v} point group. One can notice that all the parameter functions except $\Delta_i(\mathbf{k})$ belong to the A_1 irreducible representation of C_{4v}. Thus, if $\Delta_\alpha(\mathbf{k})$ and $\Delta_\beta(\mathbf{k})$ belong to different irreducible representations, namely, they have different pairing symmetries, the conventional Josephson coupling J vanishes because of the symmetry constraint. Therefore, the ground state is determined by the sign of g, that is if $g > 0$, $\Delta\theta = \pm\pi/2$ and if $g < 0$, $\Delta\theta = 0, \pm\pi$. In the Supplemental Material [47], we have shown the positive natural of $g_2(\mathbf{k})$ for all $\mathbf{k}$ in the Brillouin zone (BZ). Thus the relative phase in the ground state is always $\pm\pi/2$. There also exists a physical reason for the positive natural of g, that is the superconductivity favors a larger SC gap in the BZ. If the two pairing symmetries are different, without breaking time-reversal symmetry, the interference through the proximity effect between the two superconductors are destructive, while with breaking the time-reversal symmetry, the proximity effect can always enhance the SC gaps to save more energy in the SC state. This can be checked by comparing the two functions $|\Delta_\alpha(\mathbf{k}) \pm \Delta_\beta(\mathbf{k})|$ and $|\Delta_\alpha(\mathbf{k}) \pm i\Delta_\beta(\mathbf{k})|$. The latter has always larger gaps in the entire BZ than the former in the case of the two SC layers having different pairing symmetries.

A. Mean-field calculation

The above results can be further examined in a specific model. We consider two layered superconductors to obtain the TRB $d + id$ order. The d-wave SC state develops naturally if the SC pairing is driven by local antiferromagnetic fluctuations [48,49]. Theoretically, we can use the t-J model to model the d-wave SC state. Following the well-known result, we consider the following junction as illustrated in Fig. 2(a). The pairing interactions on each layer are attributed to antiferromagnetic exchange interactions J_α. We only consider the nearest-neighbor (NN) exchange J_1, and the next-nearest-neighbor (NNN) J_2 in the top and bottom layers respectively. Including the tunneling coupling between the two layers, the overall Hamiltonian can be written as $H = H_{J_1} + H_{J_2} + H_t$, where

$$H_{J_\alpha} = \sum_{\mathbf{k},\sigma} \xi_\alpha(\mathbf{k}) d^{\alpha\dagger}_{\mathbf{k},\sigma} d^\alpha_{\mathbf{k},\sigma} + \sum_{i,\delta=\hat{x},\hat{y}} J_\alpha\left(\vec{S}^\alpha_i \cdot \vec{S}^\alpha_{i+\delta} - \frac{1}{4}n^\alpha_i n^\alpha_{i+\delta}\right),$$

$$H_t = t\sum_{\alpha,\mathbf{k},\sigma} d^{1\dagger}_{\mathbf{k},\sigma} d^2_{\mathbf{k},\sigma} + \text{H.c.} \tag{7}$$

Here the tunneling term t is chosen to be real and independent of $\mathbf{k}$ for simplicity. We also drop the double occupancy projection operators which is required in the standard t-J model because the double occupancy projection in the mean-field level can be treated as an overall renormalization factor to the band dispersion [50,51]. Therefore, it does not affect the qualitative result. The sketch of this model is shown in Fig. 2(a).

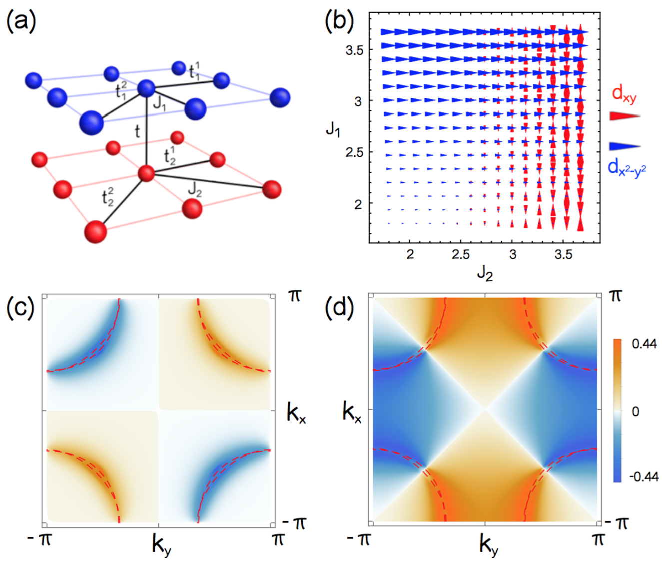

FIG. 2. The sketch of the two-layer model and calculated SC orders from the mean-field calculation: (a) two layer t-J models with the top layer J_1 and bottom layer J_2; (b) superconducting orders Δ_{xy} (red arrow) and $\Delta_{x^2-y^2}$ (blue arrow) as function of exchange strength $J_{1,2}$ with other parameters given in the main text. The magnitudes and phases of pairing orders are represented by the lengths and directions of the arrows; panels (c) and (d) show the real and imaginary parts of $d_{x^2-y^2}+id_{xy}$ order in the first layer with $J_1=3.4$ and $J_2=3$. The Fermi surfaces of the two-layer Hamiltonian are represented by the red dashed lines, which coincide with the arcs in BZ.

In the mean-field solution, we can compare the energies of the s-wave and d-wave SC states. The self-consistent mean-field solutions for the d-wave SC states are given by [47]

$$\frac{\Delta_{x^2-y^2}(\mathbf{k})}{\cos k_x-\cos k_y}=\sum_{\mathbf{k}'}-\frac{2J_1}{N}(\cos k_x'\pm\cos k_y')\langle d^1_{-\mathbf{k}',\downarrow}d^1_{\mathbf{k}',\uparrow}\rangle, \tag{8}$$

$$\frac{\Delta_{xy}(\mathbf{k})}{\sin k_x\sin k_y}=\sum_{\mathbf{k}'}-\frac{8J_2}{N}\sin k_x'\sin k_y'\langle d^2_{-\mathbf{k}',\downarrow}d^2_{\mathbf{k}',\uparrow}\rangle. \tag{9}$$

We take the band dispersion

$$\xi_\alpha=-2t^1_\alpha(\cos k_x+\cos k_y)-4t^2_\alpha\cos k_x\cos k_y+\mu_\alpha-\mu, \tag{10}$$

where $t^{1(2)}$ indicates the NN (NNN) hopping and $\alpha=1,2$, corresponding to the top and bottom layers. μ_α is the corresponding on-site energy in each layer and μ is the chemical potential. Without the tunneling, in the mean-field solution shown in the Supplemental Material [47], we find that the $d_{x^2-y^2}$-wave and d_{xy}-wave orders are favored on the top and bottom layers respectively when the parameters are set as $t^1_1=0.88$, $t^2_1=-0.35$, $t^1_2=1.67$, $t^2_2=-0.33$, $\mu_1=-0.4$, $\mu_2=-1.2$, and $\mu=0$.

Turning on the layer tunneling and taking $t=0.4$, the phase diagram is plotted in Fig. 2(b) as a function of $J_{1,2}$. The lengths of the vectors in Fig. 2(b) represent the strength of the orders and the directions relate to the phases. As J_1 and J_2 increase, both d-wave orders become stronger and the relative phase maintains to be $\pm\pi/2$. The imaginary and real parts of the order parameter in the first layer are shown in Figs. 2(c) and 2(d). Clearly, the real part has $d_{x^2-y^2}$ symmetry and the imaginary part has d_{xy} symmetry. The d_{xy} order in the first layer is induced through the proximity effect from the second layer. This result is consistent with our previous analysis.

B. Topological analysis

Now we discuss the topological properties of the above spontaneous TRB SCs. The state in the $\pi/2$ junction can be topologically nontrivial for a $d\pm id$ state but trivial for an $s\pm id$ state. To show this, we can analyze the symmetry property of the Berry curvature. Starting from the Hamiltonian (3) with $\theta_\alpha=0$ and $\theta_\beta=\pi/2$, the band dispersions $\epsilon_\alpha(\mathbf{k})$ and $\epsilon_\beta(\mathbf{k})$ both belong to the A_1 irreducible representation (IR) of C_{4v}. We consider the σ_v symmetry operation which maps $\mathbf{k}=(k_x,k_y)\to\tilde{\mathbf{k}}=(-k_x,k_y)$ or $(k_x,-k_y)$. In the $s\pm id$ state, if the tunneling term $t(\mathbf{k})$ belongs to the A_1 or B_1 IR, the Hamiltonian is invariant under σ_v operation. If the tunneling term $t(\mathbf{k})$ belongs to the B_2 IR, under a σ_v operation, the Hamiltonian becomes $\tilde{\mathcal{H}}(\tilde{\mathbf{k}})$, which can be expressed as $\tilde{\mathcal{H}}(\tilde{\mathbf{k}})=\tau_z\mathcal{H}(\mathbf{k})\tau_z$. The corresponding eigenstate becomes $|\tilde{u}_n(\tilde{\mathbf{k}})\rangle=\tau_z|u_n(\mathbf{k})\rangle$. In both cases, considering the definition of Berry curvature,

$$B(\mathbf{k})=i\sum_{n\in occ}\epsilon_{k_xk_y}\langle\partial_{k_x}u_n(\mathbf{k})|\partial_{k_y}u_n(\mathbf{k})\rangle, \tag{11}$$

one can find that the Berry curvature changes sign under σ_v operation so that the total Chern number is zero. But for the $d\pm id'$ state, the Berry curvature is invariant under C_4, σ_v, and σ_d operation. This means that the nonzero Chern number is not forbidden by any symmetry operations. Thus, under some suitable parameters, the system can be a topological $d\pm id$ SC. This is clearly shown in Fig. 3.

The above conclusion can be numerically verified. We perform numerical calculation for the model in Eq. (7). We calculate the topologically protected edge states in a stripe lattice as shown in Fig. 4. The parameters in the calculation are set to be $J_1=3.4$, $J_2=3$ in Eq. (7), and the corresponding mean-field SC orders strength are $\Delta_{x^2-y^2}=0.8183$, $\Delta_{xy}=0.5562i$, and $\Delta_{x^2+y^2}=\Delta_{x^2y^2}=0$. Under these parameters, there are four chiral modes on each edge, which correspond to a Chern number equal to -4, as shown in Fig. 4.

C. Experimental signatures

There are three smoking-gun signatures for the above topological $d+id$ $\pi/2$-Josephson junction. The first one is the topologically protected edge state as shown in Fig. 4. On the edge, one can use superconducting quantum interference microscopies to detect spontaneously generated supercurrents [23,28,52]. The second one is that the Josephson frequency doubles the conventional one. For a conventional Josephson junction, the Josephson frequency is given by $\omega_0=2eV_0/h$, where V_0 is the applied external voltage on the junction. For the $\pi/2$ junction, the modified Josephson equations are

$$I=I_0\sin 2\Delta\theta,$$
$$\frac{d\Delta\theta}{dt}=-\frac{2e}{\hbar}V_0. \tag{12}$$

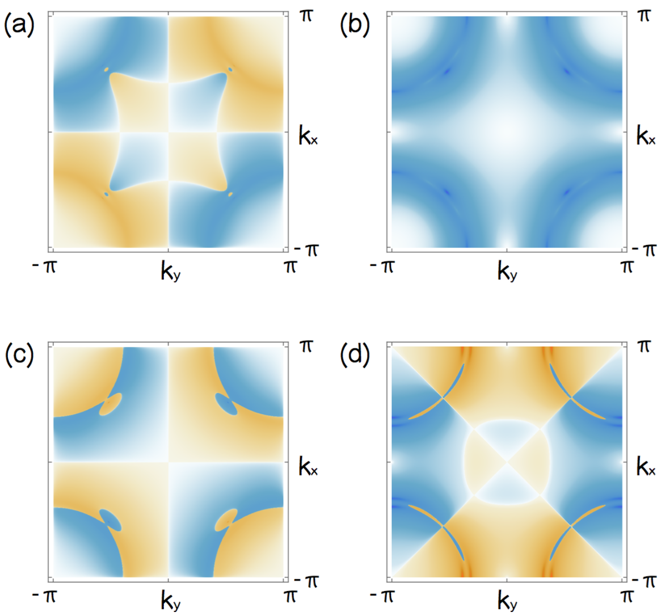

FIG. 3. The Berry curvature for different TRB orders in the Hamiltonian (3) with parameters $t_1^1 = 0.88$, $t_1^2 = -0.35$, $t_2^1 = 1.67$, $t_2^2 = -0.33$, $\mu_1 = -0.4$, $\mu_2 = -1.2$, $\mu = 0$, $t = 0.4$, $\Delta_\alpha = 0.8183$, $\Delta_\beta = 0.5562$, $\theta_\alpha = 0$, and $\theta_\beta = \pi/2$. Panels (a)–(d) represent $d + is'$, $d + id'$, $d + is$, and $d' + is$ orders, respectively. According to our analysis, the Berry field of $d \pm is(s')$ belongs to B_2 IR, $d' \pm is(s')$ belongs to B_1 IR, and only $d + id'$ belongs to A_1 IR. Only $d \pm id'$ in (b) shows a nonzero Chern number.

The ac Josephson current is $I = I_0 \sin(2\Delta\theta_0 - 4eV_0t/\hbar)$. The corresponding Josephson frequency is $\omega_i = 2\omega_0 = 4eV_0/h$, which is twice the ordinary Josephson frequency.

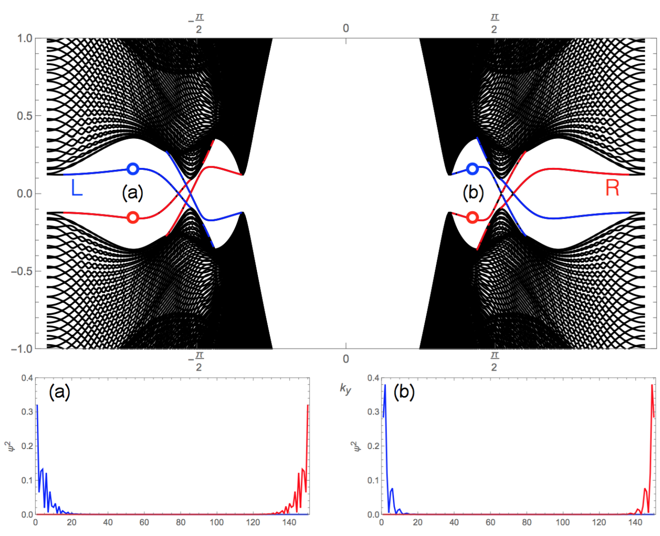

FIG. 4. Edge states of $d + id'$ superconductivity in the Hamiltonian (6); the parameter we choose is $J_1 = 3.4$, $J_2 = 3$, and the corresponding self-consistent mean-field SC orders are $\Delta_{x^2-y^2} = 0.8183$, $\Delta_{xy} = 0.5562i$. On the top panel is the band structure with left edge state blue and right red. The bottom panel shows the corresponding distribution of edge states.

The third experimental signature is the magnetic field dependence of the critical current. When a magnetic field B is applied to a conventional Josephson junction with length L and penetration depth W, the critical current is

$$I_c = j_0 \left| \frac{\sin(\pi\Phi/\Phi_0)}{\pi\Phi/\Phi_0} \right|, \tag{13}$$

where $\Phi_0 = h/2e$, is the flux quantum and $\Phi = BWL$. In the $\pi/2$-Josephson junction, it is easy to show that

$$I_c = j_0 \left| \frac{\sin(2\pi\Phi/\Phi_0)}{2\pi\Phi/\Phi_0} \right|. \tag{14}$$

The oscillation pattern is changed. Notice the last two experimental signatures are valid for all $\pi/2$-Josephson junctions.

D. Engineering TSC

Previously, the TSCs have already been proposed in p-wave superconductors [21], TI surface states [29–31], and semiconductor nanowires [22,32–37]. These proposals all focus on the 0d Majorana bound states, not the 1d Majorana chiral edge states. Recently, the chiral Majorana modes have been observed in the quantum anomalous Hall insulator-superconductor structure [53]. Compared to the previous work, the major advantage here is that TSC and the corresponding chiral edge state can be realized with conventional d-wave superconductors, such as cuprates, and no external magnetic field [54] or topological nontrivial band structures are needed. Thus, in principle, our method allows TSC to operate at very high temperature because of the high SC transition temperature of cuprates.

Recently, the advances in vdW heterostructure technology provide an effective way to engineer the rotation angle between the two SC layers with a high accuracy [55,56]. Furthermore, the vdW junction is defect-free contacted and has a strong proximity coupling [1,2], which renders a larger value of g in Eq. (1). Thus an explicit design of a TSC $\pi/2$ junction is to align two identical d-wave superconductors along the z direction with a relative $\pi/4$ in-plane angle [47]. This design can be implemented by recent rapid technological progress in engineering heterostructures.

E. Engineering TRB state

The result also allows us to engineer exotic SC states with TRB, e.g., a $s + id$ pairing state. Superconductors with this type of pairing state have been widely studied. However, success has been very limited. So far, spontaneous TRB in SC states have been rarely observed.

The above physics can also be potentially realized in bulk materials. For example, it has been theoretically suggested that the FeAs layer, the building block in iron-based superconductors, and the CuO_2 layer, the building block in cuprates, can be hybridized to form a hybrid crystal [57]. Following our results, in such a hybrid crystal, the time-reversal symmetry must be broken as FeAs [51,58] and CuO_2 [48,49] are known to favor s-wave and d-wave pairing symmetries respectively. The superconducting state in such a material must be $s \pm id$.

III. APPLICATIONS

The $\pi/2$-Josephson junction can be used to determine the pairing symmetry of an unknown superconductor. This is based on the fact that the Josephson frequency will be doubled if two SC layers have different pairing symmetries. The $\pi/2$-Josephson junction can also be used to make a quantum qubit because the free energy has two minima. It becomes a natural two-level system to form a qubit. Other excited states have much higher energy so that the two-level system is well protected.

In summary, we have shown that the $\pi/2$-Josephson junction is an inevitable result when a Josephson junction is formed by two superconductors with different pairing symmetries. The Josephson frequency is doubled. A TSC with a $d \pm id$ pairing symmetry can be achieved in this way. The result also provides a method to design a TRB $s + id$ superconductor.

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