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Axionic superconductivity in three-dimensional doped narrow-gap semiconductors

Pallab Goswami
National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32306, USA

Bitan Roy
National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32306, USA
and Condensed Matter Theory Center, Department of Physics, University of Maryland, College Park, Maryland 20742, USA

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We consider the competition between the conventional s-wave and the triplet Balian-Werthamer or the B-phase pairings in doped three-dimensional narrow-gap semiconductors, such as $Cu_xBi_2Se_3$ and $Sn_{1-x}In_xTe$. When the coupling constants of the two contending channels are comparable, we find a simultaneously time-reversal and parity violating $p+is$ state at low temperatures, which provides an example of a dynamic axionic state of matter. In contradistinction to the time-reversal invariant, topological B phase, the $p+is$ state possesses gapped Majorana fermions as surface Andreev bound states, which give rise to an anomalous surface thermal Hall effect. The anomalous gravitational and electrodynamic responses of the $p+is$ state can be described by the θ vacuum structure, where $\theta \neq 0$ or π.

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The notion of θ vacuum is a venerable concept of modern quantum field theory, which has profound implications for the vacuum structure of gauge theories [1]. Physically, the θ term for the gauge theory is a *pseudoscalar magnetoelectric coefficient*, and an arbitrary θ violates the time-reversal ($\mathcal{T}$) and parity ($\mathcal{P}$) symmetries. In order to preserve $\mathcal{P}$ and $\mathcal{T}$ symmetries in the strong interaction regime and also to account for the violation of these fundamental discrete symmetries in the lower energy scales, the existence of a very light pseudoscalar boson, dubbed *axion*, was postulated almost three decades ago [2–4]. Thus far the axion has eluded experimental detection.

However, the interest in the θ vacuum and its tangible experimental consequences has been revived due to the discovery of time-reversal symmetric (TRS), three-dimensional Z_2 topological insulators [5–7]. It has been recognized that the topological invariant of the Z_2 topological insulator couples to the electromagnetic gauge field as a θ term, and as a consequence of the $\mathcal{T}$ symmetry and the nondegeneracy of the underlying ground state, the magnetoelectric coefficient θ is quantized to be π [5]. When the criterion of the nondegenerate ground state is relaxed while maintaining the $\mathcal{T}$ symmetry, the θ can acquire fractional values, which reflect the degeneracy of the ground state on a torus [8,9]. Similar θ vacuum structures have also been found for the spin gauge field [10] and the gravitational field [11,12], respectively, for the TRS topological superconductors (TSCs) in the classes CI and DIII [13].

The Balian-Werthamer or the B phase of superfluid ^{3}He [13] and the pseudoscalar pairing of the four component charged Dirac fermions [14] are experimentally pertinent examples of TSCs in the class DIII. Upon projecting onto the low energy quasiparticles in the vicinity of the Fermi surface, the pseudoscalar pairing also maps onto the B phase [15]. Following the suggestion of Ref. [14], that the DIII TSC may be realized in doped three-dimensional, strongly spin-orbit coupled narrow-gap semiconductors, there has been considerable experimental interest in the superconducting $Cu_xBi_2Se_3$ and $Sn_{1-x}In_xTe$ [16–25].

A natural question arises whether there is a condensed matter realization of $\mathcal{P}$ and $\mathcal{T}$ breaking dynamic axionic state of matter [26]. Recently, the possibility of such a phase has been proposed for some magnetic insulators [27–29], which is yet to be experimentally found. In this Rapid Communication, we demonstrate that a spontaneously $\mathcal{P}$ and $\mathcal{T}$ breaking *axionic superconducting* state can be realized in doped three-dimensional narrow-gap semiconductors. This axionic paired state has $p+is$ pairing symmetry, and emerges due to the competition between the conventional s-wave and the triplet B-phase pairings, and lacks any analog in the superfluid ^{3}He [30]. According to the Altland-Zirnbauer classification scheme, the $p+is$ state is a member of the class D [31]. Both the electromagnetic gauge field and the axion fields are massive inside this phase. As a consequence of the broken $\mathcal{T}$ symmetry, the $p+is$ state possesses gapped Majorana fermions as surface Andreev bound states (SABS), and supports an anomalous surface thermal Hall (STH) conductivity.

The low energy quasiparticle dispersion in many narrow-gap semiconductors is succinctly captured by a massive Dirac equation, which describes the Kramer's degenerate quasiparticles in the conduction and valence bands. In the presence of strong spin-orbit coupling, electron-phonon scattering can lead to attractive interactions in both the singlet and triplet channels [14]. As these materials are weakly correlated, perhaps it is not a strong assumption that the retarded pairing interaction will generically emerge in the vicinity of the Fermi surface. Therefore, the completely filled (or empty) bands can be safely integrated out for determining the low energy pairing physics. We consider a situation when the Fermi level lies in the conduction band, and begin with the following interacting Hamiltonian,

$$H_{\text{qp}} = \sum_{\mathbf{k},s} \xi_k\, c^*_{\mathbf{k},s} c_{\mathbf{k},s} + \sum_{\mathbf{k}} V(\mathbf{k}) \hat{n}_{\mathbf{k}}\, \hat{n}_{-\mathbf{k}}. \tag{1}$$

In the above equation the number operator $\hat{n}_{\mathbf{k}} = \sum_{\mathbf{q},s} c^*_{\mathbf{q}+\mathbf{k},s} c_{\mathbf{q},s}$, and $c^*_{\mathbf{k},s}$, $c_{\mathbf{k},s}$ are, respectively, the creation and annihilation operators of the quasiparticles with momentum $\mathbf{k}$. The index s represents the Kramer's pair, and $\xi_{\mathbf{k}} = \sqrt{v^2k^2+\Delta_g^2} - \mu$ describes the quasiparticle energy with respect to the Fermi level μ. The band gap is denoted by Δ_g and the band parameter v has the dimension of velocity. When $|\mu - \Delta_g| \ll \Delta_g$, a nonrelativistic approximation $\xi_{\mathbf{k}} \approx k^2/(2m) - \tilde{\mu}$ can be applied, where $m = \Delta_g$ and $\tilde{\mu} = \mu - \Delta_g$. If we choose a simplified interaction potential $V(\mathbf{k})$, which is a sum of the attractive interactions in the s- and p-wave channels, the pertinent reduced BCS Hamiltonian for the mean-field description becomes $H = \frac{1}{2}\sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger \hat{H}_{\mathbf{k}} \Psi_{\mathbf{k}}$. The four component Nambu spinor $\Psi_{\mathbf{k}}^\dagger = (c^*_{\mathbf{k},\uparrow}, c^*_{\mathbf{k},\downarrow}, c_{-\mathbf{k},\downarrow}, -c_{-\mathbf{k},\uparrow})$, and the operator

$$\hat{H}_{\mathbf{k}} = \begin{pmatrix} \xi_k\, \sigma_0 & \Delta_s\, \sigma_0 + \Delta_t\, \mathbf{d}_{\mathbf{k}}\cdot\boldsymbol{\sigma} \\ \Delta_s^*\, \sigma_0 + \Delta_t\, \mathbf{d}^*_{\mathbf{k}}\cdot\boldsymbol{\sigma} & -\xi_k\, \sigma_0 \end{pmatrix}, \quad (2)$$

where σ_0 and $\boldsymbol{\sigma}$ respectively denote the identity and the conventional Pauli matrices operating on the Kramer's indices. We have introduced the complex s-wave pairing amplitude Δ_s, and a triplet amplitude Δ_t. In a weak coupling approach, the time-reversal symmetric (TRS), fully gapped B phase is energetically most favorable in the triplet channel, and is characterized by $\mathbf{d}_{\mathbf{k}} = \mathbf{k}/k_F$, where k_F is the Fermi momentum.

When we focus on the competition between the s-wave and the B-phase pairings, the associated reduced BCS Hamiltonian is described by

$$\hat{H}_{\mathbf{k}} = \xi_{\mathbf{k}}\gamma_0 + \frac{\Delta_t}{k_F}\gamma_0\gamma_j k_j + \mathrm{Re}(\Delta_s)\gamma_5 + i\,\mathrm{Im}(\Delta_s)\gamma_0\gamma_5, \quad (3)$$

where $\gamma_0 = \sigma_0 \otimes \tau_3$, $\gamma_j = i\sigma_j \otimes \tau_2$, $\gamma_5 = \sigma_0 \otimes \tau_1$, and τ_j are the Pauli matrices operating in the Nambu space. A pristine s-wave phase only breaks $U(1)$ gauge symmetry leaving all the discrete symmetries intact, and is topologically trivial. In the absence of any s-wave pairing, the Hamiltonian for the B phase takes the form of a *band inverted, massive Dirac equation in three dimensions*. Due to the presence of $\mathcal{T}$ and the broken spin rotational symmetry, this phase belongs to the class DIII, and is characterized by an integer topological invariant

$$\mathcal{N} = \tfrac{1}{2}[1 + \mathrm{sgn}(m\tilde{\mu})]\,\mathrm{sgn}(\Delta_t), \quad (4)$$

which for weak or BCS pairing $[\mathrm{sgn}(m\tilde{\mu}) = 1]$ reduces to $\mathcal{N} = \mathrm{sgn}(\Delta_t)$. On the other hand, for strong pairing or BEC limit $[\mathrm{sgn}(m\tilde{\mu}) = -1]$, and $\mathcal{N} = 0$. As a consequence of the $\mathcal{T}$ symmetry, the Hamiltonian of the B phase involves only four mutually anticommuting Dirac γ matrices, and the gravitational response of the B phase is characterized by an *axion angle* $\theta^0_{\mathrm{ax}} = \pi$ [11,12]. It is important to note that the B-phase pairing breaks the inversion symmetry of the normal state. However, due to the spin-orbital locking in the B phase, there is an emergent parity symmetry $\mathcal{P}$ defined by $\mathbf{k} \to -\mathbf{k}$ and $\Psi_{\mathbf{k}} \to \gamma_0\Psi_{\mathbf{k}}$. When both pairings coexist, the imaginary part of the s-wave amplitude appears as a *pseudoscalar mass*, with the fifth anticommuting γ matrix, and enhances the gap on the Fermi surface. On the other hand, the real part of the s-wave pairing commutes with the B-phase operator, and only anticommutes with the kinetic energy. Therefore, the real part appears as an *axial chemical potential* [32]. In the coexisting

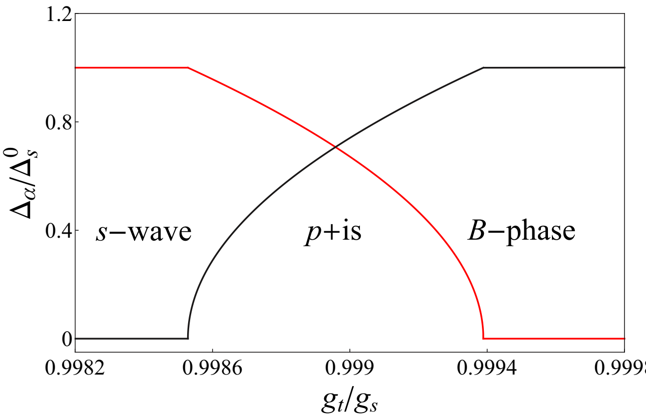

FIG. 1. (Color online) A cut of the zero temperature phase diagram for $g_s\rho(\mu) = 1$, showing the normalized pairing amplitudes, as a function of the ratio g_t/g_s, where $\rho(\mu)$ is the density of states at the Fermi level. The amplitudes are normalized by Δ_s^0, the s-wave amplitude, when the triplet channel is turned off. The s-wave and the B-phase amplitudes are respectively shown as the red and the black lines.

phase, both $\mathrm{Re}(\Delta_s)$ and $\mathrm{Im}(\Delta_s)$ break the $\mathcal{P}$ symmetry, and only the pseudoscalar mass simultaneously breaks $\mathcal{P}$ and $\mathcal{T}$.

The emergence of the $p + is$ state can be justified in the following manner. The quasiparticle spectra corresponding to $\hat{H}_{\mathbf{k}}$ in Eq. (3) are given by $E_{\mathbf{k}} = \pm E_{\alpha,\mathbf{k}}$, and

$$E_{\alpha,\mathbf{k}} = \sqrt{\xi_k^2 + |\Delta_s|^2 + \Delta_t^2\frac{k^2}{k_F^2} + 2\alpha\Delta_t\frac{k}{k_F}\,\mathrm{Re}(\Delta_s)}, \quad (5)$$

where $\alpha = \pm 1$. From the above dispersion relations it becomes clear that the gap at the Fermi surface is maximized, when Δ_s is purely imaginary, and leads to an axion angle $\theta^0_{\mathrm{ax}} = \pi + \tan^{-1}\mathrm{Im}(\Delta_s)/\mu$, for the gravitational response. Due to the simultaneous violation of $\mathcal{T}$ and the spin rotational symmetries, the $p + is$ state belongs to the class D [13,31].

The stabilization of the $p + is$ phase can be further substantiated via a minimization of the free energy,

$$f_s = \frac{|\Delta_s|^2}{2g_s} + \frac{|\Delta_t|^2}{2g_t} - 2T\sum_{\alpha,\mathbf{k}}\log\left[2\cosh\frac{E_{\alpha,\mathbf{k}}}{2T}\right] + \sum_{\mathbf{k}}\xi_{\mathbf{k}}, \quad (6)$$

where g_s,g_t are, respectively, the coupling constants in the s-wave and p-wave channels, and T is the temperature (throughout this Rapid Communication we are using the units $e = c = \hbar = k_B = 1$). We illustrate our findings through a cut of the phase diagram at $T = 0$, as a function of the ratio g_t/g_s, in Fig. 1. For simplicity, we have chosen the same energy cutoff $\omega_D = 0.1\mu$ in both pairing channels, and we are demonstrating the results for $g_s\rho(\mu) = 1$, where $\rho(\mu)$ stands for the density of states at the Fermi level. Since both the s wave and the B phase are fully gapped states, the coexistence occurs only in a sliver of the entire phase diagram, when $g_t/g_s \sim 1$. This phase diagram suggests the presence of two stage thermal phase transitions in the vicinity of $g_t/g_s \sim 1$. As the temperature is gradually lowered, one first enters the dominant pure phase depending on the relative strength of the couplings, and the $\mathcal{T}$ breaking occurs only at a lower temperature.

When the transition temperatures of the two pairings are comparable, the coexistence can be addressed by using the phenomenological Landau-Ginzburg free energy. The condensation energy density $\Delta f = f_s - f_n$ can be written as

$$\Delta f = \sum_{\alpha=s,t}\left[\frac{c_\alpha}{2}|(\nabla - 2i\mathbf{A})\Delta_\alpha|^2 + r_\alpha|\Delta_\alpha|^2 + u_\alpha|\Delta_\alpha|^4\right] + u_{st1}|\Delta_s|^2|\Delta_t|^2 + u_{st2}|\Delta_s|^2|\Delta_t|^2\cos 2\theta_- + \frac{\mathbf{B}^2}{8\pi}, \tag{7}$$

where f_n is the normal state's free energy density, $\theta_- = (\theta_t - \theta_s)$ is the relative phase between the two complex amplitudes $\Delta_\alpha = |\Delta_\alpha|\exp(i\theta_\alpha)$, and $\mathbf{B} = \nabla \times \mathbf{A}$ is the magnetic field strength. The constants c_α have the dimension of inverse mass and the individual superfluid stiffness can be defined as $\rho_\alpha = c_\alpha|\Delta_\alpha|^2$. As shown in the Supplemental Material [33], all the quartic coefficients for this problem turn out to be positive definite, and consequently the free energy is minimized for $\theta_- = \pm\pi/2$ in the coexisting phase. Deep inside the $p + is$ phase we can ignore the amplitude fluctuations, and in the absence of any singularity in the phase fields we can also shift the vector potential as $\mathbf{A} \to \mathbf{A} - \frac{\nabla\theta_+}{2e} + \frac{\rho_-}{2\rho_+}\nabla\theta_-$, where $\rho_\pm = \rho_s \pm \rho_t$ and $\theta_+ = (\theta_s + \theta_t)/2$. After this shift, the explicit form of the free energy in the London limit becomes

$$\Delta f = \frac{\rho_+^2 - \rho_-^2}{8\rho_+}\left[(\nabla\delta\theta_-)^2 - \frac{2u_{st2}\rho_+}{c_s\, c_t}\cos(2\delta\theta_-)\right] + \frac{\rho_+}{2}\mathbf{A}^2 + \frac{\mathbf{B}^2}{8\pi}, \tag{8}$$

where $\delta\theta_-$ is the deviation of θ_- from $\pm\pi/2$. After expanding the periodic sine-Gordon term $\cos(2\delta\theta_-)$ as $1 - (\delta\theta_-)^2/2$, this equation shows that both the gauge field and the *axionic excitaions* are massive.

Next we focus on the physical implications of this axionic superconductor. We begin by demonstrating the existence of the *gapped* SABS, which in turn lead to an anomalous STH effect. For concreteness, we assume that the semi-infinite regions with $z < 0$ and $z > 0$ are respectively occupied by the $p + is$ superconductor and the vacuum. Therefore, the spinor wave function must satisfy the boundary conditions $\psi(z = 0) = \psi(z \to -\infty) = 0$. For simplicity, we also choose $\xi_{\mathbf{k}} = k^2/(2m) - \mu$ and set $\mathrm{Re}(\Delta_s) = 0$ in Eq. (3). Now assuming $\psi(z) \sim \exp(\lambda z)$, and subsequently setting $k_z \to i\lambda$ in Eq. (5), we obtain the following secular equation for λ,

$$\lambda^4 + 2\lambda^2\left[k_F^2 - k_\perp^2 - 2m^2v_\Delta^2\right] + (k_F^2 - k_\perp^2)^2 + 4m^2\left(v_\Delta^2k_\perp^2 + \mathrm{Im}(\Delta_s)^2 - E^2\right) = 0, \tag{9}$$

where $v_\Delta = \Delta_t/k_F$, and E is the energy of the SABS. The solutions of this equation are of the form $\pm\lambda_j$, with $j = 1,2$. In order to satisfy $\psi(z \to -\infty) = 0$, we require $\mathrm{Re}(\lambda_j) > 0$. After imposing the condition $\psi(z = 0) = 0$, we obtain the constraints

$$4mv_\Delta^2\Delta e^{-i\phi}(m\Delta e^{-i\phi} - k_\perp^2 - \lambda_1\lambda_2) + 2E^2\lambda_1\lambda_2 + v_\Delta^2\left(k_\perp^4 + \lambda_1^2\lambda_2^2\right) + \left(E^2 - v_\Delta^2k_\perp^2\right)\left(\lambda_1^2 + \lambda_2^2\right) = 0, \tag{10}$$

where $\Delta = \sqrt{\mu^2 + \mathrm{Im}(\Delta_s)^2}$, and $\tan\phi = \tan\theta_{\mathrm{ax}}^0$. By using Eqs. (9) and (10), we obtain the following spectra of the SABS,

$$E = \pm\sqrt{v_\Delta^2k_\perp^2 + \mathrm{Im}(\Delta_s)^2}. \tag{11}$$

The explicit form of $\psi(z)$, as shown in the Supplemental Material [33], together with the dispersion E, demonstrate that the SABS are massive Majorana fermions, and the gap is given by $\mathrm{Im}(\Delta_s)$. On the other hand, we recover the gapless SABS of the pure B phase [34,35] by setting $\mathrm{Im}(\Delta_s) = 0$ in Eq. (11). The gapped SABS have an interesting consequence on the tunneling current measurements. In contrast to the B phase, there is no zero bias conductance peak (ZBCP) for the $p + is$ state. Rather, a two gap structure will be found, where the smaller gap stems from the SABS. In the absence of $\mathcal{T}$, the gapless Majorana fermion bound states can only be found along a domain wall between the $p + is$ and the $p - is$ states.

For the characteristic physical response functions of the $p + is$ phase, we first consider the correlation functions of the conserved quantities described by the energy-momentum tensor. In classes D and DIII, the anomalous response of the energy-momentum tensor may be attributed to the gravitational anomaly formula

$$\mathcal{S}_g = \frac{1}{1536\pi^2}\int d^4x\,\epsilon^{\alpha\beta\rho\lambda}\,\theta_{\mathrm{ax}}(x)\mathcal{R}^\eta_{\sigma\alpha\beta}\,\mathcal{R}^\sigma_{\eta\rho\lambda}, \tag{12}$$

where $\mathcal{R}_{\eta\sigma\alpha\beta}$ is the Riemann curvature tensor [11,12]. Recalling that $\theta_{\mathrm{ax}}^0 = \pi + \tan^{-1}\mathrm{Im}(\Delta_s)/\mu$, we note that the π part is tied to the SABS's contribution, whereas the $\tan^{-1}\mathrm{Im}(\Delta_s)/\mu$ part comes from the scattered states. Recently, it has been argued that the gravitational anomaly may be responsible for a STH effect [11,12,36,37], and some additional cross-correlated responses of the DIII TSC [38], when the $\mathcal{T}$ symmetry is broken on the surface by a weak external Zeeman coupling. The STH conductivity of the massive two-dimensional Majorana fermions in the low temperature limit is given by $\kappa_{xy} = \mathrm{sgn}[\mathrm{Im}(\Delta_s)]\pi T/24$. For the $p + is$ state, the $\mathcal{T}$ symmetry is spontaneously broken and consequently no external Zeeman coupling is required to induce this effect. The dimensionless quantities $\hbar\kappa_{xy}/(k_B^2T)$ and $\hbar\kappa_{xy}/(k_B\mu)$ for the SABS of the $p + is$ state, obtained within a linear response calculation [39], are shown in Fig. 2, as a function T/T_F, where T_F is the Fermi temperature. We note that the contribution from the SABS will be generically much larger than that from the scattered states.

Now we briefly discuss the electrodynamic response of this exotic phase. It is natural to anticipate that the signature of the broken $\mathcal{T}$ symmetry can be found through the polar Kerr effect measurements [40]. In addition, there will be *dynamic magnetoelectric effects*, which can be demonstrated by following the calculations in Refs. [41,42]. In these papers, by employing the s-wave and the pseudoscalar pairings of the Dirac fermions, it has been established that the topological electrodynamic response of a TSC is captured by the following magnetoelectric term,

$$\mathcal{S}_{\mathrm{em}} = -\frac{e^2}{64\pi^2}\int d^4x\,\epsilon^{\mu\nu\rho\lambda}\,\theta_{\mathrm{ax}}(x)\mathcal{F}_{\mu\nu}\mathcal{F}_{\rho\lambda}, \tag{13}$$

for the massive gauge fields. In the context of $\mathcal{T}$ preserving TSC in class DIII, the observable effect can only come through the surface state contributions (where θ_{ax} jumps). On the other

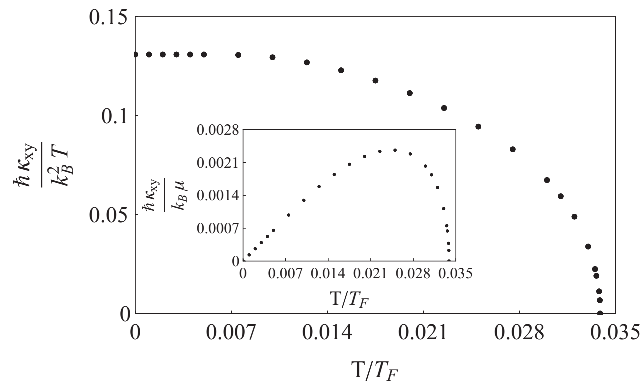

FIG. 2. The dimensionless quantity $\hbar\kappa_{xy}/(k_B^2 T)$ for the gapped SABS, as a function of T/T_F, where κ_{xy} and $T_F = \mu/k_B$ are, respectively, the thermal Hall conductivity and the Fermi temperature. We have used the mean-field gap amplitudes for $g_s\rho(\mu) = 1$ and $g_t/g_s = 0.998\,95$. This quantity saturates to the universal number $\pi/24$ in the zero temperature limit. Inset: The dimensionless thermal Hall conductivity $\hbar\kappa_{xy}/(k_B\mu)$ for the gapped SABS, as a functions of T/T_F.

hand, there are contributions from both the bound and the scattered states for the $p + is$ phase, and $\theta_{\rm ax}(x)$ is dynamical. We also note that in contrast to the *topological magnetic insulators* [43], the massive nature of the gauge field and the axion provides additional stability of this phase. In addition, the gapless one-dimensional modes along the line vortex of the B phase and the pseudoscalar pairings [15,44] acquire a gap in the $p + is$ state.

We conclude this Rapid Communication by discussing the experimental prospect of realizing the $p + is$ state. The current experimental status regarding the nature of the paired state in $Cu_xBi_2Se_3$ is confounding. In Refs. [20–22], a ZBCP in point contact spectroscopy measurements has been reported, which is consistent with the existence of the gapless SABS of a TSC. However, the subsequent tunnel spectroscopy measurements on $Cu_xBi_2Se_3$, with lower copper concentrations, have not found any ZBCP, and the results have been interpreted in terms of the conventional s-wave pairing [23]. This discrepancy in the spectroscopic measurements on compounds with different copper concentrations may be an indicator of an underlying competition between the singlet and the triplet pairings. But, the lower quality of the sample currently prohibits a systematic study of the paired state, as a function of the copper concentration. In this direction, the superconducting $Sn_{1-x}In_xTe$ seems to be a promising material, with a higher superfluid fraction [19]. In Ref. [19] a ZBCP has been reported for $x = 0.045$. More recent measurements by Novak *et al.* [24] have indicated the existence of a competition between the odd and even parity pairings in $Sn_{1-x}In_xTe$, which is the crucial ingredient for realizing the $p + is$ phase. In particular, they have argued for a change of the pairing symmetry around $x = 0.038$. We note that the superconductivity in $Sn_{1-x}In_xTe$ is also realized in the ferroelectric phase, which naturally lacks inversion symmetry. The absence of inversion symmetry is conducive for the coexistence of odd and even parity pairings. However, it remains to be seen if a $\mathcal{T}$ symmetry broken state is indeed realized in this system, which, for example, can be confirmed through surface thermal Hall effect, polar Kerr rotation and Faraday rotation measurements.

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