Surface chiral superconductivity in odd-parity nematic superconductors with magnetic impurities

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We study odd-parity nematic superconductivity in doped topological insulators in the presence of surface magnetic impurities. The peculiar surface subgap spectrum, characterized by a Majorana flat band, nodal cones, and the surface states of the parent topological insulator, gives rise to overall ferromagnetic RKKY interactions between the surface impurities. An additional coupling between the impurities and a preemptive chiral order parameter promote a surface time-reversal symmetry breaking solution at the surface of the system. We discuss the relevant scenarios and suggest to engineer surface chiral superconductivity by properly choosing magnetic adatoms with highly anisotropic exchange coupling.

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I. INTRODUCTION

Chiral superconductivity is a highly interesting and long sought unconventional state of matter that spontaneously breaks time-reversal symmetry through the development of a Cooper pair finite angular momentum [1,2]. It represents an instance of topological superconductivity [3–5] that has attracted great interest thanks to its potential for hosting Majorana fermions in vortex cores [6–8], and in topological quantum computation [9–11]. Intrinsic chiral superconductivity is an unstable state of matter and its occurrence has been suggested in particular conditions, such as layered material like UPt_3 [12], Li_2Pt_3B [13], Sr_2RuO_4 [14,15], SrPtAs [16], and 4Hb-TaS_2 [17]. However, its detection relies on observation of spontaneous magnetization or generation of local magnetic fields [18] that is usually hindered by Meissner screening, and its unequivocal demonstration still remains controversial.

Quantum design has become a very attractive and promising way to attain unconventional and fascinating states of matter. This is the case of engineered topological superconductors [3,6,7], where by bringing together materials with different properties it is possible to engineer the resulting compound at will. It is then natural to wonder whether chiral superconductivity can be stabilized by suitable quantum design. To this end the relevant ingredients that need to be brought together are the quasi-two-dimensional character, a time-reversal symmetry breaking (TRSB) phase trigger, and a multicomponent order parameter (OP) [1]. A bulk two-component OP can choose two solutions, either a rotation symmetry breaking solution, the nematic state, or a chiral TRSB solution. In three-dimensional Dirac materials with a closed weakly anisotropic Fermi surface, the nematic solution is more stable [19–22]. Nevertheless, C_3 crystal symmetry [22] and two dimensionality [23,24] help in stabilizing a chiral solution, and magnetic fluctuations [21,25] can provide a mechanism that triggers a TRSB phase. However, none of them alone is sufficient nor fully practical.

In this work we consider an odd-parity nematic superconductor in the presence of surface magnetic impurities. The system is schematized in Fig. 1(a). We study the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction mediated by the surface gapless states. Three main actors contribute to the interaction: (i) states close to the nodes of the gap, (ii) a flat band of Majorana surface states extending between the nodes, and (iii) the surface states of the parent TI. We find that for an impurity ensemble dilute on the scale of the Fermi wavelength a ferromagnetic interaction is mediated by the surface gapless modes and the system, albeit disordered, is expected to show ferromagnetic order. Close to the surface, a preemptive chiral OP couples to the out-of-plane magnetization. For an in-plane magnetic order, fluctuations of the out-of-plane magnetizations generated by the chiral OP itself promote a phase transition to a TRSB surface state for sufficiently strong coupling. For an out-of-plane order, the chiral OP always condenses at the surface. Due to the small scales provided by the SC gap and in the dilute impurity ensemble approximation, the RKKY mediated in-plane order scenario turns out to be quite fragile and in general the out-of-plane order is realized. These results open the way to engineering surface chiral superconductivity in bulk nematic odd-parity superconductors and provide a mechanism to stabilize the chiral phase in thin samples.

A promising platform for the realization of surface chiral superconductivity is provided by doped Bi_2Se_3 [23,24]. Early experiments [26–29] and recent measurements [30–39] have by now established the odd-parity nematic character of the superconducting state, characterized by a C_2 symmetry. The latter is consistent with the two-component E_u representation of the D_{3d} crystal point group of the material [20,40–42], possibly triggered by odd-parity fluctuations [43,44], density wave fluctuations [45], structural distortion [46], nematicity above T_c [47], and ferroelectric fluctuations [48].

The results presented are generic of odd-parity nematic superconductors, and can be extended to other systems such as UPt_3 [49,50], Sr_2RuO_4 [51–53], or topological semimetals [54], rendering these systems an ideal platform for quantum designing of unconventional physics.

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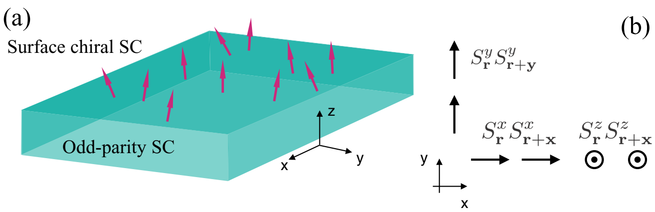

FIG. 1. (a) Schematics of the setup considered: a bulk odd-parity nematic superconductor with surface magnetic disorder and chiral surface solution. (b) Directional dependence of the RKKY interaction experienced by magnetic impurities.

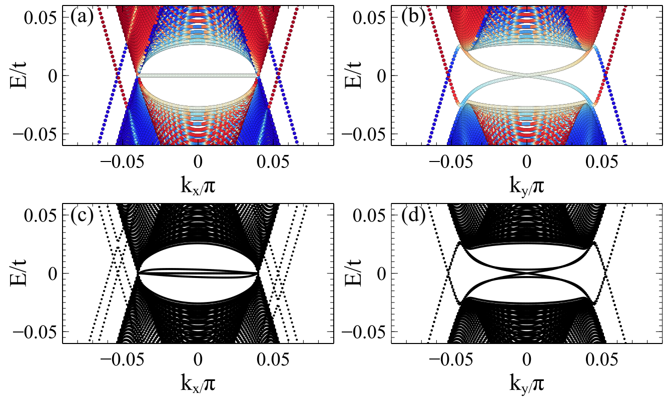

FIG. 2. Surface spectrum of the system obtained with a tight-binding model for a slab of 200 bilayers [55]. (a) and (b) The nematic phase along the k_x and k_y, respectively. The color code represents the charge (red electrons, blue holes). (c) and (d) The nematic phase with surface chiral solution and finite magnetization $\langle S_y \rangle$ only on one surface, along the k_x and k_y, respectively.

II. THE SYSTEM

We start the analysis considering the $k \cdot p$ unperturbed Hamiltonian describing doped Bi_2Se_3. The latter is well described by a 3D anisotropic massive Dirac equation ($\hbar = 1$) [40]

$$\mathcal{H}^0_{\mathbf{k}} = m\sigma_x + v(k_x s_y - k_y s_x)\sigma_z + v_z k_z \sigma_y, \tag{1}$$

where the Pauli matrices σ_i span a twofold orbital subspace and s_i are spin Pauli matrices. Superconductivity is studied by means of the Bogolyubov–deGennes (BdG) Hamiltonian $H = \frac{1}{2}\sum_{\mathbf{k}} \psi^\dagger_{\mathbf{k}} \mathcal{H}_{\mathbf{k}} \psi_{\mathbf{k}}$. In the Nambu basis $\psi_{\mathbf{k}} = (\mathbf{c}_{\mathbf{k}}, is_y \mathbf{c}^\dagger_{-\mathbf{k}})^T$, with $\mathbf{c}_{\mathbf{k}}$ a vector of fermion operators in spin and orbital basis, the BdG Hamiltonian reads

$$\mathcal{H}_{\mathbf{k}} = (\mathcal{H}^0_{\mathbf{k}} - \mu)\tau_z + \hat{\Delta}\tau_+ + \hat{\Delta}^\dagger \tau_-. \tag{2}$$

In the E_u odd-parity channel the gap matrix reads $\hat{\Delta} = -\psi_x \sigma_y s_y + \psi_y \sigma_y s_x$, where $\boldsymbol{\psi} = (\psi_x, \psi_y)$ is the two-component OP. The ground state admits two possible solutions: (i) a TR invariant nodal nematic phase $\boldsymbol{\psi} \propto (1, 0)$ and (ii) a chiral phase $\boldsymbol{\psi} \propto (1, \pm i)$ that breaks TR symmetry. The chiral phase has Weyl nodes in 3D and is fully gapped in 2D systems. Consistently with experiments, we choose a bulk nematic phase.

We assume the system to occupy the $z > 0$ region of space. The full surface spectrum obtained by a tight-binding model [55] is shown in Figs. 2(a) and 2(b) and nodes are present in the spectrum. At the surface of the system, a topologically protected, doubly degenerate Majorana flat band appears for $|k_x| < k_F = \sqrt{\mu^2 - m^2}/v$, extending between the surface projection of the bulk nodes at $\pm k_F$. Additional crossing takes places at momentum $\pm\mu/v$. These states are gapless modes originating from the TI surface states that cross the Fermi level at finite momentum and are hybridized but not gapped by the odd-parity OP [56].

We then place magnetic impurities on the $z = 0$ surface of the system and assume coupling to the electrons via an anisotropic exchange interaction

$$\mathcal{H}_Z = -\frac{1}{n}\sum_i \left[J_z S^z_i s_z + J_\parallel \left(S^x_i s_x + S^y_i s_y\right)\right], \tag{3}$$

where $\mathbf{S}_i$ is the spin of the impurity located at position $\mathbf{r}_i$, $\mathbf{s}$ is the electronic spin operator, J_z and $J_\parallel$ are out-of-plane and in-plane exchange couplings, and $n = N/V$ is the electron density [57]. Impurities also induce scattering via the scalar part of their potential. This typically has detrimental effects of unconventional pairing due to momentum randomization. Nevertheless, for sufficiently diluted impurities, such that the mean free path $\ell_{\rm mf}$ is much larger than the Fermi wavelength λ_F but comparable to the coherence length ξ, $\lambda_F \ll \ell_{\rm mf} \sim \xi$, we neglect their effect.

III. RKKY INTERACTION

For relatively weak exchange coupling, we integrate away the fermionic degrees of freedom and obtain the RKKY interaction experienced by the magnetic impurities [57],

$$\chi_{\mu\nu}(\mathbf{r}) = \frac{J_\mu J_\nu}{n^2} T \sum_{i\omega_n} \mathrm{Tr}[s_\mu G_{i\omega_n}(\mathbf{r}) s_\nu G_{i\omega_n}(-\mathbf{r})], \tag{4}$$

where $G_{i\omega_n}(\mathbf{r}) = \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}}(i\omega_n - \mathcal{H}_{\mathbf{k}})^{-1}$ is the Green's function of the BdG Hamiltonian Eq. (2). Three main actors mediate the interaction: (i) the states around the nodes, (ii) the Majorana flat band, and (iii) the surface hybridized TI modes. The TI states contribution can be estimated by neglecting the hybridization induced by the gap. In this case, well know results for doped TI surface states apply [58–64]. The different terms that arise show oscillations at $2k_F$ and decay as $1/r^2$. In addition, above critical temperature, conduction band electrons provide an additional term that oscillates at $2k_F$ and decays as $1/r^3$. We neglect fast decaying terms in a dilute impurity ensemble approximation.

A. RKKY Majorana flat band

The effective Hamiltonian describing the Majorana flat band is written as

$$h_{\mathbf{k}} = -v_M k_y \hat{\alpha}_y, \tag{5}$$

with $v_M = vm\Delta/\mu^2$ the velocity of the Majorana modes [56] and $\hat{\alpha}_i$ a set of Pauli matrices spanning the subspace defined by $|\phi_\pm\rangle = \frac{1}{2}\binom{1}{\pm i}_s \otimes \binom{1}{\mp i}_\tau$. The RKKY interaction mediated by Majorana fermions has been discussed in Ref. [65] and it represents a particular case of surface TI fermions, with two peculiar differences: (i) the zero chemical potential condition is satisfied exactly and (ii) only one spin component, the s_y in this case, has nonzero projection on the Majorana wave function $|\phi_\pm\rangle$, resulting in the Pauli matrix $\hat{\alpha}_z$. This is the well known Ising property of Majorana Kramers pairs [58]. The flat-band-mediated RKKY interaction reads [55]

$$\chi_{yy}^{\mathrm{FB}}(x,y) = -\frac{J_\parallel^2 \rho_{2\mathrm{D}}}{2\pi v_M n^2}\frac{\sin^2(k_F x)}{(k_F x)^2}\frac{f(k_F y)}{y}, \tag{6}$$

with $f(x) = (2/\pi)\int_0^x dz_1 dz_2 \cos(z_1 - z_2)/(z_1 + z_2)$ and $\rho_{2\mathrm{D}} = k_F^2/\pi^2$ the surface density. The flat band extending between momenta $\pm k_F$ along x generates a contact interaction that dies on a scale $1/k_F$. Along the y direction, $f \to 1$ for large argument, so that for an ensemble dilute on the scale of the Fermi wavelength $\lambda_F = 1/k_F$, the interaction has a purely ferromagnetic long range character. As for the case of TI, magnetic order along the direction dictated by the relevant Majorana operator opens a gap in the Majorana spectrum [Figs. 2(c) and 2(d)]. The RKKY interaction survives also in presence of a gap, self-consistently sustained by the interaction itself [62,64]. Additionally, a magnetization along y acts as a tilting field [66,67] on the TI surface modes along k_x.

B. RKKY nodes

We then consider the RKKY interaction generated by the nodes at the surface. The BdG Hamiltonian Eq. (2), projected onto the conduction band states $\{|\psi_{\mathrm{cb}}^1\rangle_{\mathbf{k}}, |\psi_{\mathrm{cb}}^2\rangle_{\mathbf{k}}\}$ and expanded around the nodal points at $\pm k_F$, reads [55]

$$\mathcal{H}^\pm = \begin{pmatrix} \pm v_x k_x & \delta(k_y\tilde{s}_z - k_z\tilde{s}_y) \\ \delta(k_y\tilde{s}_z - k_z\tilde{s}_y) & \mp v_x k_x \end{pmatrix}, \tag{7}$$

with $v_x = v^2 k_F/\mu$, $\delta = \Delta v/\mu$, and $\tilde{s}_i$ are Pauli matrices spanning the conduction band states. Introducing the rescaled position $\boldsymbol{\rho} = \mu[x/(k_F\xi), y, zv/v_z]/v$, the resulting spin susceptibilities are given by

$$\chi_{xx}(\rho) = -\chi_0 J_\parallel^2\big[(m/\mu)^2 A_0(\rho) + A_x(\rho)\cos(2k_F x)\big],$$
$$\chi_{yy}(\rho) = -\chi_0 J_(1 - (m/\mu)^2 A_z(\rho)\cos(2k_F x),$$
$$\chi_{zz}(\rho) = -\chi_0 J_z^2[1 - (m/\mu)^2]A_y(\rho),$$

where $\chi_0 = 8\nu_F^2\Delta(\mu/k_F v)^4/n^2$ and $\nu_F = \mu k_F/(2\pi^2 v v_z)$ is the density of states at the Fermi level of the bulk Hamiltonian. The functions $A_i(\rho)$ [55] carry a weak dependence on the direction $\hat{\mathbf{r}}$ and are well approximated by $A_i(\rho) = \sin^3(\rho/2)/(3\rho^3)$. The terms oscillating with frequency $2k_F$ originate from internode scattering, whereas the others come from intranode scattering. Along the nodal direction x, the length scale is provided by the superconducting coherence length $\xi = v/\Delta$, whereas along the other directions it is given by λ_F. Assuming a bulk gap $\Delta \sim 1$ K and a velocity $v = 0.6\times 10^8$ cm/s, we have $\xi \sim 5\ \mu$m. In turn, assuming $\mu = 0.33$ eV and $m = 0.3$ eV, we have $\lambda_F \sim 3$ nm. This way, for an impurity ensemble dilute on the scale of λ_F, the RKKY interaction mediated by the nodes acts only along the x direction and its character is mainly ferromagnetic.

IV. IMPURITY-CHIRAL ORDER PARAMETER COUPLING

As shown in Refs. [21,25], magnetic impurities couple to the chiral OP $i\boldsymbol{\psi}\times\boldsymbol{\psi}^*$, that transforms as a pseudovector and can be regarded as an electron spin polarization [68] or Cooper pair spin. Although in the nematic state the chiral OP is zero, a coupling to magnetic impurity can trigger a finite value in proximity of the surface. Including the RKKY interaction arising from the Majorana flat band and the nodes in a total susceptibility $\chi^{\mu\mu}$, the free energy describing magnetic impurities coupled to the order parameter reads

$$F_m = \sum_{ij}\chi^{\mu\mu}(i,j)S_i^\mu S_j^\mu + i\frac{J_z\kappa}{n}\sum_i S_i^z(\boldsymbol{\psi}_{0,i}\times\boldsymbol{\psi}_{0,i}^*)_z, \tag{8}$$

where $\kappa \simeq \mu\nu_F/T_c^2$ is calculated in the normal state [21].

The magnetic interaction is of XX and ZZ type along the x direction and of YY type along the y direction (see Fig. 1) and the chiral OP plays the role of an external field pointing about the $\hat{z}$ direction. Whereas the oscillations with frequency $2k_F$ tend to randomize the XX and YY coupling, the ZZ coupling is practically constant for $x < \xi$. The impurity ensemble is in general disordered, so that the values of $\chi^{\mu\mu}(i,j)$ can be thought as random in magnitude, distributed about different nonzero negative average values. The system belongs to the widely studied class of spin glass models with ferromagnetic random couplings [69,70]. The ground state is ferromagnetic [71], with the total magnetization pointing about the direction of largest average coupling $\chi^{\mu\mu}$. We study two cases: (i) nonnegligible interaction with in-plane order, for $\ell_{\mathrm{mf}} \lesssim \xi$ and (ii) negligible interaction for $\ell_{\mathrm{mf}} \gtrsim \xi$ or out-of-plane order.

For $J_\parallel > J_z$, in-plane ferromagnetic order is expected. The preemptive chiral OP tends to destroy the in-plane order and establish a nonzero expectation value of $\langle S_i^z\rangle$, proportional to the chiral OP itself, $\langle S_i^z\rangle = (\Theta\kappa J_z/n)|\boldsymbol{\psi}_0\times\boldsymbol{\psi}_0^*|$, where Θ is the zero field susceptibility in the ferromagnetic phase. This yields a second order correction to the superconductor free energy

$$F = F_\psi - \Theta(\kappa J_z/n)^2 n_{\mathrm{imp}}|\boldsymbol{\psi}_0\times\boldsymbol{\psi}_0^*|^2, \tag{9}$$

with n_{imp} the impurity concentration. For a spin chain with nearest neighbor coupling $\chi_0 J_\parallel^2$, the susceptibility is $\Theta = 1/(2\chi_0 J_\parallel^2)$, and the interaction is $\propto (J_z/J_\parallel)^2$.

For $J_z > J_\parallel$ the ferromagnetic order is out-of-plane, the ground state has already a finite $\langle S_i^z\rangle$ and the correction to the free energy is linear in the chiral OP,

$$F = F_\psi - i\kappa J_z(n_{\mathrm{imp}}/n)\boldsymbol{\psi}_0\times\boldsymbol{\psi}_0^*, \tag{10}$$

where we assumed the ground state with all impurities pointing about the $\hat{z}$ direction. This scenario also applies to the experimentally relevant case in which the impurity can be considered as noninteracting.

V. SURFACE CHIRAL SOLUTION

In a semi-infinite system it is natural to expect a TRSB solution in proximity of the surface, so that $\boldsymbol{\psi}$ acquires a position dependence that matches two asymptotic solutions, a nematic one at infinity and a TRSB one at $z = 0$. We describe the modulation of the OP via a Ginzburg-Landau (GL) free energy whose form is dictated by symmetry arguments,

$$F_\psi = \int \frac{d^3\mathbf{r}}{V}\left[a|\boldsymbol{\psi}|^2 + b|\boldsymbol{\psi}|^4 + b'|\boldsymbol{\psi}\times\boldsymbol{\psi}^*|^2 + \beta_z|\partial_z\boldsymbol{\psi}|^2\right], \tag{11}$$

where V is the volume of the system and we neglect in-plane gradients [68]. Below T_c, a becomes negative and a finite $b > 0$ ensure a stable finite solution. The two possible nematic and chiral solutions are favored by $b' > 0$ and $b' < 0$, respectively. In the absence of TRSB perturbations, the condition $b' > 0$ is met for bulk 3D systems.

We then parametrize $\boldsymbol{\psi}$ in terms of real valued amplitude $\psi(z)$ and relative phase $\varphi(z)$, $\boldsymbol{\psi} = \psi(e^{-i\varphi/2}, e^{i\varphi/2})/\sqrt{2}$ [72]. We rescale the amplitude by the bulk value $\psi_\infty \equiv \sqrt{|a|/(2b)}$, the position by the GL coherence length $\xi = \sqrt{\beta_z/(2|a|)}$, and set $\eta = b'/b$. For $\eta \ll 1$ we assume constant amplitude and the GL free energy is written as [55]

$$\delta F \propto \int_0^\infty dx[\mathcal{F}(\varphi,\varphi') - gU(\varphi)\delta(x)], \tag{12}$$

where $\mathcal{F} = (\varphi')^2/4 + \eta U(\varphi)$. The potential U depends on the boundary interaction.

A. $J_\parallel > J_z$

In case the magnetic order is in-plane, we have $g = \chi(\kappa J_z)^2 n_{\rm imp}/(2|a|\psi_\infty^2 n^2)$ and $U(\varphi) = \sin^2(\varphi)/4$ that provide the boundary condition $\varphi_0' = -g\sin(2\varphi_0)/4$. The solution for the phase reads

$$\varphi(x) = 2\arctan\left[\tan(\varphi_0/2)e^{-\sqrt{\eta}x}\right] \tag{13}$$

that represents a kink that matches the solution φ_0 at the origin with the asymptotic one $\varphi_\infty = 0$. The boundary condition is solved by $\varphi_0 = \arccos(2\sqrt{\eta}/g)$ and the associated free energy reads $\delta F = -g(1-2\sqrt{\eta}/g)/4$. A critical line $g_c = 2\sqrt{\eta}$ separates a nematic solution $\varphi_0 = 0$ for $g < g_c$ and a TRSB solution $\varphi_0 = \arccos(2\sqrt{\eta}/g)$ for $g > g_c$, as shown in the phase diagram Fig. 3(a). This way, for sufficiently strong coupling, a surface TRSB state occurs with surface solution $\boldsymbol{\psi}_0 \propto (1, e^{i\varphi_0})$. We numerically solve the coupled equations for amplitude and phase, and find an excellent agreement [55]. The critical coupling $g_c = 2\sqrt{\eta}$ is matched exactly. The solution for the phase is shown in Fig. 3(b) and closely matches Eq. (13), especially for small η. The amplitude is shown in Fig. 3(c) and as expected varies on the scale ξ, whereas the phase varies on the scale $\xi/\sqrt{\eta} \gg \xi$. The purely chiral solution $\varphi_0 = \pi/2$ is asymptotically reached for large g. By inspection of Fig. 3(c) we also conclude that for a quasi 2D system satisfying $\xi > L$, $\boldsymbol{\psi}$ can be assumed constant and the results of Ref. [21] apply.

B. $J_z > J_\parallel$

In case the magnetic order is out-of-plane (or in the noninteracting case) the boundary interaction is $U(\varphi) = \sin(\varphi)$ and $g = \kappa J_z n_{\rm imp}/(2n|a|\psi_\infty^2)$. It is clear that the out-of-plane magnetization favors a surface chiral OP and a TRSB solution always exists, as long as $g \neq 0$. The kink solution Eq. (13) applies and the value of the surface phase φ_0 is found by the boundary conditions $\varphi_0' = -2g\cos(\varphi_0)$, so that $\varphi_0 =$

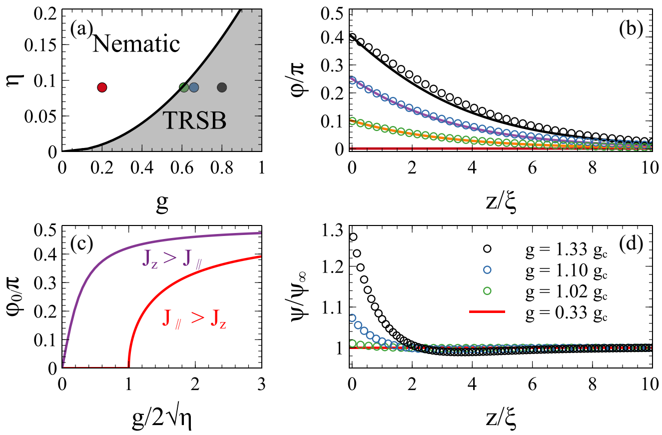

FIG. 3. (a) Phase diagram for the onset of a surface TRSB phase. The separatrix $g_c = 2\sqrt{\eta}$, marked in black, divides the diagram in a nematic phase for $g < g_c$ and a TRSB phase for $g > g_c$. (b) Phase φ and (c) amplitude ψ versus the transverse direction z for $\eta = 0.09$: empty dots refer to the exact numerics and continuous lines in (b) to Eq. (13). (d) Surface phase φ_0 versus coupling for boundary conditions Eqs. (9) and (10).

$\arctan(2g/\sqrt{\eta})$, that is nonzero for every $g > 0$ and asymptotically reach the chiral solution $\varphi_0 = \pi/2$. Furthermore, comparison to the $J_z < J_\parallel$ case shows that, for nominally equal coupling g, the chiral solution is obtained for much weaker coupling in the case $J_z > J_\parallel$ [see Fig. 3(d)].

VI. DISCUSSION

For Cr adatoms an almost isotropic spin exchange is predicted on Bi_2Se_3 [73]. Considering that magnetic adatoms tend to sit on precise microscopic lattice sites, either substitutional or interstitial, and that the nematic phase favors the crystallographic directions, the RKKY interaction cannot be completely ruled out on the basis of its peculiar directional dependence. In this case, an interacting picture applies and a minimum density is required to trigger a surface TRSB solution if the order is in-plane. On the other hand, for magnetic adatoms characterized by $J_z \gg J_\parallel$, like Fe on Bi_2Te_3 [74], a surface TRSB solution always arises and a relatively high impurity concentration can also be tolerated, owing to the predicted out-of-plane order. In this case, $g \simeq \mu J_z n_{\rm imp}/T_c^2$. Assuming $J_z/a^2 \simeq 1$ meV, we find $g \simeq 10^{-2} n_{\rm imp}\xi^2$. It is important to stress that when magnetic impurities align no pair-breaking spin randomization takes place. In conclusion, we show how magnetic impurities on the surface of a nematic odd-parity superconductor can stabilize a surface chiral solution.

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