

Self-consistent surface superconductivity in time-reversal symmetric Weyl semimetals

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Weyl semimetals host topologically protected surface states, the so-called Fermi arcs, that have a penetration depth into the bulk that depends on surface-momentum, and diverges at the Weyl points. It has recently been observed in PtBi₂ that such Fermi arc states can become superconducting, with a critical temperature larger than that of the bulk. Here we introduce a general variational method that captures the interplay between surface and bulk superconductivity for any bulk Hamiltonian that harbors (topological) surface states with varying penetration depth. From the self-consistent solutions, we establish that the surface state localization length of Weyl semimetals leads to characteristic features in the surface superconductivity, with a gap depending on surface momentum and a penetration length for the order parameter that is temperature-dependent due to competition with the bulk superconductivity.

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The possibility of superconductivity at surfaces being enhanced compared to that of the bulk has a long history, and was considered already by Ginzburg and Kirzhnits 60 years ago [1,2]. During the past couple of decades, this theoretical possibility found experimental confirmation with the observation of two-dimensional (2D) superconductivity, naturally realized in quasi-2D electronic gases at interfaces between oxides, the paradigm being the LaAlO₃/SrTiO₃ interface [3–6], or in ultrathin materials [7–12].

More recently, an interplay of bulk electronic topology and surface superconductivity has emerged. Tunneling [13] and photoemission spectroscopy [14] have confirmed that the surface of trigonal PtBi₂ superconducts, with a critical temperature up to one order of magnitude larger than that of the bulk [15–18]. Interestingly, this material is a time-reversal (TR) invariant Weyl semimetal, and the surface states gapped out by superconductivity are its topological Fermi arcs [14,18,19].

Given the potential implications of these findings for topological superconductivity, it is essential to understand what drives the pairing from which the surface superconductivity emerges as well as to address the large difference between surface and bulk critical temperatures. This requires a self-consistent theoretical framework that allows for an order parameter that depends on the position in the surface Brillouin zone (BZ).

Here we introduce a variational approach that meets this challenge. A previous approach [20] adopted an effective

surface Green's function method, with an order parameter included in the first surface layer. In contrast, we argue that the penetration of the surface states into the bulk is an essential component of the emergent superconductivity. Therefore, we introduce an independent method that generally allows for a self-consistent assessment of the superconducting gap opened both at the surface and in the bulk of the material.

In the context of Weyl semimetals, given the recent observation of surface superconductivity, the application of our method is of fundamental importance for two reasons. First, their surface Fermi arcs are anomalous: They can only be realized at the surface of a 3D bulk, and not in any stand-alone 2D system [20,21]. Second, their penetration depth is tied to their topology, given that their localization length diverges as they merge with the projections of the Weyl points on the surface BZ [18,22–28].

We apply our approach to a TR symmetric model for a type-I Weyl semimetal, and we find that the resulting surface superconductivity is not a 2D phenomenon, but rather the emerging manifestation of a full 3D phase transition on the surface. We show that the numerical determination of the surface state wave functions is sufficient to deduce the self-consistent order parameter as a function of temperature. The enhanced density of states (DOS) leads to an increased surface superconductivity via an interplay between the spatial penetration of the surface states into the bulk and the spatially homogeneous order parameter opened in the bulk, which is determined by a conventional 3D Bardeen-Cooper-Schrieffer (BCS) theory [29]. The resulting gap opened at the surface has an intrinsic dependence on the quasimomentum, which comes from the k -dependent penetration of the states enhancing or damping the surface order parameter. Such an effect can only be described by a self-consistent approach such as the one presented here, making it essential for any study aiming at a description of the surface gap opening throughout the BZ.

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I. SEMI-INFINITE SUPERCONDUCTOR

We consider a TR symmetric Hamiltonian H_0 possessing some surface states when translational invariance is broken along the $\hat{z}$ direction. We consider a finite slab as a series of layers with an in-plane, 2D quasimomentum $\mathbf{k}$, with the Hamiltonian

$$H_0 = \sum_{\mathbf{k}, \alpha, \beta, \sigma, \sigma'} h_{\sigma\sigma'}^{\alpha\beta}(\mathbf{k}) c_{\mathbf{k}\alpha}^{\sigma\dagger} c_{\mathbf{k}\beta}^{\sigma'}, \quad (1)$$

where $\sigma = \uparrow, \downarrow$ is the spin label, and the indices α and β contain the orbital (l) and the layer position (z) degrees of freedom of $h(\mathbf{k})$. We will consider the Hamiltonian referred to as a unit surface, so that $\sum_{\mathbf{k}} = \int d^2\mathbf{k}/(2\pi)^2$. Time-reversal symmetry allows for a local s -wave pairing so that the interaction Hamiltonian is $H = H_0 + V$ with [29,30]

$$V = -U \sum_{\mathbf{r}, \alpha} c_{\mathbf{r}\alpha}^{\downarrow\dagger} c_{\mathbf{r}\alpha}^{\uparrow\dagger} c_{\mathbf{r}\alpha}^{\downarrow} c_{\mathbf{r}\alpha}^{\uparrow}, \quad (2)$$

where we are now evaluating the creation and annihilation operator in real space as a function of the 2D coordinate $\mathbf{r}$ (the third coordinate, z , is included in α), and U is the interaction strength. For simplicity, the pairing is assumed to be local on each layer and not mixing different orbitals. The order parameter is $\Delta_\alpha = \langle c_{\mathbf{r}\alpha}^{\downarrow\dagger} c_{\mathbf{r}\alpha}^{\uparrow\dagger} \rangle$ (independent of $\mathbf{r}$ due to translational invariance), and in mean field one finds, after going back to quasimomentum space, an effective pairing term (per unit surface)

$$V = -U \sum_{\mathbf{k}, \alpha} \Delta_\alpha^* c_{-\mathbf{k}\alpha}^{\downarrow\dagger} c_{\mathbf{k}\alpha}^{\uparrow\dagger} - U \sum_{\mathbf{k}, \alpha} \Delta_\alpha c_{\mathbf{k}\alpha}^{\downarrow\dagger} c_{-\mathbf{k}\alpha}^{\uparrow\dagger} + U \sum_{\alpha} |\Delta_\alpha|^2, \quad (3)$$

and the Hamiltonian is

$$H = H_{\text{BdG}} + \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}, \alpha} (h_{\uparrow\uparrow}^{\alpha\alpha}(-\mathbf{k}) + h_{\downarrow\downarrow}^{\alpha\alpha}(-\mathbf{k})) + \sum_{\alpha} U |\Delta_\alpha|^2, \quad (4)$$

where the sum is over the half of the BZ and with its full expression provided in Appendix A. One may neglect the trace of $h(-\mathbf{k})$ in Eq. (4) as it does not depend on Δ_α . H_{BdG} in the previous equation can be diagonalized in order to find the eigenvalues as

$$H_{\text{BdG}} = \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}, i} \lambda_i(\mathbf{k}) \Gamma_{\mathbf{k}i}^\dagger \Gamma_{\mathbf{k}i}, \quad (5)$$

where λ_i are the eigenvalues of the system, and $\Gamma_{\mathbf{k}i}$ are the annihilation operators of the states that, in principle, mix all the previous degrees of freedom and depend on Δ_α . To find the self-consistent values of Δ_α as a function of temperature T , one can minimize the free energy F as a function of Δ_α , where

$$F = -k_B T \log(\mathcal{Z}) = U \sum_{\alpha} |\Delta_\alpha|^2 - \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}, i} k_B T \log(1 + e^{-\beta \lambda_i(\mathbf{k})}), \quad (6)$$

where $\mathcal{Z} = \text{Tr} e^{-\beta H}$ is the partition function of the system, and k_B is the Boltzmann constant. The minimization of Eq. (6)

for all the values of Δ_α is not trivial, since surface states will cause an enhanced order parameter close to the surface. To deal with the broken homogeneity, we introduce the *Ansatz*

$$\Delta_\alpha = \Delta_0 + \Delta_1 e^{-z/z_0}, \quad (7)$$

where Δ_0 , Δ_1 , and z_0 are real and positive variational parameters. The exponential decay of the gap function reflects the exponential decay of the surface states; we comment later on the assumption of choosing all parameters real and positive.

For a semi-infinite system with $N \rightarrow \infty$ layers, electrons in the normal state can either form delocalized bulk states $|b_{\mathbf{k}}^n\rangle$ whose annihilation operator can be defined as $b_{\mathbf{k}}^n = \sum_{\sigma l z} \mathcal{U}_{\mathbf{k}\sigma l z}^n c_{\mathbf{k}\sigma l z}$ with $|\mathcal{U}_{\mathbf{k}\sigma l z}^n| \sim 1/\sqrt{N}$ and eigenvalues $\xi_{\mathbf{k}}^n$, or they may form localized surface states $|e_{\mathbf{k}}^n\rangle$, whose associated annihilation operator is $e_{\mathbf{k}}^n = \sum_{\sigma l z} \tilde{\mathcal{U}}_{\mathbf{k}\sigma l z}^n c_{\mathbf{k}\sigma l z}$ with $|\tilde{\mathcal{U}}_{\mathbf{k}\sigma l z}^n| \sim \rho_{\mathbf{k}\sigma l}^{-z}$, $|\rho_{\mathbf{k}\sigma l}| < 1$, and eigenvalues $\xi_{\mathbf{k}}^n$. We now rewrite V in terms of the bulk and surface eigenstates, separating the terms depending on Δ_0 and $\Delta_1 e^{-z/z_0}$. The homogeneous Δ_0 does not mix bulk and surface states, since it is a constant, and basis changes maintain the property of not mixing different states. The inhomogeneous term contains z , hence the change of basis could cause mixing among eigenstates. However, for a semi-infinite system, a decoupling is induced by the different spatial extensions of the states. When expressed in the new basis, the terms containing the projection over the bulk states vanish since $\sum_z \mathcal{U}_{\mathbf{k}\sigma l z}^n \mathcal{U}_{-\mathbf{k}\sigma' l' z}^m e^{-z/z_0} \sim \sum_z e^{-z/z_0}/N \rightarrow 0$, similarly for the mixing of a bulk and a surface state, $\sum_z \mathcal{U}_{\mathbf{k}\sigma l z}^n \tilde{\mathcal{U}}_{-\mathbf{k}\sigma' l' z}^m e^{-z/z_0} \sim \sum_z e^{-z/z_0}/N \rightarrow 0$. Only surface states are sizeably affected, since $\sum_{\sigma\sigma' l z} \Delta_1 e^{-z/z_0} c_{\mathbf{k}\sigma l z} c_{-\mathbf{k}\sigma' l z} = \sum_{\sigma\sigma' l z} \sum_{nm} \Delta_1 (\tilde{\mathcal{U}}_{\mathbf{k}\sigma l z}^n \tilde{\mathcal{U}}_{-\mathbf{k}\sigma' l z}^m e^{-z/z_0}) e_{\mathbf{k}}^n e_{-\mathbf{k}}^m = \sum_{nm} \Delta_1 f_{\mathbf{k}}^n(z_0) e_{\mathbf{k}}^n e_{-\mathbf{k}}^m$ (the potential mixing matrix elements among different edge eigenstates can be neglected unless the Fermi surface includes a point where said eigenstates are nearly degenerate, as we discuss in detail in Appendix A), where

$$f_{\mathbf{k}}^n(z_0) = \sum_{\sigma l z} |\tilde{\mathcal{U}}_{\mathbf{k}\sigma l z}^n|^2 e^{-z/z_0}. \quad (8)$$

In light of the above, one may separate the two blocks related to bulk and surface states with quasiparticles in the two blocks having energies $\lambda_n^b(\mathbf{k}) = \pm \sqrt{(\xi_{\mathbf{k}}^n)^2 + U^2 \Delta_0^2}$ for the bulk and $\lambda_n^e(\mathbf{k}) = \pm \sqrt{(\xi_{\mathbf{k}}^n)^2 + U^2 [\Delta_0 + \Delta_1 f_{\mathbf{k}}^n(z_0)]^2}$ for the surface. Replacing the *Ansatz* in Eq. (7) and the eigenvalues in Eq. (6), we find $F = NF_E + F_1$, where

$$F_E = U \Delta_0^2 - \quad (9)$$

$$\frac{k_B T}{2} \int \frac{d^2\mathbf{k} dk_z}{8\pi^3} \sum_i \log(1 + e^{-\beta \Lambda_i^b(\mathbf{k}, k_z)}), \quad (10)$$

$$F_1 = 2U \frac{\Delta_0 \Delta_1}{1 - e^{-1/z_0}} + U \frac{\Delta_1^2}{1 - e^{-2/z_0}} - \frac{k_B T}{2} \int \frac{d^2\mathbf{k}}{4\pi^2} \sum_n \log(1 + e^{-\beta \lambda_n^e(\mathbf{k})}), \quad (11)$$

where in F_E the sum over the layers for the bulk states is converted as usual into an integral over the k_z quasimomentum, having introduced the 3D bulk eigenvalues $\Lambda_i^b(\mathbf{k}, k_z)$. The free

energy separates into an extensive portion NF_E , growing with the number of layers N , and an intensive portion F_I dominated by surface physics. The extensive terms reproduce the conventional BCS free energy depending only on Δ_0 and the bulk states. Minimizing the extensive free energy reproduces Δ_0 from bulk BCS theory. The intensive free energy is now affected by the bulk and surface states in two ways: (i) due to the mixing term $\Delta_0\Delta_1$ and the impact of Δ_0 on the eigenvalues of surface states, bulk superconductivity in general affects the surface superconductivity; (ii) the extension of the surface states into the first layers of the bulk, naturally encapsulated in the function $f_{\mathbf{k}}^n(z_0)$, renormalizes the effective order parameter to which each surface state is subjected. The function $f_{\mathbf{k}}^n(z_0)$ is always less than 1, and is higher for well-confined surface states, which, therefore, for a fixed interaction strength are affected more strongly by the superconducting instability.

Notice that the use of short-range interaction implies a summation over momentum states even far from the Fermi surface. However, this does not signal a logical inconsistency in the theory, since the dependence on deep Fermi sea states is only logarithmic, and—as is standard in weak-coupling treatments—it is removed by renormalizing the coupling constant to match physical observables (see, e.g., Ref. [31]). In this sense, our approach is fully consistent with the weak-coupling limit.

The method described above has the advantage of disentangling the bulk from the surface problem, emphasizing at the same time that the bulk order parameter heavily influences the value of the surface order parameter. Moreover, it suggests an operative way to determine the surface properties of real systems. In fact, one can infer the strength of interaction U from the critical temperature in the bulk. Assuming that the interaction strength is the same also at the surface, the surface physics is thus entirely determined. Should specific interaction channels be modified at the surface, one may include them by introducing an inhomogeneous interaction strength.

II. SURFACE SUPERCONDUCTIVITY IN A WEYL SEMIMETAL

We demonstrate our approach on a generic TR symmetric model for a Weyl semimetal proposed in Refs. [32,33] (in Appendix D, we also confirm our numerical approach by applying it to a simpler, analytically solvable model),

$$h(\mathbf{k}, k_z) = a(\sin k_y \tau_y \sigma_x + \sin k_z \tau_z \sigma_0) + \beta \tau_z \sigma_z + d \tau_z \sigma_x + [t \cos k_x + 2b(2 - \cos k_y - \cos k_z)] \tau_x \sigma_0 + \alpha \sin k_z \tau_y \sigma_z + \lambda \sin k_x \tau_0 \sigma_y - \mu \tau_0 \sigma_0, \quad (12)$$

where σ_i and τ_i are the Pauli matrices along the direction i , respectively, for the spin and orbital degrees of freedom, and the subscript 0 refers to the identity in their subspace. Terms in λ , d , and β break the inversion symmetry, while TR symmetry is always preserved. Fermi arcs appear when translational invariance is broken along the z axis. Bulk and surface spectral functions and the Fermi surface of this model for a benchmark choice of parameters are shown in Appendix B.

We build a finite slab of 40 layers, and numerically diagonalize it in the normal state. We separate the surface states based on their penetration in the bulk (specifically, such that

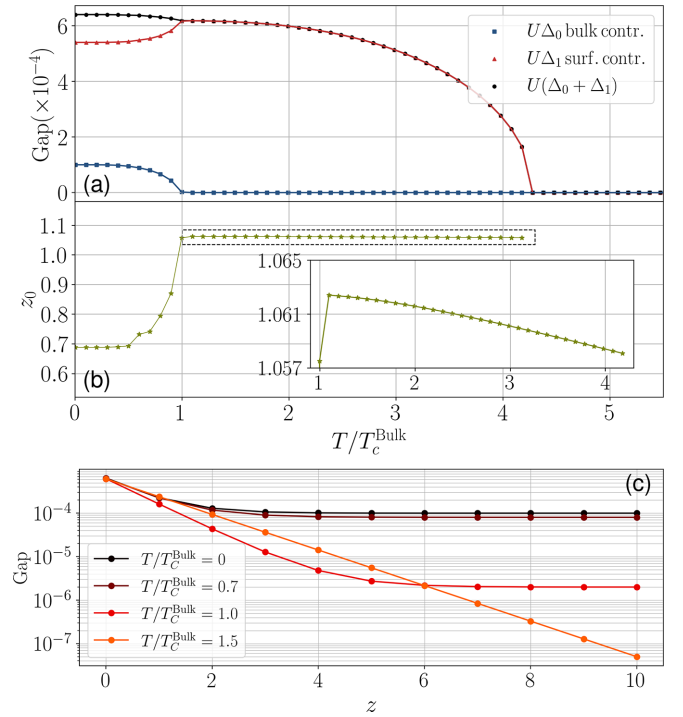


FIG. 1. (a) Temperature dependence of the bulk and surface order parameters. Blue squares, red triangles, and black points correspond to the superconducting gap for bulk ($g_0 = U\Delta_0$), surface ($g_1 = U\Delta_1$) and total order parameter of the first layer [$g_s = U(\Delta_0 + \Delta_1)$], respectively. (b) Penetration depth z_0 of the surface order parameter as a function of the temperature. The inset details of the behavior of $1/z_0$ when the bulk order parameter has already vanished, showing that in this regime there is an opposite tendency of increasing penetration depth with increasing temperature. Temperature is scaled to the bulk critical temperature T_c^{Bulk} . (c) Total gap $U(\Delta_0 + \Delta_1 e^{z/z_0})$ as a function of layer position, for different temperatures. The curve for $T/T_c^{\text{Bulk}} = 1.0$ approaches a finite value since $T \rightarrow T_c^{\text{Bulk}}$ from left.

$\sum_{\sigma l, z=0}^{z=5} |\tilde{u}_{\mathbf{k}\sigma l z}^n|^2 \geq 0.7$). For a 4×4 Hamiltonian, we expect only two surface states on the top layer and two on the bottom layer. For each of them, we determine the function $f_{\mathbf{k}}^n(z_0)$ using Eq. (8) for $z_0 \in [0.1, 10]$.

We first minimize the bulk free energy to obtain the conventional BCS results for the bulk order parameter as a function of temperature. We use this as a scale, fixing the interaction strength U to the value required to open a gap in the bulk $g_0 = U\Delta_0 = 10^{-4}$ at zero temperature. With this value of U , and the self-consistent order parameter $\Delta_0(T)$, we identify $\Delta_1(T)$ and $z_0(T)$ by minimizing the surface free energy. Figure 1 shows the results for chemical potential $\mu = -0.03$ (the role of chemical potential is further discussed in Appendix C). We find a nonvanishing surface order parameter Δ_1 , which is in fact higher than Δ_0 at zero temperature, due to the higher local DOS at the surface. The penetration depth z_0 has a nontrivial temperature dependence, increasing with the decreasing bulk order parameter Δ_0 . We interpret this as a competition between bulk and surface superconductivity: With decreasing Δ_0 , electrons at the surface tend to compensate for the reduction by extending the surface order parameter

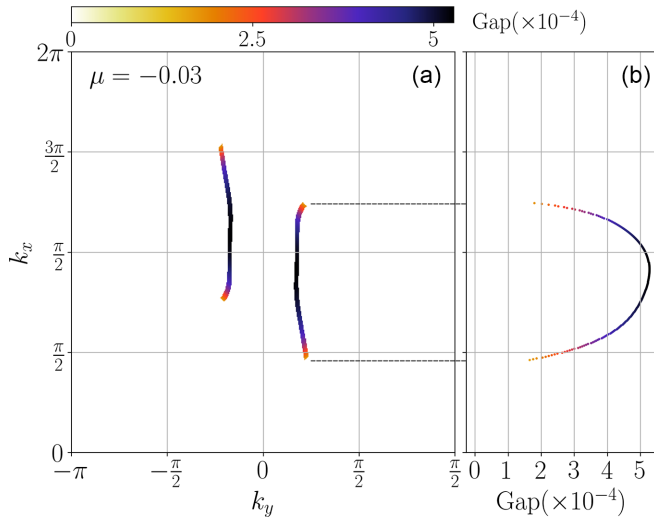


FIG. 2. (a) Fermi arcs with their gap $U[\Delta_0 + \Delta_1 f(z_0)]$ (color scale) at $\mu = -0.03$ in the 2D BZ spanned by k_x and k_y . (b) Momentum dependence of the energy gap along the Fermi arc. The highlighted Fermi surface has been found by selecting an energy window of $2\delta w = 5 \cdot 10^{-2}$ centered on the Fermi arcs, i.e. by computing $U[\Delta_0 + \Delta_1 f(z_0)]\Theta(\mu - \bar{\xi} + \delta w)\Theta(-\mu + \bar{\xi} + \delta w)$, with $\Theta(\cdot)$ the Heaviside function.

into the bulk, an effect that can only be captured from the 3D approach. Indeed, its origin can be traced to the mixed product term $\Delta_0\Delta_1$ in Eq. (11): A nonvanishing bulk parameter Δ_0 introduces an additional energy cost for Δ_1 , disfavoring surface superconductivity. The explicit spatial dependence of the order parameter is shown in Fig. 1 for varying temperatures. As the temperature is increased above the critical value for the bulk, the order parameter far from the interface decreases as expected, yet in the very first layers there is an overall spreading, due to the increasing value of z_0 . Finally, the surface states contribute to the free energy weighted by the factor $f_{\mathbf{k}}(z_0)$, measuring the degree of penetration of each state into the material. This weighting factor depends on $\mathbf{k}$, so that a momentum dependence of the surface-related gap emerges, despite the absence of long-range components of the electron-electron interaction. This is visible in Fig. 2, from which one sees that the gap at the first layer is very strongly $\mathbf{k}$ -dependent.

III. SUMMARY AND CONCLUSIONS

We considered superconductivity at surfaces of TR invariant materials which possess surface states, where one may expect the modified density of states in the vicinity of the surface to cause superconductivity with a critical temperature different from the bulk one. By introducing a variational method, we tackled this problem that takes into account the interplay between surface and bulk states in the formation of a superconducting order parameter. It enables us to quantitatively determine the extent to which surface superconductivity penetrates through multiple layers away from the surface, and is able to capture its emergence out of the underlying 3D model. We have applied this approach on a generic Weyl Hamiltonian as a prototype of a system for which an effective,

analytical surface Hamiltonian does not exist, and the emerging surface states are found numerically. The method can be directly extended to more complex Hamiltonians with a larger number of degrees of freedom. Despite the lack of a long-range component, we find that the pairing interactions still induce the opening of a gap on the surface which varies across the BZ, where the variability is induced by the changing penetration depth of the surface states into the bulk, a feature that clearly can only be captured by a 3D approach. We also showed that bulk superconductivity at sufficiently low temperatures competes with the emerging surface superconductivity. The results suggest a route towards the explanation of the recently observed surface superconductivity in PtBi_2 without a corresponding bulk superconductivity, and they provide the tools to investigate this emerging behavior with quantitative accuracy.

Interestingly, the presence of an inhomogeneous order parameter, which forms the basis of our modeling, opens intriguing avenues for further exploration (for example, in Appendix E we have tested more complex patterns for the spatial dependence of the order parameter). Here we have explored the starting point that the inhomogeneous part of the order parameter is real and with the same sign as the bulk order parameter. However, the inhomogeneous component of the order parameter may in principle have its own phase, which may also be space-dependent. Our variational approach is particularly suited for such generalizations, since it only requires minimization with respect to the space-varying phase as well as the amplitude of the order parameter.

We adopt a short-range interaction, as in the original BCS treatment. A potential longer-range component may definitely be present, in which case, however, the method we have introduced can be applied in the same way, with the only additional complexity of a dependence of the order parameter on the wave vector. This should cause no conceptual complication in our method, and we plan to consider this case in future work. Overall, the method we propose allows for a systematic and flexible investigation of the penetration of the order parameter inside the material. Our framework relies on a controlled approximation—its convergence can be tested by simply extending the variational *Ansatz*—and it can be easily generalized to arbitrary models, offering the possibility of a systematic characterization of surface superconductivity. Such a task, which until recently may have been of mainly theoretical interest, has become extremely relevant given the recent observations of surface superconducting states.

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APPENDIX A: DETAILS OF THE BOGOLIUBOV-DE GENNES HAMILTONIAN

In this Appendix, we give the details of the BdG Hamiltonian presented in the main text and the explicit form of the diagonalized Hamiltonian. By referring to the notation introduced in the main text, and by using the particle-hole basis as

$$\gamma_{\mathbf{k}\alpha} = \begin{pmatrix} c_{-\mathbf{k}\alpha}^{\uparrow\dagger} \\ c_{-\mathbf{k}\alpha}^{\downarrow\dagger} \\ c_{\mathbf{k}\alpha}^{\downarrow} \\ c_{\mathbf{k}\alpha}^{\uparrow} \end{pmatrix}, \quad (\text{A1})$$

the full Hamiltonian $H = H_0 + V$ can be written in a mean-field approach in the following way:

$$H = H_{\text{BdG}} + \sum_{\alpha} U |\Delta_{\alpha}|^2, \quad (\text{A2})$$

where

$$H_{\text{BdG}} = \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}, \alpha, \beta} \gamma_{-\mathbf{k}\alpha}^{\dagger} \begin{pmatrix} h_{\uparrow\uparrow}^{\alpha\beta}(\mathbf{k}) & h_{\uparrow\downarrow}^{\alpha\beta}(\mathbf{k}) & 0 & -U \Delta_{\alpha} \delta_{\alpha\beta} \\ h_{\downarrow\uparrow}^{\alpha\beta}(\mathbf{k}) & h_{\downarrow\downarrow}^{\alpha\beta}(\mathbf{k}) & U \Delta_{\alpha} \delta_{\alpha\beta} & 0 \\ 0 & U \Delta_{\alpha}^* \delta_{\alpha\beta} & -h_{\downarrow\downarrow}^{\beta\alpha}(-\mathbf{k}) & -h_{\downarrow\uparrow}^{\beta\alpha}(-\mathbf{k}) \\ -U \Delta_{\alpha}^* \delta_{\alpha\beta} & 0 & -h_{\uparrow\downarrow}^{\beta\alpha}(-\mathbf{k}) & -h_{\uparrow\uparrow}^{\beta\alpha}(-\mathbf{k}) \end{pmatrix} \gamma_{\mathbf{k}\beta} \quad (\text{A3})$$

and having neglected the trace of $h(-\mathbf{k})$ in Eq. (A2) which does not depend on Δ_{α} . H_{BdG} in the previous equation can be diagonalized in order to find the eigenvalues as

$$H_{\text{BdG}} = \sum_{\mathbf{k} > 0, i} \lambda_i(\mathbf{k}) \Gamma_{\mathbf{k}i}^{\dagger} \Gamma_{\mathbf{k}i}, \quad (\text{A4})$$

where λ_i are the eigenvalues of the system, and $\Gamma_{\mathbf{k}i}$ are the annihilation operators of the states that, in principle, mix all the previous degrees of freedom and depend on Δ_{α} . However, as we showed in the main text, the diagonalization can be performed easily by using the variational *Ansatz* $\Delta_{\alpha} = \Delta_0 + \Delta_1 e^{-z/z_0}$. Due to the separation among the surface and bulk states, the Hamiltonian (A4) admits the following block form in the basis of the edge and bulk eigenstates of the normal Hamiltonian, respectively $|e_{\mathbf{k}}^n\rangle$ and $|b_{\mathbf{k}}^n\rangle$, corresponding to the annihilation operators $e_{\mathbf{k}}^n$ and $b_{\mathbf{k}}^n$ in second quantization. Using the basis

$$\tilde{\gamma}_{\mathbf{k}n} = \begin{pmatrix} e_{\mathbf{k}}^{n\dagger} \\ e_{-\mathbf{k}}^n \\ b_{\mathbf{k}}^{n\dagger} \\ b_{-\mathbf{k}}^n \end{pmatrix}, \quad (\text{A5})$$

the BdG Hamiltonian reads

$$H_{\text{BdG}} = \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}, n} \tilde{\gamma}_{-\mathbf{k}n}^{\dagger} \begin{pmatrix} \tilde{\xi}_{\mathbf{k}}^n & -U[\Delta_0 + \Delta_1 f_{\mathbf{k}}^n(z_0)] & 0 & 0 \\ U[\Delta_0 + \Delta_1 f_{\mathbf{k}}^n(z_0)] & -\tilde{\xi}_{-\mathbf{k}}^n & 0 & 0 \\ 0 & 0 & \xi_{\mathbf{k}}^n & -U\Delta_0 \\ 0 & 0 & U\Delta_0 & -\xi_{-\mathbf{k}}^n \end{pmatrix} \tilde{\gamma}_{\mathbf{k}n}, \quad (\text{A6})$$

where the matrix elements of the off-diagonal block terms belonging to the subspace of the edge states have been computed as

$$\sum_{\sigma\sigma'lz} \Delta_1 e^{-z/z_0} c_{\mathbf{k}\sigma lz} c_{-\mathbf{k}\sigma'lz} = \sum_{\sigma\sigma'lz} \sum_{nm} \Delta_1 (\tilde{\mathcal{U}}_{\mathbf{k}\sigma lz}^n \tilde{\mathcal{U}}_{-\mathbf{k}\sigma'lz}^m e^{-z/z_0}) e_{\mathbf{k}}^n e_{-\mathbf{k}}^m = \sum_n \Delta_1 f_{\mathbf{k}}^n(z_0) e_{\mathbf{k}}^n e_{-\mathbf{k}}^n, \quad (\text{A7})$$

with

$$f_{\mathbf{k}}^n(z_0) = \sum_{\sigma lz} |\tilde{\mathcal{U}}_{\mathbf{k}\sigma lz}^n|^2 e^{-z/z_0} \quad (\text{A8})$$

having used the relation $\tilde{\mathcal{U}}_{-\mathbf{k}\sigma'l}^n = (\tilde{\mathcal{U}}_{\mathbf{k}\sigma l}^n)^*$. The potential mixing among different edge states, induced by matrix el-

ements of the form $\sum_{nm} \Delta_1 f_{\mathbf{k}}^{nm}(z_0) e_{\mathbf{k}}^n e_{-\mathbf{k}}^m$, with $f_{\mathbf{k}}^{nm}(z_0) = \sum_{\sigma lz} \tilde{\mathcal{U}}_{\mathbf{k}\sigma lz}^n (\tilde{\mathcal{U}}_{\mathbf{k}\sigma lz}^m)^* e^{-z/z_0}$, would be relevant only if the chemical potential intersects both eigenstates simultaneously at the same value of $\mathbf{k}$, i.e., if the Fermi surface includes a point of degeneracy among the two eigenstates. This is because in the limit $U \ll \mu$, only states within a small energy interval

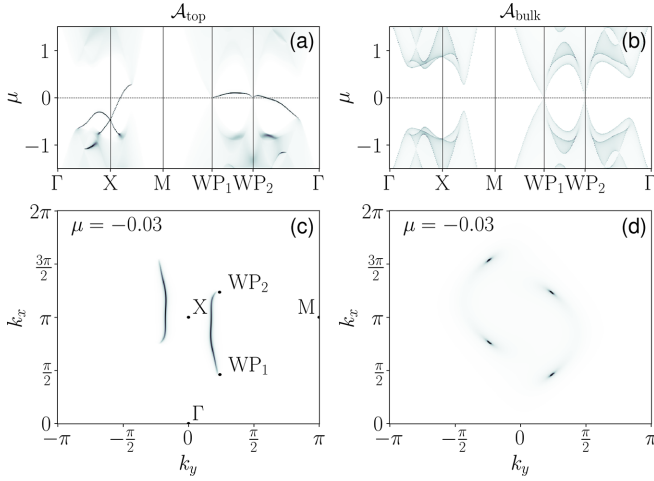


FIG. 3. (a) Spectral function $\mathcal{A}_i$ of the top surface and (b) of the bulk of Hamiltonian in Eq. (11) of the main text. The color maps of (a) and (b) are the same, and they have been cut off at $\mathcal{A}^{\max} = 10$ to improve the contrast. (c) Spectral function of the surface and (d) of the bulk for a benchmark choice of the chemical potential $\mu = -0.03$. The color maps of the two plots have been chosen to enhance the contrast. The parameters of the Hamiltonian are $a = b = 1$, $t = 1.5$, $\alpha = 0.9$, $d = 0.2$, $\lambda = 0.8$, $\beta = 0.7$.

of order U from the Fermi surface are affected by the superconducting coupling, so only if the two states are close in energy can their mixing induce any consequence. Mathematically, unless $|\tilde{\xi}_{\mathbf{k}}^n| - |\tilde{\xi}_{\mathbf{k}}^m| \sim U$, a potential off-diagonal term of order U would negligibly affect the eigenstates of the BdG Hamiltonian. For the regime we study here, this is never the case; it is straightforward to extend our method to include the off-diagonal mixing of edge states if the Fermi surface includes a degeneracy point. The eigenvalues of the blocks, therefore, are $\lambda_n^b(\mathbf{k}) = \pm \sqrt{(\tilde{\xi}_{\mathbf{k}}^n)^2 + U^2 \Delta_0^2}$ for the bulk and $\lambda_n^e(\mathbf{k}) = \pm \sqrt{(\tilde{\xi}_{\mathbf{k}}^n)^2 + U^2 [\Delta_0 + \Delta_1 f_{\mathbf{k}}^n(z_0)]^2}$ for the surface. We note that we rewrite the bulk eigenvalues by explicitly taking into account the out-of-plane momentum k_z dependency included in the label n , so that we can define $\Lambda_n^b(\mathbf{k}, k_z) = \pm \sqrt{(\Xi_{\mathbf{k}, k_z}^n)^2 + U^2 \Delta_0^2}$, where $\Xi_{\mathbf{k}, k_z}^n$ are the single-particle bulk eigenvalues for which we made explicit k_z .

APPENDIX B: ELECTRONIC BAND STRUCTURE IN THE NORMAL STATE

The model presented in the main text exhibits Weyl points with a characteristic structure in the Brillouin zone (BZ). In this Appendix, we show explicitly the band structure of the Hamiltonian in the normal state, i.e., for vanishing interaction strength $U = 0$.

Figure 3 shows the band structure and the spectral function for the Hamiltonian. To obtain the latter, we define a bare Green's function $\mathcal{G}^{-1} = h(\mathbf{k}) + i\delta$, and we use it to find the exact surface ($\mathcal{G}_{\text{top}} = \mathcal{G}_0$) and the bulk ($\mathcal{G}_{\text{bulk}} = \mathcal{G}_{N/2}$) Green's functions using the iterative method of Ref. [34]. The imaginary part of this Green's function is connected to the spectral density as $\mathcal{A}_i = -\frac{1}{\pi} \text{ImTr} \mathcal{G}_i$.

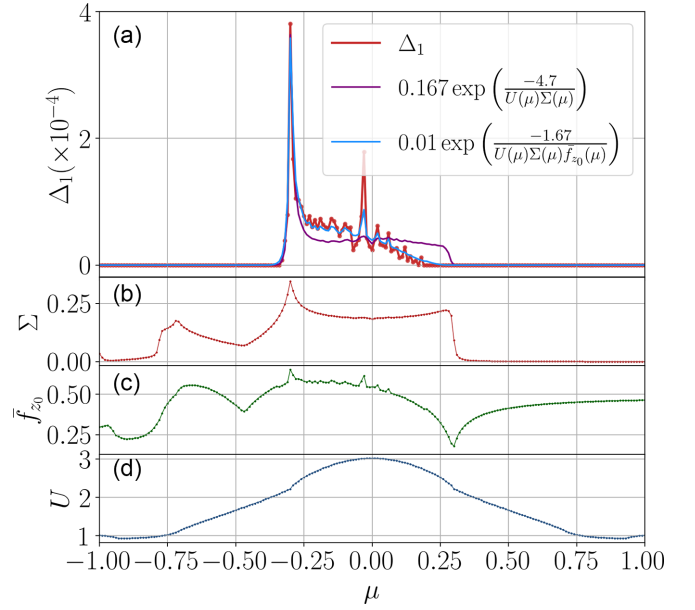


FIG. 4. Chemical potential analysis. (a) Δ_1 as a function of the chemical potential and fit of the BCS expression of the order parameters. (b) Surface density of states. (c) Average of the penetration function f_{z_0} according to the self-consistent value of z_0 which depends on μ . (d) Value of U which opens a gap in the bulk of $U \Delta_0 = 10^{-4}$.

APPENDIX C: DENSITY OF STATES AND ORDER PARAMETER DEPENDENCE

In this Appendix, we discuss the role of the chemical potential (μ) and the surface density of states (Σ) in determining the surface contribution to the order parameter. According to the BCS theory [35], the bulk order parameter follows the equation

$$\Delta_0 = 2\hbar\omega e^{-\frac{1}{U\Sigma_0}}, \quad (\text{C1})$$

where $2\hbar\omega$ is the bandwidth of the single-particle spectrum centered on μ for a system whose density of states Σ_0 is almost constant in that interval. We want to test how such an expression qualitatively fares with the description of the surface contribution. In Fig. 4(a), we show the self-consistent values of Δ_1 as a function of the chemical potential, constraining the value of the bulk gap $g_0 = U \Delta_0 = 10^{-4}$. In Fig. 4(b), we show the value of the surface local DOS computed as

$$\Sigma(\mu) = \int_{\text{BZ}} \frac{d^2\mathbf{k}}{4\pi^2} \mathcal{L}_{\delta\tau}(\tilde{\xi}_{\mathbf{k}}^n - \mu), \quad (\text{C2})$$

where we highlighted the chemical potential dependence on the surface eigenvalues, and we defined $\mathcal{L}(x) = \frac{1}{\pi} \frac{\delta\tau}{x^2 + \delta\tau^2}$ as a Lorentzian approximation for the δ function, with $\delta\tau = 0.005$. In Fig. 4(d), the value of U as a function of the chemical potential is shown. We use these quantities to define a function analogous to Eq. (C1) as

$$\delta_1(\mu) = a e^{\frac{-b}{U(\mu)\Sigma(\mu)}}, \quad (\text{C3})$$

and we fit the coefficients a and b by minimizing the function $\chi^2 = \sum_i [\Delta_1(\mu_i) - \delta_1(\mu_i)]^2$ as a function of the parameters.

The result of the fit is shown in Fig. 4(a) with a purple line. As we can see, the density of states is able to reproduce the peak of Δ_1 , but it is unable to fit the oscillations and the correct decrease as a function of the chemical potential. This is because Eq. (C3) does not contain information on the penetration depth of the surface states within the slab of the material, which is crucial to correctly reproduce the behavior. To clarify this, we define an average value of the penetration function $\bar{f}_{z_0}$ as the weighted average,

$$\bar{f}_{z_0}(\mu) = \frac{\sum_n \int \frac{d^2\mathbf{k}}{4\pi^2} f_{\mathbf{k}}^n(z_0) \mathcal{L}_{\delta\tau}(\xi_{\mathbf{k}}^n - \mu)}{\sum_n \int \frac{d^2\mathbf{k}}{4\pi^2} \mathcal{L}_{\delta\tau}(\xi_{\mathbf{k}}^n - \mu)}. \quad (\text{C4})$$

Replacing the density of states by the product of Σ and Eq. (C4) in Eq. (C3), we obtain the blue line in Fig. 4(a). Including the information on the penetration of the surface states results in a better agreement between the fit and the self-consistent data. We conclude that the density of states by itself cannot explain the superconductivity on the surface, but needs to be complemented with the information on the penetration of the Fermi arcs within the slab.

APPENDIX D: ANALYTICAL TIGHT-BINDING MODEL

Here we discuss another tight-binding Hamiltonian of a time-reversal Weyl semimetal, one that can be analytically solved in the normal state, and thus can validate our numerical analysis. The Hamiltonian that we are taking into account is a generalization to a spinful Hamiltonian of the lattice model in Ref. [36],

$$h(\mathbf{K}) = \begin{pmatrix} h_{\uparrow\uparrow}(\mathbf{K}) & 0 \\ 0 & h_{\downarrow\downarrow}(\mathbf{K}) \end{pmatrix}, \quad (\text{D1})$$

where

$$h_{\uparrow\uparrow}(\mathbf{K}) = -t[\cos(k_x)\tau_x + \cos(k_y)\tau_y - \sin(k_z)\tau_z] + \alpha[1 - \cos(k_z)][\cos(\gamma)\tau_x + \sin(\gamma)\tau_y] - \mu\tau_0, \quad (\text{D2})$$

$$h_{\downarrow\downarrow}(\mathbf{K}) = [h_{\uparrow\uparrow}(-\mathbf{K})]^*, \quad (\text{D3})$$

with matrices τ_i acting on the orbital degrees of freedom. This model has the advantage that Weyl cones are always positioned at the four points $\mathbf{K}_{\text{Weyl}} = (\pm\pi/2, \pm(\kappa)\pi/2, 0)$, with $\kappa = \pm 1$. In the following, we fix the hopping amplitude to $t = 1$, and note that α and γ do not affect the location of the Weyl points but only the spectrum of the Fermi arcs. The eigenvalues of the Hamiltonian Eq. (D2) can be written analytically for both the bulk (since its diagonal blocks have the form $h = \mathbf{v} \cdot \boldsymbol{\tau} - \mu\tau_0$) and the xy surface as

$$\tilde{\xi}_{\pm}^e(\mathbf{k}) = \pm[\cos(k_y)\cos(\gamma) - \cos(k_x)\sin(\gamma)], \quad (\text{D4})$$

within the domain defined by the expression $|r(\mathbf{k})| \leq |\alpha|$, with $r(\mathbf{k}) = \alpha - \cos(k_x)\cos(\gamma) - \cos(k_y)\sin(\gamma)$. The spectral function for the surface and the bulk, together with the analytical Fermi surface, are shown in Fig. 5. The $\pm$ sign in Eq. (D4) refers to the bottom and top surfaces of a finite slab. For our purpose, we will focus only on the top surface of a slab, which corresponds to the negative sign.

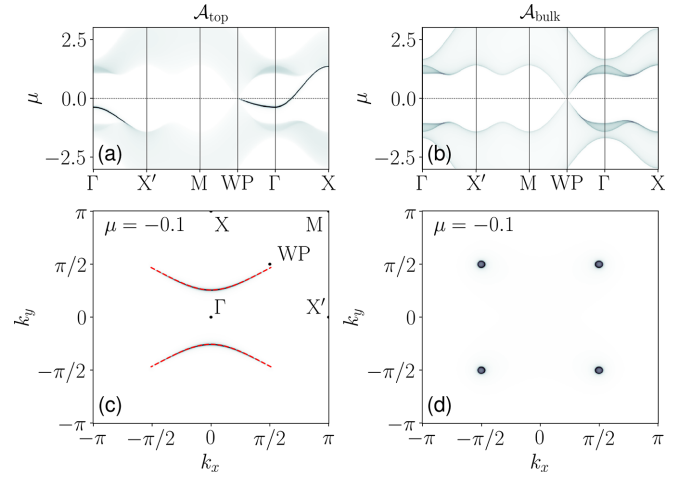


FIG. 5. (a) Spectral function $\mathcal{A}_i$ of the top surface and (b) of the bulk of Hamiltonian Eq. (D1). The color maps of (a) and (b) are the same, and they have been cut off at $\mathcal{A}^{\max} = 10$ to improve the contrast. (c) Spectral function of the surface and (d) of the bulk for a benchmark choice of the chemical potential $\mu = -0.1$. In dashed lines, the analytical eigenvalues match the positions of the Fermi arcs found with the numerical spectral function. Color maps of the two plots have been chosen to enhance the contrast. The parameters of the Hamiltonian are $t = 1$, $\alpha = 1.5$, $\gamma = \pi/6$, and the smearing of the Green's function $\delta = 10^{-3}$.

We write analytical expressions for the surface eigenstates $|e_{\mathbf{k}}^n\rangle$ after verifying that the *Ansatz*

$$|e_{\mathbf{k}}^n\rangle = \sum_{l_z} \tilde{A}_{\rho_{\mathbf{k}}} c_l [\rho_{\mathbf{k}}^{z+1} - (\rho_{\mathbf{k}}^{z+1})^*] |c_{\mathbf{k}l_z}\rangle \quad (\text{D5})$$

correctly satisfies the Schrödinger equation, where $\tilde{A}_{\rho_{\mathbf{k}}}$ is the normalization of the vector, c_l is a coefficient that depends only on the orbital degrees of freedom, and $|\rho_{\mathbf{k}}| < 1$. The value of ρ can be found by using the eigenvalues Eq. (D4), solving the Schrödinger equation with the *Ansatz* above. We obtain

$$\rho_{\mathbf{k}} = \frac{r(\mathbf{k}) \pm \sqrt{1 - \alpha^2 + r(\mathbf{k})^2}}{1 + \alpha}, \quad (\text{D6})$$

where, again, $\pm$ refers to the bottom and top eigenvalues. We can get the expression of $\tilde{A}_{\rho_{\mathbf{k}}}$ by normalizing the eigenvectors, obtaining $|\tilde{A}_{\rho_{\mathbf{k}}}|^2 = 1/G_{\mathbf{k}}$, where

$$G_{\mathbf{k}} = \left(\frac{2|\rho_{\mathbf{k}}|^2}{1 - |\rho_{\mathbf{k}}|^2} - \frac{\rho_{\mathbf{k}}^2}{1 - \rho_{\mathbf{k}}^2} - \frac{\rho_{\mathbf{k}}^{*2}}{1 - \rho_{\mathbf{k}}^{*2}} \right). \quad (\text{D7})$$

In this way, we obtain $f_{\mathbf{k}}(z_0) = T_{\mathbf{k}}/G_{\mathbf{k}}$, where

$$T_{\mathbf{k}} = \left(\frac{2|\rho_{\mathbf{k}}|^2}{1 - |\rho_{\mathbf{k}}|^2 e^{-1/z_0}} - \frac{\rho_{\mathbf{k}}^2}{1 - \rho_{\mathbf{k}}^2 e^{-1/z_0}} - \frac{\rho_{\mathbf{k}}^{*2}}{1 - \rho_{\mathbf{k}}^{*2} e^{-1/z_0}} \right). \quad (\text{D8})$$

We can now exploit the fact that the BdG Hamiltonian can be decoupled in two distinct blocks since there is no spin-mixing in the single-particle Hamiltonian in order to further simplify the problem. From the expression of the free energy in the main text, we derive the self-consistent expression of

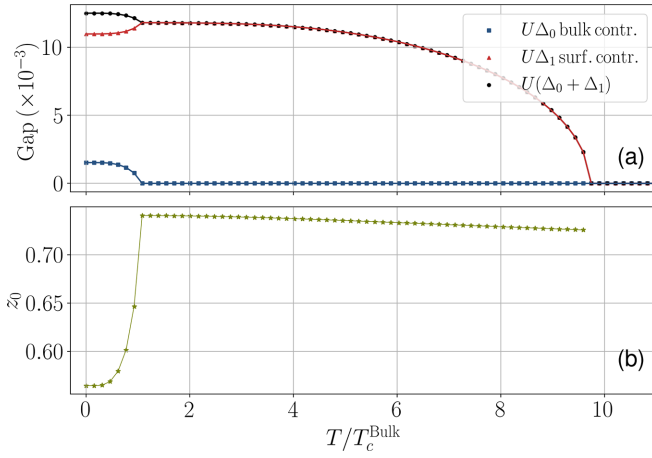


FIG. 6. (a) Temperature dependence of the order parameter contributions. The blue, red, and black points correspond to the energy gap for the bulk ($g_0 = U\Delta_0$), the surface ($g_1 = U\Delta_1$), and for the total order parameter on the very first layer [$g_s = U(\Delta_0 + \Delta_1)$], respectively. (b) Penetration depth of the surface order parameter as a function of the temperature. Above the critical temperature of the surface, z_0 loses meaning. All temperatures are rescaled to the bulk critical temperature T_c^{Bulk} .

the coupling strength U of the bulk from the equation

$$1 = \frac{U}{2} \sum_{\pm} \int \frac{d^3k}{(8\pi^3)} \frac{1}{\Lambda_{\pm}} \left(1 - \frac{2e^{-\beta\Lambda_{\pm}}}{1 + e^{-\beta\Lambda_{\pm}}} \right), \quad (\text{D9})$$

where now $\pm$ refer to the two positive BdG eigenvalues $\Lambda_{\pm} = \sqrt{(\Xi_{\mathbf{k},k_z} \pm \mu)^2 + U^2\Delta_0^2}$, where $\Xi_{\mathbf{k},k_z}$ refers to the eigenvalues of the bulk Hamiltonian without taking into account the chemical potential, and where we already summed over the two degenerate BdG eigenvalues due to the two spin-branches.

Using a value of U that opens a gap within the bulk of $g_0 = 10^{-3}$, we can perform the self-consistent calculation of the surface free energy with the analytical value of the eigenvalues and the penetration function $f_{\mathbf{k}}(z_0)$. The results are collected in Fig. 6. In this case, we obtain an order parameter on the surface which is more than 10 times higher than the one in the bulk, as in the PtBi_2 case, showing that such an enhancement of the critical temperature is within reach of an explanation based on the surface states as we propose here.

APPENDIX E: GENERALIZED VARIATIONAL ANSÄTZE

The main advantage of the method we introduce in this work is that it is easily scalable to arbitrarily large systems; a direct numerical solution for the self-consistency of a spatially varying order parameter would otherwise be quite challenging to obtain, especially when multiple degrees of freedom and a large number of layers is considered. The principle by which we circumvent the difficulty is to incorporate the

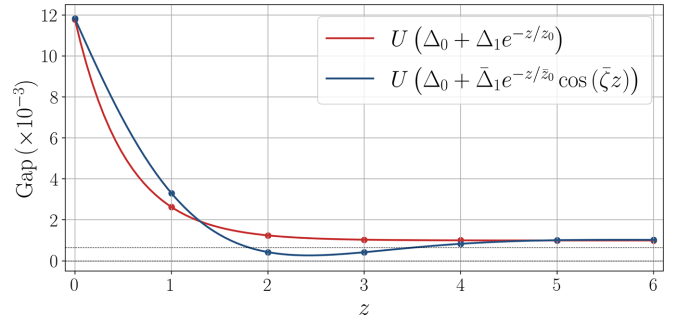


FIG. 7. Superconducting gap as a function of the layer position using (red) exponentially decaying and (blue) exponentially and oscillatory decaying surface order parameters at zero temperature. The value that minimizes the free energy is $\Delta_1 = 7.17 \times 10^{-3}$, and $z_0 = 0.56$ for the first model (red) and $\tilde{\Delta}_1 = 7.17 \times 10^{-3}$, $\tilde{z}_0 = 0.98$, and $\tilde{\zeta} = 0.92$ for the second model (blue). The values of U and Δ_0 refer to the minimum found in Appendix D.

space dependence in a small number of variational parameters which are *intensive*, independent of the size of the system. The main advantage that such a method offers is that it can be easily improved in precision by constructing generalized variational *Ansätze* for the order parameter. In this Appendix, we perform such a test for the tight-binding model introduced in Appendix D.

Throughout the main text and the Appendixes, we have restricted ourselves to an exponential *Ansatz* for the spatially varying part of the order parameter $\Delta = \Delta_0 + \Delta_1 e^{-z/z_0}$. A simple nonexponential behavior that can be constructed involves an additional modulation by means of an oscillating factor $\Delta = \Delta_0 + \Delta_1 e^{-z/z_0} \cos(\zeta z)$. Such oscillations might not be entirely unexpected, since the individual eigenstates generally exhibit a combined exponential and oscillating behavior in space [24]. To test for this more general *Ansatz* for the order parameter, we follow the same strategy as in the main text with an additional variational parameter ζ .

Figure 7 shows the minimum-free-energy solution for the order parameter at zero temperature. We compare the generalized solution, in which we minimize the surface free energy over Δ_1 , z_0 , and ζ , with the simple exponential *Ansatz*, in which ζ is fixed to 0 and the other parameters Δ_1 and z_0 minimize the free energy. We find that a slow oscillation that modulates the spatial variation is indeed preferred, yet the qualitative picture remains the same as in the simple exponential *Ansatz*. This retroactively justifies our results in the main text at the qualitative level; on the other hand, it also makes it clear that the order parameter in a realistic model can penetrate with a nontrivial structure within the surface layers of the crystal. The method we propose here offers a systematic way to obtain precise theoretical predictions for this structure, which is encoded in the variational parameters of the *Ansatz*.

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