Electrically Tunable Superconductivity Through Surface Orbital Polarization

Maria Teresa Mercaldo,1 Paolo Solinas,2 Francesco Giazotto,3 and Mario Cuoco4,1
1Dipartimento di Fisica “E. R. Caianiello”, Università di Salerno, IT-84084 Fisciano (SA), Italy
2SPIN-CNR, Via Dodecaneso 33, 16146 Genova, Italy
3NEST, Istituto Nanoscienze-CNR and Scuola Normale Superiore, Piazza San Silvestro 12, I-56127 Pisa, Italy
4SPIN-CNR, IT-84084 Fisciano (SA), Italy

We investigate the physical mechanisms for achieving an electrical control of conventional spin-singlet superconductivity in thin film by focusing on the role of orbital polarization occurring at the surface. Assuming a multi-orbital description of the metallic state, due to screening effects, the electric field acts by uniquely modifying the strength of the surface potential and, in turn, the amplitude of the orbital Rashba coupling due to the enhanced inversion asymmetry effects. The resulting orbital polarization at the surface and in its close proximity is shown to have a dramatic impact on superconductivity. We demonstrate that, by varying the strength of the induced asymmetric interactions, the superconducting phase can be either suppressed, i.e. turned into normal metal, or undergo a 0–π transition with the π phase being marked by non-trivial sign change of the superconducting order parameter between different orbitals. These findings unveil a rich scenario to design heterostructures with superconducting orbitronics effects.

Introduction — Because of the screening effect, a static electric field cannot penetrate inside a metal deeper than a few Thomas-Fermi lengths (0.1–1 nm) [1–3]. As a consequence, the behaviors and features of a metal, e.g., its transport properties, are practically unaffected by the application of static electric fields.

Analogously, the interaction of a static electric fields with a superconductor is a long time standing problem that has been discussed in several papers with many different approaches [4–7]. It turns out that for standard metallic superconductors, that are well described by the Bardeen-Cooper-Schrieffer theory [8, 9], the penetration length of an electrostatic field is roughly unchanged with respect to the normal metal phase [10]. Still, recent experiments have shown that a strong static electric field can dramatically affect the properties of superconducting wires and planes [11–15] suppressing the dissipationless critical current, and inducing a superconductor-to-normal metal transition. This superconducting field effect (SFE) seems to be quite ubiquitous since it has been observed in different materials [11], in Dayem bridges [12, 13], in superconductor-normal metal-superconductor mesoscopic junctions [14], and in superconducting quantum interference devices [15]. These experimental evidences suggest that the SFE is a genuine new phenomenon rooted in the foundations of superconductivity which cannot be explained or framed in terms of other well-known effects such as, for instance, charge accumulation or depletion [13, 15].

First, to reconcile the assumption of short electric field penetration with the destruction of superconductivity on larger scales, we must invoke a key-feature of superconductors: the long-range spatial correlation typical of Cooper pairs. Since the Cooper pairs forming the superconductor condensate are correlated over distances (r0) much longer than the electric field screening length (e.g., 100 nm for titanium [11]), any perturbation occurring on the edge of the superconductor is felt and affects the system within a distance comparable to the pair correlation length. This vision seems to be confirmed by the fact that the SFE is observable only on structures with characteristic dimensions of a few coherence lengths, and then vanishes exponentially [11]. Besides this, our understanding of the physics at the origin of the SFE is somewhat limited. A few phenomenological models [11, 13, 15] have been proposed so far but they are rather incomplete by nature, and a fully microscopic theory allowing a deeper insight into the SFE and to exploit its potential is still missing.

Motivated by the above experimental results [11–15], in this Letter we propose a theoretical model which is able to grasp some of the observed features typical of the SFE and to provide a physical scenario to account for the modification of the superconducting order parameter due to the applied electric field at the surface. Our key idea is to consider the effects of the electric field as a source of inversion symmetry breaking at the surfaces of the superconductor and to focus on the consequences of the induced orbital polarization on the electron pairing. It has been recently recognized that an orbital analogue of the spin Rashba effect [16] can be achieved on the surfaces [17–19] even in the absence of atomic spin-orbit coupling [20]. The emergent orbital Rashba (OR) interaction allows for mixing of orbitals on neighboring atoms that would not overlap in an inversion symmetric configuration. For instance, in an electronic tight-binding description of a layered system with p− bands, px and py orbitals would couple to the pz due to the inversion asymmetric surface potential and atomic displacements that lead to unequal overlap of the p|x/p| and p|y with the positive and negative lobes of the p|z configuration (similar considerations apply for d-orbitals). Such coupling generally leads to non-vanishing orbital polarization that form chiral patterns in the momentum space. Remarkably, the OR coupling is quite ubiquitous in metals and semiconductors since it occurs either in pure p- and d-orbitals [17–19] or sp- or pd-hybridized systems [20]. For instance, evidences of anomalous electronic splitting and of the fundamental role played by the orbital degrees of freedom have been found on a large variety of surfaces, i.e. Au(111), Pb/Ag(111) [21], Bi/Ag(111) [22], etc. as well as in transition-metal oxide interfaces [23, 24]. Here, we con-
sider how the induced orbital polarization at the surface is able to significantly modify the amplitude and phase of conventional spin-singlet superconducting order parameter (OP) in thin films. Through a multi-orbital description we show that the electric field can suppress the superconducting state at the surface by inducing a substantial orbital polarization into the electronic states close to the Fermi level. Then, the occurrence of electrically driven orbital-polarized surface states can guide a complete breakdown of the superconducting state in the whole system or an unconventional $0$-$\pi$ transition with a non-trivial sign change of the superconducting order parameter between different bands, resembling the unconventional $s_\pm$ pairing proposed in iron based superconductors [25, 26]. Such phase transitions manifest themselves as a consequence of the interplay of two fundamental electronic processes which can be induced by the electric field in the surface layers whereas it effectively penetrates: i) the orbital Rashba coupling ($a_{\text{OR}}$) due to inversion symmetry breaking and electric field surface potential, ii) an inter-layer hybridization coupling ($\lambda$) that locally breaks time and inversion symmetry and is uniquely active between the surface and the first neighbor layer (Fig. 1). The latter can effectively mimic an orbital dependent current through the Peierls reconstruction of the charge transfer amplitude due to the application of a constant electric field or equivalently via a time dependent vector potential. Our study uncovers fundamental mechanisms for an electrical control of conventional superconductors based on the modification of the orbital polarization at the surface and its mismatch with respect to the orbital configuration in the inner side of the superconductor.

Model and methodology — We simulate the superconducting thin film by assuming a conventional spin-singlet pairing for a geometry with a variable number, $n_z$, of layers (see Fig. 1). In order to assess the impact of the orbital polarization on superconductivity, we consider a basic electronic description based on $d$-orbitals belonging to the $t_{2g}$ sector in a tetragonal symmetry, i.e. $(yz, xz, xy)$, in the presence of an orbital dependent asymmetric coupling at the surface layer. For convenience and clarity of notation we indicate as $(a, b, c)$ the $(yz, xz, xy)$ $d$–orbitals. Then, we introduce the creation $d_{a, \sigma}(\mathbf{k}, i_z)$ and annihilation $d_{a, \sigma}(\mathbf{k}, i_z)$ operators with momentum $\mathbf{k}$, spin ($\sigma = \uparrow, \downarrow$), orbital ($\alpha = (a, b, c)$), and layer $i_z$, to construct a spinorial basis $\Psi^\dagger(\mathbf{k}, i_z) = (\Psi^\dagger_{a, \sigma}(\mathbf{k}, i_z), \Psi^\dagger_{b, \sigma}(\mathbf{k}, i_z), \Psi^\dagger_{c, \sigma}(\mathbf{k}, i_z))$ with $\Psi^\dagger_{\alpha, \sigma}(\mathbf{k}, i_z) = (d^\dagger_{a, \sigma}(\mathbf{k}, i_z), d^\dagger_{b, \sigma}(\mathbf{k}, i_z), d^\dagger_{c, \sigma}(\mathbf{k}, i_z))$. In this representation, the Hamiltonian can be expressed in a compact way as:

$$\mathcal{H} = \frac{1}{N} \sum_{\mathbf{k}, i_z, j_z} \Psi^\dagger(\mathbf{k}, i_z) \hat{H}(\mathbf{k}) \Psi(\mathbf{k}, j_z),$$

with

$$\hat{H}(\mathbf{k}) = \sum_{\alpha = (a, b, c)} [\tau_z \epsilon_\alpha(\mathbf{k}) + \Delta_\alpha(i_z) \delta(i_z, j_z)][(L^2 - 2L^2_z)] \delta(i_z, j_z) + a_{\text{OR}} \tau_z \delta(i_z, j_z) \delta(i_z, n_z)] + \tau_x \delta(i_z, 1) \delta(i_z, n_z) + \lambda \tau_z \delta(i_z, 1) \delta(j_z, 2) \delta(i_z, n_z) \delta(j_z, n_z - 1),$$

where the orbital angular momentum operators $\hat{L}$ have components

$$\hat{L}_x = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & i \\ 0 & i & 0 \end{bmatrix}, \hat{L}_y = \begin{bmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix}, \hat{L}_z = \begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

within the $(yz, xz, xy)$ subspace, and $\tau_z (i = x, y, z)$ are the conventional Pauli matrices for the electron-hole sector. This Hamiltonian is a standard description of layered superconductors with multi-bands at the Fermi level and conventional intra-orbital spin-singlet pairing including inversion asymmetric OR couplings close to the surface. The orbital dependent kinetic energy for the in-plane electron itinerancy is basically due to the symmetry allowed [28] nearest neighbor hopping, thus, one has that $\epsilon_a(\mathbf{k}) = -2||[\eta \cos(k_x) + \cos(k_y)]$, $\epsilon_b(\mathbf{k}) = -2||[\cos(k_x) + \eta \cos(k_y)]$, and $\epsilon_c(\mathbf{k}) = -2||[\cos(k_x) + \cos(k_y)]$, $\epsilon_c(\mathbf{k}) = -2||[\cos(k_x) + \eta \cos(k_y)]$.
FIG. 2. (a) Phase diagram in the $(\alpha_{\text{OR}}, \lambda)$-plane, with the three possible states: conventional superconducting state (0-SC), unconventional (\(\pi\)-SC), and normal state. The parameters used are: \(n_z = 6, \mu = -0.4t, t_z = 1.5t, \eta = 0.1\). (b)-(c) Behavior of the order parameter in the inner side of the system \(\Delta_{\alpha}(\alpha = a, b, c)\) (i.e. in the central layer at \(t_z = n_z/2\)) as function of the surface interlayer coupling \(\lambda\) in the regimes of weak (panel (b)) and strong (panel (c)) orbital Rashba interaction, namely \(\alpha_{\text{OR}} = 0.2t\) and \(\alpha_{\text{OR}} = 3.0t\), respectively. In (b) we observe a sharp transition to an unconventional superconducting state (\(\pi\)-SC), in which there is a sign change in \(\Delta\), and hence a relative \(\pi\)-phase between the \(c\) and \(a, b\) orbital dependent OPs, while in (c) we observe that the three orbital dependent OPs all go to zero. In the insets, the profile of \(\Delta\) along the \(z\) direction is shown for different values of \(\lambda\). In panels (d) and (e) we show the analogous transitions of (b) and (c), but for doubling the size of the system \((n_z = 12)\). In (f) and (g) we present the profile of \(\Delta\) for \(n_z = 30\), for weak and strong \(\alpha_{\text{OR}}\), respectively. Indeed, in (f) we see the sign change (for \(\lambda = 0.3t\) and \(0.4t\)), while in (g) the OP tends to be suppressed by increasing \(\lambda\). In (b)-(g) \(\Delta_0\) is the superconducting OP for a monolayer without the OR and sets its scale [27].

\(-2t_\parallel[\cos(k_y) + \eta \cos(k_z)]\), with \(\eta\) being a term that takes into account deviations from the ideal cubic symmetry. We assume that the layer dependent spin-singlet order parameter is non-vanishing only for electrons belonging to the same band and it is expressed as \(\Delta_{\alpha}(i_z) = \mathcal{N} \sum_{k} \langle d_{\alpha, \uparrow}(k, i_z) d_{\alpha, \downarrow}(-k, i_z) \rangle\) with (...) being the expectation value of the ground state. Here, \(\mathcal{N} = n_x \times n_y\) sets the dimension of the layer in terms of the linear lengths \(n_x\) and \(n_y\), while we assume translation invariance in the \(xy\)-plane and a finite number \(n_z\) of layers along the \(z\)-axis (Fig. 1). The computational analysis is performed by determining the superconducting order parameters corresponding to the minimum of the free energy employing a self-consistent iterative procedure until the desired accuracy is achieved. We assume the planar hopping amplitude as energy unit, \(t_\parallel = t\), while the interlayer hopping is independent of the orbital index, i.e. \(t_{\perp, \alpha} = t_{\perp}\), and the pairing coupling is \(g = 2t\). Variation of the parameters do not affect the qualitative outcomes of the analysis. It is worth pointing out that, due to symmetry arguments, a similar model description can be also obtained for \(p\)-orbitals or in \(sp\)- or \(pd\)-hybridized systems.

**Results** — The effect of the orbital Rashba coupling is to induce an orbital polarization at the surface and to form chiral orbital textures in the Brillouin zone close to the Fermi level (see Supplemental Material (SM) [27]). Moreover, the orbital polarization is generally associated to an orbital configuration with non vanishing angular momentum components and thus it tends to reduce the superconducting OP amplitude [27] because activates orbital mixing which are not compatible with the intra-orbital attractive interaction. On the other hand, both the interlayer electronic processes, i.e. \(\lambda\) and \(t_\perp\), allow for a transfer of the orbital polarization into the inner layers of the superconducting films [27]. Moreover, due to the symmetry breaking for complex conjugation of the orbital processes induced by \(\lambda\), there is also a tendency to link the surface electronic couplings with the development of an orbital dependent phase of the superconducting order parameter. Such relation indeed can be deduced by employing an orbital dependent gauge transformation at the layers which are involved in the \(\lambda\) coupling. The combination of these processes hence indicates that the surface couplings can significantly alter the superconducting phase of the whole system.

To get more insight into such competition it is instructive to start with the phase diagram of the heterostructure for the \(n_z = 6\) multilayer, in terms of the orbital Rashba coupling \(\alpha_{\text{OR}}\) and the surface inter-layer interaction \(\lambda\) (Fig. 2(a)) for a representative value of the electron hopping along the out-of-plane.
direction \((t_\perp = 1.5 t)\). The evolution of the conventional superconducting state \((0-SC)\) clearly indicates that, depending on the strength of the orbital Rashba coupling, one can end up in two distinct phases: i) for weak \(\alpha_{OR}\) an unconventional \(\pi-\)phase with the emergence of a non-trivial phase relation between the orbital dependent OPs, ii) a normal metal configuration with vanishing superconducting OP in the regime of large \(\alpha_{OR}\). The nature of the phase transitions can be tracked by following the layer and orbital dependent behavior of \(\Delta_n(i_{\perp})\). In the regime of weak \(\alpha_{OR}\) the increase of \(\lambda\) cannot lead to a breakdown of superconductivity. Instead, we find that there is a first order phase transition [27] between two superconducting phases with a reorganization of the relative phase between the orbital dependent pairing amplitudes. As demonstrated in Fig. 2(b), at a critical value of \(\lambda\) the superconducting order parameter associated to the band \(c\) undergoes a first order phase transition with an abrupt sign change of \(\Delta_n(i_{\perp})\) in all the layers (see inset Fig. 2(b)) while the other two OPs exhibit a discontinuous variation of the amplitude which is sign conserving. The sign change of the superconducting OP for one of the band implies an inter-orbital \(\pi\)-phase between the electron pairs within the \((a,b)\) and \(c\) orbitals. Such an orbital reconstruction of the superconducting state is an evidence of an unconventional pairing which can remarkably manifest in an anomalous Josephson coupling with non-standard current-phase relations especially in the presence of other sources of inhomogeneity (e.g. disorder, crystalline domains, etc.). The fact that the band \(c\) undergoes a sign change of the OP with respect to the \(a,b\) bands is a consequence of the structure of the asymmetric inversion couplings at the interface which allow for orbital mixing between \((a,c)\) and \((b,c)\) configurations. The presence of competing phases is also evident if one considers the free energy dependence on the superconducting OP. Indeed, in order to catch the main competing mechanisms, one can assume a uniform spatial profile as a function of the layer index by allowing for an orbital dependent phase reconstruction of the type \(\Delta_{a,c}(i_{\perp}) = \exp[i\varphi_{a,c}]\Delta_0\). Hence, one can directly observe two distinct minima in the free energy, associated with the 0- and \(\pi\) phases, whose relative energy difference can be tuned by varying the amplitude of \(\lambda\) (see SM for details [27]).

Moving to a larger value of the orbital Rashba coupling (i.e. \(\alpha_{OR}/t \geq 1\)) the surface inter-layer coupling \(\lambda\) is able to suppress the superconducting state by vanishing the OP amplitude (Fig. 2(c)). The value of the critical \(\lambda\) setting the 0-SC/normal boundary has a maximum at \(\alpha_{OR}/t \sim 1\) and then stays about unchanged by further increasing the orbital Rashba coupling. Such behavior is accompanied by a qualitative change of the superconducting order parameter at the surface which starts to get reduced once \(\alpha_{OR}\) induces a sufficiently large orbital polarization nearby the Fermi level [27]. This is consistent with the fact that the pairing is optimized in a configuration with maximal value of the quadratic component of the orbital momentum, \(\hat{L}_{\perp}^2\), rather than the orbital polarization induced by the linear terms in \(\hat{L}\). The decrease of the superconducting OP at the bottom and top layers of the superconducting thin film, then, is the seed to drive the complete suppression of the superconducting state within the whole system. The 0-SC/normal metal phase transition appears to be continuous and it occurs about simultaneously for all the orbitals involved in the pairing close to the Fermi level (Fig. 2(c)). It is interesting to notice that a closer inspection of the free energy profile with suitably selected boundary conditions of the OPs at the surfaces and uniform spatial profile in the other layers indicates a smeared type of phase transition from superconductor-to-normal state with weak first order precursors due to the competition between OP configurations with inequivalent amplitude [27]. This implies that the breakdown of the superconducting state, as driven by \(\lambda\), cannot be directly linked to that which can be obtained in a standard BCS thermal evolution of the OP.

At this stage, after having fully addressed the most favorable superconducting configurations in a thin film with \(n_z = 6\) layers, we consider whether the orbital asymmetric potential at the surface is able to be also effective in thicker layered films. Such issue is accounted by simulating the cases with \(n_z = 12\) and \(n_z = 30\). In Figs. 2(d),(e) we demonstrate that for two representative values of \(\alpha_{OR}\), corresponding to weak and strong orbital Rashba couplings, the surface interlayer interaction is able to induce the 0-\(\pi\) and superconductor-normal metal phase transitions. The phase diagram and the effects are then confirmed and observable either for doubling the system size, \(n_z = 12\) (Figs. 2(d),(e) or for superconducting thin film with \(n_z = 30\) layers (Figs. 2(f),(g)). However, one remark is relevant here concerning the amplitude of the kinetic energy along the \(z\)-axis. Indeed, to drive the superconducting state one needs to adjust the inter-layer hopping amplitude. In this way, the critical boundaries occur in the same range of strengths for the surface couplings as for the case of thinner superconductors. Such study in terms of the superconducting thin film thickness indicates that the obtained phenomenology is also dependent on the out-of-plane kinetic energy and, in turn, on the superconducting coherence length along the \(z\)-axis. Although we are dealing with a superconductor with a discrete number of layers, it is plausible to expect that the effective coherence length along the \(z\)-direction is proportional to the out-of-plane Fermi velocity, and thus one can qualitatively argue that it scales with the amplitude of the inter-layer hopping. In this context, we point out that the reduction of the inter-layer hopping can significantly alter the phase diagram with the normal state region being replaced by the \(\pi\)-SC configuration [27].

Conclusions and discussion — We have demonstrated that by tuning the orbital polarization on the surface of conventional superconducting thin films and the surface inter-layer communication channels one can control both the amplitude and the phase of the superconductor. Remarkably, starting from the idea that the electric field is responsible of the surface orbital Rashba coupling whose strength is proportional to the amplitude of the applied field, we unveil how orbital chiral textures can lead to a complete reconstruction of the superconducting state. We have identified two fundamental electronic
processes which, although active only at the surface, can account for the observation of a superconducting-normal metal transition. The character of the phase transitions is an important mark of the orbitally driven scenario. Indeed, while the $0-\pi$ phase change is first-order like, the transition from superconductor to normal metal includes distinct features such as weakly first-order precursors of the superconducting OP before it continuously goes to zero. Another striking aspect is represented by the possibility of electrically achieve an unconventional superconducting state with an intrinsic $\pi$-phase difference as it emerges when the OR coupling is smaller than the inter-layer coupling acting at the surface. This state can manifest an anomalous Josephson coupling along the $z$-direction due to layer dependence of the $\pi$-phase, and also for the in-plane behavior in the case of inhomogeneous thin films. We expect that evidences of this state can be observed by phase sensitive superconducting interferometry\cite{15}. Another important remark concerns the $\pi$-phase and the role of disorder. In the presence of non-magnetic impurity potential, as for other unconventional superconductors, the induced inter-orbital scattering between bands having opposite sign in the order parameters will tend to suppress the overall superconducting state with a resulting $\pi$-SC to normal phase transition.

The observation of the obtained phase transitions is also strongly linked to the character of the electron itinerancy of the superconducting thin film and, consequently, also to its thickness. We have indeed verified that one needs to be in a regime where the inter-layer kinetic energy is comparable to the planar one. Furthermore, the energy scales of the inversion asymmetric potentials at the surface for achieving the transitions are of the same order of magnitude of the kinetic energy. This observation sets a clear reference for the electrically and orbitally tunable of the superconducting-normal phase transition. We point out that assuming $\lambda$ and $\delta_{OR}$ to be proportional to the applied electric field, and with $t_{ij} \sim 100$ meV, one would get that an electric field $\sim 30$ meV/Å would suffice to observe the superconducting phase transitions, which is in the range of the experimental observations\cite{11-15}. Finally, the presented results indicate distinct paths for designing devices with electrically tunable superconducting orbitronics effects. In particular, we foresee a significant potential and impact for the design of heterostructures with few layers of strong orbital Rashba coupled material deposited on the surface of conventional superconductors with superconducting field effects phenomena that can be magnified and suitably controlled.

FG acknowledges the European Research Council under the European Unions Seventh Framework Programme (COMANCHE: European Research Council Grant No. 615187) and Horizon 2020 and innovation programme under grant agreement No. 800923-SUPERTED.

[1] N. Ashcroft and N. Mermin, *Solid State Physics* (Saunders College, Philadelphia, 1976).
In this Supplemental Material we start describing the impact of the orbital Rashba coupling on superconductivity for a monolayer configuration. Then, for a representative multi-layered superconductor we investigate the role of the inter-layer hopping in tuning the spatial dependence of the order parameter and the related consequences on the phase diagram. Hence, we discuss the character of the phase transitions by inspecting the free energy profile. Finally, the behavior of the layer dependent orbital polarization for the $n_z=10$ superconducting film is evaluated in terms of the orbital Rashba coupling and the surface inter-layer interaction.

ROLE OF ORBITAL RASHBA FOR THE SINGLE LAYER

For a single layer, the presence of the orbital Rashba (OR) coupling tends to reduce the strength of the superconductivity by inducing a suppression of the order parameter (OP). This behavior is explicitly demonstrated in Fig. 1, where the superconducting OP amplitude for each band, self-consistently determined, exhibits a monotonous decrease as a function of $\alpha_{OR}$. We have also considered the self-consistent value of $\Delta_{\alpha}$ while changing $\alpha_{OR}$ assuming two different values of the pairing coupling $g$. We find that the amplitudes are scaled by means of the pairing coupling $g$ and thus in the following and in the main text we have performed the calculation assuming $g=2.0t$.

![FIG. 1: Plot of the self-consistent superconducting order parameter $\Delta_{\alpha} (\alpha = a, b, c)$ as function of $\alpha_{OR}/t$ for $g=2.0t$ in a monolayer system. $\Delta_0$ is the pairing amplitude in absence of OR effect (i.e for $\alpha_{OR}=0$) for the orbitally isotropic case ($\Delta_a = \Delta_b = \Delta_c$).](image)

EFFECTS OF INTERLAYER HOPPING

Here, we analyze the influence of the interlayer hopping $t_\perp$ on the order parameter in the superconducting phase. In Fig. 2 we show the profile of $\Delta_{\alpha}(i_z)$ along the $z$ direction at $n_z=6$ for five different values of $t_\perp$, in the case of weak (panels (a) and (b)) and strong (panels (c) and (d)) orbital Rashba coupling. The surface interlayer interaction $\lambda$ is set to zero.

![FIG. 2: Spatial profile of the superconducting order parameter $\Delta_{\alpha}(\alpha = a, b, c)$ along the $z$ direction at $n_z=6$ for five different values of $t_\perp$, in the case of weak (panels (a) and (b)) and strong (panels (c) and (d)) orbital Rashba coupling. The surface interlayer interaction $\lambda$ is set to zero.](image)

The effects of $t_\perp$ can be quite relevant and indeed the phase diagram reported in the Fig.2 (a) of the main text can be modified. A large amplitude of $t_\perp$ with respect to $t$ can destroy the superconducting state, even for small values of $\alpha_{OR}$ and $\lambda$. On the other hand, in the opposite regime of small $t_\perp$ one needs a large amplification of $\lambda$ to get into the normal state. For this circumstance, one can typically obtain only $0/\pi$ superconducting transition. In Fig. 3(a)-(c) we show the behavior of the order parameter in the inner side the system $\Delta_{\alpha}$ (i.e. in the central layer $i_z=n_z/2$) for $t_\perp=0.9t$. We see that both for weak and strong values of the OR interaction the $0-\pi$-SC transition can be achieved.

COMPETING PHASES AND CHARACTER OF THE PHASE TRANSITIONS

In this section, we study the free energy of the examined model Hamiltonian by considering the order parameter $\Delta_{\alpha}(i_z)$
as uniform through the layers and isotropic in the orbital channels. The analysis is done by introducing the variable $\Delta$ which is the common amplitude of the OPs in the various orbital channels, i.e. $|\Delta_a| = |\Delta_b| = |\Delta_c| = \Delta$, while the sign is added when evaluating the $\pi$-SC configuration (i.e. $\Delta_c = -\Delta_{a,b} = -\Delta$).

Representative cases of weak and strong OR effect, namely with $\alpha_{OR} = 0.3t$ and $\alpha_{OR} = 3.0t$ respectively, are reported in Figs. 4. We have considered a system with $n_z = 6$ layers and with interlayer hopping $t_{\perp} = 1.5t$. Similarly to the full self-consistent analysis (see Fig. 2 in the main text), we find that the increase of $\lambda$ drives a transition between 0-SC and $\pi$-SC states for weak $\alpha_{OR}$ (see Fig. 4(a)) and a transition from SC to normal state for strong $\alpha_{OR}$ (see Fig. 4(b)). Indeed, for $\alpha_{OR} = 0.3t$ comparing the panels (c) and (e) of Fig. 4, we see that for $\lambda > 0.16t$ the case with $\Delta_c < 0$ has an energetically more favorable solution. This transition if of first order, since we have a discontinuity in the first derivative of the free energy.

For larger values of $\alpha_{OR}$, the free energy of the case with $\Delta_c < 0$ has a minimum only for $\Delta = 0$ (i.e. normal state solution) as can be easily deduced from Fig. 4(f). Hence, for $\alpha_{OR} = 3.0t$ the system never reaches the $\pi$-SC phase and we observe a continuous transition from SC to normal state by following the free energy minima, as shown in Fig. 4(b) and (d), for $\lambda \simeq 0.21t$. The values of the transition points are slightly different from those reported in the phase diagram (Fig. 2(a) of the main paper), since in the present analysis we are assuming an uniform and isotropic superconducting OP.

Indeed, for strong OR effect, the uniform profile of the OP is not very good since the values of $\Delta_c(i_z)$ in the outer layers are strongly suppressed, compared to those in the inner layers, as can be seen in Fig. 2(c)-(d). Hence, for $\alpha_{OR} = 3.0t$ we have also performed an analysis in which we assume that the order parameter is zero in the outer layers and uniform in the remaining ones. Results are reported in Fig. 5, where we observe the presence of multiple minima and the increase of $\lambda$ drives a weak first order transition before the continuous second order SC-normal transition is achieved.

### LAYER DEPENDENT ORBITAL POLARIZATION

Finally, we present the layer dependent orbital polarization for a superconducting heterostructure with $n_z = 10$. The analysis is performed by considering firstly the role of the OR coupling at the surface and how the obtained orbital polarization in the Brillouin zone is also transferred inside the inner layers (first and third column of Fig. 6). Starting from the case at
FIG. 5: Plot of the energy difference \( E(\Delta) - E(0) \) as a function of the order parameter \( \Delta \) (assuming that \( |\Delta_a| = |\Delta_b| = |\Delta_c| = \Delta \)) for several values of \( \lambda \) close to the critical point for strong OR coupling \( \alpha_{OR} = 3.0t \). The system has \( n_z = 6 \) layers and \( t_{\perp} = 1.5t \). The analysis has been made by assuming that the superconducting order parameter is zero in the outer layers and uniform in the inner ones. In the inset (a) there is a zoom for four values of \( \lambda \), underlying the presence of multiple minima and hence the occurrence of a first order phase transition. (b) Behavior of the order parameter as a function of the orbital mixing term \( \lambda \). After the small discontinuity the second minimum goes smoothly to zero.

\[ \lambda = 0, \text{ one can observe a chiral texture of the orbital components with windings around the high symmetry points of the Brillouin zone (BZ). In particular the winding around the } \Gamma \text{ point at } (k_x, k_y) = (0, 0) \text{ is opposite to that occurring around the } M \text{ point at } (\pi, \pi) \text{ with a domain wall in between the } (0, \pm \pi) \text{ and } (\pm \pi, 0) \text{ points. We observe that moving from the surface to the inner layers, the domain walls proliferate and there are extra structures emerging along the diagonal of the BZ with opposite orbital chirality. We notice that the presence of the orbital Rashba coupling at the surface is sufficient to induce a non-trivial orbital polarization into the inner layers of the superconductor (see first column of Fig. 6).}

The effect of \( \lambda \) is then investigated by evaluating the difference in the orbital texture with respect to the configurations with \( \lambda = 0 \) by keeping the same amplitude of \( \alpha_{OR} \). As one can see in the second and fourth column of Fig. 6, the effect of \( \lambda \) is to amplify the formation of pockets of orbital textures with inequivalent or opposite orientation of the orbital polarization thus indicating an orbital connectivity which is less regular if compared to the case without \( \lambda \). Such structure of the orbital texture in the reciprocal space contributes to reduce the superconducting pairing which is maximally favored for electron pairs without any orbital polarization.
FIG. 6: Vector plots of $\mathbf{L}(k_x, k_y)$ per layer in the Brillouin zone, for a superconducting system with 10 layers, in the case of weak ($\alpha_{OR} = 0.5t$) and strong ($\alpha_{OR} = 3.0t$) orbital Rashba effect. $L_z = 0$ everywhere. In the first and third column we show the vector plots for $\lambda = 0$, while in the second and fourth we show the difference $\Delta L$ between the vectors $\mathbf{L}$ for $\lambda = 0.3t$ and $\lambda = 0$. In each row we present the behavior in each layer labeled by $i_z$. Since the system is symmetric, layers from 6 to 10 are not shown. The arrows are colored according to the magnitude of the vector field (see the legend at the bottom), with intensities that are scaled to unity. In each panel, the magnitude of $\mathbf{L}$ is also represented by the dimension of the arrows.