Unveiling Mechanisms of Electric Field Effects on Superconductors by Magnetic Field Response

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We demonstrate that superconducting aluminium nano-bridges can be driven into a state with complete suppression of the critical supercurrent via electrostatic gating. Probing both in- and out-of-plane magnetic field responses in the presence of electrostatic gating can unveil the mechanisms that primarily cause the superconducting electric field effects. Remarkably, we find that a magnetic field, independently of its orientation, has only a weak influence on the critical electric field that identifies the transition from the superconducting state to a phase with vanishing critical supercurrent. This observation points to the absence of a direct coupling between the electric field and the amplitude of the superconducting order parameter or 2π-phase slips via vortex generation. The magnetic field effect observed in the presence of electrostatic gating is well described within a microscopic scenario where a spatially uniform inter-band π-phase is stabilized by the electric field. Such an intrinsic superconducting phase rearrangement can account for the suppression of the supercurrent, as well as for the weak dependence of the critical magnetic fields on the electric field.

In recent years, it has been shown that the superconducting (SC) properties of metallic Bardeen-Cooper-Schrieffer (BCS) superconductors can be significantly influenced via electrostatic gating [1]. The most striking effects: reduction and suppression of the critical supercurrent, have been broadly demonstrated in metallic nanowires [2] and Dayem bridges [3–5] made of titanium, aluminium and vanadium, as well as in aluminium–copper–aluminium Josephson junctions [6]. Moreover, recent experiments have probed the effect of electrostatic gating on the SC-phase in a SQUID [7], and on the nature of the current switching distributions in gated titanium Dayem bridges [8].

While these observations clearly indicate that the electric field can suppress the supercurrent, whether and how it acts on the amplitude or the phase of the SC order parameter is a question that has not yet been fully settled. In order to get a deeper insight into this fundamental problem we carefully investigate how the SC state is modiﬁed by the simultaneous presence of electric and magnetic ﬁelds. In this context, probing both the in-, and out-of-plane magnetic ﬁelds (B_Y and B_Z, respectively) is particularly relevant because the two orientations affect the SC thin ﬁlms via very different mechanisms [9]. In thin ﬁlms B_Z generally leads to screening currents and a spatially varying order parameter, marked by 2π-phase slips, as vortex cores penetrate the sample. On the other hand, B_Y ideally affects only the Cooper pairing amplitude homogeneously via the electron spin paramagnetism, inducing pair breaking and spin polarization [10, 11]. Thus, the search for magneto-electric cross-talking effects in SC thin ﬁlms can provide indications and constraints on the quantum states at SC breakdown, and reveal the origin of the unexpected coupling between electric ﬁeld and SC phase and/or pairing amplitude.

In this Letter, we demonstrate that SC Al nano-bridges can be electrically driven into a state with complete suppression of the critical supercurrent. The magnetic ﬁeld response of Al nano-bridges is investigated with the aim to single out the nature of the phase transitions and the mechanisms that mark the electric ﬁeld effects in metallic superconductors. Remarkably, we find that the magnetic ﬁeld has only a weak influence on the electric ﬁeld effect in the SC Dayem bridges. Moreover, this phenomenology is starkly independent on the magnetic ﬁeld orientation, despite the very different interactions between SC thin ﬁlms and in-, and out-of-plane magnetic ﬁelds. These ﬁndings point to the absence of a direct electric coupling between the electric ﬁeld and the amplitude of the SC order parameter or 2π phase slips generated by vortices. Both cases, in fact, would have manifested with a signiﬁcant variation of the SC/normal (N) critical boundaries due to magnetic ﬁelds. Instead, our observations are consistent with a model in which the surface electric ﬁeld is a source of inversion-symmetry breaking that strongly affects the orbital polarization only at the surface layers of a multi-band SC thin ﬁlm. [12] This results in an electric-ﬁeld-driven phase transition into a mixed SC state with a rearrangement where the relative SC phases between different bands are shifted by π. This state is hardly inﬂuenced by the applied magnetic ﬁeld, as shown in the phase diagram, thus capturing the main experimental ﬁndings.

In addition to the fundamental aspects discussed above, the full supercurrent suppression observed in our Al-based devices (up to know only a 35% reduction was achieved for Al [2]), makes our results have a large technological impact. Considering the broad application of Al-based thin ﬁlms as SC qubits [13], Josephson devices [14–16], photon detectors [17, 18] and bolometers [19], one can easily envision a new generation of SC electronics that can fully exploit the demonstrated SC electric ﬁeld effects.

Results — The samples were fabricated in a single step employing electron-beam lithography to pattern a resist mask on a sapphire substrate, see Fig. 1a. Consequently 3 nm of Ti were evaporated as a sticking layer, followed by 14 nm of Al, in an electron beam evaporator with a base pressure of \( \approx 10^{-11} \) torr. The Dayem bridge is approximately 120 nm wide, 100 nm long, and has a normal-state resistance \( R_N \approx 25 \Omega \). Gate-bridge separation is approximately 30 nm, and the leads on either side of the bridge are 2 µm wide. Re-
siderable hysteresis at low temperature (see the blue dots in Fig. 1b and the lines in Fig. 1d), with a retrapping current $I_R \approx 4.2 \mu A$ at $T = 50$ mK. The hysteresis is likely thermal in origin [27–29], and it disappears when $T > 400$ mK, which is consistent with an enhanced thermalization mediated by phonon coupling.

As in similar experiments[1, 2, 4–8], the critical current can be reduced, up to complete suppression at the critical gate voltage $V_{CG} \approx 23$ V. This is shown in Fig. 1c, for several temperatures. The effect is bipolar in $V_{Gate}$ (not shown here) and is consistent with what has been reported previously for different materials [1]. $V_{Gate}$ has little to no effect at low values until a sudden decrease close to $V_{CG}$. At higher temperatures, the region where $V_{Gate}$ has little effect widens, while $V_{CG}$ is unaffected. In Fig. 1d, we show four selected IV curves for different $V_{Gate}$, taken at $T = 50$ mK. In line with previous field effect experiments, the retrapping current $I_R$ is not affected by $V_{Gate}$ until it coincides with $I_C$ (see SM). Above $V_{CG}$, some residual non-linearity is still observable, before the device becomes completely ohmic (see the 24 V line in Fig. 1d). [1]

While the critical current is easily identified when the switch to the normal state is abrupt, this is less evident when $I_C$ is close to zero as the transition is more gradual. For this reason, we have defined $I_C$ as the value of the bias current $I$ for which the differential resistance is larger than 10 $\Omega$, which is of the same order of magnitude as the normal state resistance $R_S \approx 25 \Omega$, and can be reliably identified over the background noise. Unlike the switching process, the retrapping generally does not occur in one step, but tends to happen in two successive events (see e.g. the 18 V line in Fig. 1d). The exact origin of this ‘partial’ switching is not yet fully settled, but it is likely related to two local thermalization processes taking place in different regions of the device.

The out-of-plane critical in-plane field versus temperature follows the phenomenological profile $B_T(T) = B_Z(T) = 0/(1 − (T/T_C)^2)$ [30], which yields $T_C = 507$ mK and $B_Z(T = 0) = 16.25$ mT (see Fig. 2a and SM [20]). At $T = 50$ mK, the critical field $B_Y \approx 850$ mT. From the two critical magnetic fields, we estimate the London penetration depth $\lambda_L = B_N / \sigma L_C = 160$ nm [30] via the Ginzburg-Landau theory using $d = 17$ nm. Since the thickness of the SC film is $d \ll \lambda$, it is reasonable to assume that the in-plane field $B_Y$ penetrates the superconductor completely. Indeed, the critical in-plane field’s temperature dependence $B_Y(T)$ is consistent with the evolution of a spin-split BCS condensate with a critical Zeeman field near the Clogston-Chandrasekhar limit $\mu_B H_C = \Delta_0 / \sqrt{2}$ [11, 31, 32] (see Fig. 2d and SM [20]).

The simultaneous application of magnetic and electric fields is shown in Fig. 2, where the evolution of $I_C$ is plotted as a function of both temperature $T$ and out-of-plane magnetic field $B_Z$. The reduction of $I_C$ is monotonous in $B_Z$ and $T$. At low temperatures, there is a noticeable ‘kink’ in the $I_C$ vs. $B_Z$ curves, that disappears when $T > 150$ mK or when the supercurrent is significantly depressed via $V_{Gate}$. This behavior was not observed in a similar device, and could be related to fluxoid pinning inside or near the constriction.

FIG. 1. a) A false color SEM image of a typical device. Inset: a close up of the region indicated by the black square shows the Dayem bridge and gate electrode. b) Critical current $I_C$ and retrapping current $I_R$ versus temperature. $I_C$ follows the typical BCS evolution (gray line), see also the main text. c) $I_C$ versus gate voltage $V_{Gate}$ at four different temperatures. d) Voltage drop across the bridge versus bias current, for four values of $V_{Gate}$ at 50 mK. Sweep direction is indicated by the arrows, and the curves are horizontally offset for clarity. The black arrows in c) indicate the position of the curves with $V_{Gate} = 18$, 21 and 24 V.

Using the BCS relation, we find that $\Delta_0 = 1.764 k_B T_C = 91 \mu eV$ ($k_B$ being the Boltzmann constant). Here, $T_C$ is relatively low for Al, most likely due to an inverse proximity effect from the Ti layer. Via the conductivity $\sigma$, $\Delta_0$ and the magnetic permeability of the vacuum $\mu_0$, we can estimate the London penetration depth $\lambda_L = \sqrt{\hbar / \mu_0 \pi \sigma \Delta_0} \approx 100$ nm, and the superconducting coherence length $\xi_0 = \sqrt{\hbar \sigma / N e^2 \Delta_0} \approx 170$ nm. Here we take the electron density at the Fermi energy of aluminum to be $N_F = 2.15 \times 10^{27}$ J$^{-1}$ m$^{-3}$ [21–23].

Fig. 1b shows the critical and retrapping currents versus temperature. At the base temperature of $50$ mK, the critical current $I_C \approx 12.8 \mu A$. The evolution of $I_C$ as a function of temperature follows the conventional Bardeen’s profile [24–26] $I_C \approx I_C \left[1 - \left( \frac{T}{T_C} \right)^2 \right]^{3/2}$. The IV characteristics show a considerable hysteresis at low temperature (see the blue dots in Fig. 1b and the lines in Fig. 1d), with a retrapping current $I_R \approx 4.2 \mu A$ at $T = 50$ mK. The hysteresis is likely thermal in origin [27–29], and it disappears when $T > 400$ mK, which is consistent with an enhanced thermalization mediated by phonon coupling.

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FIG. 2. a) 3D plot of $I_C$ as a function of the out-of-plane magnetic field $B_Z$ and temperature $T$. The full black line shows $I_C(T)$ at zero field, while the dashed black line shows the critical field $B_d^C$ versus temperature. b) 3D plot of $I_C$ as a function of the out-of-plane magnetic field $B_Z$ and the gate voltage at 50 mK. The full black line indicates $I_C(V_{\text{Gate}})$ at zero field, while the dashed black line indicates the critical field $B_d^C$. c) Critical $B_d^C$ versus $V_{\text{Gate}}$ at $T = 50, 250$ and 350 mK. The error bars indicate the resolution in $B_d$. d) 3D plot of $I_C$ as a function of the in-plane magnetic field $B_Y$ and temperature $T$. The full black line shows $I_C(T)$ at zero field, while the dashed black line shows the critical field $B_d^C$ versus temperature. e) 3D plot of $I_C$ as a function of the in-plane magnetic field $B_Y$ and the gate voltage at 50 mK. The full black line indicates $I_C(V_{\text{Gate}})$ at zero field, while the dashed black line indicates the critical field $B_d^C$. f) Critical field $B_d^C$ versus $V_{\text{Gate}}$ at $T = 50, 250$ and 350 mK. The error bars indicate the resolution in $B_d$.

It is interesting to compare the dependence of $I_C$ on $T$ and $B_Z$, with the dependence on $V_{\text{Gate}}$ and $B_Z$, which is presented in Fig. 2b. While the critical magnetic field $B_d^C$ decreases continuously with temperature, the same is not true for $V_{\text{Gate}}$. For $V_{\text{Gate}} < 17$ V, the dependence of $I_C$ on $B_Z$ is unaffected. Only when $V_{\text{Gate}}$ exceeds this value, we see a reduction in both $I_C$ and a sharp decrease of $B_d^C$. The dependence of $B_d^C$ on $V_{\text{Gate}}$ is shown in Fig. 2c, for three different temperatures. Even at higher $T$, the onset of the reduction of $B_d^C$ is not significantly changed.

The complete evolution of $I_C$ as a function of both $T$ and $B_Y$ is shown in Fig. 2d. Analogous to the effect of $B_Z$, $I_C$ is reduced monotonously. Also the behavior of $I_C$ versus $T$ and $V_{\text{Gate}}$ is similar; for $V_{\text{Gate}} < 17$ V, the dependence of $I_C$ on $B_Y$ is not significantly affected. Figure 2f depicts the evolution of the critical magnetic field $B_d^C$ versus $V_{\text{Gate}}$ for several $T$. For neither $B_Z$ nor $B_Y$ does the relation between $I_C$, $B$ and $V_{\text{Gate}}$ depend on the sign of either $B$ or $V_{\text{Gate}}$.

Model — In order to capture the effects of magnetic and electric fields we introduce a microscopic model to simulate multiband thin film superconductivity. Conventional BCS spin-singlet pairing and a multilayered geometry is assumed. The electric field enters as a source of inversion symmetry breaking, introducing an orbital Rashba (OR) coupling for the intra-layer and inversion asymmetric inter-layer electronic processes, yet only at the surface layers due to screening effects [12]. Indeed, the electric field tunes the strength of the interactions that break inversion symmetry at the surface, and creates an electronic coupling that induces an orbital polarization at the Fermi level, that would not exist in an inversion symmetric environment. The details of the microscopic model are described in the SM [20]. Here, for our purposes it is sufficient to recall that the surface layer OR coupling, indicated as $\alpha_{OR}$, and the surface inter-layer inversion asymmetric interaction $\lambda$, are the key electronic parameters by which the electric field influences the SC state [12]. Starting from the zero magnetic field configuration, the coupling $\lambda$ can drive transitions of the type $0-\pi$ (i.e. conventional-to-unconventional superconducting phase) or superconducting-normal depending on whether the $\alpha_{OR}$ coupling is smaller or comparable to the planar kinetic energy scale set by the hopping amplitude $t$ [12]. Here, the $\pi$-phase means that the SC order parameter in a given band has a different sign with respect to that in the other bands contributing to the pairing at the Fermi level. Instead, in the 0-phase there is no phase difference among the bands. Within our modelling a minimal set of three bands (i.e., $a, b, c$) is sufficient to simulate OR effects, thus in the $\pi$-phase we have that $\Delta_a = \Delta_b = -\Delta_c$. We start from a representative case with $\alpha_{OR} = 0.2 t$ at zero temperature (Fig. 3). For $B = 0$ the superconductor undergoes a 0-\pi transition above a critical $\lambda$ which mimics the effect of the applied electric field. As expected, when considering a non-vanishing Zeeman field $B$, the superconductor transitions into a normal state if $B$ exceeds a critical field $B_C$. This SC-Normal transition is also obtained
in the presence of a non-vanishing $\lambda$. Remarkably, both the 0-$\pi$ phase boundary and the critical lines separating the 0- or $\pi$-phases from the normal state shows a weak interplay between the electric and magnetic fields (Fig. 3(a)). Indeed, $\lambda_c$ does not exhibit significant changes as a function of the magnetic field $B$, except for close to the transition point. A similar behavior is also observed for $B_C$. The phase diagram is determined by evaluating the behavior of the free energy at a given magnetic field for the 0 and $\pi$ phases [Fig. 3(b)-(c)]. While the free energy minimum of the 0-phase is strongly affected by the electric field, via $\lambda$, the $\pi$-phase is more resilient and at $\lambda \sim \lambda_c$ there is a transition from 0- to $\pi$-phase due to the crossing of the corresponding free energies (Fig. 3(d)). This transition is starkly unaffected by the magnetic field $B$ and it varies only close to the critical point where both 0- and $\pi$ phases can be brought into the normal state. The weak dependence of $\lambda_c$ on the magnetic field can be ascribed to the character of the $\pi$-phase, characterized by only a rearrangement of the relative phases between the band-dependent SC order parameters, while their amplitudes do not significantly vary across the transition. Instead, when considering the transition from the 0-SC to N state by varying $\lambda$ at a larger value of the $a_{OR}$ coupling one finds that the effective critical electric field is strongly dependent on the intensity of the magnetic field (see SM [20]). Furthermore, we have also investigated the evolution of the phase diagram in temperature. The 0-$\pi$ phase boundary is unaffected by the temperature, indicating that the electric field threshold is not altered by thermal effects until reaching the critical superconducting temperature.

Comparing the above theoretical results with the experimental observations we argue that the $\pi$-phase can qualitatively reproduce the phenomenology of the magnetic field response of the SC nano-bridges in the presence of an electrostatic gating. We note that in the $\pi$-phase, the presence of inter-band $\pi$-phase slips can naturally account for a suppression of the supercurrent, due to a cancellation between positive and negative pair currents in disordered metallic films. Moreover, since the $\pi$-phase does not exhibit spatial modulations or gradients of the superconducting order parameter we expect a weak influence from the formation of a vortex phase, as induced by the out of plane magnetic field $B_z$. Thus, this supports the observation that the electric field is able to disrupt the superconducting state by primarily inducing $\pi$-phase slips between the electronic states that contribute to the pairing at the Fermi level. This remark is also consistent with the enhancement of non-thermal phase fluctuations that have been recently observed in the switching current distributions of Ti Dayem bridges.[8]

**Summary** — We have investigated the suppression of supercurrent effected by the electric field combined with and in-plane, or out-of-plane magnetic field, and ascertained that the two are weakly coupled: the critical magnetic fields are only affected for gate voltages close to the critical gate voltage. These findings are consistent with a microscopic, multiband model recently proposed [12], that posits that the electric field induces an orbital Rashba effect at the surface of metallic superconductors, ultimately leading to a transition to a normal or $\pi$ state, suppressing the supercurrent. Furthermore, we have for the first time realized a complete suppression of the critical current in an aluminium-based Dayem bridge via electrostatic gating. Since aluminium is an important material from the technological point of view, this paves the way for future applications of the electric field effect.

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[20] In the Supplemental Material we provide further details about the Al-based Dayem bridge fabrication process and extra characterizations related to the supercurrent behavior. Then, we present the microscopic modelling and the magnetic-electric field phase diagram for the 0-normal critical boundary. Finally, we show the temperature dependence of the 0-π phase boundary in the presence of Zeeman magnetic field.

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Supplemental Material for “Unveiling Mechanisms of Electric Field Effects on Superconductors by Magnetic Field Response”

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In this Supplemental Material we report the methodology for the fabrication of the nano-bridges and further characterization concerning the evolution of the superconducting critical current in the presence of electric and magnetic fields. Then, we present the microscopic modelling and the magnetic-electric field phase diagram for the superconducting-normal critical boundary. Finally, we show the temperature dependence of the superconducting 0-π phase boundary in the presence of an applied Zeeman field.

METHODS AND SAMPLE CHARACTERIZATION

The samples were fabricated by using single step electron beam lithography to pattern a resist mask on a sapphire substrate. Hereafter titanium and aluminium were evaporated at room temperature in an electron beam evaporator with a base pressure of $10^{-11}$ Torr. 3 nm of titanium was deposited at 1 Å/s (to improve adhesion), after which 14 nm of aluminium was deposited at 2.5 Å/s. The critical current measurements were performed in a He-3 He-4 dilution refrigerator at temperatures ranging from 50 to 600 mK, using a standard four wire set-up, biasing with a current. The voltage drop was amplified using a room temperature differential pre-amplifier, while the gate voltage was supplied by a low noise sourcemeter. To determine the critical current $I_c$, current-voltage $I-V$ measurements were repeated 30 to 50 times.

Fig. 1 shows the device’s resistance as a function of temperature, measured with a Picowatt resistance bridge. The transition from the superconducting to the normal state takes place between 560 and 630 mK.

The leakage current between the gate and device was carefully measured by applying a voltage to the gate in the usual manner, and amplifying the current flowing into the device using a room temperature current amplifier over a long period of time. At $V_{Gate} = 25$ V, the leakage current $I \approx 7 \times 10^{-11}$ A, giving a gate-device resistance of $R \approx 0.63$ Ω. This is of the same order of magnitude as reported in previous works.[1]

It is well known, that magnetic fields suppress superconductivity. In Fig. 2a, we show the critical out of plain field versus temperature, where the error bars indicate the resolution in $B_Z$. The data is fitted with the phenomenological expression $B_Z(T) = B_Z(T=0)(1 - (T/T_C)^2)[2]$, which yields $T_C = 507$ mK and $B_Z(T=0) = 16.25$ mT. Fig. 2b, shows the critical in-plane field $B_Y(T)$, fitted with a calculation of the temperature dependence of the critical in-plane field assuming a homogenous spin splitting, also minimizing the free energy.[3–5]

For completeness we also report the behavior of the critical and retrapping currents as a function of $V_{Gate}$. $B_Z$ and $B_Y$ while for the magnetic fields, both $I_C$ and $I_R$ are immediately suppressed, this is not the case when a gate voltage is applied. Moreover, when considering the effect of $V_{Gate}$, the retrapping current is initially unaffected, even when $I_C$ is already significantly reduced. Only when $I_C$ is reduced to a value comparably to $I_R$, the latter is affected.
FIG. 3: a) The critical and retrapping current versus gate voltage, b) out-of-plane field \(B_Z\) and c) in-plane field \(B_Y\).

MODEL AND PHASE DIAGRAMS

We simulate the superconducting thin film by assuming a conventional \(s\)-wave spin-singlet pairing for a geometry with \(n_z\) layers [6]. We consider an electronic description based on three bands, e.g. \(d\)-orbitals belonging to the \(t_{2g}\) sector in a cubic symmetry which are usually indicated as \((yz, zx, xy)\). In this subspace, one can encode the effects of inversion symmetry breaking by introducing orbital dependent asymmetric couplings at the surface layers. For convenience and clarity we indicate as \((a,b,c)\) the \((yz, zx, xy)\) \(d\)-orbitals. Then, assuming translational invariance in the \(xz\) planes, we introduce the creation \(d_{a\sigma}^\dagger(k, i_z)\) and annihilation \(d_{a\sigma}(k, i_z)\) operators with momentum \(k\), spin \((\sigma = \uparrow, \downarrow)\), orbital \((\alpha = (a, b, c))\), and layer \(i_z\), to construct a spinorial basis \(\Psi^\dagger(k, i_z) = (\Psi^\dagger_{\uparrow}(k, i_z), \Psi^\dagger_{\downarrow}(-k, i_z))\) with \(\Psi^\dagger_{\alpha}(k, i_z) = (d_{a\sigma}^\dagger(k, i_z), d_{b\sigma}^\dagger(k, i_z), d_{c\sigma}^\dagger(k, i_z))\). In this representation, the Hamiltonian can be expressed in a compact way as:

\[
H = \frac{1}{N} \sum_{k, i_z, j_z} \Psi^\dagger(k, i_z) \hat{H}(k) \Psi(k, j_z),
\]

with

\[
\hat{H}(k) = \hat{H}_{SC} + \hat{H}_{ISB} + \hat{H}_M
\]

where \(\hat{H}_{SC}\) is the superconducting part related with the multilayered configuration assuming an intra-orbital singlet pairing,

\[
\hat{H}_{SC} = \sum_\alpha [\tau_\alpha \varepsilon_\alpha(k) + \Delta_\alpha(i_z) \tau_\alpha (L^2 - 2L^2_{\alpha})] \delta(i_z, j_z) + \tau_{\perp, \alpha} \tau_\alpha (L^2 - 2L^2_{\alpha}) \delta(i_z, j_z \pm 1),
\]

the term \(\hat{H}_{ISB}\) arises from the inversion symmetry breaking at the surface of the superconducting thin film due to the presence of the electric field and is expressed as

\[
\hat{H}_{ISB} = \omega_{OR} \tau_\alpha \delta(k_z, \pm L_z) \delta(i_z, j_z) \delta(i_z, 1) + \delta(i_z, n_z) + \lambda \sum_{\alpha} \lambda \delta(k_z, \pm L_z) \delta(i_z, 1) \delta(j_z, 2) - \delta(i_z, 2) \delta(j_z, 1) + \delta(i_z, n_z) \delta(j_z, n_z - 1) - \delta(i_z, n_z - 1) \delta(j_z, n_z)],
\]

and finally the Zeeman term is given by

\[
\hat{H}_M = B \sum_\alpha \tau_0 \otimes (L^2 - 2L^2_{\alpha}) \delta(i_z, j_z)
\]

Here, the orbital angular momentum operators \(\hat{L}\) have components \(\hat{L}_x = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & -i & 0 \end{bmatrix}, \hat{L}_y = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix}, \hat{L}_z = \begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\) within the \((yz, zx, xy)\) subspace, and \(\tau_i (i = x, y, z)\) are the Pauli matrices for the electron-hole sector, and \(\tau_0\) the identity matrix. 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 orbital Rashba couplings close to the surface and a Zeeman spin coupling related to the applied planar magnetic field \(B\). The kinetic energy for the in-plane electron itinerancy is linked to the symmetry allowed [7] nearest-neighbor hopping. Indeed, due to the anisotropy of the \((yz, zx, xy)\) orbitals, they are mostly directional and the main hopping amplitude occurs for homologous orbitals. Thus, one has that \(\varepsilon_{\alpha}(k) = -2\eta \delta(k_z, \pm L_z) \cos(k_x) + \eta \cos(k_y),\) \(\varepsilon_{\beta}(k) = -2\eta \delta(k_z, \pm L_z) \cos(k_x) + \eta \cos(k_y),\) and \(\varepsilon_{\gamma}(k) = -2\eta \delta(k_z, \pm L_z) \cos(k_x) + \eta \cos(k_y),\) with \(\eta\) being a term that takes into account the in-plane bond distortions that lead to deviations from the ideal cubic symmetry. The bond distortions with respect to the ideal cubic symmetry typically can lead to off-diagonal terms that mix non-homologue bands. For the problem upon examination we assume that the electric field can yield distortions at the surface layers and that those are responsible of the orbital Rashba coupling and of the orbital dependent inter-layer hybridization terms. The way the inversion symmetry breaking modifies the electronic structure is such that allows for nearest-neighbors orbital overlap that would not be allowed in inversion symmetric microscopic conditions. We assume that the layer dependent spin-singlet OP is non-vanishing only for electrons belonging to the same band and it is expressed as \(\Delta_\alpha(i_z) = \frac{1}{N} \sum_k g(d_{\alpha\dagger}(k, i_z)d_{\alpha}(\pm k, i_z))\), with \(g\) the pairing strength and \(\langle ... \rangle\) being the expectation value on the ground state. This assumption is not affecting the conclusions, and (4) even in the presence of inter-band spin-singlet pairing the
overall phenomenology is not qualitatively modified. Here, $N = n_x \times n_y$ 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 $n_z$ layers along the $z-$axis. The planar hopping amplitude sets the energy unit, $t_{\parallel} = t$, while the interlayer hopping is orbital independent, i.e. $t_{\perp,\alpha} = t_{\perp}$, and the pairing coupling is $g = 2t$. Since the electronic bandwidth is of the order of $\delta t$, the amplitude of the superconducting pairing interaction is in the weak coupling regime. Variations of the electronic parameters do not affect the qualitative outcomes of the analysis.

Hence, let us consider the consequences of the applied magnetic field on the superconducting phase. We have performed the analysis by assuming that a uniform amplitude of the superconducting order parameter develops along the $z$-direction and we determine the minimum of the free energy. This approach is justified because typically the variation of the superconducting order parameters occurs only within few layers nearby the surface and the corresponding contribution can be negligible with respect to the total free energy. Full self-consistent analysis yield the same competing phases in the phase diagram [6]. In the main text we have shown that for weak orbital Rashba couplings compared to $t$, i.e. the in-plane kinetic energy scale, the increase of the asymmetric inter-orbital interaction $\lambda$ can drive a rearrangement of the interband superconducting phase difference resulting into a $\pi$-phase above a critical threshold for $\lambda$. We have also demonstrated that this transition is substantially unaffected by the presence of a magnetic field. In this supplemental part of the Letter, for completeness we also investigate another interesting regime that refers to values of the orbital Rashba coupling that are comparable to $t$, where the $\pi$-phase is not stable and the variation of the amplitude of the inter-layer asymmetric coupling $\lambda$ drives a transition from the superconducting to the normal metal. In Fig. 4(a) we observe that at zero magnetic field, for $\alpha_{\text{OR}} = 1.0t$, the superconducting state with uniform orbital phase (0-SC) can be tuned into a normal state by increasing the amplitude of $\lambda$ above a critical amplitude $\lambda_c$. The type of transition is continuous as one can notice by inspection of the superconducting order parameter (Fig. 4(d)). Here, we recall that the amplitude of $\lambda$ measures the strength of the applied electric field. The evolution of the critical line separating the 0-SC state from the normal metallic state indicates that one can destroy the superconducting phase with a smaller amplitude of the $\lambda$ coupling in the presence of an applied magnetic field. In particular, close to the critical magnetic field the strength of the $\lambda$ coupling can be tuned to be vanishingly small. This would imply that the threshold of the electric field to disrupt the superconductivity can be tuned to zero by the simultaneous presence of an applied magnetic field. In this respect, the behavior of the critical line is not compatible with the experimental observation that the electric field amplitude to disrupt the superconducting phase is weakly dependent on the strength of the applied magnetic field. It is reasonable to expect such behavior for the 0-SC/normal transition line because the magnetic field acts as a source of spin pair-breaking and thus it tends to reduce the energy of the superconducting phase and in turn it favors the stability of the normal state. We point out that the transition line is first order and there is net jump at the boundary of the superconducting order parameter which grows with the increase of the magnetic field strength. We have also verified that a change in the number of layers does not alter the outcomes of the results by explicitly evaluating the case with $n_z = 12$.

In order to assess the role of the thermal fluctuations we have also determined the phase diagram at finite temperature for the case of small orbital Rashba coupling. In Fig. 5 we report the phase diagram with the evolution of the transition lines among the 0-, $\pi$-SC phases and the normal metallic state by considering the effects of the temperature and of the effective electric field through the $\lambda$ coupling. We compare the zero magnetic field case with one representative configuration corresponding to $B \sim 0.6B_c$. There are two relevant observations to highlight: firstly, the critical boundary from the 0-SC state to the $\pi$-phase is substantially unaffected by the temperature and by the applied magnetic field; then, it is also interesting to notice that the critical temperature for the superconducting-normal transition is also independent on the 0- or $\pi$-character of the superconducting phase. Remarkably, this phenomenology agrees very well with the electromagnetic effects observed experimentally. The evolution of the superconducting order parameters in temperature demonstrate a conventional trend with a weak dependence on the electric (via $\lambda$) and magnetic fields ($B$).

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FIG. 4: (a) Phase diagram in the $(B, \lambda)$ plane for $\alpha_{OR} = 1.0t$ assuming an in-plane magnetic field (e.g. $B_y$). For this value of $\alpha_{OR}$, there is no stable ($\pi$-SC) state. Indeed the free-energy for the $\pi$-SC case has a higher value with respect to the normal phase. $B_{c0}$ is the critical field when $\alpha_{OR} = \lambda = 0$ and the other parameters are: $n_z = 6$ (number of layers); $t_\perp = 1.5t$, $\mu = -0.4t$, $\eta = 0.1$. (b-c) Behavior of the free-energy as function of the SC-order parameter $\Delta$ for two different values of the magnetic field $B$ (marked by the yellow lines in (a)). $\Delta_{G_0}$ is the energy gap when $\alpha_{OR} = \lambda = 0$ and $B = 0$. The free energy is shown for several values of $\lambda$, ranging from 0 to $\sim 0.9 \lambda_c$ ($\lambda_c \simeq 0.33t$). (d) Behavior of the SC order parameter $\Delta$ as a function of $\lambda$ for different applied magnetic field.

FIG. 5: (a) Phase diagram in the $(\lambda, T)$ plane showing three different states: conventional superconducting state (0-SC), unconventional ($\pi$-SC), and normal state for $\alpha_{OR} = 0.2t$ for $B = 0.615B_{c0}$. We assume an in-plane magnetic field orientation, e.g. $B_y$. The critical $\lambda$ amplitude for the 0-$\pi$ transition (black line) does not change as a function of temperature. The transition from the SC to normal state (blue line) is of second order. The gray dashed lines are the transition lines in absence of magnetic field ($B = 0$). (b-c) Behavior of the order parameter as a function of temperature for $B = 0$ (panel (b)) and $B \neq 0$ (panel(c)) and for several values of $\lambda$. 

