

Robust Two-Dimensional Surface Superconductivity and Vortex Lattice in the Weyl Semimetal γ -PtBi₂

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The layered compound γ -PtBi₂ is a topological semimetal with Fermi arcs at the surface joining bulk Weyl points. Recent work has found signatures of surface superconductivity consisting of gap openings compatible with a critical temperature orders of magnitude larger than the bulk value. However, no superconducting vortices have been identified, raising questions about the robustness of the phase coherence. Here, we use very low temperature STM and find robust superconductivity with $T_c = 2.9$ K and $H_{c2} \approx 1.8$ T linked to the Fermi arcs. We observe quantized superconducting vortices and the Josephson effect, demonstrating two-dimensional macroscopic quantum phase coherence.

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Many semimetals present Weyl or Dirac nodes in their electronic band structures [1,2]. The bulk-boundary correspondence often dictates the presence of topologically protected bound states at the surface, which are of particular interest in superconductors because of possible non-Abelian modes [3]. Nevertheless, few of these semimetals are superconductors in the bulk. This has led to proposals to study heterostructures consisting of a thin layer of a topological semimetal on top of a bulk superconductor [2,4–8]. Superconductivity is then induced in the surface state of the semimetal by proximity. Contrasting with these proposals, it could also happen that superconductivity is intrinsically realized on the very same topologically protected surface state of a bulk material with Dirac or Weyl nodes [9–11]. Recent experiments and calculations suggest that this situation could be realized in the layered Weyl semimetal γ -PtBi₂ [12–18]. The realization of such surface two-dimensional superconductivity with topologically protected states could have important applications in quantum devices [19–21] and complement efforts to understand two-dimensional superconductivity in rhombohedral, twisted

bilayer and trilayer graphene [22–24], few-layer single crystals [25,26], and ultrathin films [27–30].

γ -PtBi₂ crystallizes in a layered trigonal structure without inversion symmetry [31]. The band structure contains wide bands crossing the Fermi level at the upper and lower borders of the Brillouin zone. However, the center of the Brillouin zone is gapped. This provides a semimetallic character presenting a huge and nonsaturating magnetoresistance [32]. The electronic band structure contains Weyl points at approximately 50 meV above the Fermi level [1,2,32]. The calculated electronic band structure agrees well with the angle-resolved photoemission (ARPES), quasiparticle scattering (QPI), and quantum oscillation experiments [13,18,33,34]. Furthermore, ARPES finds Fermi arcs in surface bands located close to the projection of the Weyl points to the surface plane [13,33]. Specific heat of bulk γ -PtBi₂ does not present traces of superconductivity down to 0.5 K. There are large resistive transitions with T_c well below 1 K in thin flakes of γ -PtBi₂ obtained by exfoliation [14,16,31,35,36]. Other indications for surface superconductivity present T_c 's well above 10 K, associated with superconducting gap values of several tens of meV and are based on ARPES and STM measurements [13,15–18]. The values of the superconducting gap obtained from ARPES and STM vary widely among different experiments, with recent ARPES measurements showing no signature of a superconducting gap opening on the Fermi arcs at temperatures of 3 K and within an energy range down to below 1 meV [16,33]. What is

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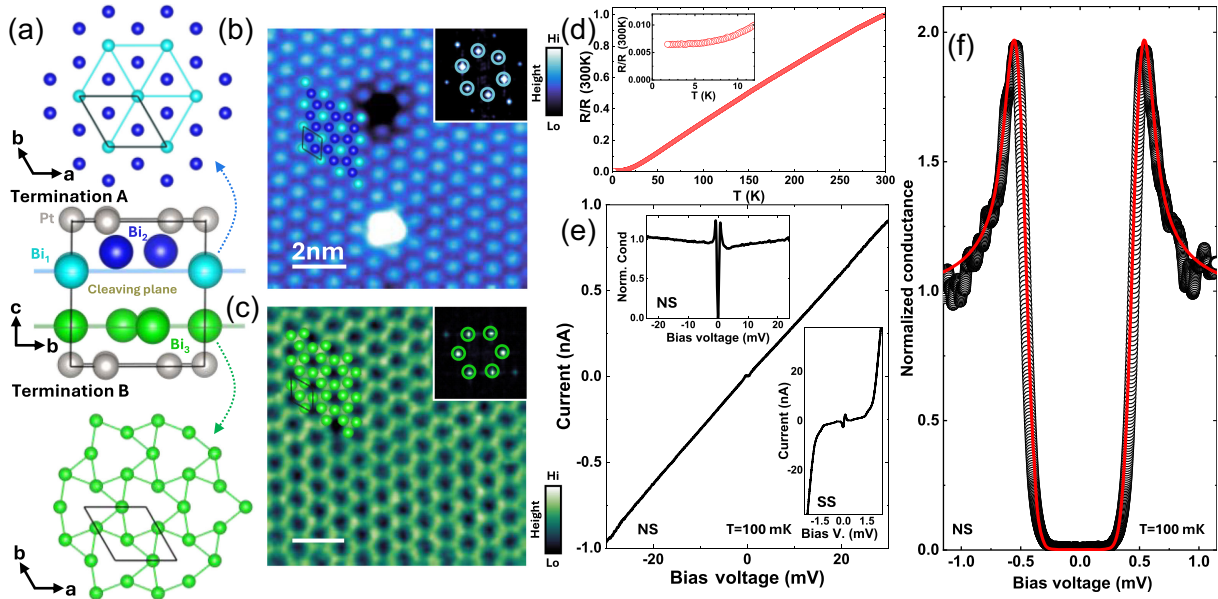


FIG. 1. (a) Unit cell of γ -PtBi₂ (space group No. 157), with Bi atoms in blue and green and Pt atoms in gray. The system is composed of layers where Pt is surrounded by Bi, so that the surface after cleaving is always composed of Bi. (b),(c) Atomically resolved STM topography taken on the two possible terminations of γ -PtBi₂. White scale bar corresponds to 2 nm. Color map is shown to the right. Inset shows the fast Fourier transform with the Bragg peaks marked with circles. The atomic arrangement of Bi atoms is schematically shown by colored circles in the main panels. The surface unit cell is shown as a black rhombus. Notice that the hexagonal pattern observed in the STM image for termination A (b) is formed by the Bi₁ atoms. The Bi₂ atoms are located farther below the surface, as schematically shown in (a). The hexagonal pattern observed in the STM image for termination B (c) is formed by groups of three Bi₃ atoms that lie at the surface plane. These two surfaces are similar to the ones found previously in Ref. [16], although, as shown below, the tunneling conductance found here on these surfaces is very different. (d) Resistivity measurement as a function of temperature on γ -PtBi₂ (red circles). (e) Current vs voltage curve obtained with a normal (PtIr) tip. Top inset shows conductance vs voltage curve in the same voltage range ($T = 0.1$ K, $V = 30$ mV, and $I = 1$ nA). Bottom inset shows a current vs voltage curve obtained with a superconducting (Pb) tip. We observe a sharp zero bias signal showing the Josephson effect ($T = 0.1$ K, $V = 3.5$ mV, $I = 90$ nA). (f) Tunneling conductance vs bias voltage obtained with the same parameters as in (e) and $V = 1.2$ mV. Red line is a fit to a modified BCS density of states described in the text and in Fig. 2.

more, the gaps observed around the Fermi level in STM often persist up to very strong magnetic fields (up to 12 T) and there are no reported observations of a vortex lattice or the Josephson effect [16,37]. To better understand the eventual connection of superconductivity and Fermi arcs, it is important to demonstrate the presence of vortices and thus flux quantization in the surface superconductivity of γ -PtBi₂.

Here, we use dilution refrigerator STM under magnetic fields to study γ -PtBi₂. We find homogeneous superconductivity down to atomic scale with a critical temperature of $T_c = 2.9$ K and the gap size and temperature dependence following BCS theory. We observe superconducting vortices and study the behavior of the vortex lattice as a function of magnetic field and temperature. QPI shows features of the Fermi arcs at the superconducting quasiparticle peaks.

The cleaving plane of γ -PtBi₂ is located in between two Bi layers, as shown in Fig. 1(a). The two possible surfaces present a trigonal arrangement of Bi atoms, one in which Bi atoms located at two nonequivalent sites (Bi₁ and Bi₂, termination A) form intertwined corrugated hexagons, and another where the Bi atoms are located on a single plane

(Bi₃, termination B). The resulting STM topographic images show hexagonal patterns [Figs. 1(b) and 1(c)], characteristic of each termination, as found previously in Ref. [16]. Both terminations provide the same superconducting and vortex lattice properties described below.

In Fig. 1(e) we show a characteristic current vs voltage curve for γ -PtBi₂ at 0.1 K obtained on atomically flat areas. We note that the current is linear as a function of the voltage, suggesting excellent metallic behavior. We also note the clear and small superconducting gap that opens and a vanishing current inside the gap. Furthermore, when we use a superconducting Pb tip, we find a sharp signal at zero voltage [bottom inset in Fig. 1(e)] showing the Josephson effect. Further details are provided in Supplemental Material [38]. To better visualize the superconducting gap, we show the tunneling conductance vs bias voltage in Fig. 1(f). We find large quasiparticle peaks and vanishing density of states inside the gap. We fit the tunneling conductance using a modified BCS density of states $DOS(E)$ with a Gaussian distribution of gap values $\gamma_i(\Delta_i)$ (see End Matter). We find a Gaussian distribution centered around $\Delta_0 = 0.48$ meV with a width of 0.07 meV.

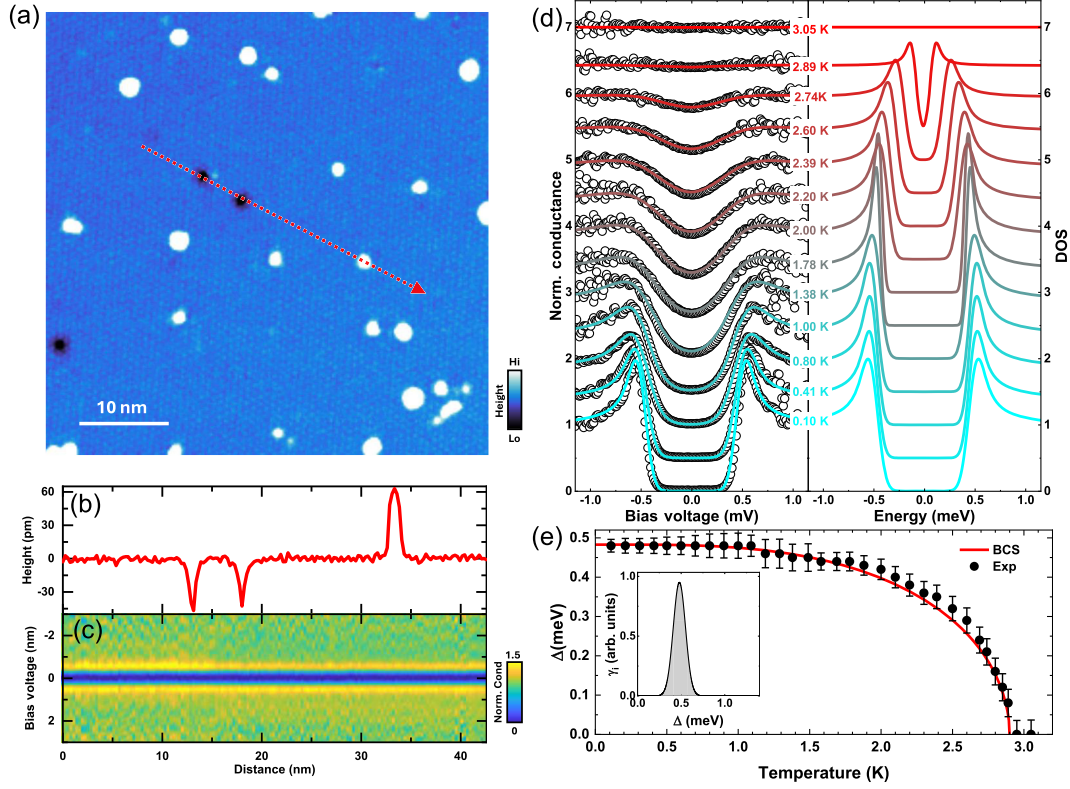


FIG. 2. (a) Atomically resolved STM topography presenting a few defects on termination A. White scale bar corresponds to 10 nm. Color map is shown to the right. (b) Profile along the red arrow on the topography shown in (a). (c) Conductance as a function of bias voltage taken simultaneously as the topography, following the same profile as (b). Color map is shown to the right. (d) On left panel we show the tunneling conductance vs bias voltage as a function of temperature (dots) and calculated conductance curves obtained after convoluting the BCS DOS (shown on the right panel) with the derivative of the Fermi function at each temperature, as described in the text. Curves are shifted vertically for clarity. (e) Temperature dependence of the superconducting gap, obtained as described in the text. The solid line is the BCS temperature dependence of the superconducting gap. Inset shows the Gaussian distribution $\gamma_i(\Delta_i)$ centered around $\Delta_0 = 0.48$ meV with a width of 0.07 meV used for the $T = 100$ mK fit.

The spatial dependence of the superconducting tunneling conductance at atomic scale is shown in Figs. 2(a)–2(c). The topography shown in Fig. 2(a) provides an atomically flat surface, containing a few defects, which appear as atomic size adatoms or interstitials (white spots) and as atomic vacancies (black spots). In Fig. 2(b) we represent a line scan of the height measured by STM. We see that the atomic corrugation is small, of a few pm. At the atomic size defects, there are peaks or troughs in the surface of a few tens of pm. Along the whole path, the superconducting gap slightly fluctuates [Fig. 2(c)]. The defects do not strongly influence the tunneling conductance.

The temperature dependence of the superconducting tunneling conductance is shown in Figs. 2(d) and 2(e). We can fit the whole temperature dependence using the DOS(E) discussed previously, convoluted by the derivative of the Fermi function at the temperature of the measurement, without any adjustment parameters up to about 1 K. Above that temperature, the value of the peak is reduced in $\gamma_i(\Delta_i)$, which gives the DOS(E) provided in the right panel of Fig. 2(d). We plot the peak position in $\gamma_i(\Delta_i)$ as a

function of temperature in Fig. 2(e) and find an excellent agreement with BCS theory (red line), obtained by solving the self-consistent gap equation with $T_c = 2.9$ K and $\Delta_{\text{BCS}} = 1.76 k_B T_c = 0.44$ meV. The latter coincides with Δ_0 better than within 10%.

Whereas we observe superconductivity for temperatures below 3 K, compatible with the recent ARPES experiments of Ref. [33] and the STM work of Ref. [89], the temperature range for earlier claims of superconductivity in γ -PtBi₂ is reported to be even far above the T_c we observe here [13–17,90]. The repeated observation of the same gap values in our experiment shows that the superconducting properties observed here are reproducible. As we have now shown for a superconducting state that exists on the surface and is robust, it is also interesting to ask about the relationship with the topological properties of the electronic band structure.

To investigate the relationship between surface two-dimensional superconductivity and the Fermi arcs found in ARPES [13,33], we discuss now QPI experiments made in small areas with atomic resolution. We show the Fourier

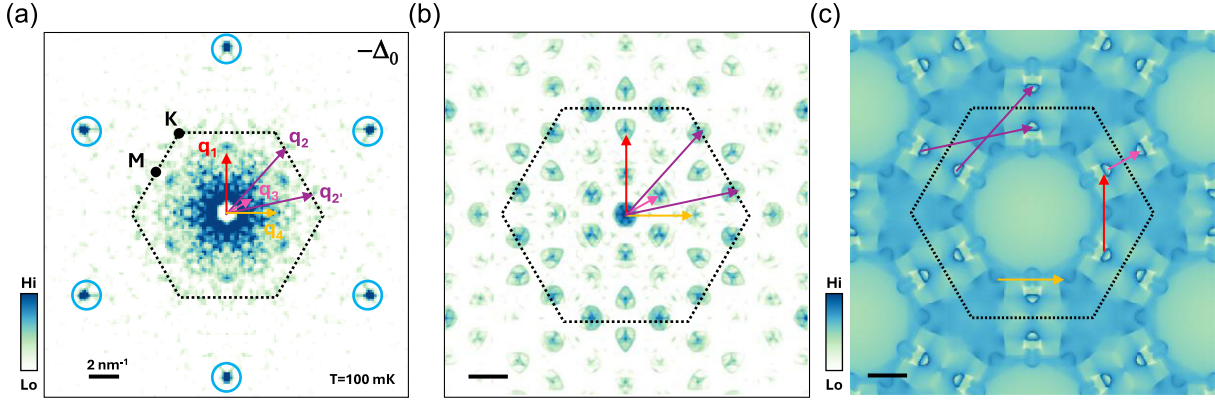


FIG. 3. (a) Quasiparticle interference pattern in γ -PtBi₂ measured on the region shown in Fig. 2(a) at 0.1 K. The scattering intensity is shown on a color scale, following the bar on the left. The dashed line is the first Brillouin zone. K and M denote high symmetry points. We mark the position of Bragg peaks with blue circles. We mark relevant scattering wave vectors q_1 , q_2 , q_2' , q_3 , and q_4 by colored arrows. The pattern is obtained from a tunneling conductance map at the bias voltage of the BCS quasiparticle peak, $V = -\Delta_0/e = -0.48$ mV. (b) Joint density of states obtained from density-functional theory calculations of the Fermi surface [shown in (c)]. The colored arrows are the scattering wave vectors found in (a). (c) Fermi surface of γ -PtBi₂. The Brillouin zone is shown by a dashed black line in (b),(c).

transform of the tunneling conductance map at $V = -\Delta_0/e$ in Fig. 3(a). A similar result is found for $V = +\Delta_0/e$. We find several scattering wave vectors agreeing with previous QPI and ARPES experiments at energies above the superconducting gap edge [18,33]. By comparing with density-functional theory calculations [Figs. 3(b) and 3(c)] we find, at the superconducting gap edge, scattering wave vectors (q_1 , q_2 , q_2' , and q_3) that coincide with wave vectors joining

Fermi arcs that connect the projections of Weyl points to the surface.

We present the superconducting vortex lattice, obtained by mapping the zero bias conductance as a function of the position over large size areas, for different values of the magnetic field and at 0.1 K in Fig. 4. In Fig. 4(a) we show a zero bias conductance map with a single vortex (see Supplemental Material [38]). In Figs. 4(b)–4(d) we show

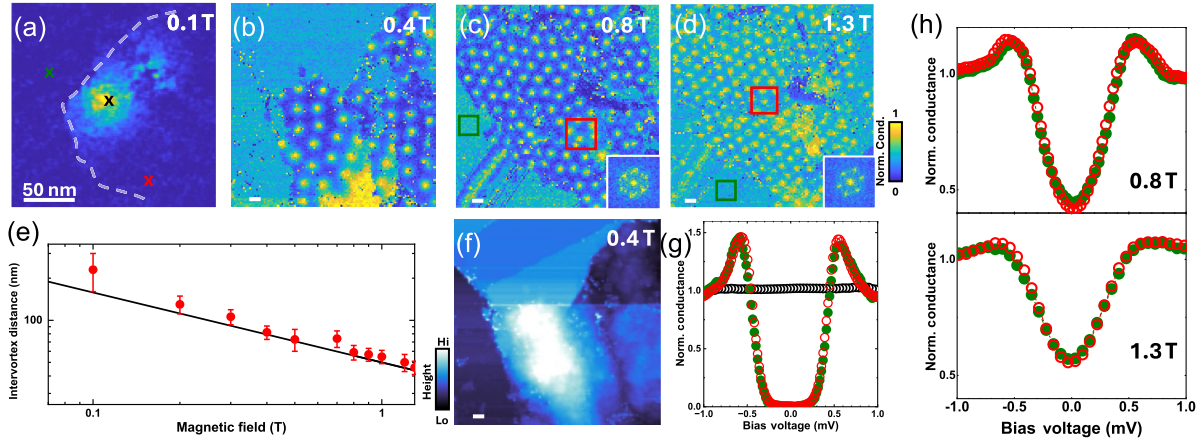


FIG. 4. (a) Zero-bias tunneling conductance map of a single vortex at 0.1 T. White dashed line shows the boundary between an atomically flat surface on the left and a nanometer-sized flakes (see Supplemental Material [38]). (b)–(d) Tunneling conductance maps at zero bias showing the vortex lattice as a function of the magnetic field at 0.4 (b), 0.8 (c), and 1.3 T (d). The maps shown in (c),(d) were taken on the same field of view. White scale bars correspond to 50 nm in the main panels. Insets in (c),(d) on the bottom right corner show the Fourier transform. (e) Intervortex distance as a function of the magnetic field is shown as red disks. The error bars are obtained by Delaunay triangulating vortex positions and measuring the width of the distance distribution. Solid line corresponds to the Abrikosov prediction for an hexagonal vortex lattice $d_{\text{vortex}} \sim 1.075(\Phi_0/B)^{1/2}$. (f) Topography taken simultaneously to the conductance map shown in (b). (g) Normalized tunneling conductance curves obtained at the core of the vortex [black dots, black cross in (a)] and away from the vortex (red and green cross, taken far from the vortex core in an atomically flat region, green, and on a flake, red). (h) Top (bottom): average of conductance curves over the regions marked with rectangles in (c),(d) are shown by circles: in red the regions presenting a vortex lattice and in green the regions presenting no apparent vortex lattice in STM measurements at 0.8 T (1.3 T). Data taken at 0.1 K.

zero bias tunneling conductance maps obtained at 0.4, 0.8, and 1.3 T, respectively. The intervortex distance as a function of the magnetic field [Fig. 4(e)] follows Abrikosov behavior for a hexagonal vortex lattice [91,92]. The Fourier transforms of vortex lattice images mostly present a hexagonal patterns, although the vortex lattice is disordered [insets of Figs. 4(c) and 4(d)]. Interestingly, we find extended areas where we do not observe vortices under magnetic fields; see left part of Fig. 4(a) and portions of Figs. 4(b)–4(d). Previous work focused on such areas and did not find indications for a vortex lattice [16,18,89]. In Fig. 4(f) we show the topography of the field of view shown in Fig. 4(b); see Supplemental Material [38] for topographies in other fields of view. The areas where we do not observe vortices are atomically flat, and the areas where we observe vortices present a surface with a corrugation of the order of nanometers. The superconducting tunneling conductance is equivalent in both areas. We show the tunneling conductance far from a single isolated vortex in both areas in Fig. 4(g) (red and green circles). Furthermore, the superconducting tunneling conductance averaged over a single unit cell of the vortex lattice in the areas where we observe vortices is equivalent to the tunneling conductance found in the areas where we do not observe vortices [Fig. 4(h)]. We provide further line scans in Supplemental Material [38]. All this, together with scanning electron microscopy structural and elemental characterization provided in Supplemental Material [38], shows that the areas with an irregular topography are nanometer-sized flakes of the same sample produced during the cleave.

We do not have a clear answer to the apparent absence of vortices in these atomically flat surfaces and the results found at high magnetic fields in Refs. [16,18,89]. The lack of observable vortices does not necessarily preclude their presence, however. We hypothesize effects related to enhanced vortex mobility through thermal excitation [93,94] or tip-sample interaction [95] and discuss further experiments in Supplementary Material [38]. In the areas where we do observe vortices we find (i) enhanced vortex mobility; (ii) thermally activated vortex motion, leading to vortex lattice melting when increasing temperature well below T_c ; and (iii) little or no dependence of the vortex density from the magnetic field history, suggesting a very large magnetic field penetration. These findings (described in detail in Supplemental Material [38]) are hallmarks of two-dimensional vortex lattices [91–93,96–100] and suggest that we could be observing two-dimensional vortices at the surface of γ -PtBi₂.

We have thus established that robust two-dimensional superconductivity with $T_c = 2.9$ K exists on the surface of γ -PtBi₂. We observe the Josephson effect when using superconducting tips (see Supplemental Material [38]), implying a well-defined phase difference across the junction. Furthermore, the Cooper pair wave function is uniquely valued, as shown by the observation of flux quantization and the vortex lattice.

Our QPI measurements show that the superconducting gap opens at the Fermi arcs, in agreement with theoretical predictions [101]. For superconducting Fermi arcs that are projections of bulk Weyl points, Majorana modes are expected inside vortices. These should be visible by a peak in the tunneling conductance inside the vortex core [2–8]. But even in absence of Majorana modes, superconductors in the clean limit present a large density of states inside vortex cores [98,102–107]. We do find a flat tunneling conductance inside vortex cores that suggests that the electronic mean free path on nanometer-sized flakes is comparable to or smaller than the coherence length ($\xi_{a,b} \sim 13$ nm), smearing localized vortex core states despite surface superconductivity being present. In contrast, vortices on atomically flat surfaces are most likely in the clean limit and may host bound states, potentially including Majorana modes [11].

In summary, we have found robust surface superconductivity in γ -PtBi₂ connected to the Fermi arcs joining the projection of bulk Weyl points. An interesting prospect is to better understand the pairing mechanism leading to two-dimensional surface superconductivity. Changes in the phonon structure close to the surface could play a role [9,10]. Interestingly, monolayer γ -PtBi₂ has been proposed to be a ferroelectric metal due to phonon softening [108]. Furthermore, it has been recently proposed that metals with Weyl points can show topological surface superconductivity [11]. Superconductivity could be favored by the enhanced electronic density of surface states, in a distinct but somewhat comparable situation to the flat band surface state in rhombohedral graphene [23,109–111]. γ -PtBi₂ provides an exceptional case of two-dimensional superconductivity arising at the surface state of a topological semimetal.

Note added—We recently became aware of an independent similar work that reports the observation of bulk superconductivity with $T_c \sim 0.35$ K and surface superconductivity with $T_c \sim 3$ K. Only atomically flat surfaces are found, without an observable vortex lattice under magnetic fields [89].

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Data availability—The data that support the findings of this article are openly available [112].

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End Matter

Appendix A: Experimental details—High-quality single crystals of γ -PtBi₂ were grown using a Bi-rich flux [33], as described in Supplemental Material [38]. When probed macroscopically either by resistivity or by magnetization, there are no hints of bulk superconductivity down to

1.8 K [see Fig. 1(d) and Supplemental Material [38]]. The residual resistance ratio is of $RRR \sim 245$, showing that the crystals have minimal disorder scattering. We used a dilution-refrigerator STM with an energy resolution below 8 μ eV cooling to ~ 0.1 K, described in Refs. [113,114],

and the data treatment software described in Refs. [115–117]. Further details on the STM data acquisition are described in Supplemental Material [38].

Appendix B: Temperature convolution—We fit the tunneling conductance using a modified BCS density of states with $\text{DOS}(E) \propto \sum_i \gamma_i(\Delta_i) \text{Re}\left(E/\sqrt{E^2 - \Delta_i^2}\right)$, convoluted with the derivative of the Fermi function at the corresponding temperature. For a perfectly isotropic s -wave BCS superconductor with gap Δ_{BCS} , $\gamma_i(\Delta_i) = 1$ when $\Delta_i = \Delta_{\text{BCS}}$ and zero elsewhere. In systems with an anisotropic band structure, however, the superconducting gap is often not perfectly homogeneous and there is a certain distribution of gap values over the Fermi surface [104,118–122].

Appendix C: Estimation of critical field and coherence lengths—As we discuss in Supplemental Material [38], we measure the upper critical field using the magnetic field dependence of the tunneling conductance in between vortices and the magnetic field dependence of the radial behavior of the tunneling conductance around vortices, finding $H_{c2} \approx 1.8$ T and an in-plane coherence length of about $\xi_{a,b} = 13$ nm.

There is no signature of superconductivity in bulk measurements in the field and temperature range discussed in Figs. 2 and 4. The anisotropy of the upper critical field ($\xi_{a,b} \sim 20\xi_c$) [35] shows that superconductivity in γ -PtBi₂ is confined to a thin surface layer. The very short coherence length along the c axis suppresses proximity effects from the normal bulk, leaving the surface density of states essentially unaffected.