

Anomalous Residual Surface Conductivity in a Superconductor with Strong Spin-Orbit Coupling

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Conventional BCS superconductors are expected to exhibit a conductivity with vanishing dissipation with decreasing temperature. While bulk physical properties measurements indicate PdPb₂ is a conventional superconductor with a T_c of 3.0 K, measurements of surface impedance through the microwave cavity perturbation technique indicate a large, non-vanishing dissipative component below T_c that is at odds with conventional superconductivity. We demonstrate PdPb₂ to be a possible topological superconductor with a fully gapped bulk and a dissipative Majorana fluid surface.

Topological superconductors can host Majorana fermions inside vortex cores and on their surfaces [1]. They have been proposed as platforms for topological quantum computing [2, 3]. There are a number of ways of realizing such Majorana fermions, such as topological insulator-superconductor heterostructures [4–8], doping-induced superconductivity in topological insulators [9–12], and superconductivity-proximitized nanowires [13–16]. There are also recent proposals of intrinsic topological superconductors, such as in β -Bi₂Pd [17–19] and FeTe_{1-x}Se_x [20–22].

Analogous to a topological insulator, an intrinsic topological superconductor is expected to show surface behavior markedly different from that of the bulk. Topological surface states, if present in the normal state, as is the case in β -Bi₂Pd and FeTe_{1-x}Se_x, may be proximitized by the superconducting bulk, and are expected to possess an unconventional, odd-parity gap function that may give rise to Majorana bound states within vortices. Alternatively, under the right symmetry conditions, the bulk of the superconductor may itself be topological, and can give rise to topological surface states below T_c [8]. This kind of topological superconductor is fully gapped in the bulk, but is expected to show non-superconducting surface metallicity. The metallic surfaces in such cases are expected to host Majorana fermions as opposed to massless Dirac fermions, as in topological insulators [8].

In this Letter, we present evidence for the existence of non-superconducting metallic surface states in centrosymmetric, tetragonal PdPb₂ below its bulk superconducting transition temperature of $T_c = 3$ K. Measurements of the bulk indicate a fully gapped superconducting state, which are in contrast to measurements of the

surface impedance through the microwave cavity perturbation technique that indicate significant low frequency dissipation by surface carriers below T_c , an observation contrary to the expectations for a conventional superconductor. Through extensive sample characterization, we rule out extrinsic origins for the observed phenomenology, and propose PdPb₂ to be a candidate topological superconductor based on an analysis of the $\mathbb{Z}_2$ invariant of the normal state band structure.

Data from measurements of heat capacity, magnetization, and resistivity of a single crystal of PdPb₂ grown by the Bridgman technique (details in Supplementary Material [23]) are shown in **Figure 1**. The heat capacity at zero applied magnetic field, shown in **Figure 1 A**, demonstrates a superconducting phase transition at $T_c = 3.0$ K, as per an equal-entropy approximation, in agreement with previous reports [24, 25]. An applied field of $\mu_0 H = 2$ T is sufficient to quench this transition down to the lowest measured temperature of $T = 0.2$ K. Fits to the $\mu_0 H = 2$ T data, using $C/T = \gamma + \beta_3 T^2 + \beta_5 T^4$, result in an electronic term $\gamma = 6.32(6)$ mJ/mol K², and two phonon terms $\beta_3 = 1.62(1)$ mJ/mol K⁴ and $\beta_5 = 0.0184(2)$ mJ/mol K⁶. The second phonon term arises from the presence of an Einstein mode with $T_E = 49.5(4)$ K, with fits shown in the Supplementary Material [23]. By subtracting these phonon terms from the $\mu_0 H = 0$ T data, we are able to obtain the electronic heat capacity of PdPb₂, C_{el} . After normalizing C_{el} by γ and T_c , we may fit the data to $C_{el}/T = A[\exp(-BT_c)/T]$ in order to obtain the magnitude of the superconducting gap $\Delta(0)$, where A and B are two constants [26], and B is given by $B = \Delta(0)/k_B T_c$ at temperatures $T_c/T \geq 2$. Our fits, shown in the inset of **Figure 1 A** on a log scale, indicate a bulk superconducting gap of $\Delta(0) = 0.370(2)$ meV and a $2\Delta(0)/k_B T_c = 2.86$. The origin of this anomalously small value of $2\Delta(0)/k_B T_c$ in relation to the expected BCS value of 3.53 is unknown. The gap value is in agree-

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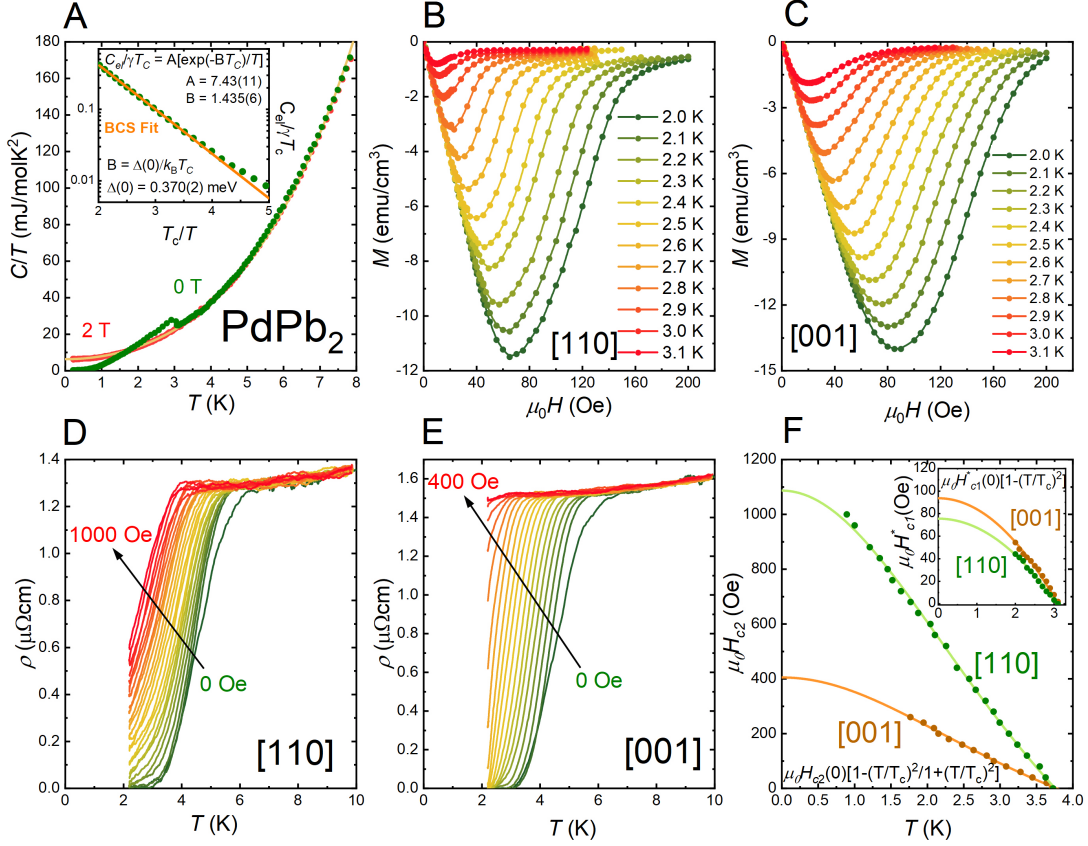


FIG. 1. **A** Heat capacity data of PdPb₂ at both $\mu_0 H = 0$ T and 2 T. The 2 T data has been fit to determine the electronic and phonon contributions to the heat capacity in the normal state. The inset shows a BCS fit, described in the text, to the $\mu_0 H = 0$ T electronic heat capacity of PdPb₂, which indicates a bulk gap of $\Delta(0) = 0.370(2)$ meV. **B** and **C** Field-dependent magnetization curves of PdPb₂ at various temperatures, with applied fields along the [110] and [001] directions, respectively. **D** and **E** Temperature-dependent resistivity curves of PdPb₂ under various applied fields, with applied fields along the [110] and [001] directions, respectively. **F** Extracted upper critical fields along both directions, using values determined from the fastest slope of the resistivity curves, shown in **D** and **E**. Fits are to the Ginzburg-Landau equation. The inset shows the lower critical fields for both directions, as determined from the magnetization curves after considering demagnetization effects as explained in the Supplementary Material.

ment with the value of $\Delta(0) \approx 0.4$ meV obtained via microwave cavity measurements, as discussed below and in the supplementary material [23]. Furthermore, the Sommerfeld coefficient $\gamma \rightarrow 0$ as the temperature $T \rightarrow 0$ K, marking the absence of a finite residual electronic contribution to the heat capacity in the bulk, as expected for a fully gapped superconductor.

The results of magnetization and resistivity measurements of PdPb₂ are shown in **Figure 1 B-F**. Both measurements show a higher onset T_c into a superconducting state compared to heat capacity and microwave perturbation. It is possible that the higher T_c and breadth of the transition are due to superconducting fluctuations above T_c . In order to rule out extrinsic origins for this behavior, as well as for the surface behavior discussed later in this manuscript, we have performed rigorous measurements of crystal and sample quality, such as powder and single crystal X-ray diffraction, scanning electron microscope imaging, energy-dispersive X-ray spectroscopy,

TABLE I. Superconducting parameters of a PdPb₂ crystal oriented along two crystallographic directions relative to the applied field. These include the lower critical field H_{c1}^* , upper critical field H_{c2} , thermodynamic critical field H_c , coherence length $\xi(T = 0)$, penetration depth $\lambda(T = 0)$, and the Ginzburg-Landau parameter κ . More information is available in the Supplementary Material [23].

Parameter	[110]	[001]
H_{c1}^* (Oe)	75(6)	93(4)
H_{c2} (Oe)	1086(26)	404(17)
H_c (Oe)	191	163
$\xi(T = 0)$ (nm)	55	90
$\lambda(T = 0)$ (nm)	221	158
κ	4.02	1.75

Laue diffraction, and X-ray microtomography, the details of which can be found in the Supplementary Material [23]. Both X-ray diffraction and energy-dispersive X-ray

spectroscopy indicate a nominal stoichiometry of PdPb_2 , with the latter measurement indicating atomic ratios of 33.42(13)% Pd and 66.58(22)% Pb in the best crystal.

Measurements were performed on two samples, cut from the same Bridgman-grown crystal, with either the [110] or [001] faces exposed, as determined through Laue diffraction. The extracted superconducting parameters are shown in **Table 1**. The H_{c2} values for both directions, $H_{c2}[110] = 0.1086$ T and $H_{c2}[001] = 0.0404$ T, are much lower than the expected Pauli limit for PdPb_2 , which through $H_P(T) \sim 1.85T_c(K)$, should be approximately $\mu_0 H = 5.5$ T. PdPb_2 , therefore, does not appear to show signs of triplet pairing or otherwise unconventional superconductivity in the bulk. The derived Ginzburg-Landau parameter $\kappa = \lambda(0)/\xi(0)$, exceeds $\kappa \geq 1/\sqrt{2}$ in both crystallographic directions, indicating PdPb_2 is a type-II superconductor.

The microwave cavity perturbation technique was used to determine the temperature-dependent complex surface impedance, $\tilde{Z}_s(T)$, of a single crystal of PdPb_2 . Measurements were performed in a superconducting NbTi, $T_c \approx 10$ K, cylindrical cavity resonator with a resonance frequency of the TE_{011} mode at 18.66 GHz. The cavity perturbation shifts, marked by a change in complex resonance frequency $\Delta\tilde{\omega}/\omega_0$, were measured upon insertion of the sample. For a sample in the skin depth regime, $\Delta\tilde{\omega}/\omega_0$ can be related to the impedance $\tilde{Z}_s = R_s + iX_s$ via $\frac{\Delta\tilde{\omega}}{\omega_0} = \xi\tilde{Z}_s + \lim_{|\tilde{\sigma}| \rightarrow \infty} \frac{\Delta\tilde{\omega}}{\omega_0}$, where ξ is the resonator constant and $\lim_{|\tilde{\sigma}| \rightarrow \infty} \frac{\Delta\tilde{\omega}}{\omega_0}$ is the metallic shift [23]. The decomposed surface impedance components, surface resistance R_s and surface reactance X_s , for PdPb_2 are shown as a function of temperature in **Figure 2 A**. Both R_s and X_s show a sharp change at $T = 3.0$ K, consistent with a transition into a superconducting state, and reduce with decreasing temperature to plateaued values of $R_s \sim 0.16 \mu\Omega$ and $X_s \sim 1.60 \mu\Omega$. The non-zero surface resistance of $\sim 0.16 \mu\Omega$ implies the presence of a significant dissipative channel that is odds with conventional electrodynamics. As discussed below, it may be consistent with a normal surface fluid when the sample is in superconducting state.

For a bulk homogeneous system, the low temperature dissipation is best quantified via the complex conductivity $\tilde{\sigma}(T)$, which can be extracted in the local electrodynamics regime, from the complex surface impedance by $\tilde{Z}_s = \sqrt{i\omega\mu_0/\tilde{\sigma}}$. The decomposed complex conductivity $\tilde{\sigma} = \sigma_1 - i\sigma_2$, where σ_1 and σ_2 are the normal fluid and superfluid responses, respectively, is shown in **Figure 2 B**. For a conventional superconductor, σ_1 is expected to show a Hebel-Schlichter peak near T_c before decreasing with temperature to a minimum, such that $\sigma_1(0)/\sigma_n = 0$. For d-wave and bulk nodal superconductors, the ratio $0 \leq \sigma_1(0)/\sigma_n \leq 1$ can arise as a result of an underlying nodal gap function. PdPb_2 , in contrast, shows an anomalous ratio of $\sigma_1(0)/\sigma_n > 1.4$. Even at the lowest measured temperatures of $T_c/T > 3$, the effective value of σ_1 is greater than its value in the normal state, indicating that PdPb_2 hosts a significant normal

dissipative fluid that coexists with fully gapped superconductivity.

The normal fluid components of the effective complex conductivity for several superconductors are shown in **Figure 2 D**, as extracted from S. Bae, *et. al.* [27]. These are effective conductivities, since they are calculated as if the system is homogeneous. In the case of elemental Ti, which is an s-wave superconductor with $T_c = 0.39$ K, σ_1 decreases monotonically to zero as all of $\sigma_n \rightarrow \sigma_2$, i.e. all of the normal fluid becomes superfluid below T_c . In the case of CeCoIn_5 , a known d-wave superconductor, the normal fluid component of the complex conductivity decreases to a finite, non-zero value, as the superconducting gap function possesses nodes. These nodes are sources of dissipation and low energy excitations. Both PdPb_2 and topological superconductor candidate UTe_2 show an anomalous response, where σ_1 increases with decreasing temperature and subsequently plateaus. In order to determine whether PdPb_2 possesses non-trivial topology in its electronic band structure, we performed density functional theory (DFT) calculations that are discussed below.

The anomalous residual low temperature dissipation could arise either from the bulk or the surface of this material. We investigate each of these possibilities in turn. To further explore the anomalous behavior of the complex conductivity in the case of a homogeneous system, we can describe it using a two-fluid model in terms of the normal fluid and superfluid responses, respectively:

$$\tilde{\sigma}(T) = \frac{ne^2}{m^*} \left(\frac{f_n(T)}{i\omega} + \frac{f_n(T)\tau(T)}{1+i\omega\tau(T)} \right)$$

where n is the total electron density, m^* is the effective mass of the electrons, τ is the effective normal fluid scattering lifetime, f_n is the normal fluid fraction and $f_s = 1 - f_n$ is the superfluid fraction. As τ is always a real positive number, the normal fluid fraction must satisfy $f_n \geq 2/(1 + \sqrt{1 + (\sigma_2/\sigma_1)^2})$. For conventional superconductors, as $T \rightarrow 0$ K, the ratio $\sigma_2/\sigma_1 \rightarrow \infty$, implying $f_n \rightarrow 0$. As shown in **Figure 2 B**, this ratio approaches $\sigma_2/\sigma_1 \rightarrow 5$ in PdPb_2 as $T \rightarrow 0$, giving a minimum residual normal fluid fraction $f_n \geq 33\%$, i.e. one-third of the normal state value, as shown in **Figure 2 C**. This result is similar to one observed recently in the topological superconductor candidate UTe_2 [27]. Again, this is an effective density calculated from a model for the conductivity that assumes that system has a homogeneous electrodynamic response. Similar to UTe_2 , PdPb_2 possesses significant dissipation on the surface against the backdrop of a fully gapped bulk superconducting state. Furthermore, as shown in the inset of **Figure 2 C**, we determine from the above equation the inverse scattering rate for the anomalous fluid, which decreases to a plateau at low temperature to a value of $\tau^{-1} \approx 0.14$ THz. This value is too low to create an impurity-induced normal fluid scenario.

A possible origin for the anomalous effective conductivity and significant dissipative response is a surface ori-

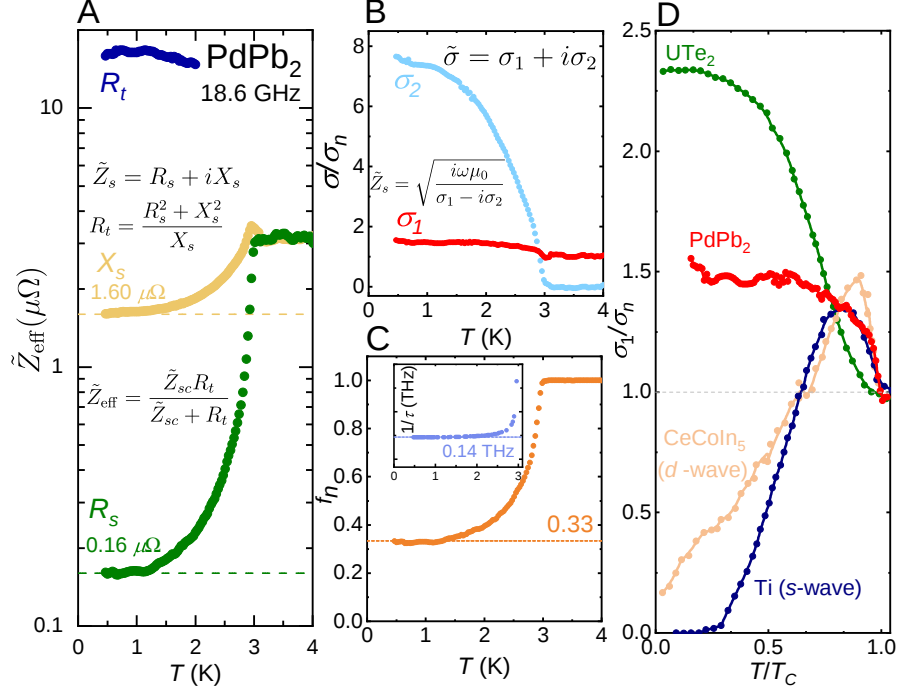


FIG. 2. **A** Temperature-dependent complex impedance $\tilde{Z}_{\text{eff}}$ of PdPb₂. A surface metal sheath with thickness t is considered, giving rise to an effective impedance which arises from both a metal sheath impedance R_t and the impedance of the underlying superconductor Z_{sc} . The real (R_s) normal fluid component of $\tilde{Z}_s$ reaches a finite value of $\sim 0.16\mu\Omega$, contrary to the expectations for a fully gapped superconductor. **B** The effective surface complex conductivity of PdPb₂ decomposed as given by $\tilde{\sigma} = \sigma_1 - i\sigma_2$, where σ_1 and σ_2 are the normal fluid and superfluid responses, respectively. σ_1 does not go to zero, as expected for a fully gapped superconductor, and instead increases. **C** The temperature dependence of the fraction of effective normal fluid in PdPb₂. This value approaches 33% instead of zero as $T \rightarrow 0$ K. The inset shows the extracted value of the scattering rate, which plateaus at 0.14 THz at low temperatures. **D** The real part of the conductivity is shown for several materials, including topological superconductor UTe₂ [27], s -wave superconductor Ti [28], d -wave superconductor CeCoIn₅ [29], and PdPb₂. Just as in UTe₂, σ_1 anomalously increases with decreasing temperature. The data for Ti, CeCoIn₅, and UTe₂ were extracted from S. Bae, *et.al.* [27]

gin of a dissipative normal conduction channel. If the surface of the bulk superconductor has a normal conductor sheath then the expression for extracting a bulk conductivity from the surface impedance becomes invalid. The extracted conductivity can naturally show anomalous features due to contributions from the normal conduction channel. One would need to account for such a thin normal fluid layer at the surface in the effective impedance of the sample. For a thin normal conductor layer with 2D resistance R_t on top of a bulk superconductor with impedance Z_{sc} the effective impedance of the system is given to good approximation by $Z_{\text{eff}} = Z_{sc}R_t/(Z_{sc} + R_t)$ [23]. We assume that the bulk superconductor has a purely imaginary impedance $Z_{sc} = iX_{sc}$ (reasonable for a fully gapped 3D superconductor at low temperatures). Then normal channel's response follows from the measured signal as $R_t = \frac{R_s^2 + X_s^2}{X_s}$. **Figure 2 A** shows the impedance R_t of the thin normal layer with $R_t \sim 16 \mu\Omega$ for $T \rightarrow 0$ K. R_t reduces monotonically with increasing temperature. One can see

that it is reasonably temperature independent in the low temperature region where this analysis applies.

The DFT calculated electronic band structure, with spin-orbit coupling included, is shown in **Figure 3**. PdPb₂ has been predicted to be a symmetry-enforced semimetal with line crossings between the two high symmetry points of the first Brillouin zone Γ and M [30, 31]. It has also been predicted to be a topological insulator based on the absence of a linear combination (NLC) of the elementary band representations (EBRs) across the Brillouin zone [30], though it is not an insulator. Our calculated band structure, as well as those previously calculated, indeed show band crossings along the Λ path along k_z between Γ and M , with point group $4mm$. Since PdPb₂ possesses time-reversal symmetry, its bulk topology is characterized by a $\mathbb{Z}_2$ invariant [32–34]. As it further possesses inversion symmetry, the $\mathbb{Z}_2$ invariant can be calculated readily by considering the product of the parities of all of the bands up to and including the ones that cross the Fermi level at both Γ and M [35]. Only

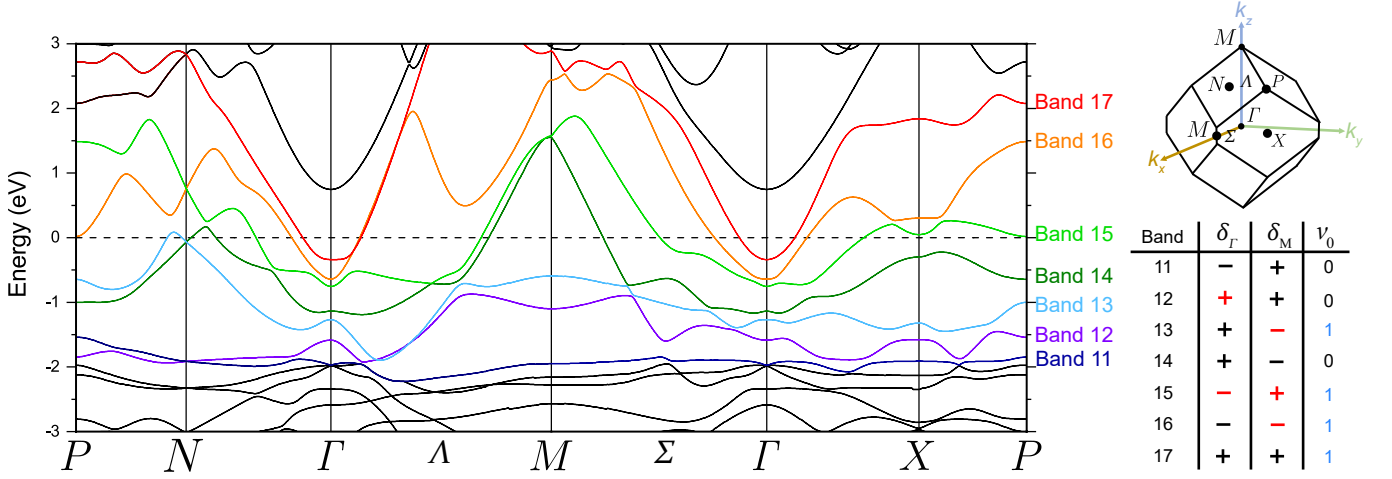


FIG. 3. The calculated electronic band structure of PdPb₂, calculated using the local density approximation and with spin-orbit coupling considered. The first Brillouin zone for body-centered tetragonal is shown, under the $a > c$ lattice parameter condition. The parities at $\Gamma(\delta_\Gamma)$ and $M(\delta_M)$ are shown as positive and negative signs. A change in parity from one band to the next is shown in red. The $\mathbb{Z}_2$ topological invariant ν_0 is shown for each band, which is calculated as the product of the parities at Γ and M for that band, along with the product of those two parities for all previous bands. Non-trivial invariants of $\nu_0 = 1$ are shown in blue.

these two special points in the Brillouin zone determine the topological invariant for body-centered tetragonal, as of the eight time-reversal invariant momentum (TRIM) points in three-dimensions, Γ and M are the only two with odd coefficients ($1\Gamma, 4N, 2X, 1M$). The band parities at Γ and M , and the resulting topological invariant ν_0 , which also considers the product of the parities of all bands beneath the one in question, are shown in **Figure 3**. Our calculations indicate that four of the five bands that cross the Fermi level have non-trivial invariants of $\nu_0 = 1$. PdPb₂ is therefore a $\mathbb{Z}_2$ metal in the normal state, a designation also used to describe other topological superconductor candidates such as FeTe_{1-x}Se_x [20–22], β -Bi₂Pd [17–19], doped Bi₂Se₃ [12, 36], Tl₅Te₃ [37], and more recently the AV₃Sb₅ ($A = \text{K, Rb, and Cs}$) family of materials [38–41]. More in depth theoretical studies are needed to disentangle the influence of the topological band crossings near the Fermi level on the topology and physical properties.

While our calculations do not reveal the presence of

surface states near the Fermi level in the normal state (not shown), the non-trivial invariant of bands that cross the Fermi level in PdPb₂ could result in a topological superconductor [42]. Strong spin-orbit coupling arising from the heavy elements Pd and Pb can drive the system into a topological ground state characterized by a fully gapped bulk superconducting state with topologically non-trivial Majorana surface states. We believe that low temperature microwave dissipation is a generic signature of such surface states. PdPb₂ therefore adds on to a short list of three-dimensional topological superconductor candidates.

In conclusion, we have discovered an anomalous, large, dissipative surface electrodynamic response in the centrosymmetric superconductor PdPb₂. A natural explanation of this phenomenon is that this material is a topological superconductor with surface Majoranas states. The ease with which large, high quality single crystals can be grown can make this material ideal for understanding the origin of this anomalous behavior.

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