Heat Capacity analysis of Superconducting Pb₂Pd having non-trivial band Topology

M.M. Sharma¹,², N.K. Karn¹,², Poonam Rani³, R.N. Bhowmik⁴, V.P.S. Awana¹,²

¹National Physical Laboratory (CSIR), Dr. K. S. Krishnan Road, New Delhi 110012, India.
²Academy of Scientific and Innovative Research, Ghaziabad, U.P. 201002, India
³Materials Science Division, Inter-University Accelerator Centre, New Delhi-110067, India
⁴Department of Physics, Pondicherry University, R. Venkataraman Nagar, Kalapet, Pondicherry-605014, India

Abstract:

A perfect topological superconductor can be realized by simultaneous existence of bulk superconductivity along with topological non-trivial surface states. In this article, we report single crystal growth of superconducting binary compound Pb₂Pd. The crystal is well characterized through X-Ray Diffraction (XRD), Selected Area electron diffraction (SAED), Transmission Electron Microscopy (TEM), Field emission Scanning electron Microscopy (FESEM) and X-Ray Photoelectron spectroscopy (XPS). The bulk superconducting nature of the studied crystal is determined through AC susceptibility and magneto-heat capacity measurements. The specific heat jump at superconducting transition suggests Pb₂Pd to be a moderately coupled s-wave superconductor. The topological non-trivial character of Pb₂Pd is evidenced through bulk electronic band structure and Z2 invariants, which are calculated under the protocols of Density Functional Theory (DFT). This is the first report on topological invariant of Pb₂Pd, showing Pb₂Pd to possess non-trivial topological character and thereby, a possible topological superconductor.

Key Words: Topological Superconductors, Crystal Growth, Structural Details, Heat capacity, Density Functional Theory.

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Introduction:

The discovery of topological insulators (TIs) has renewed the field of quantum condensed matter. Since then, a flurry of research has been carried out on the materials with symmetry protected topological states, which are characterized by nontrivial topological states viz. Z2, Z4 invariants and the Chern numbers [1-4]. Topological materials are classified in different categories based on their band structure and the symmetry such as topological insulators, topological crystalline insulators (TCIs) and topological semimetals (TSMs) etc. [1,5]. Moreover, TSMs are further sub-classified as Dirac Semimetal (DSM), Weyl Semimetal (WSM) and Nodal line semimetal (NLSM) [5-7]. Among them, DSMs has fourfold degenerate band crossings and WSMs have pairs of Weyl nodes, which are generated due to broken Time Reversal symmetry (TRS) and Inversion symmetry [6]. Apart from these three categories semimetals are classified into two other categories such high-symmetry-point semimetal (HSPSM) and high-symmetry-line semimetal (HSLSM) [8]. These materials show band degeneracy at high symmetry points in the bulk electronic band structure calculated without considering Spin Orbit Coupling (SOC) [8]. TSMs show a variety of properties such as extremely large magneto-resistance (MR) [9,10], high career mobility [11] and negative MR induced from Chiral anomaly [12,13].

Apart from the above exotic properties, when these materials are accompanied with bulk superconductivity, a new class of materials is evolved, which is termed as Topological Superconductors (TSc) [2]. TSc fulfills all required conditions to realize Majorana Fermions [2,14], and these can be the key materials for futuristic quantum computations. There are very few topological materials that show superconductivity in their intrinsic form due to their low carrier density. TIs are made superconducting by means of doping of suitable elements [15-18] that increases carriers in parent compound. Interestingly, some of the TSMs e.g. PdTe2 [19], PbTaSe2 [20], SnTaS2 [21], NbC [22], AuSn4 [23], Au2Pb [24] show superconductivity in their intrinsic form as they possess high carrier density. Apart from these materials some binary compounds like BiPd, Bi2Pd, Sb2Pd are found to show topological states with bulk superconductivity [25-27]. Pb2Pd is another similar compound, and its superconducting nature has been known since 1962 [28]. Pb2Pd is not much studied in the context of topological superconductivity as there is only one theoretical report available in which Pb2Pd is considered as a TSM [8]. Interestingly, superconducting properties of Pb2Pd also have not been explored to that extent. There are only two reports available in literature about superconducting properties of Pb2Pd [29,30]. The first report was on the effect of Bi substitution in Pb2Pd [29]
and the other one is a very recent report showing type-I superconductivity in the same [30]. It is important to study Pb$_2$Pd in the context of topological superconductivity as it is supposed to show superconductivity along with topological nontrivial surface states [8,30].

In this article, we report the synthesis of Pb$_2$Pd single crystal using solid state reaction route, this sample is well characterized through various characterization techniques. Bulk superconductivity is the key issue for topological superconductors, and the same with $T_c^{onset}$ at 2.86K is confirmed in the synthesized Pb$_2$Pd crystal through AC susceptibility and magneto-heat capacity measurements. This is the first report showing magneto-heat capacity measurements for Pb$_2$Pd crystal. Further, the topological properties are studied through DFT calculations and theoretical simulation of topological invariants. The $\mathbb{Z}_2$ invariants of Pb$_2$Pd shows the presence of strong topology. It is worth mentioning here that this is the first report on such topological invariants for Pb$_2$Pd.

**Experimental:**

Solid state reaction route is used to synthesize a single crystalline sample of Pb$_2$Pd. 4N pure powders of Pb and Pd were mixed in 2:1. The mixture was made homogenous by grinding it inside MBraun Glove Box filled with Argon gas. This homogenous mixture was then palletized and vacuum encapsulated in quartz ampoule under the pressure of $5 \times 10^{-5}$ mbar. This ampoule was heated to 900°C at 120°C/h for 24 hours. Then the sample is slowly cooled to 460°C at 2°C/h and then kept at this temperature for 24 hours. After that, the sample is quenched at 460°C in water. The step of quenching at 460°C is very crucial as the phase formation temperature of Pb$_2$Pd and PbPd is very close and the both phases have simultaneous existence at around 445°C [31]. The schematic of heat treatment is shown in Fig. 1.

Rigaku Miniflex II table top XRD equipped with Cu-K$_\alpha$ (1.5406Å) was used for phase purity characterization of synthesized Pb$_2$Pd single crystal. Surface morphology and elemental composition were analyzed using MIRA II LMH from TESCAN made Field Emission Scanning Electron Microscope (FESEM) equipped Energy Dispersive X-Ray (EDX) detector. JEOL/JEM-F200 with One view CMOS camera (4K*4K) Transmission Electron Microscope (TEM) image is used to get Side Angle Electron Diffraction (SAED) pattern and to visualize atomic planes. XPS spectra was recorded by using X-ray Photoelectron Spectrometer (XPS) (Model: K-Alpha-KAN9954133, Thermo Scientific) with monochromated and micro-focused Al K$_\alpha$ radiation (1486.6 eV). The spectrometer has been designed with a dual-beam flood source to provide a charge compensation option and calibration is confirmed by the position of
C 1 s line at the binding energy 284.8 eV. The working pressure was maintained < 6 × 10^{-8} mbar. The scans for Pb 4f and Pd 3d were recorded with the step size of 0.1 eV. AC susceptibility and Heat capacity measurements were carried out using Quantum Design Physical property measurement system (QD PPMS) in temperature range from 5K to 2K. FullProf software and VESTA software were used for Rietveld refinement of PXRD pattern and the unit cell construction respectively.

To study and understand the topological properties of synthesized Pb_2Pd crystal, the first principle simulations and Z2-invariants are calculated to categorize the topology present in the system. For this, Density Functional Theory (DFT) based on first principle calculations are executed in QUANTUM ESPRESSO [32,33] to obtain the bulk electronic band structure and density of states (DOS). For calculation of Z2 invariants, DFT generated Bloch wave function is wannierized using the WANNIER90 software [34]. The WANNIER90 uses the Wannier functions which are the representation of Bloch wave functions in real space [35,36]. The band structure is again obtained using the Maximally localized Wannier Function (MLWFs) and it is verified by the band structure generated through first principle method. An effective tight-binding (TB) model for Pb_2Pd crystal was obtained using MLWFs after wannierization of Block wavefunction. This effective TB model is further processed and applied in WANNIER-TOOLS [37]. The whole Brillouin Zone is sampled on a 8×8×8 K-mesh, for all calculations. The states of Z2-invariants are determined by the evolution of Wannier charge centers in Brillouin zone planes.

**Result & Discussion:**

Fig. 2 represents the Rietveld refinement results of PXRD pattern of synthesized Pb_2Pd single crystal. The synthesized Pb_2Pd crystal crystallizes in tetragonal structure with I4/mcm space group. All peaks of PXRD pattern are well fitted with the phase parameters and confirms that the synthesized crystal is phase pure. The quality of refinement is determined through $\chi^2$ value which is found to be 4.95, which is reasonably good. The constituent elements viz. Pb and Pd occupies the atomic positions viz. (0.165,0.665,0) and (0,0,0.25) in a tetragonal unit cell. The Rietveld refined lattice parameters are $a = b = 6.863(5)\text{Å}$ & $c = 5.847(5)\text{Å}$ and $\alpha = \beta = \gamma = 90^\circ$. All these parameters are in good agreement with previous report on the same compound [30]. The inset of Fig. 2 represents the VESTA drawn unit cell of synthesized Pb_2Pd crystal. Crystallographic information file (CIF) generated from Rietveld refinement is used to
draw the unit cell. The synthesized crystal is shown to have a body-centered tetragonal unit cell with Pd atoms at the body center.

Fig. 3(a) shows SAED pattern of synthesized Pb$_2$Pd single crystal. SEAD pattern was taken on powdered sample. Spots from (002), (004) and (220) are encircled in Fig. 3(a). Crystalline nature of synthesized Pb$_2$Pd crystal is evident from TEM image, shown in Fig. 3(b). Fig. 3(b) is showing the stacking of (002) planes. Interplanar spacing of these (002) planes is found to be 3.74Å, which is in accordance with the XRD data. This suggests that the crystal of Pb$_2$Pd is grown in (00l) direction, it is also consistent with previous report [30]. To get more insight about crystalline nature of synthesized Pb$_2$Pd, surface morphology is visualized through FESEM image. Fig. 3(c) shows the FESEM image of the synthesized Pb$_2$Pd single crystal. The FESEM image is showing terrace type morphology which represents layer by layer growth of the sample. No grain is visible in the FESEM image which also confirms the single crystalline nature of the synthesized crystal. Elemental composition is determined through Energy Dispersive X Ray Analysis (EDAX) measurements and the results are shown in Fig. 3(d). The EDAX spectra shows the presence of constituent elements viz. Pb and Pd. No impurity element is detected in EDAX spectra which shows the purity of the synthesized crystal. Elemental composition is shown in inset of Fig. 3(d), both the elements are found to be in near stoichiometric ratio.

Chemical states of the constituent elements of Pb$_2$Pd are analyzed through XPS spectra. XPS peak of C 1s is taken as reference to calibrate all XPS peaks. Fig. 4(a) and 4(b) show XPS spectra of synthesized Pb$_2$Pd crystal in Pb 4f and Pd 3d regions respectively. These spectra are fitted with Gaussian distribution function. The XPS spectra in Pb 4F region is deconvoluted in four peaks viz. P1, P2, P3 and P4. Peaks P1 and P3 correspond to spin orbit doublet of Pb 4f viz. Pb 4f$_{5/2}$ and Pb 4f$_{7/2}$. These peaks occurred due to core levels of Pb$^{2+}$ cations and the corresponding binding energies are found to be 138.55eV and 143.42eV for Pb 4f$_{7/2}$ and Pb 4f$_{5/2}$ respectively. These values are shifted from the binding energy values for metallic Pb [38], this shift in binding energies is occurred due to bonding with Pd atoms. This suggests strong covalent bonding between Pb and Pd. The remaining peaks viz. P2 and P4 corresponds to Pb$^{2+}$ cations resulting from PbO formation. These peaks occurred due to surface oxidation of the synthesized sample due to air exposure. The peaks of Pb$^{2+}$ ions generated due to PbO formations is less pronounced in comparison to that observed due to Pb$^{2+}$ cations of synthesized Pb$_2$Pd sample. The energy separation between the spin orbit doublet of Pb is found to be 4.87eV, which is very close to the standard value (4.86eV) [38]. Fig. 4(b) shows the XPS
spectra in Pd 3d regions, which consists of XPS peaks due to spin orbit doublet of Pd 3d core levels viz. Pd 3d_{5/2} and Pd 3d_{3/2}. The peaks of spin orbit doublets viz. Pd 3d_{5/2} and Pd 3d_{3/2} are observed at 335.85eV and 341.16eV respectively. These values are slightly shifted from the standard value due to bonding of Pd with Pb atoms [38]. The energy separation of these peaks is found to be 5.31eV which is very close to the standard value (5.26eV) [38]. These XPS results suggest that the valency of Pb atoms in Pb$_2$Pd is +2.

Fig. 5 shows the results of AC susceptibility measurements carried out at different AC magnetic field viz. 3Oe, 5Oe, 7Oe, 9Oe and 11Oe. The frequency of the AC field was set at 333Hz and the background DC field was made stable to 0Oe while performing the experiment. Real part of AC magnetic moment (M') is shown in the lower plot while the imaginary part (M'') of the same is shown in upper one. Both the real and imaginary parts of the magnetic moment show presence of bulk superconductivity in the synthesized Pb$_2$Pd crystal. The onset superconducting transition temperature (T$_{c\text{onset}}$) is found to be 2.9K. AC susceptibility measurements give the hint about the granularity of the sample. For granular superconductors, T$_c$ tends to shift to lower temperature with increasing the amplitude of the AC field. This effect occurs due to the intergranular coupling of superconductors. The shift of T$_c$ with respect to the change in AC field amplitude determines the strength of intergranular coupling [39]. Here, T$_c$ remains constant when the amplitude of the AC field is increased. This suggests that grains are absent due to which intergranular T$_c$ could be evolved. This also determines the crystalline character of the synthesized Pb$_2$Pd crystal.

Heat capacity measurements are the most reliable method to determine bulk superconductivity in a superconducting sample. Heat capacity measurements are carried out on synthesized Pb$_2$Pd crystals at different magnetic fields viz. 0Oe, 20Oe, 30Oe and 50Oe. Heat capacity divided by T i.e., C/T is plotted against T and the same is shown in inset of Fig. 6(a). A clear jump in specific heat is visible with T$_{c\text{onset}}$ at 2.86K. This confirms the presence of bulk superconductivity in the synthesized Pb$_2$Pd single crystal. Heat capacity of a material is mainly contributed by two terms; first one is electronic term (C$_e$) and the other is phonon term (C$_p$). These are described by the following equation

\[ \frac{C}{T} = \gamma_e T + \beta_n T^3 \text{ or } C = \gamma_e T + \beta_n T^3 \]  

In the above equation the first term $\gamma_e T$ represents electronic contribution to specific heat and the second term $\beta_n T^3$ phonon contribution to specific heat. The coefficient associated with these terms can be determined by fitting the C/T vs T$^2$ plot with equation (1). The Fig. 6(a) represents
a linearly fitted C/T vs T^2 plot of synthesized Pb_2Pd single crystal. The coefficient of C el i.e. γ_n is known as the Sommerfeld coefficient and it is found to be 6.62 ± 0.50 mJ mol^{-1} K^{-2}. The other constant term β_n is found to be 3.12 ± 0.02 mJ mol^{-1} K^{-4}. The value of γ_n used to determine the density of states at Fermi level [D_c(E_F)] using the formula,

\[ \gamma_n = \pi^2 k_B^2 D_c(E_F)/3 \]  

(2)

Here, k_B is Boltzmann constant. The obtained value of D_c(E_F) is 2.71 states eV^{-1}f.u.^{-1}. This value will be further checked with theoretical calculations under DFT protocols in the later part. The coefficient associated with phonon contribution term i.e., β_n is related to Debye temperature with the following formula

\[ \theta_D = \left(\frac{12\pi^4 R n}{5\beta_n}\right)^{1/3} \]  

(3)

Here R=8.314 J mol^{-1} K^{-2}, n=3 (for Pb_2Pd). The obtained value of θ_D is 123 ± 1 K. Now the values of θ_D and T_c are used to determine the electron phonon coupling constant λ_{e-ph} using the McMillan formula [40] given below

\[ \lambda_{e-ph} = \frac{1.04 + \mu^* ln(\theta_D/1.45T_c)}{(1 - 0.62 \mu^*)ln(\theta_D/1.45T_c) - 1.04} \]  

(4)

Here, the value of μ* is taken to be 0.13 and it is known as the screened repulsive Coulomb potential. The obtained value of λ_{e-ph} is 0.71 and it is higher than the usual values that are obtained for weakly coupled superconductors. This suggests moderate coupling in synthesized Pb_2Pd single crystal.

Now, the electronic heat capacity (C_el) is calculated by subtracting the phonon term from the total heat capacity. The normalized electronic specific heat is plotted against T/T_c and is shown in Fig. 6(b). This is used to determine the magnitude of specific heat jump i.e., ΔC_el/γ_nT_c, which is found to be 1.83. This value is higher than the BCS weak coupling limit which is 1.43. This also suggests that the synthesized Pb_2Pd is a moderately coupled superconductor which agrees with the obtained value of λ_{e-ph}. Normalized electronic specific heat also gives the information about the superconductivity of the sample whether it is a conventional superconductor or an unconventional superconductor. Also, it is used to determine the value of α = Δ(0)/k_B T_c. For this the normalized specific heat data is fitted with the s wave equation and it is found to be well fitted. This suggests Pb_2Pb to be a bulk superconductor with conventional s-wave pairing. The obtained value of α from the fitted plot
is 2.84, which is also higher than the BCS value for weakly coupled superconductors. All above discussed parameters suggest that synthesized Pb₂Pd single crystal is a moderately coupled superconductor with conventional s-wave pairing; these results are in agreement with the previous report on the same compound [30].

Fig. 6(c) shows the results of heat capacity vs T measurements at different field viz. 0Oe, 20Oe, 30Oe and 50Oe. In this plot, C/T is plotted against T to visualize the heat capacity jump more clearly. These measurements give the value of T_c on set to be 2.86K, 2.75K, 2.66K and 2.40K at 0Oe, 20Oe, 30Oe and 50Oe respectively. It can be clearly seen that the T_c is decreasing with increasing field. Critical field is plotted against the temperature and fitted with equation

\[ H_c = H_c(0) \times \left[ 1 - T^2 / T_c^2 \right] \]  \hspace{1cm} (5)

where T_c is taken to be 2.86K. The fitted plot is shown in inset of Fig. 6(c), this fitted plot gives the value of critical field H_c(0) to be 260Oe, which is comparable to that was obtained in previous report [30].

Moreover, the C_el is determined at the various applied fields. C_el/T is plotted against T and is shown in Fig. 6(d). This gives the value of \( \gamma \) at different fields at T=2K. The values of \( \gamma \) are normalized with \( \gamma_n \) and are plotted against the applied field. This gives the important information regarding the low energy excitations which exists in the proximity the Abrikosov vortex line. In conventional superconductors, these low energy excitations take place inside the vortex cores in normal states having the radius which are proportional to penetration depth (\( \xi \)). For this, specific heat in superconducting state is proportional to vortex density and linearly depends on magnetic field giving \( \gamma(H) \propto H \) [41]. While, in case of superconductors having nodes in energy gap, the DOS are found in the neighborhood of gap nodes. Due to this, the low energy excitations occur outside the vortex core and the specific heat is found to show square root dependence on magnetic field given as \( \gamma(H) \propto H^{1/2} \), this is known as Volovik effect [42]. In the present case, \( \gamma(H) \) is found to have a linear relationship with the applied field as shown in inset of Fig. 6(d). This also confirms that the observed bulk superconductivity in synthesized Pb₂Pd single crystal is conventional superconductivity.

To determine topological non-trivial character of synthesized Pb₂Pd crystal, bulk electronic band structure and DOS are calculated using DFT. The band structure calculation is performed using the cell parameters obtained from the Rietveld refinement. Both the SOC and
without SOC protocols are executed in calculations as these are applied in Quantum Espresso with Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [32,33]. The k-path which is followed for calculations is determined from the SeeK-path: the k-pathfinder and visualizer [43], which eventually suggests that the k-path \( Z \rightarrow \Gamma \rightarrow M \rightarrow \Gamma \rightarrow N \rightarrow P \rightarrow X \) is an optimized path. Fig. 7(a) depicts this particular k-path as it is marked in the First Brillouin zone. The DFT calculated bulk electronic band structure is shown as the left hand side image of Fig. 7(b) while the DOS are shown in the right one. From this analysis, it is found that bands near Fermi level have major contributions from the d-orbitals of Pd and p-orbitals of Pb atoms. There are four bands which cross Fermi level, confirming Metallic/Semi-metallic behavior of the crystal. The Fermi surfaces corresponding to these four bands are calculated using WANNIER TOOLS and plotted in XCRYSDEN [44], the same are shown in Fig. 7(a). The DOS at Fermi level are found to be 2.74 states eV\(^{-1}\)f.u.\(^{-1}\), this value is in agreement with that was obtained from heat capacity analysis. This shows that the theoretical results agree with the experimental one. Moreover, bands are found to show line degeneracy which is eventually lifted when SOC parameters are included in the calculations. This indicates the effectiveness of SOC in the studied Pb\(_2\)Pd system. Particularly, along the path \( N \rightarrow P \), the bands near fermi level are doubly degenerate forming nodal lines. Interestingly, when SOC parameters are included in calculation, the line degeneracy has been lifted except from a few points where double degeneracy remains intact. The region showing the bands along path \( N \rightarrow P \), is shaded in Fig. 7(b) and Fig. 7(c), which represents the zoomed view of the same. Nodal lines are clearly visible in Fig. 7(c) and the line degeneracy is found to be removed when SOC parameters are considered. The presence of line degeneracy in w/o SOC bands suggest that Pb\(_2\)Pd can be classified as a high symmetry line semimetal which is also in agreement with the previous report [8], which is a catalogue of all possible topological materials.

It is important to characterize the topology present in a topological material in terms of a topological invariant. These topological invariants depend on the symmetry preserved by the material. The observed splitting of bands with inclusion of SOC parameters in bulk electronic band structure, suggests that the system respects the TRS and these kinds of topological systems are characterized in terms of Z2-invariants [45]. We follow the Soulyanov-Vanderbilt [45] method of Wannier Charge Centers (WCC), calculated from MLWFs. The evolution of WCC take place in the 6-planes in the Brillouin zone, namely \( k_1, k_2, k_3 = 0 \) and \( k_1, k_2, k_3 = 0.5 \). The calculated Z2 invariants for the Pb\(_2\)Pd crystal in the mentioned six planes are

(a) In \( k_x = 0.0 \) i.e. \( k_y-k_z \) plane: \( Z2 = 1 \).
(b) In $k_x = 0.5$ i.e. $k_y-k_z$ plane: $Z_2 = 0$.
(c) In $k_y = 0.0$ i.e. $k_x-k_z$ plane: $Z_2 = 1$.
(d) In $k_y = 0.5$ i.e. $k_x-k_z$ plane: $Z_2 = 0$.
(e) In $k_z = 0.0$ i.e. $k_x-k_y$ plane: $Z_2 = 1$
(f) In $k_z = 0.5$ i.e. $k_x-k_y$ plane: $Z_2 = 0$

The topological $Z_2$ index is represented by four parameters as $(v_0; v_1 v_2 v_3)$, where $v_0$ represents the strong index and $v_1, v_2, v_3$ represent the weak index. The weak index is the values of $Z_2$ numbers for $k_i=0.5$ planes ($i=x, y$ and $z$). The strong index may have some redundancy as the strong index is given by $Z_2$ value for all three planes $k_i=0$($i=x, y$ and $z$) and $Z_2$ value may not be the same for all three planes. But here we find that for all three planes, $v_0 = 1$, indicating a topologically non-trivial state and strong topology present in the system. Here the weak index has no redundancy, and thus $Z_2$ index is $(1; 000)$ for the studied Pb$_2$Pd system. The same $Z_2$ index is reported for Bi$_2$Te$_3$, which is considered as a strong topological material [46]. Thus, on the basis of theoretical calculations, it can be concluded that Pb$_2$Pd has strong non-trivial band topology.

**Conclusion:**

Summarily, we presented heat capacity analysis of superconducting properties of Pb$_2$Pd single crystal. Pb$_2$Pd is first time characterized through TEM and XPS techniques in this paper. Pb is found to be Pb$^{2+}$ state in Pb$_2$Pd. The synthesized Pb$_2$Pd crystal is found to be a bulk superconductor with $T_c$ $^{onset}$ at 2.86K. The calculated parameters viz. electron phonon coupling constant $\lambda_{e-ph}$, $\Delta C_{el}/\gamma T_c$ and $\alpha$ confirm that Pb$_2$Pd is a moderately coupled s wave superconductor. Conventional superconductivity is also confirmed through magnetic field dependence of the Sommerfeld coefficient in superconducting state. The calculated bulk electronic band structure gives the glimpses of band inversion with including SOC parameters and suggest Pb$_2$Pd to be a possible topological material with bulk superconductivity. Topological properties of Pb$_2$Pd are also confirmed by calculating $Z_2$ invariants for the same which suggest the presence of strong topology in Pb$_2$Pd. This is the first report on calculation of topological invariants for Pb$_2$Pd. This report strongly represents Pb$_2$Pd material as a possible topological superconductor.
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Conflict of Interest statement:

Authors have no conflict of interest.

Figure Captions:

Fig. 1: Schematic of heat treatment followed to synthesize Pb$_2$Pd single crystal.

Fig. 2: Rietveld refined PXRD of synthesized Pb$_2$Pd single crystal inset is showing the unit cell of the same drawn by using VESTA software.

Fig. 3: (a) SAED pattern of synthesized Pb$_2$Pd crystal (b) HRTEM image of Pb$_2$Pd crystal showing stacking of (002) planes. (c) SEM image of surface morphology of synthesized Pb$_2$Pd crystal (d) EDAX spectra of Pb$_2$Pd crystal in which inset is showing the atomic percentage of the constitute elements.

Fig. 4: XPS spectra of synthesized Pb$_2$Pd crystal in (a) Pb 4f region (b) Pd 3d region.

Fig. 5: AC magnetic moment vs temperature plot of synthesized Pb$_2$Pd single crystal.

Fig. 6(a): Zero Field C/T vs $T^2$ plot of synthesized Pb$_2$Pd single crystal where solid black line represents a linear fit to $C/T = \gamma + \beta T$. The inset is showing a C/T vs T plot showing a $T_c$ onset at 2.86K.

Fig. 6(b): Normalized electronic specific heat vs $T/T_c$ plot of synthesized Pb$_2$Pd single crystal where the solid black line represents s-wave fitting of the same.

Fig. 6(c): Variation of C/T with temperature at different fields viz. 0Oe, 20Oe, 30Oe and 50Oe of Pb$_2$Pd single crystal, the inset is showing variation of $T_c$ with applied field.

Fig. 6(d): $C_v/T$ vs $T$ plot of synthesized Pb$_2$Pd single crystal, the inset is showing the linear fitted plot of normalized Sommerfeld coefficient against the applied field.

Fig. 7(a): The first image shows the first Brillouin zone with high symmetric points, the green arrow shows the path chosen for the Band structure calculation. The other four images show the fermi surface of synthesized Pb2Pd crystal corresponding to the bands that are crossing the Fermi level.

Fig. 7(b): Calculated bulk electronic band structure along with the Density of States(DOS) w/o and with SoC within the protocols of Density Functional Theory(DFT).

Fig. 7(c): The zoomed view of the bands of the shaded region in bulk electronic band structure in Fig. 7(b).
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Fig. 1

![Graph with temperature and time intervals](image)

- Fig. 2

![X-ray diffraction pattern](image)

- a=b=6.863(5) Å
- c=5.847(5) Å
- α=β=γ=90°
- \( \chi^2 = 4.95 \)
- Space group: I4/mcm

- Fig. 3
Fig. 4

(a) Graph showing intensity versus B.E. (eV) for Pb$_2$Pd, with peaks labeled P1, P2, P3, and P4.

(b) Graph showing intensity versus B.E. (eV) for Pd 3d and Pb 2Pd, with peaks labeled Pd 3d$_{3/2}$ and Pb 2Pd.

Fig. 5

Table showing element weight and atomic percentage for Pb$_2$Pd:

| Element | Weight% | Atomic% |
|---------|---------|---------|
| Pd L    | 22.67   | 38.34   |
| Pb M    | 77.33   | 61.66   |
| Totals  | 100.00  |         |
Fig. 6
Fig. 7(a)

Fig. 7(b)
Fig. 7(c)