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# Electrical Transport Properties of half-Heusler ScPdBi single crystals Under Extreme Conditions

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## Abstract

We synthesize single-crystals of ScPdBi, a Half-Heusler compound by self-flux growth technique and report its physical properties such as magneto-transport and specific heat down to 2 K. Resistivity measurements were performed on these single-crystals at ambient and high-pressure conditions. Temperature-dependent resistivity measurements reveal that ScPdBi shows the metallic character at ambient pressure and without applied magnetic field. The metallic character of ScPdBi was un-altered even in extreme conditions such as high pressure (up to 19 GPa) and magnetic field (up to 9 T). We observe an upturn in the resistivity which persists even at high pressure. We rule-out the presence of the Kondo effect by performing the specific heat measurements down to 2 K which resulted in a low Sommerfeld coefficient ( $\gamma \approx 2.6 \pm 0.9$  mJ mol<sup>-1</sup> K<sup>-2</sup>). This anomaly in resistivity below 30 K could be attributed to an electron-hole scattering process or a carrier imbalance effect.

**Keywords:** Single crystals; Flux-growth; Electrical transport; High-Pressure

## 1. Introduction

Half-Heusler compounds with general formula  $XYZ$  ( $X$  and  $Y$  = transition or rare-earth elements and  $Z$  = post-transition metal) attract much attention in the field of topological quantum materials because of their narrow band gap [1–3], tunability of spin-orbit coupling (SOC) to induce the band inversion and possibility to close the band gap and form nearly linear dispersive bands resembling the Dirac cone by introducing heavier elements [4–6]. This tunability of band gap makes these materials particularly suitable for various applications based on spintronic, thermoelectric, and magneto-caloric effects [7–9]. Among all the ternary half-Heusler compounds, special attention has been given to  $REPdBi$  and  $REPtBi$  ( $RE$ : rare-earth) compounds because of their potential topological superconductivity [10,11]. A classification (topologically nontrivial and topologically trivial) has been proposed from NMR studies on a range of these half-Heusler systems  $RETBi$  ( $RE = Sc, Y, \text{ and } Lu; T = Ni, Pd, \text{ and } Pt$ ) [12,13]. Notably,  $ScPtBi$  and  $ScPdBi$  compounds were found to be topologically nontrivial and trivial, respectively [12–14]. Among these two compounds, the non-trivial insulating nature of  $ScPtBi$  was well studied with help of first principle calculations which reveal that the  $ScPtBi$  has an inverted band gap of 0.7 – 0.9 eV [3,13–17]. Apart from these,  $ScPtBi$  was reported for its poor thermoelectric properties with a negative Seebeck coefficient and  $n$ -type carrier behavior [18,19]. Apart from numerous theoretical studies, experimental studies on  $ScPtBi$  was least explored. Hou et al. reported the synthesis of  $ScPtBi$  single crystal by self-flux growth technique, and the non-magnetic semi-metallic behavior was observed from the low field magneto-transport studies [20]. Interestingly,  $ScPdBi$  substituted by Pt is claimed to be a superconductor below 1.2 K and paving a path to reach the topological non-trivial superconductivity [21].  $ScPdBi$  was reported to have a trivial band topology with a semiconducting band gap in the range of 0.3 – 0.7 eV [3,6,14]. It has been reported that the band inversion strength ( $\Delta = \Gamma_6 - \Gamma_8$ ) of  $ScPdBi$  was close to the trivial to non-trivial insulating phase transition region [3, 6, 14]. Nowak et al. confirmed the trivial band topology of  $ScPdBi$  with help of  $^{209}Bi$  NMR [17]. It was reported that the uniaxial pressure induces the trivial to non-trivial insulating phase transition in half-Heusler compounds [3,15]. To understand the effect of pressure on the band order, Narimani et al. explored the pressure-dependent electronic band structure of  $ScPdBi$  and could not observe any band inversion. Apart from this, the energy difference between  $\Gamma_6$  and  $\Gamma_8$  was enhanced with pressure [22]. Narimani studied the band

topology of ScPdBi by incorporating the heavy elements (Gd, Np, and Cm) at the Sc site and could not observe any band inversion [23]. These studies motivated us to grow the single crystals of ScPdBi and study their electrical transport properties at ambient and extreme conditions such as magnetic field up to 7 T and pressure up to 19 GPa assuming that these can cause a change in the electronic band structure, which may lead into interesting topological properties.

## 2. Experimental sDetails

Single crystals of ScPdBi were prepared by self-flux growth technique in which stoichiometric amount of Sc (31.2 mg, Alfa Aesar, 99.9% pure chunks), Pd (73.8 mg, Alfa Aesar, 99.99% pure wire) and Bi (145 mg, Sigma Aldrich, 99.9% pure shots) metals were taken in a tantalum ampule. Extra Bi (2.5 gm) was added to these reagents which act as a self-flux. The tantalum ampule was placed in a 13 mm quartz tube and was flame sealed under a vacuum of  $10^{-5}$  torr for preventing oxidation during the reaction. The reactant mixture was heated to 1173 K in 8 h and maintained at that temperature for 48 h. Finally, the sample was cooled down to room temperature with a rate of 3 °C/h. Excess flux present in the sample was removed by heating the sample at 773 K and subsequently centrifuged through a coarse frit. To remove the flux present over the crystals, these crystals were soaked in dilute HNO<sub>3</sub> solution for a period of 16 h. Finally, these crystals were rinsed with acetone and sonicated for several times. The obtained ScPdBi crystal was having a cube shape with an average size of 200  $\mu\text{m}$ .

Single crystal X-ray diffraction measurements were done on the as cleaned ScPdBi single crystals with the help of Bruker Smart Apex 2- CCD diffractometer having a normal focus, 2.4 kW sealed tube X-ray source with graphite monochromatic Mo K $\alpha$  radiation ( $\lambda = 0.71073$  Å). SAINT was used for the integration of diffraction profiles and SADABS was used for the absorption correction. The structure was solved by direct and Fourier methods and refined by full-matrix least-squares, using standard crystallographic software: SHELXT-2014, SHELXL- 2014 [24,25]. Olex2 software package was used for these refinements. The crystal structure of ScPdBi was built with help of DIAMOND software.

Temperature-dependent resistivity measurements down to 2 K were done on ScPdBi single crystals at ambient and different magnetic fields with help of physical property measurement system (PPMS, Quantum Design). The dependence of electrical transport on temperature and the magnetic field is systematically investigated between 2 K and 300 K up to 9

T. Electrical transport measurements at ambient and different pressure values (2–19 GPa) were performed using a linear four-probe technique in which we use Keithley 220 and Keithley 2182 as a source and nano voltmeter respectively. For this pressure-dependent measurements, we used a sintered diamond Bridgman anvil apparatus in which a pyrophyllite gasket and two steatite disks act as a pressure transmitting medium [26].

### 3. Results and Discussion

The X-ray diffraction pattern of ScPdBi is depicted in **Fig. 1a**. It has been seen that the compound is phase pure and forming in the MgAgAs structure type having  $F\bar{4}3m$  space group. The inset of **Fig. 1a** shows the unit cell of half Heusler ScPdBi compound within which Sc, Pd, and Bi atoms occupy  $4b$ ,  $4c$ , and  $4a$  Wyckoff sites, respectively. The lattice parameters ( $a = b = c = 6.449 \text{ \AA}$ ; and  $\alpha = \beta = \gamma = 90^\circ$ ) obtained from the single crystal refinement are in good agreement with reported values [13]. The cubic morphology of the as-synthesized ScPdBi single crystals was confirmed using a scanning electron micrograph (inset of **Fig. 1a**). The dimensions of these single crystals are in the range of  $150 \mu\text{m}$  as shown in the inset of **Fig. 1a**. EDAX and elemental color mapping studies were done on ScPdBi single crystals and are shown in **Fig. 1b** and **Fig. 1c** respectively. It has been seen that all the Sc, Pd, and Bi elements are uniformly distributed throughout these single crystals. Apart from this, a slight deficiency in Sc was observed from the composition analysis.

**Fig. 2a** shows the temperature-dependent resistivity of ScPdBi single crystals at ambient pressure. Resistivity decreases with the decreasing temperature down to 2 K, confirming the metallic nature of the system. However, it contradicts the expected semiconducting behavior as predicted from the previous theoretical reports [3,6,14]. Results of the temperature-dependent resistivity under constant magnetic field ranging from 1 T to 9 T are depicted in **Fig. 2b**. We could not observe any appreciable effect on the resistivity with the application of an external magnetic field which is expected for a trivial band system. Moreover, Sc, Pd, and Bi atoms probably do not contribute any magnetic moment to the system which is in agreement with the reported theoretical prediction [23].

Apart from the metallic nature, we could observe an upturn in the resistivity below 30 K (**Fig. 2a**). In general, this kind of behavior in resistivity is expected in the case of  $f$ -electron-based systems because of the hybridization of conduction bands with the localized  $f$ -electrons at low temperature and giving rise to the Kondo effect [27]. It was reported that Kondo behavior

can also be observed in 3d-electron-based systems [28–31]. To understand the origin of the upturn in the resistivity of ScPdBi, we have performed the temperature-dependent specific heat measurements down to 2 K. **Fig. 3** shows the  $C/T$  vs  $T^2$  and this variation is fitted with the usual quadratic equation  $C/T = \gamma + \beta T^2$ . From this, we extract a Sommerfeld coefficient ( $\gamma \approx 2.59 \pm 0.87$  mJ mol<sup>-1</sup> K<sup>-2</sup>) and a Debye term ( $\beta \approx 0.87 \pm 0.01$  mJ mol<sup>-1</sup> K<sup>-4</sup>). These results ruled out the possibility of Kondo behavior in the ScPdBi compound.

To further understand the electrical transport properties under extreme conditions, we have carried out the temperature-dependent resistivity at different hydrostatic pressures ranging from 2 – 19 GPa (see **Fig. 4**).

These measurements were performed with a four-probe technique and observed a ground of 0.55 k $\Omega$  above 10 GPa in the electronic set-up which affects the absolute value of the resistivity without changing the general trends. For completeness, we present all the temperature sweeps at constant pressure where the results above 10 GPa have been normalized by multiplying the factor (0.97+0.00000381) (**Fig. 4a**). These results indicate that the metallic nature of ScPdBi persists even up to 19 GPa. Interestingly, we also observe an upturn in the resistivity at low temperature in all the measured pressure range. As we rule out the presence of Kondo state in ScPdBi by specific heat measurements, we ascribe this upturn in resistivity to the conduction in strongly disordered systems.

Considering the very low RRR of the sample that suggests defects and impurities, we turn to a model that describes conduction in strongly disordered systems with localized charge carrier states, i.e. the Mott variable-range hopping model. Analyzing a model in which charge is transported via phonon-induced tunneling of electrons between localized states which are randomly distributed in energy and position, Mott [32] has obtained an electrical conductivity of the form

$$\rho = \rho_0 \exp[2(T_0/T)^{1/4}]$$

In general, this type of temperature dependence has been observed in a large number of systems [32], including high-temperature superconductors [33]. **Fig. 4(c)** presents the resistivity versus temperature as  $\ln(\rho)$  versus  $T^{-1/4}$ . The linear fit of the low-temperature part (i.e.  $T < 25$  K) determines  $T_0 = \alpha^3 / k_B N(E_F)$ , where  $\alpha$  is the localization distance, which can be assumed to be

constant with pressure, and  $N(E_F)$  is the density of states at the Fermi level. On **Fig. 4(b)** we show a decreasing variation of  $T_0^{-1}$ , i.e. increasing for  $N(E_F)$ , as a function of pressure. According to band structure calculations [22], the valence  $\Gamma_8$  band is almost on the Fermi level, at all pressures. Thus, the increase of  $N(E_F)$  that we observe, would indicate an increase of hole charge transfer from defects and impurities to this band. This analysis also supports an explanation for the metallicity of the samples: doping of the valence band by impurities. The low-temperature deviation from 40 K can be attributed due to percolative transport via interlayer coupling or electronic disorders [34,35].

#### 4. Conclusions

Single crystals of ScPdBi were successfully grown by the self-flux growth technique and observed to crystallize in MgAgAs structure type with  $F\bar{4}3m$  space group. The temperature dependences of the resistivity of ScPdBi was measured at ambient pressure at different magnetic field up to 9 T and under pressure up to 19 GPa. ScPdBi observed to have metallic nature down to 2 K and persists its behavior even under extreme conditions (up to 9 T and 19 GPa). ScPdBi did not show a noticeable magneto-transport effect. The Sommerfeld coefficient ( $\gamma$ ) and Debye term ( $\beta$ ) were extracted from the specific heat measurements and found to be  $2.59 \pm 0.87$  mJ mol<sup>-1</sup> K<sup>-2</sup> and  $0.87 \pm 0.01$  mJ mol<sup>-1</sup> K<sup>-4</sup>, respectively. The low  $\gamma$  value and smooth temperature dependence of the specific heat indicate the absence of the Kondo effect in ScPdBi. The upturn in resistivity observed in the low-temperature region at all pressure probably due to either electron-hole scattering process or carrier imbalance.

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#### References

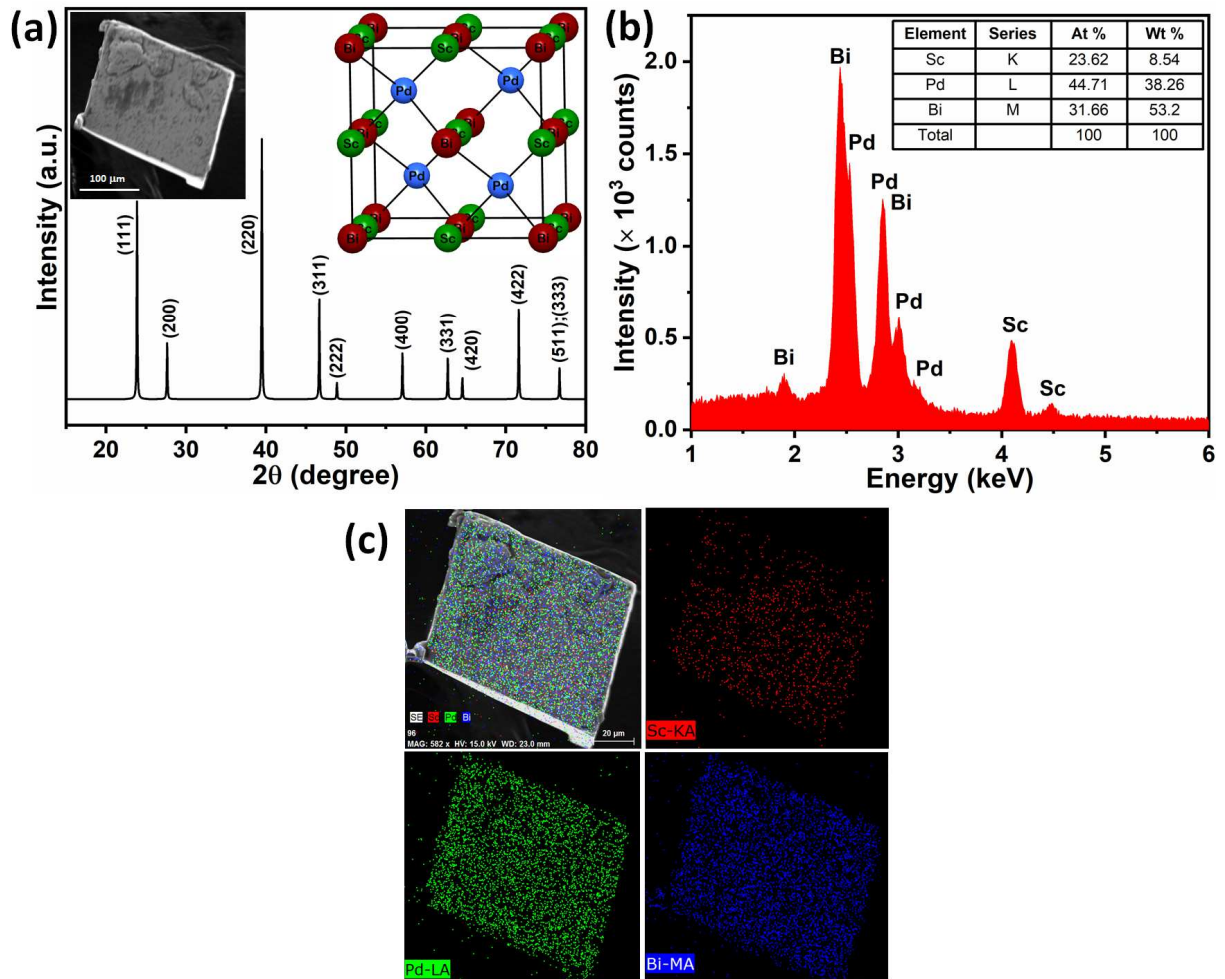
- [1] D. Xiao, Y. Yao, W. Feng, J. Wen, W. Zhu, X.-Q. Chen, G.M. Stocks, Z. Zhang, Phys. Rev. Lett. 105 (2010) 096404.

- [2] H. Lin, L.A. Wray, Y. Xia, S. Xu, S. Jia, R.J. Cava, A. Bansil, M.Z. Hasan, *Nat. Mater.* 9 (2010) 546–549.
- [3] S. Chadov, X. Qi, J. Kübler, G.H. Fecher, C. Felser, S.C. Zhang, *Nat. Mater.* 9 (2010) 541–545.
- [4] J.C. Tung, G.Y. Guo, *New J. Phys.* 15 (2013) 033014.
- [5] T. Suzuki, R. Chisnell, A. Devarakonda, Y.T. Liu, W. Feng, D. Xiao, J.W. Lynn, J.G. Checkelsky, *Nat. Phys.* 12 (2016) 1119–1123.
- [6] F. Casper, T. Graf, S. Chadov, B. Balke, C. Felser, *Semicond. Sci. Technol.* 27 (2012) 063001.
- [7] C.J. Palmstrøm, *Prog. Cryst. Growth Charact. Mater.* 62 (2016) 371–397.
- [8] T. Zhu, C. Fu, H. Xie, Y. Liu, X. Zhao, *Adv. Energy Mater.* 5 (2015) 1500588.
- [9] A. Planes, L. Māosa, M. Acet, *J. Phys. Condens. Matter* 21 (2009) 233201.
- [10] Y. Nakajima, R. Hu, K. Kirshenbaum, A. Hughes, P. Syers, X. Wang, K. Wang, R. Wang, S.R. Saha, D. Pratt, J.W. Lynn, J. Paglione, *Sci. Adv.* 1 (2015) e1500242.
- [11] M. Meinert, *Phys. Rev. Lett.* 116 (2016) 137001.
- [12] B. Nowak, O. Pavlosiuk, D. Kaczorowski, *J. Phys. Chem. C* 119 (2015) 2770–2774.
- [13] X. Zhang, Z. Hou, Y. Wang, G. Xu, C. Shi, E.K. Liu, X. Xi, W. Wang, G. Wu, X.-X. Zhang, *Sci. Rep.* 6 (2016) 23172.
- [14] W. Feng, D. Xiao, Y. Zhang, Y. Yao, *Phys. Rev. B* 82 (2010) 235121.
- [15] W. Al-Sawai, H. Lin, R.S. Markiewicz, L.A. Wray, Y. Xia, S.Y. Xu, M.Z. Hasan, A. Bansil, *Phys. Rev. B* 82 (2010) 125208.
- [16] H.J. Zhang, S. Chadov, L. MÜchler, Y. Yan, X.L. Qi, J. Kübler, S.C. Zhang, C. Felser, *Phys. Rev. Lett.* 106 (2011) 156402.
- [17] B. Nowak, D. Kaczorowski, *J. Phys. Chem. C* 120 (2016) 21797–21801.
- [18] G. Ding, G. Y. Gao, L. Yu, Y. Ni, K. Yao, *J. Appl. Phys.* 119 (2016) 025105.
- [19] K. Kaur, S. Dhiman, R. Kumar, *Phys. Lett. Sect. A* 381 (2017) 339–343.
- [20] Z. Hou, Y. Wang, E. Liu, H. Zhang, W. Wang, G. Wu, *Appl. Phys. Lett.* 107 (2015) 202103.
- [21] O. Prakash, M. Klemm, M. Aronson, *APS March Meet. 2018, Abstr. Id.F31.004* (2018).
- [22] M. Narimani, Z. Nourbakhsh, *Mod. Phys. Lett. B* 30 (2016) 1650159.
- [23] M. Narimani, Z. Nourbakhsh, *J. Magn. Magn. Mater.* 434 (2017) 62–67.

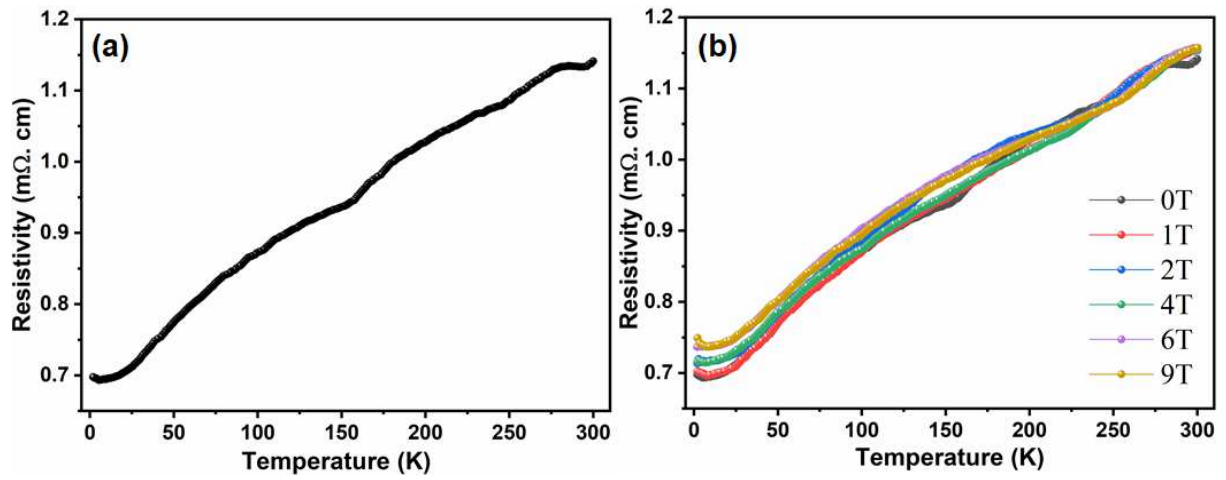


- [24] G.M. Sheldrick, *Acta Crystallogr. Sect. A Found. Crystallogr.* 71 (2015) 3–8.
- [25] G.M. Sheldrick, *Acta Crystallogr. Sect. C Struct. Chem.* 71 (2015) 3–8.
- [26] S. Sanfilippo, H. Elsinger, M. Núñez-Regueiro, O. Laborde, S. LeFloch, *Phys. Rev. B* 61 (2000) R3800.
- [27] G.R. Stewart, *Rev. Mod. Phys.* 56 (1984) 755–787.
- [28] V.I. Anisimov, M.A. Korotin, M. Zöfl, T. Pruschke, K. le Hur, T.M. Rice, *Phys. Rev. Lett.* 83 (1999) 364–367.
- [29] W. Kobayashi, I. Terasaki, J.I. Takeya, I. Tsukada, Y. Ando, *J. Phys. Soc. Japan* 73 (2004) 2373–2376.
- [30] A. Shimoyamada, S. Tsuda, K. Ishizaka, T. Kiss, T. Shimojima, T. Togashi, S. Watanabe, C.Q. Zhang, C.T. Chen, Y. Matsushita, H. Ueda, Y. Ueda, S. Shin, *Phys. Rev. Lett.* 96 (2006) 026403.
- [31] Y.P. Wu, D. Zhao, A.F. Wang, N.Z. Wang, Z.J. Xiang, X.G. Luo, T. Wu, X.H. Chen, *Phys. Rev. Lett.* 116 (2016) 147001.
- [32] N.F. Mott, E.A. Davis, Clarendon Press, Oxford (1979).
- [33] M.A. Kastner, R.J. Birgeneau, C.Y. Chen, Y.M. Chiang, D.R. Gabbe, H.P. Jenssen, T. Junk, C.J. Peters, P.J. Picone, T. Thio, T.R. Thurston, H.L. Tuller, *Phys. Rev. B* 37 (1988) 111–117.
- [34] M. Gu, S.A. Wolf, J. Lu, *J. Phys. D Appl. Phys.* 51 (2018) 10LT01.
- [35] L.J. Zhu, J.H. Zhao, *Sci. Rep.* 7 (2017) 42931.

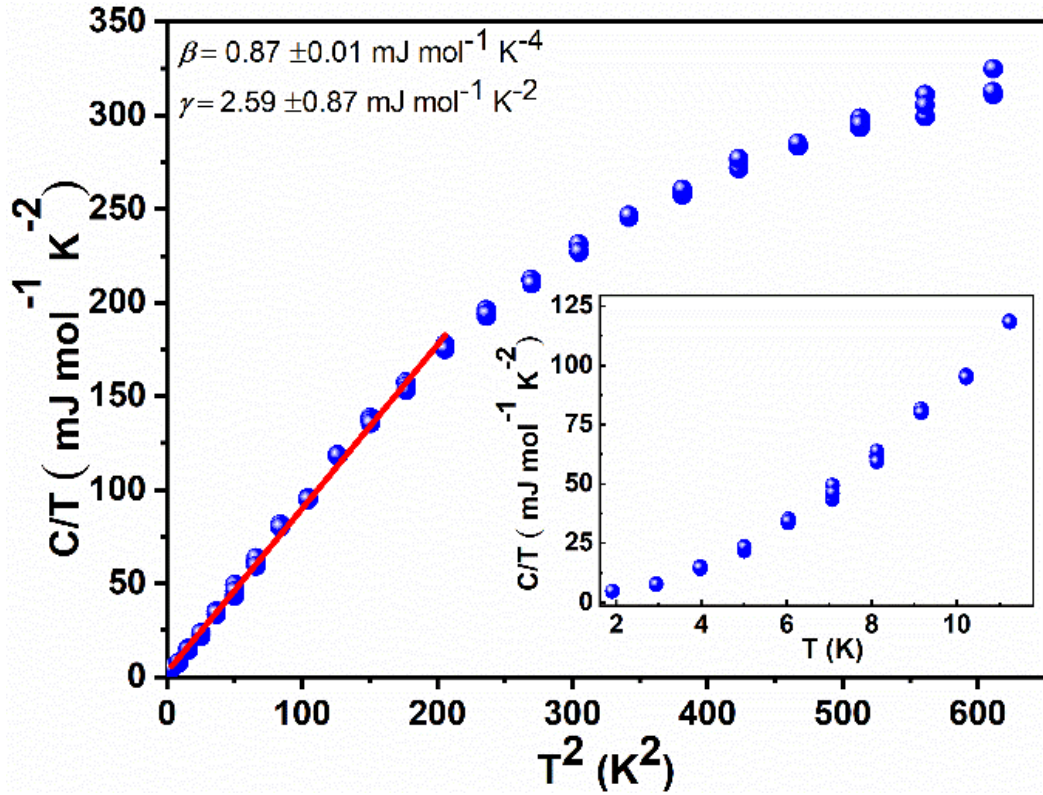
## Figures



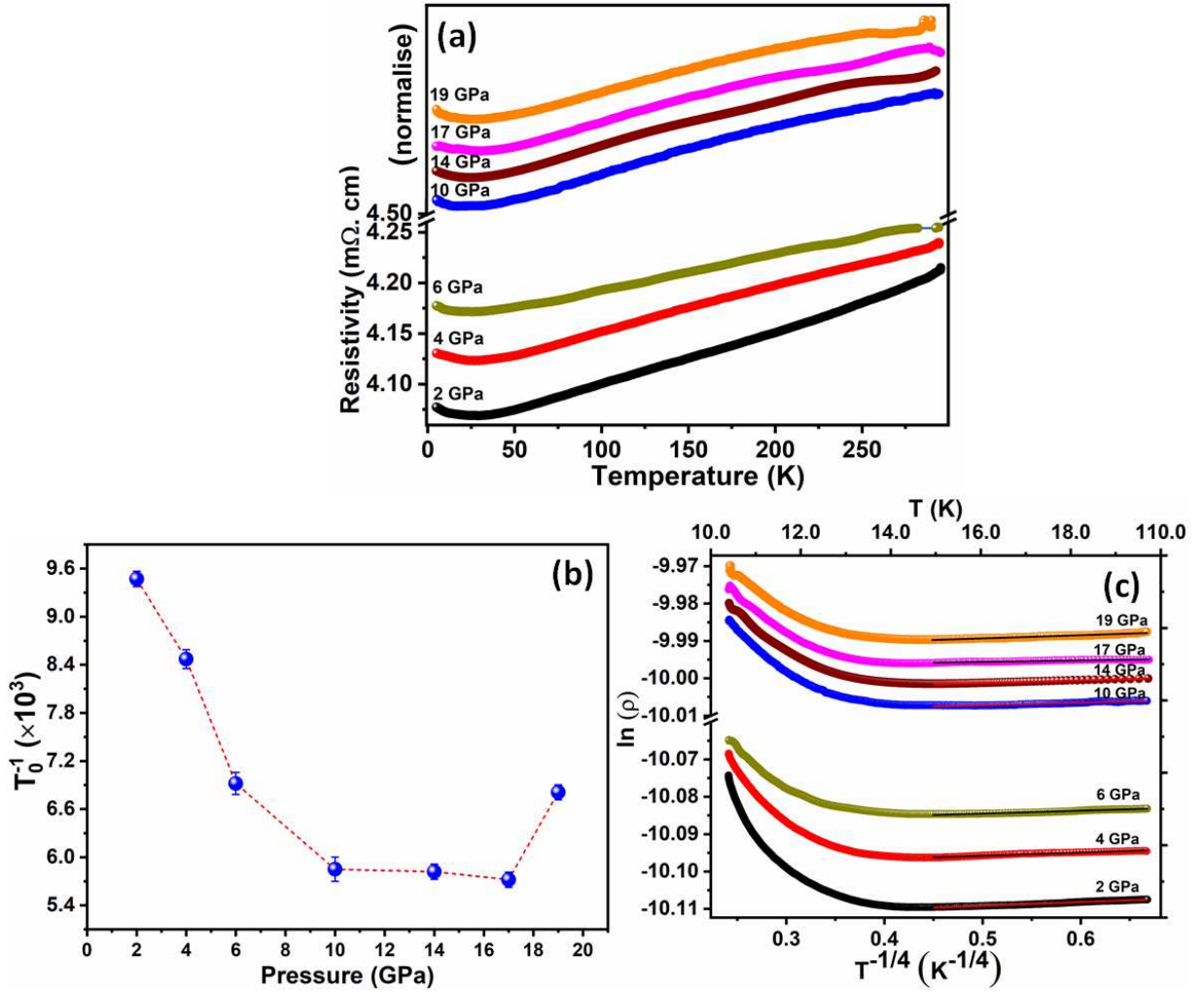
**Figure 1.** (a) Powder X-ray diffraction pattern of ScPdBi, insets show the crystal structure and scanning electron microscopic image of ScPdBi single crystal (b) Compositional analysis on ScPdBi single crystals (c) Elemental colour mapping on ScPdBi single crystal, showing uniform distribution of Sc, Pd and Bi elements.



**Figure 2.** (a) Temperature dependent resistivity of ScPdBi showing the metallic nature down to 2 K (b) Temperature dependent resistivity of ScPdBi at different magnetic fields, indicating there is no appreciable change in the resistivity with external magnetic field.

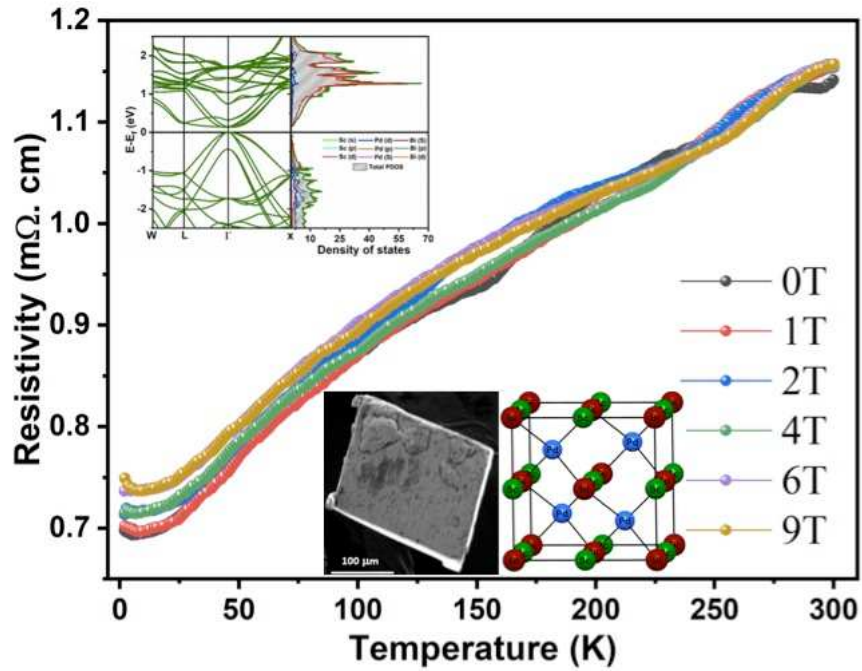


**Figure 3.** Temperature dependent specific heat of ScPdBi. A linear fit to the low temperature region resulted in a low Sommerfeld coefficient ( $\gamma \approx 2.59 \pm 0.87 \text{ mJ mol}^{-1} \text{K}^{-2}$ ) and Debye term ( $\beta \approx 0.87 \pm 0.01 \text{ mJ mol}^{-1} \text{K}^{-4}$ ).



**Figure 4.** (a) Temperature dependence of the resistivity of ScPdBi at different pressure (2 – 19 GPa), (b) Variation of  $T_0^{-1}$  with pressure, and (c) 3D Mott variable-range hopping (VRH) model.

## Graphical abstract



## Graphical abstract legend

Resistivity measurements at extreme conditions on ScPdBi single crystals reveal the metallic nature, which further confirmed by band structure calculations.