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# Synthesis and Thermo-Electric Properties of $Nb_2PdS_5$ Superconductor In Bulk.

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## Abstract

We report synthesis and thermal transport properties of ternary chalcogenide, $Nb_2PdS_5$ superconducting compound at low and room temperatures via thermo-electric measurements. Polycrystalline sample was synthesized via conventional solid-state reaction method. Seebeck coefficient (S) abruptly changes near superconducting transition temperature ($T_c \sim 6.5$ K). The positive value of S indicates the presence of holes like charge carriers in majority in normal state. The figure of merit, an essential qualitative parameter of thermo-electric materials, increases from low temperature towards the room temperature.

**Keywords:** Polycrystalline, Thermoelectric material, Seebeck coefficient, Superconductivity

## 1. INTRODUCTION

Nb-based non- superconducting ternary chalcogenide $Nb_2Pd_{0.81}Se_5$ compound exhibits superconductivity at transition temperature, $T_c \sim 6.5$K by substituting S in place of Se [1], and crystallized in monoclinic iso-structure like $Nb_2PdS_5$ of C2/m space group [1–3]. This superconducting compound has been got attention due to showing extremely large upper critical field ($H_{c2}$) among reported Nb-based superconductors [1–6]. The highest upper critical field ($H_{c2}$), at which superconductivity



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sustain, has been reported ~240 kOe in polycrystalline $Nb_2PdS_5$ superconductor [1, 3], higher than Pauli Paramagnetic limit ($H_{c2}$ ~$1.84T_c$) [7]. Andreev reflection spectroscopic measurements revealed its two band nature of superconductor like $MgB_2$ [7, 8] and nodeless superconducting gap in quasi 1-D, preserved time reversal symmetry [9]. These superconductors Cooper pair have *s*-wave superconducting order parameter [10]. Fermi surface of compound consists multiple sheets of both electron and holes thus theoretically also reported through DFT band structure calculations as multi-band superconductor [1, 11]. It is further corroborated in Andreev spectroscopy and transport measurements [12, 13]. The type of Fermi surface is resembled to the Fe-based superconductors whereas contradicts to the cuprate superconductors [3, 4]. Various research groups have tried to enhance the transition temperature via doping or substitution of chemical elements such as Ir, Pt, Ru and Ni at Pd site, these have slightly modified the $T_c$, and however Ag, and Ni completely suppressed superconductivity [1, 14–18]. Vinay et al., have reported the enhancement of the upper critical field by doping of Te at S site [19]. Jennifer et al., have studied doping concentration of Pd thoroughly in $Nb_2Pd_xSe$ structure and reported that Pd is intercalated into the structure, also confirmed from DFT [20]. During single crystal formation, this compound in form of fiber, one group has tried to make flexible fiber by coating it on paper [21]

Keeping in mind above studies, thermal-transport nature of the compound have been left to explore by which one may predict type of majority of charge carriers present in Fermi-surface before entering in superconducting phase. We have performed electrical and thermal transport measurements within superconducting as well as in normal states. This study also explore figure of merit of this compound which concern with conversion of heat energy into electrical energy.

## 2. EXPERIMENTAL TECHNIQUES

Polycrystalline sample of $Nb_2PdS_5$ were synthesized via conventional solid state reaction method as reported in references [3, 6]. Initial ingredients of high purity Nb(4N), Pd(4N) and S(5N) were taken from Alfa Aesar in desired stoichiometry and homogenized in agate/mortar. Fine homogenized grinded powder was filled in double wall quartz tube followed by evacuation up to $10^{-5}$ torr. Sealed evacuated quartz tube was loaded in a high temperature furnace and heated with the rate of 10 ºC/h for solid state reaction at temperature 800 ºC for 24 hours. Powder X-ray diffraction (XRD) was performed in Philips X'Pert Pro diffractometer with Cu-K radiation for phase purity analysis. Thermal transport and heat capacity measurements were performed in PPMS (Quantum Design Inc., USA).

## 3. RESULTS AND DISCUSSIONS

### 3.1 Structure Analysis

FIGURE 1 shows the powder XRD pattern of $Nb_2PdS_5$ sample along with Rietveld refinements results. The Rietveld refinement was performed in X'Pert plus software using monoclinic crystal lattice of C2/m space group as reference (ICSD: 53315) with atomic positions: Nb1: 4i(0.582, 0, 0.183), Nb2: 4i(0.156, 0, 0.378), Pd1:2a(0, 0, 0), Pd2:2c(0, 0, 0.5) and S5 (x,y,z) and temperature parameter B = 0.5. All intensity peaks are well matched with reference pattern. The lattice

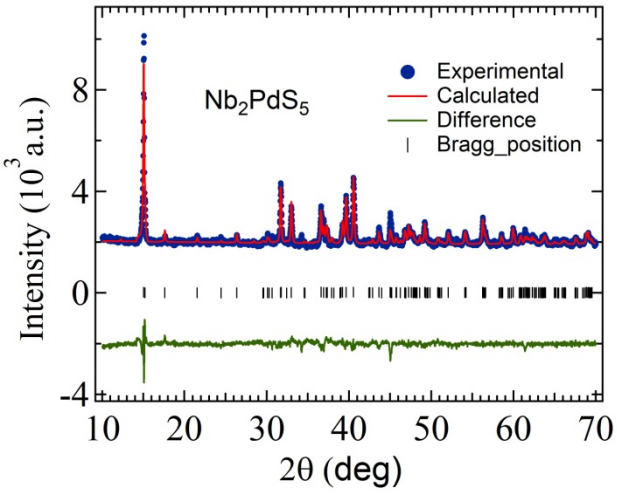


Figure 1: X-ray diffraction patterns of $Nb_2PdS_5$ polycrystalline sample. Peaks are well matched with monoclinic crystal structure of *C2/m* space group.

parameters are coming out from refinement and found to be, $a = 12.14(7)$ Å, $b = 3.28(1)$ Å and $c = 15.08(4)$ Å with $\chi^2 = 2.640$, comparable to previous published results [1–3].

### 3.2 Thermal Transport Properties

FIGURE 2(a) depicts the thermal conductivity ($\kappa$) measurements as a function of temperature in the range 2 – 300 K of $Nb_2PdS_5$ compound. In low temperature regime, $\kappa(T)$ gradually increases with increasing temperature and attains maximum $\kappa$ near 120 K. This is the typical behavior in thermal conductivity found for the metal and alloys, further limits arises due to the scattering of conducting electrons from grain boundaries [22]. The nature of $\kappa(T)$ curve is found reminiscent with the $MgB_2$ superconductor [23]. In metal, electrons and phonons, both are responsible for the transport of thermal energy. Contribution from electron only can be explained by Wiedemann-Franz Law *i.e.*, $K_e = L_0 T/\rho$, where $L_0$ (= $2.45\times10^{-8}$ W/M-K) is the Lorentz constant. Inset of FIGURE 2(a) shows the thermal conductivity at zero and 90 kOe magnetic fields within superconducting state. The higher field the $\kappa(T)$ curve deviates from zero field curve as magnetic field break the Cooper pairs and generate free electrons [23]. FIGURE 2(b) depicts the Seebeck coefficient ($S$) measurements as function of temperature in the range 2 - 300 K at zero magnetic fields. The variation of $S$ with $T$ is comparable with metallic compound. In superconducting state, initial $S(T)$ increases sharply with temperature and reach maximum $S \sim 17.8\mu$V/K at 43 K further, $S$ decrease gradually up to 100 K and rises again to attain maximum $S \sim 20.0\mu$V/K at 300 K.

This thermoelectric trend can be explained using the concept of diffusion ($S_d$) and phonon-drag ($S_{ph}$) components [22, 24]. In low temperature regime, say below Debye temperature, the $S_{ph}$ varies as third power of temperature in addition with diffusion component and above this temperature, $S_d$ varies inversely with temperature [24]. The sign $S$ can be negative or positive that refers

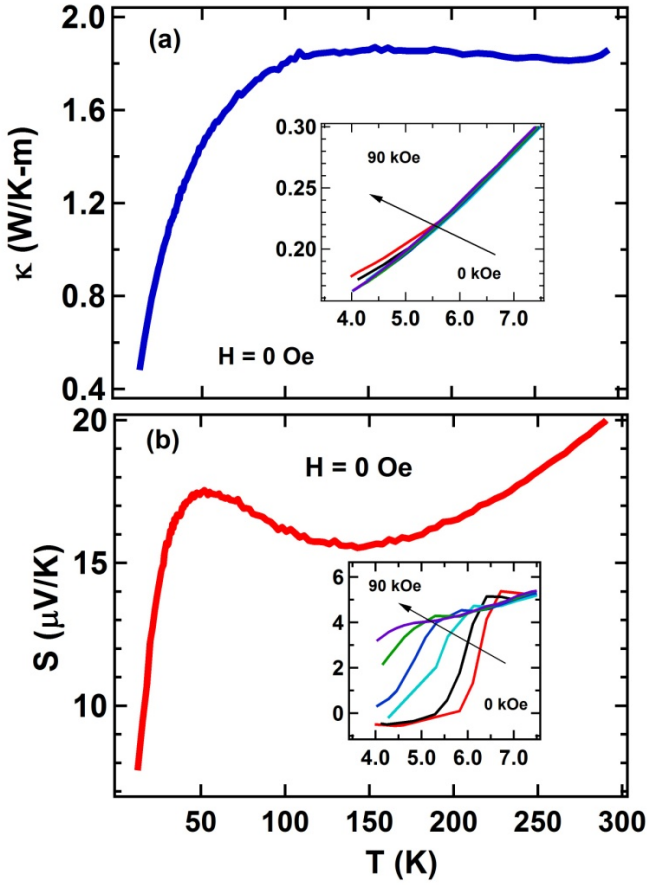


Figure 2: (a) Thermal conductivity vs temperature plot at zero magnetic field (b) Seebeck coefficient ($S$) vs temperature plot at zero magnetic field.

type of charge carriers in normal state [25], here, positive sign of $S$ suggests that holes are dominant charge carriers available for thermal conduction in normal state [1, 11, 26].

Thermo power relation with Fermi temperature is used to probe the correlation strength of system. Following the Fermi-liquid theory, Behania *et al.*, [26] have reported that diffusive Seebeck component should be linear with temperature in low temperature limit and magnitude at zero temperature provide strength of electronic correlation,

$$S/T = (\pm\pi^2 k_B)/2eT_F$$

where $k_B$ is Boltzmann's constant, $e$ electron charge and $T_F$ is Fermi temperature, it is similar to electronic specific heat response with temperature i.e. $C_{el}/T = \gamma$, at zero temperature limit, where $\gamma$ tells about electronic correlation strength. To get the value of $S/T$ at zero-temperature limit, the data of $S/T$ at higher temperature fitted linearly and further extrapolated lower to zero temperature. The quantitative value of $S/T$ at zero-temperature limit was found to be 0.79 $\mu V/K^2$ from fitting parameters and corresponding estimated Fermi temperature ($T_F$) is 538 K using above equation.

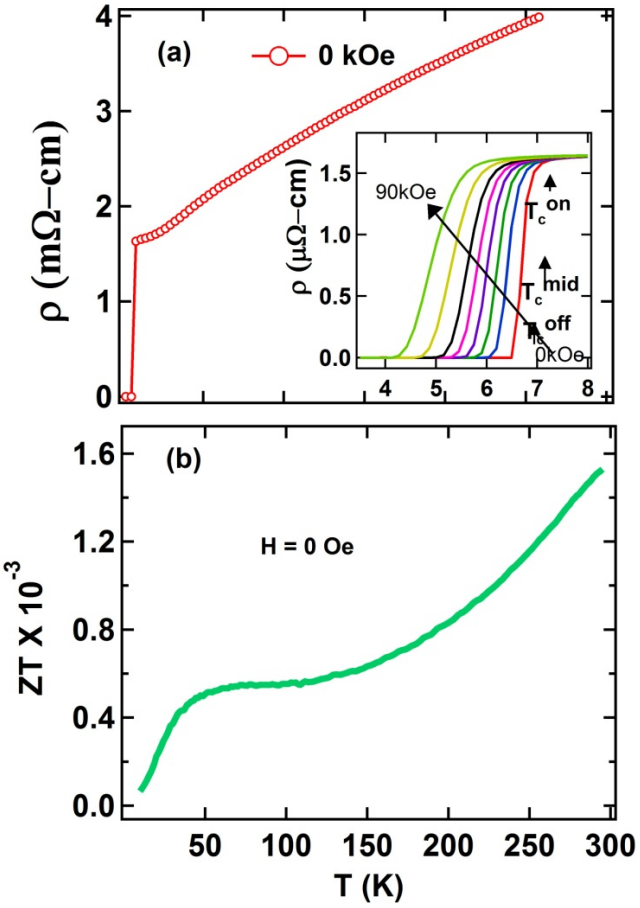


Figure 3: (a) shows $\rho$ vs $T$ plots at zero field in superconducting state. Inset shows resistivity plots at various magnetic fields ranges from 0-90kOe. (b) Figure of merit ($Z$) as a function of temperature.

Now, electron-phonon coupling strength is described by the ratio of $T_c/T_F$, [27] that is ~ 0.01 for this compound, this may suggest weakly correlated superconductor. Compound $KFe_{1-x}Se_y$ is also an example of weakly correlated iron superconductor, its ratio is found to be 0.04 [27]. Inset of FIGURE 2(b) shows temperature dependent of Seebeck coefficients plots in superconducting state at different applied fields, the transition temperature from normal to superconducting state shifts to lower temperature as applied magnetic fields increases, like resistivity behavior in superconducting state. The Seebeck coefficients changes abruptly from positive to zero value of S at $T_c$ in zero field measurement. FIGURE 3(a) shows electrical transport curves at zero magnetic fields with temperature from 2K to 300K. Resistivity drop abruptly to zero at transition temperature ($T_c$ ~ 6.5K). Inset shows transport measurement under magnetic fields, the transition temperature shifts to lower temperature as field increases, similar to Seebeck coefficient measurements. Figure of merit for thermoelectric materials defines the thermal to electrical conversion efficiency, FIGURE 3(b) displays figure of merit ($ZT$) as function of temperature, calculated by using formula $ZT = S^2\rho/\kappa$. The moderate value of $ZT \sim 1.5 \times 10^{-3}$ is observed at room temperature.

## 4. CONCLUSIONS

In summary, polycrystalline sample of $Nb_2PdS_5$ was prepared successfully via solid state reaction method. Thermal conductivity curve show typical trend of metal. Seebeck coefficient measurements confirm positive charge carriers (holes) are present in normal state for thermal transport and at transition temperature S changes abruptly. In electrical-magneto transport study, metallic nature is to be observed in normal state and superconducting transition temperature suppresses with increasing magnetic fields. A lower value of figure of merit is estimated for this compound.

## 5. ACKNOWLEDGMENTS

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