Probing the topological surface states in superconducting Sn$_4$Au single crystal: a magneto transport study

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Abstract

Materials exhibiting bulk superconductivity along with magnetoresistance (MR) in their normal state have emerged as suitable candidates for topological superconductivity. In this article, we report a flux free method to synthesize single crystal of topological superconductor candidate Sn$_4$Au. The phase purity and single crystalline nature are confirmed through various characterizations viz. x-ray diffraction, field emission scanning electron microscopy, selected area electron diffraction, and transmission electron microscopy. Chemical states of the constituent element viz. Sn and Au are analysed through x-ray photoelectron spectroscopy. Superconductivity in synthesized Sn$_4$Au single crystal is evident form $\rho$-$T$ plot, for which the critical field ($H_c$) is determined through $\rho$-$H$ plot at 2 K i.e. just below critical temperature $T_c$. A positive MR is observed in $\rho$-$H$ measurements at different temperatures above $T_c$, viz. at 3 K, 5 K, 10 K and 20 K. Further, the magnetoconductivity (MC) is analysed by using Hikami–Larkin–Nagaoka formalism, which signifies the presence of weak antilocalization (WAL) effect in Sn$_4$Au. Angle dependent magneto-transport measurement has been performed to detect the origin of observed WAL effect in Sn$_4$Au single crystal. Normalized MC vs $H \cos \theta$ plot shows presence of topological surface states in the studied system. It is evident that Sn$_4$Au is a 2.6 K topological superconductor.

Keywords: topological superconductors, topological semimetal, magneto-resistance, weak antilocalization, Hikami–Larkin–Nagaoka model

(Some figures may appear in colour only in the online journal)

1. Introduction

The quest on search for topological materials with superconducting properties has revolutionized the field of quantum condensed matter. The unique combination of bulk superconducting gap and topological surface states (TSS) enables topological superconductors to host Majorana fermions [1–4].

Recently, some of the Dirac semimetals (DSMs) have gathered significant interest in this regard, as they are found to show simultaneous existence of bulk superconducting gap and TSS [5–7]. DSMs have emerged as transitional state of topological trivial insulators to topological insulators (TIs) and vice versa [8, 9]. In DSMs, the Dirac point is not gapped due to intrinsic spin orbit coupling (SOC), instead this is the only point where the conduction band and valence band meet each other. These electronic bands are linearly dispersed in all directions around the Dirac point, thus forming a Dirac cone [9]. The spins of the
surface electrons circulate around the Dirac cone, and create π Berry phase in electron wavefunction \([10, 11]\). Presence of π Berry phase in a topological material, is considered to be the vital property of the same, which provides immunity to TSS against non-magnetic impurities \([12]\). Observation of quantum oscillations viz. Shubnikov-de Haas or de Haas-van Alphen along with weak anti-localization (WAL) effect in magneto-transport measurements signify the presence of π Berry phase in a topological material \([13–16]\). The electrical conduction through surface states of a TI is vulnerable to applied magnetic field, and as a consequence, the same results in occurrence of linear magnetoresistance (LMR) in magneto-transport measurements \([5, 17, 18]\).

Study of normal state transport properties of possible topological superconductors is an essential part to probe TSS. The transport phenomenon in normal state of a topological superconductor must principally be dominated by TSS, resulting in occurrence of significant MR above \(T_c\). Some of the recently studied superconducting topological semimetals (TSMs) such as Au\(_2\)Pb \([5]\), Pd\(_2\)Te \([17]\), Pt\(_2\)TaSe\(_4\) \([19]\), Sn\(_3\)Au \([20]\) etc have shown a large MR in their normal state, creating possibility of realization of topological superconductivity. Among these, Sn\(_3\)Au is isostuctural to recently discovered TSMs viz. Sn\(_3\)Pd \([21]\) and Sn\(_3\)Pt \([22]\). Superconducting properties of Sn\(_3\)Au were though discovered a long time ago \([23, 24]\), but any detailed analysis of both its superconducting and normal state properties is reported only very recently \([20]\). Sn\(_3\)Au exhibits large MR similar to its isostuctural compounds Sn\(_3\)Pd and Sn\(_3\)Pt, which are considered to have Dirac nodal arc semimetal like band structure \([21, 22]\). Sn\(_3\)Au also lie in category of TSMs based on available theoretical reports \([25, 26]\), while experimental confirmation of the same by various physical property measurements is yet to be done. On this front, Sn\(_3\)Au has been reported to exhibit some interesting superconducting and normal state properties \([20, 27]\). The low temperature magnetoconduction (MC) of Sn\(_3\)Au in its normal state, is seemingly contributed by TSS as shown in \([20]\). The exotic properties of Sn\(_3\)Au inspire one to study more about its superconducting and normal state properties and to compare with only available reports \([20, 27]\), as well to further extend the analysis. In current article not only the available experimental results \([20, 27]\) are reproduced but further the normal state angle dependent magneto transport is included and thus the presence of normal state topology along with low temperature superconductivity is ascertained. Besides, our magneto-transport studies in superconducting regime conclude the Sn\(_3\)Au to be a type-I superconductor instead of type-II as concluded from magnetization measurements in \([20]\).

The single crystal of Sn\(_3\)Au is synthesized using a flux free method and is well characterized using x-ray diffraction (XRD), field emission scanning electron microscopy (FESEM), transmission electron microscopy (TEM) and x-ray photoelectron spectroscopy (XPS) measurements. \(\rho\)-\(T\) measurements show the presence of superconductivity with onset critical temperature \(T^{\text{onset}}_c\) at 2.6 K. Magneto-transport measurements shows significant MR, in an applied field of \(\pm 12\) T. Though, in previous study on normal state properties of Sn\(_3\)Au, MC was fitted with simple WAL transport formula, here we used Hikami–Larkin–Nagaoka (HLN) equation to fit the same, and confirmed that the normal state transport is clearly dominated by TSS. This dominance of TSS is also evidenced through angle dependent magneto-transport measurements. This report clearly shows that normal state transport in Sn\(_3\)Au is dominated by TSS, while the same material shows superconductivity below its \(T_c\), establishing Sn\(_3\)Au to be a topological superconductor.

2. Experimental

Single crystal of Sn\(_3\)Au is synthesized using a flux free method, here it is worth mentioning that all previous methods to synthesize Sn\(_3\)Au single crystal were based on flux method, in which excess of Sn amount is taken as flux. Here, high quality powders of Sn and Au were taken in stoichiometric ratio \((4:1)\). These powders were grinded thoroughly in inert atmosphere and pelletized using hydraulic press. This palletized sample was vacuum encapsulated in a quartz ampoule. This ampoule was heated to 900 \(^\circ\)C at a rate of 120 \(^\circ\)C \(h^{-1}\) and dwelled at this elevated temperature for 24 h. This temperature is well above the melting temperature of Sn\(_3\)Au, the sample is heated to a higher temperature to melt the constituent element. The melted sample is cooled to 310 \(^\circ\)C at a cooling rate of 10 \(^\circ\)C \(h^{-1}\), and kept at 310\(^\circ\)C for 6 h. Further, the sample is cooled down to 240 \(^\circ\)C at a relatively slow cooling rate of 1 \(^\circ\)C \(h^{-1}\). This slow cooling rate provides ample time for atoms to attain their energetically stable positions, which is a necessary step in crystal growth. A hold period of 48 h is applied at 240 \(^\circ\)C, at which stabilization of phase took place. After that the sample is allowed to cool normally to room temperature. Though, all heating protocols are same as in \([20]\), the important difference is that the initial heating temperature in our case is 900 \(^\circ\)C, instead of 630 \(^\circ\)C in \([20]\). This higher heating temperature removes the need of adding extra Sn as a flux. Thus, obtained crystal is silvery shiny and cleavable along its growth axis. The schematic of heat treatment is shown in figure 1. This method is relatively easier method to synthesize single crystal of Sn\(_3\)Au, than the previously suggested flux method \([20]\), as the same does not require centrifuging of extra Sn flux.

XRD pattern on mechanically cleaved flake and powder XRD (PXRD) pattern were recorded using Rigaku mini flex-II table top XRD equipped with Cu-K\(_{\alpha}\) radiation. MIRA II LMH from TESCAN made FESEM equipped with energy-dispersive x-ray spectroscopy (EDS) is used to visualize the surface morphology and to determine elemental composition. Selected area electron diffraction (SAED) pattern and atomic planes are visualized using JEOL/JEM-F200 with One view CMOS camera \((4K \times 4K)\) TEM. XPS spectra was recorded by using XPS (Model: K-Alpha-KAN9954133, Thermo Scientific) with mono-chromated 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 1s line at the binding energy 284.8 eV. The working pressure was maintained \(<6 \times 10^{-8}\) mbar. The scans for Au 4f and...
Sn 3d were recorded with the step size of 0.1 eV. Low temperature magneto-transport measurements were carried out using Quantum Design physical property measurement system (QD-PPMS) equipped with sample rotator. Standard four probe method is used for electrical transport measurements. Hall measurements are carried out using Vander Paw geometry. Rietveld refinement of PXRD pattern is performed using Full Proof software and unit cell is drawn in VESTA software, which uses crystallographic information file (CIF) obtained from Rietveld refinement.

3. Result and discussion

Phase purity of the synthesized Sn₄Au single crystal is confirmed through Rietveld refined PXRD pattern, and the observed and fitted plots are shown in figure 2. Rietveld refinement is performed taking parameters of an orthorhombic crystal structure with Aba2 (41) space group symmetry. All observed peaks in PXRD pattern are well fitted with the applied parameters. Quality of fit is given by $\chi^2$ value, which is found to be 1.81 and is in an acceptable range. The lattice parameters obtained from Rietveld refinement are $a = 6.514(5)\ \text{Å}$, $b = 6.529(1)\ \text{Å}$ and $c = 11.726\ (6)\ \text{Å}$ and $\alpha = \beta = \gamma = 90^\circ$. These lattice parameters and atomic co-ordinates are listed in table 1, along with the refinement parameters. No impurity peak is visible in PXRD pattern, this shows that the synthesized Sn₄Au single crystal is phase pure. PXRD pattern is more effective to determine space group symmetry and phase purity. Here PXRD pattern of Sn₄Au, eventually confirms that Sn₄Au crystallizes in orthorhombic phase with Aba 2 space group symmetry. XRD pattern taken on mechanically cleaved crystal flake of synthesized crystal is shown in right inset of figure 2. High intense and sharp peaks are observed for some specific planes i.e. (002) planes, where $n = 1, 2, 3, 4$. This shows that the synthesized crystal is grown uni-directionally along c-axis, confirming the single crystalline nature of synthesized crystal. Unit cell of synthesized Sn₄Au drawn in VESTA software, is shown in left inset of figure 2. Each unit cell consists of two blocks of Sn₄Au crystal, which is in accordance with the observed peaks for (002n) planes in XRD pattern taken on crystal flake. 

Surface morphology of synthesized crystal is viewed through FESEM image, and is shown in figure 3(a). The synthesized crystal has typical terrace type morphology and the planes are arranged in a regular array. This shows the layer-by-layer growth of synthesized Sn₄Au single crystal, confirming the crystalline nature of the same. EDS spectra is recorded to determine stoichiometry and purity of the sample and the same is shown in figure 3(b). EDS spectra shows that only two elements viz. Sn and Au are present in the synthesized Sn₄Au.
sample and no other impurity element is seen. This shows the purity of the sample. Both the elements viz. Sn and Au are found to be in near stoichiometry ratio as shown in the inset of figure 3(b). Further, the single crystalline nature of the synthesized sample is verified by taking SAED pattern, as shown in figure 3(c). Typically, SAED pattern of a single crystal contains symmetrical spots due to diffraction from regularly arranged atomic planes. The SAED pattern of Sn$_4$Au shows symmetrical diffraction spots, showing crystalline nature of the same. Spots from (002), (004), (006) and (008) planes are marked in figure 3(c). Figure 3(d) shows the TEM image of Sn$_4$Au single crystal, clearly exhibiting stacking of (004) atomic planes with a regular interval. These planes are separated by 2.95 Å, which matches with the inter-atomic distance obtained for (004) planes in XRD measurement. The stacking of (004) planes as observed in TEM image is in agreement with XRD pattern taken on crystal flake, which also shows the unidirectional growth of crystal along (002n) direction.

XPS spectroscopy is used to determine chemical states of constituent elements of synthesized Sn$_4$Au crystal. XPS peaks are calibrated with C 1s peak, which is considered as reference. XPS spectra recorded in Sn3d and Au4f regions is shown in figures 4(a) and (b). Gaussian fitting formula is used to fit the XPS peaks. Figure 4(a) shows the fitted spin orbit doublet of Sn 3d core levels. These peaks are deconvoluted in four peaks two for SnO$_2$ which corresponds to Sn$^{4+}$ state and the other two for Sn$^{2+}$ state [29, 30]. Peaks for SnO$_2$ are observed due to air exposure of the sample. Peaks for Sn$^{2+}$ spin orbit doublets viz. Sn 3d$_{5/2}$ and Sn 3d$_{3/2}$ are found to be at 484.61 ± 0.009 eV and 493.05 ± 0.008 eV. The separation between these two peaks is found to be 8.44 eV which is comparable with standard value 8.4 eV [30]. Figure 3(b) shows XPS spectra recorded in Au 4f region. This shows the peaks due to spin orbit doublet of Au 4f core levels viz. Au 4f$_{7/2}$ and Au 4f$_{5/2}$. These peaks are occurred at 84.91 ± 0.004 eV (Au 4f$_{7/2}$) and at 85.99 ± 0.005 eV (Au 4f$_{5/2}$). The separation between these peaks is found to be 3.68 eV, which is comparable to standard value 3.67 eV [30]. In [30], XPS peak corresponding to Au 4f$_{5/2}$ is specifically mentioned for Sn$_4$Au compound. It is shown to be occurring in proximity of 85 eV, and is observed in our case also. The observed values of XPS peaks in Au 4f regions confirm the formation of Sn$_4$Au phase. The peak position along with the full width half maximum (FWHM) for various spin–orbit doublets of Sn and Au are given in table 2.

Figure 5(a) depicts $\rho$-$T$ measurements plot without magnetic field. A clear superconducting transition is observed with $T_c$ at 2.6 K, which is close to previously reported value of 2.5 K [20, 23, 24]. Superconducting transition is sharp as can be seen in inset of figure 5(a). Zero resistivity is attained at 2.36 K, providing a transition width of 0.25 K. This signifies the quality of studied crystal. The $\rho$-$T$ data is fitted in whole range above $T_c$ (5 K–250 K) using Bloch–Grüneisen (B-G) formula. $\rho(T)$, which is described by the following formalism,

$$
\rho(T) = \left[ \frac{1}{\rho_s} + \frac{1}{\rho_i(T)} \right]^{-1}.
$$

![Figure 3. (a) FESEM image of synthesized Sn$_4$Au single crystal. (b) EDS spectra of synthesized Sn$_4$Au single crystal in which inset is showing the atomic composition of the same. (c) SAED pattern of synthesized Sn$_4$Au single crystal. (d) TEM image of synthesized Sn$_4$Au single crystal.](image-url)
Table 2. XPS peaks position and FWHM of constituent elements of synthesized Sn₄Au single crystal.

| Element | Spin–orbit doublet | Binding energy | FWHM   |
|---------|-------------------|---------------|--------|
| Sn      | 3d_{5/2}          | 484.61 ± 0.009 eV | 0.76 ± 0.03 eV |
|         | 3d_{3/2}          | 493.05 ± 0.008 eV | 0.77 ± 0.04 eV |
| Au      | 3d_{5/2}          | 84.91 ± 0.004 eV  | 0.82 ± 0.09 eV  |
|         | 3d_{3/2}          | 88.59 ± 0.005 eV  | 0.81 ± 0.01 eV  |

Here, \( \rho_s \) represents temperature independent saturation resistivity and \( \rho(T) \) is given by following equation,

\[
\rho(T) = \rho(0) + \rho_{el-ph}(T). \tag{2}
\]

Here \( \rho(0) \) represents residual resistivity arising due to impurity scattering, the second term \( \rho_{el-ph}(T) \) represents temperature dependent term, which depend on electron–phonon scattering. Further, \( \rho_{el-ph}(T) \) is given by the following formula

\[
\rho_{el-ph} = \alpha_{el-ph} \left( \frac{T}{\theta_D} \right)^n \int_0^{\frac{2\pi}{\theta_D}} \frac{x^n}{(1 - e^{-x}) \times (e^x - 1)} \, dx. \tag{3}
\]

Here \( \alpha_{el-ph} \) is electron–phonon coupling parameter, \( \theta_D \) represents Debye temperature and \( n \) is constant. \( \rho-T \) data is well fitted with the above equation for \( n = 5 \), signifying dominant electron–phonon scattering. Residual resistivity ratio is found to be 19.06, which shows high metallicity of the synthesized crystal. The obtained value of Debye temperature \( \theta_D \) from above fitting formula is 127.50 ± 0.77 K.

Figure 5(b) shows the \( \rho-T-H \) measurements plot of synthesized Sn₄Au single crystal. A clear suppression in \( T_c \) is observed with applied magnetic field. It has been observed that onset of \( T_c \) remains above 2 K up to 120 Oe, while zero resistivity is not achieved above 100 Oe. Variation in critical field \( H_c \) is plotted against the temperature in inset of figure 5(b). This plot is fitted with conventional quadratic equation as given below

\[
H_c(T) = H_c(0) \times \left( 1 - \frac{T^2}{T_c^2} \right). \tag{4}
\]

Here, \( H_c(0) \) is critical field at absolute zero, which is found to be 248 Oe. Another approach to determine \( H_c(0) \) is using Werthamer–Helfand–Hohenberg formula, which is as follows:

\[
H_c(0) = \frac{0.693T_c}{T_c} \left( \frac{dH_c}{dT} \right)_{T_c}. \tag{5}
\]

Linearly fitted plot is shown by blue curve in inset of figure 5(b), which is further extrapolated to \( T = 0 \). \( H_c(0) \) is estimated by multiplying extrapolated value by 0.693. Thus obtained \( H_c(0) \) is found to be 285 Oe.

Hall resistivity \( (\rho_H) \) is measured with respect to applied field in a range of ±4 T at 3 K and the same is shown in figure 5(c). Field dependent Hall resistivity gives the information about carrier density and nature of the carriers. \( \rho_H \) is found to have linear relationship with applied magnetic field. Hall coefficient is determined by taking the slope of linearly fitted \( \rho_H \) vs \( H \) plot. Hall coefficient is found to be \(-4.05 \pm 0.02 \times 10^{-10} \Omega \, \text{m} \, \text{T}^{-1}\). The negative sign of Hall coefficient shows that the electrons are the dominant charge carriers. The charge carrier density is calculated by using the formula, \( n = \frac{1}{R_H} \), where charge carrier density is given by \( n \) and \( e \) is the electronic charge. The calculated value of charge carrier density is found to be 1.54 ± 0.03 × 10¹⁹ m⁻³. Further, this value is verified by calculating charge carrier density theoretically. Sn₄Au unit cell contains four formula units, in which each formula unit provides two electrons. In this way, Sn₄Au has eight electrons per unit cell. The total electron density is calculated by dividing number of electrons per unit cell by volume of unit cell. Volume of Sn₄Au unit cell obtained from Rietveld refinement is \( V = 498.783 \, \text{Å}^3 \). The electron density is found to be \( 1.60 \times 10^{28} \, \text{m}^{-3} \), this value...
closely matches with the value obtained from the experiment. The value of charge carrier density is used to determine Fermi wave vector by considering a spherical Fermi surface. Fermi wave vector is given by \( k_F = \frac{3n^2}{\pi^2} \), where \( n \) is charge carrier density. Here, the value of \( n \) obtained from Hall calculations is used, and the corresponding Fermi wave vector \( k_F \) is found to be 0.78 Å\(^{-1}\). These values are used to calculate the superconductivity characteristic lengths viz. London penetration depth \( \lambda \) and BCS coherence length \( \xi \). The value of London penetration depth \( \lambda \) can be calculated using the relation \( \lambda = \frac{m^*}{\pi e^2} \), where \( m^* \) is taken as free electron mass for simplicity. The obtained value of \( \lambda \) is 42.86 nm. The BCS coherence length is calculated using the formula \( \xi = \frac{\hbar^2}{2m^*k_B}\frac{1}{\pi^2} \), and it is found to be 480.7 nm. The values of both the superconducting lengths are used to determine Ginzberg Landau (GL) \( \kappa \) parameter. \( \kappa \) parameter is given by \( \kappa = \frac{\lambda_0}{\Lambda} \), and its value is found to be 0.08, which is less than cut-off value for type-I superconductivity. This shows Sn\(_4\)Au to be a type-I superconductor. \( \kappa \) parameter can also be determined by calculating the value of mean free path. The mean free path \( l \) can be calculated using the formula \( l = \frac{v_F}{\omega_F} \), where \( v_F \) is Fermi velocity and given by \( v_F = \frac{\hbar}{m^*} \), and \( \tau \) is scattering time and given by \( \tau = \frac{m^*}{\pi^2} \), here \( \rho \) is taken as residual resistivity \( \rho = 1.08 \times 10^{-7} \) Ω m. The value of mean free path \( l \) is found to be 119.2 nm. Certainly, \( \xi(0) \) is very large as compared to the mean free path, suggesting Sn\(_4\)Au to be a dirty limit superconductor. For a dirty limit superconductor, GL \( \kappa \) parameter can be calculated as \( \kappa = \frac{0.75\lambda_0(0)}{\Lambda} \), the obtained value of \( \kappa \) is 0.27 which is lower than the limit of type-I superconductivity, suggesting Sn\(_4\)Au to be a type-I superconductor. In [20], Sn\(_4\)Au is shown to have type-II superconductivity with very close lower critical field \( (H_{c1}) \) and upper critical field \( (H_{c2}) \), as seen from magnetization measurements. In present article Sn\(_4\)Au is seen to be a type-I superconductor based on \( \kappa \) parameter, which is deduced from magneto transport measurements. Similar feature has been observed in \( \beta \)-IrSn\(_4\) and KBi\(_2\), in which magnetization measurements suggest the same to be a type-II superconductor, while the value of \( \kappa \) parameter obtained by considering spherical Fermi surface, indicated the superconductivity to be of type-I [31, 32]. Incidentally, there are some other type-I superconductors also that shows similar type of \( M-H \) curve [33, 34]. Also need to be considered is the fact that magnetization measurements strongly depends on sample shape and geometry and this could be the reason for the observed distinct \( H_{c1} \) and \( H_{c2} \) in [20]. In this regard, magneto transport measurements [31, 32] are safer to conclude the type of superconductivity in such systems having closer values of \( H_{c1} \) and \( H_{c2} \). Our magneto transport data analysis shows that Sn\(_4\)Au is a type-I superconductor. For our sample, various superconducting parameters for present self-flux grown Sn\(_4\)Au are given in table 3.

Figure 6(a) shows \( \rho \) vs \( H \) plot of Sn\(_4\)Au single crystal at 2 K, 3 K, 5 K and 10 K in a field range of ±0.8 T. At 2 K, sample
is in its superconducting state and its resistivity remains zero up to 120 Oe, and sharply increases when the field is further increased. This shows $H_c$ to be 120 Oe at 2 K, which is in accordance with the $\rho TH$ measurements. $\rho H$ plots show no indication of superconductivity at 3 K, 5 K and 10 K. An interesting feature is observed in terms of increase of resistivity with field, hinting towards significant MR in normal state. The study of normal state of a topological superconductor is very crucial to determine normal state topological properties of the same. To analyse normal state properties of synthesized Sn₄Au single crystal, $\rho H$ measurements are carried out in a field range of $\pm 12$ T in normal state i.e. at temperatures 3 K, 5 K, 10 K and 20 K, which are shown in figure 6(b). MR% has been calculated by using the following formula

$$MR\% = \frac{\rho(H) - \rho(0)}{\rho(0)} \times 100.$$  

MR% is found to be around 150%, 120%, 110% and 65% at 3 K, 5 K, 10 K and 20 K respectively. This positive MR in normal state, indicates towards possible surface states driven transport in Sn₄Au single crystal. MR seems to be linear and non-saturating in complete magnetic field range. There can be two possible reasons for the observed LMR. The first one has the classical origin and emerges due to impurity scattering [35] in an inhomogeneous sample. This reason is not valid here as the synthesized Sn₄Au crystal is highly crystalline and there is no indication of inhomogeneity in the sample. The other reason for LMR has the quantum origin, which is observed in topological materials [36–38]. In topological materials, occurrence of LMR is attributed to the surface states dominated transport phenomenon, while a deviation from linear to quadratic dependency of MR on magnetic field indicates bulk dominated transport properties. Here, high field MR at 3 K is fitted with power law i.e. $MR\% = kH^n$ (inset of figure 6(b)), where the value of exponent $\gamma$ determine whether the conduction is dominated by bulk states or the TSS. The value of $\gamma$ near to 1 (linear) shows surface states dominated conduction mechanism while the value of $\gamma$ near to 2 (quadratic) shows that the conduction is dominated by bulk states and has the classical origin. For synthesized Sn₄Au single crystal, this value of $\gamma$ is found to be 0.8, which is close to 1, and shows that the conduction is dominated by TSS [38]. Interestingly a V type cusp has also been observed in low field MR data, which signifies possible existence of WAL effect in synthesized Sn₄Au single crystal [39]. This V type cusp broadens at higher temperatures, this happens due to shortening of coherence length $\xi$. This possible WAL effect is verified by analysing MC at different temperatures.

In figure 6(c), MC is plotted against the applied field in a temperature range of $\pm 1$ T at 3 K, 5 K, 10 K and 20 K. Presence of WAL effect in a topological material, can be confirmed by fitting low temperature MC with HLN formalism [40]. In HLN formalism, the difference in MC $\Delta \sigma(H)$ is taken as $\sigma(H) - \sigma(0)$, and the same can be described with the following equation

$$\Delta \sigma(H) = -\frac{\alpha \pi^2}{\pi^2} \ln \left(\frac{B_\phi}{H}\right) - \Psi\left(\frac{1}{2} + \frac{B_\phi}{H}\right).$$  

Here, $B_\phi$ is the characteristic field and is given by $B_\phi = \frac{\hbar}{8\pi e^2\xi}$, $\xi$ is phase coherence length and $\Psi$ is digamma function. The phase coherence length $\xi$ is defined as the maximum length travelled by electron while maintaining its phase. The value of pre-factor $\alpha$ determines which type of localization is present in the system, a positive value of $\alpha$ signifies weak localization (WL) effect, while a negative value of $\alpha$ indicates the presence of WAL effect. In topological materials, WAL is supposed to be induced by TSS. These TSS are protected by $\pi$ Berry phase. For a material with $\pi$ Berry phase, the value of pre-factor $\alpha$ should be $-0.5$ [39]. Intriguingly, the value of $\alpha$ depends upon number of TSS, for each TSS, $\alpha$ takes the value to be $-0.5$. If $\alpha$ takes the value to be equal to $-1$, it shows that there are two distinct TSS are present in the system, one is at the top and other at the bottom [39, 41]. The value of $\alpha$ between $-0.5$ and $-1$, shows that the top and bottom surface states are connected through bulk conducting channels [42]. This suggest that the conduction is contributed by the both viz. bulk conducting channels and TSS. In figure 6(c), the solid black curves show the HLN fitting results at 3 K, 5 K, 10 K and 20 K. The obtained value of pre-factor $\alpha$ and phase coherence length $\xi$ are listed in table 4. The value of $\alpha$ at 3 K is found to be $-1.028$, which is near to $-1$, and suggest that two distinct TSS contribute to the conduction mechanism in synthesized Sn₄Au single crystal. Also, it has been observed that as the temperature is increased, the value of $\alpha$ is decreased, and it is found to be $-0.2966$ at 20 K as shown by encircled dots in figure 6(d). The deviation of the value of $\alpha$ from the standard values viz. $-0.5$ and $-1$, suggest that bulk states start to contribute in conduction at higher temperatures. The variation of inverse of square of phase coherence length $\xi^{-2}$ with respect to temperature is shown by blue symbols in figure 6(d). Temperature dependence of $\xi$, gives the information about the scattering and dephasing mechanism. According to Nyquist theory, if $\xi^{-2}$ varies linearly with temperature, this shows that there is only electron–electron (e-e) scattering present in the system [43, 44]. In general, at low temperatures, e-e scattering is the only dephasing mechanism, while at higher temperatures this scattering is assisted by electron–phonon (e-p) scattering [44]. In figure 6(d), the solid red line shows linearly fitted $\xi^{-2}$ vs T plot, and it is clear that $\xi^{-2}$ is not well fitted with linear equation, suggesting the presence of both scattering processes viz. e-e and e-p. To determine nature of WAL effect, whether

| Table 3. Parameters obtained from heat capacity measurements. |
|-------------------|------------------|
| Parameter         | Obtained value   |
| Superconducting critical temperature ($T_c^{\text{net}}$) | 2.6 K            |
| Debye temperature ($\theta_D$)                           | 127.5 $\pm$ 0.77 K |
| Critical field at absolute zero, $H_c(0)$                 | 248 Oe           |
| Charge carrier density ($n_c$)                           | $1.54 \pm 0.03 \times 10^{29}$ m$^{-3}$ |
| London penetration depth $\lambda(0)$                     | 42.86 nm         |
| BCS coherence length $\xi(0)$                            | 480.7 nm         |
| Mean free path ($l$)                                      | 119.2 nm         |

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Figure 6. (a) Resistivity vs field plot at 2 K, 3 K, 5 K and 10 K. (b) MR% vs H plot at 3 K, 5 K, 10 K and 20 K of synthesized Sn$_4$Au single crystal, in which inset is showing the power law fitted high field MR% at 3 K. (c) Low field ($\pm$ 1 T) HLN fitted MC of synthesized Sn$_4$Au single crystal. (d) Variation of parameters obtained from HLN fitting with respect to temperature, wine-coloured symbols are showing variation of $\alpha$ with respect to temperature, while blue symbols are showing variation of $l^{-2}_\phi$ with respect to temperature. Solid red and black curve are showing linear fitted and power law fitted $l^{-2}_\phi$ vs $T$ plot respectively.

Table 4. Low field (up to 1 T) HLN fitted parameters of Sn$_4$Au single crystal.

| Temperature (K) | $\alpha$  | $l_\phi$ |
|----------------|-----------|----------|
| 3              | -1.028    | 113.0303 |
| 5              | -0.7973   | 101.6673 |
| 10             | -0.7473   | 87.4037  |
| 20             | -0.2966   | 61.6531  |

it is 2D or 3D, the $l^{-2}_\phi$-$T$ plot is fitted with the power law as follows [45],

$$\frac{1}{l^2_\phi(T)} = \frac{1}{l^2_\phi(0)} + A_{e-e}T^p + A_{e-p}T^q.$$ (8)

Here, $l_{\phi}(0)$ is the dephasing length at absolute zero and $A_{e-e}T^p$ and $A_{e-p}T^q$ represents the contribution from e-e scattering and e-p scattering respectively. The fitted plot is shown with solid black line in figure 6(d). Here, the value of $p$ and $q$ are found to be 1 and 2 respectively. The values of $A_{e-e}$ and $A_{e-p}$ are found to be $3.101 \times 10^{-6}$ and $3.30 \times 10^{-7}$. This result shows that both the scattering process viz. e-e and e-p scattering play significant roles in dephasing mechanism of Sn$_4$Au. Also, the obtained values of $p$ and $q$ show that the both e-e and e-p scattering are 2D in nature. This signifies the presence of 2D WAL effect in synthesized Sn$_4$Au single crystal. This because in 3D WAL effect, 3D e-p scattering dominates, and the power law is changed from $l^{-2}_\phi \propto T^2$ to $l^{-2}_\phi \propto T^3$ [44, 46]. The WAL effect can be originated due to two reasons, the first one is the existence of TSS in case of 2D and the second one is the high SOC in the bulk states of the system in case of 3D.

To confirm TSS to be the reason of observed 2D WAL effect, angle dependent magneto transport measurements have been carried out, in which angle between the applied field and direction of current is varied.

Figure 7(a) is showing anisotropic magnetoresistance of synthesized Sn$_4$Au single under a magnetic field of 12 T at 3 K. The transverse field direction is taken as the initial position and tilt angle is measured by taking the same as reference or 0$^\circ$ position. The geometry of current and field direction is shown in inset of figure 7(b). Resistivity at 12 T, is found to be periodic with the tilt angle, with regular occurring maxima and minima. Maxima of resistivity is obtained when the field is perpendicular to current (at 0$^\circ$ and 180$^\circ$ tilt angle) and minima is obtained when the field is parallel to current (at 90$^\circ$ and 270$^\circ$ tilt angle). It has been well explained in literature that the TSS dominated electric transport depends only on the perpendicular component of the field [47, 48]. This periodicity of MR, with maxima values at perpendicular field shows that the electrical transport is governed by TSS. Further, MR% has been calculated at different tilt angles viz. 0$^\circ$, 30$^\circ$, 45$^\circ$, 60$^\circ$, 75$^\circ$ and the same is shown in

It is important to note that the above explanation is based on the assumption that the observed effect is solely due to the presence of TSS. Other factors such as the presence of defects or impurities in the crystal could also contribute to the observed phenomenon, and further experiments are necessary to rule out these possibilities.
Figure 7. (a) Variation is resistivity with respect to the tilt angle under a field of 12 T and at 3 K. (b) Magnetoresistance vs $H$ plot of synthesized Sn$_4$Au single crystal at 3 K in a field range of ±12 T at various tilt angles, in which inset is showing the geometry used for the measurements. (c) Normalized MC vs $H$ plot of synthesized Sn$_4$Au single crystal at 3 K in a field range of ±12 T at various tilt angles. (d) Normalized MC vs $H \cos \theta$ plot of synthesized Sn$_4$Au single crystal at 3 K in a field range of ±12 T.

In figure 7(b), MR% is found to decrease as the tilt angle is increased. Also, the sharp V-type cusp at lower magnetic field starts to flatten with increment in tilt angle. This shows that WAL is suppressed with increased tilt angle. Angle dependent MC is supposed to be the most prominent source to determine whether the observed WAL effect is due to TSS or the strong SOC of bulk of material [49–51]. To ascertain the same, in figure 7(c), the normalized MC is plotted against the applied magnetic field at 3 K with various tilt angles viz. 0°, 30°, 45°, 60° and 75°. Further, the normalized MC is plotted against $H \cos \theta$, and the same is shown in figure 7(d). All normalized MC plots are found to merge in a single plot at low magnetic fields, this confirms that the observed WAL is 2D in nature and arises due to the presence of TSS in synthesized Sn$_4$Au single crystal [42, 49]. For a material, in which WAL effect arise due to strong bulk SOC, normalized MC vs $H \cos \theta$ plots are found to be well separated from each other [50, 51]. Our angle dependent magneto-transport measurements result support the findings of HLN model, showing that the electrical transport in synthesized Sn$_4$Au single crystal is governed by TSS at low temperature and low magnetic fields. More recently, in theoretical calculations, Sn$_4$Au is shown to have topological non-trivial bulk electronic band structure and the presence of topology is also verified by calculating $Z_2$ invariants [26].

4. Conclusion

In conclusion, single crystal of Sn$_4$Au is synthesized through a relatively easy flux free method following the solid-state reaction route and its quality is verified through host of characterization techniques viz. XRD, FESEM, TEM and XPS. Superconducting properties of Sn$_4$Au being studied through $\rho$-$T$ and $\rho$-$TH$ measurements, showed the same to be a type-I superconductor. Detailed $\rho$-$TH$ measurements being examined though detailed HLN analysis of MC resulted in occurrence of WAL effect due to TSS dominated transport phenomenon. This is further, verified through normalized MC vs $H \cos \theta$ at different tilt angle. This report shows that Sn$_4$Au exhibits superconducting properties below $T_c$ as well TSS dominated topological properties in its normal state. This unique feature of Sn$_4$Au establishes this material as a good choice to get more understanding in the field of topological superconductivity.
Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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