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Fast Track Communication

Effect of pressure on superconductivity in the indium-doped topological crystalline insulator SnTe

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Abstract

We report on the impact of hydrostatic pressure on the superconductivity of optimally (indium)-doped SnTe which is established to be derived from a topological crystalline insulating phase. Single crystals of $Sn_{1-x}In_xTe$ were synthesized by a modified Bridgman method that exhibited maximum superconducting T_c of 4.4 K for $x = 0.5$. Hydrostatic pressure up to 2.5 GPa was applied on the crystals of $Sn_{0.5}In_{0.5}Te$, and electrical resistivity as a function of temperature and pressure was measured. We observed a decrease in the onset superconducting transition temperature from 4.4 K to 2.8 K on increasing pressure from ambient to 2.5 GPa. The normal state resistivity also decreased abruptly by an order of magnitude at 0.5 GPa but for higher pressures, it decreased marginally. From onset, offset and zero resistivity values, dT_c/dP of ~ -0.6 K GPa^{-1} was confirmed. The low temperature normal state resistivity followed T^2 dependence suggesting Fermi liquid behaviour both for ambient and high pressure data. This increase in metallic characteristics accompanied by normal state Fermi liquid behaviour is in accordance with a 'dome structure' for T_c variation with varying carrier concentration.

Keywords: topological crystalline superconductor, superconductor phase diagram, high pressure measurements

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

Introduction

Superconductors derived from topological insulators (TIs) and topological crystalline insulators (TCIs) have attracted considerable attention recently [1–3]. From a theoretical perspective they could bring to fruition the search for the elusive Majorana fermions (MFs) and from the technological point of view they are expected to have a significant impact on topological quantum computation [4, 5]. An example is the indium (In)-doped many-valley semiconductor tin-telluride (SnTe) where a maximum superconducting transition temperature of ~4.4 K is reported for $x \sim 0.5$ in the series $Sn_{1-x}In_xTe$ [3, 6, 7]. Several theoretical studies and detailed angle-resolved photo-electron spectroscopic (ARPES) measurements have established SnTe to be a TCI phase due to the underlying mirror symmetry of its crystalline lattice [8, 9]. Towards developing an understanding of how these bulk superconductors are different from BCS or cuprate superconductors, in this communication we report the effect of hydrostatic pressure on the superconducting properties of optimally doped $Sn_{0.5}In_{05}Te$.

The basic premise of measurements under high pressure is that they can effectively tune the electronic and phononic band structure. This has led to the discovery of superconductivity

in myriad compounds at high pressure [10–12]. With regard to optimally doped superconductors, such studies could reflect if there is a 'dome structure' associated with phase transitions *vis á vis* carrier concentration that relates to quantum criticality and correlation effects [13]. Further, if pressure could revert to the insulating bulk phase, then in principle one can have superconductor–topological insulator interfaces leading to the emergence of MFs. Moreover, a dome structure of increasing T_c dependence with varying concentration of indium up to 50% was indicated earlier in SnTe [6], but the compounds were reportedly multiphase beyond $x = 0.5$ and therefore high pressure studies are essential to elucidate the full phase diagram of $Sn_{1-x}In_xTe$.

In particular, topological surface states have been investigated in Bi_2Se_3 and Bi_2Te_3 which have been driven to a superconducting state with application of external pressure [10–12]. In undoped topological insulators, superconductivity is achieved at relatively high pressure. For example, Bi_2Se_3 shows a turnover from semiconducting to metallic behaviour at ~8 GPa accompanied by a structural phase transition [14]. Superconductivity appears in Bi_2Se_3 at ~13.5 GPa at a transition temperature of 0.5 K, which gradually increases to a maximum of 7 K on increasing pressure up to 30 GPa. At higher pressures, the T_c remains almost constant up to 50 Gpa [10]. Bi_2Te_3 is another topological insulator, for which superconductivity is reported around 3 K under 3 GPa pressure that increases to 8 K at 15 GPa and for further higher pressures the T_c exhibits a decreasing trend [12]. Like Bi_2Se_3, Bi_2Te_3 also undergoes several structural transformations from a rhombohedral (R-3m) phase to monoclinic (C2/m) at 3 GPa, to monoclinic (C2/c) at 8 GPa and finally to a bcc Im-3m structure above 16 GPa [12].

Superconductors derived from topological insulators through intercalation have also been subjected to various pressure studies [15]. $Cu_xBi_2Se_3$ is a well-known topological insulator-based superconductor with a maximum T_c of around 3.8 K [15]. Point contact spectra on the surface of Cu intercalated Bi_2Se_3 exhibit signs of unconventional superconductivity [16]. On applying pressure on $Cu_xBi_2Se_3$ a gradual decrease in the superconducting transition temperature is reported and as pressure is increased further superconductivity disappears at ~6.3 GPa [17]. With regard to TCI systems, a first-principles study on SnTe predicts a maximum superconducting $T_c = 7.16$ K by pressure tuning the electron–phonon coupling parameters in the bcc phase (Pm-3m) [18]. Further, very recently Wang *et al* have shown unconventional superconductivity in 3D Dirac semi-metal Cd_3As_2 which belongs to a new class of topological superconductors [19]. In this communication, we focus on the impact of pressure on superconducting and normal state conduction of the recently discovered optimally doped TCI superconductor $Sn_{0.5}In_{0.5}Te$. We found that the superconducting transition temperature (T_c) decreased monotonically with pressure (~0.6 K GPa^{-1}) and the normal state resistivity also decreased by an order of magnitude at 0.5 GPa. Such behaviour shows a surprising resemblance with the curious case of over-doped cuprates.
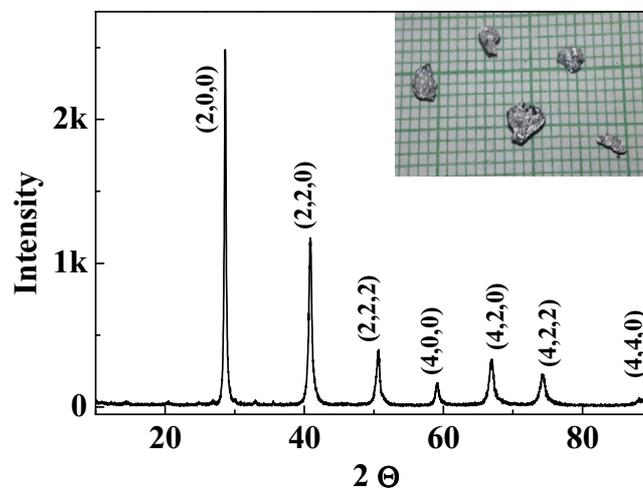
**Figure 1.** Powder XRD pattern of crystals of $Sn_{0.5}In_{0.5}Te$. The inset shows crystal flakes.

Experimental methods

Single crystals of $Sn_{0.5}In_{0.5}Te$ were prepared by a modified Bridgman method. A series of compounds with varying indium concentration were prepared and optimum superconducting T_c was achieved for $Sn_{0.5}In_{0.5}Te$ [7]. We studied the electrical resistivity at high pressure on this composition. Single crystals were obtained by melting stoichiometric amounts of high purity elemental powder of Sn (99.99%), Te (99.999%) and shots of In(99.99%) at 900 °C for 5 d in sealed evacuated quartz tubes. Intermittent shaking was performed for the homogeneity of the melt sample. The sample was cooled to 770 °C over a period of 72 h followed by annealing at 770 °C for 48 h. Silvery–shiny single crystals were cleaved along the z axis. X-ray diffraction was carried out on the powdered samples by a *RIGAKU* powder x-ray Diffractometer (Miniflex 600) [7].

Pressure-dependent resistivity measurements were performed in the Physical Property Measurements System (*PPMS*-14T, *Quantum Design*) using an HPC-33 Piston type pressure cell with Quantum Design DC resistivity option. Hydrostatic pressures were generated by a BeCu/NiCrAl clamped piston-cylinder cell. The sample was immersed in a fluid (Daphne Oil) with pressure transmitting medium of Fluorinert in a Teflon cell. Annealed Pt wires were affixed to gold-sputtered contact surfaces on each sample with silver epoxy in a four-probe configuration.
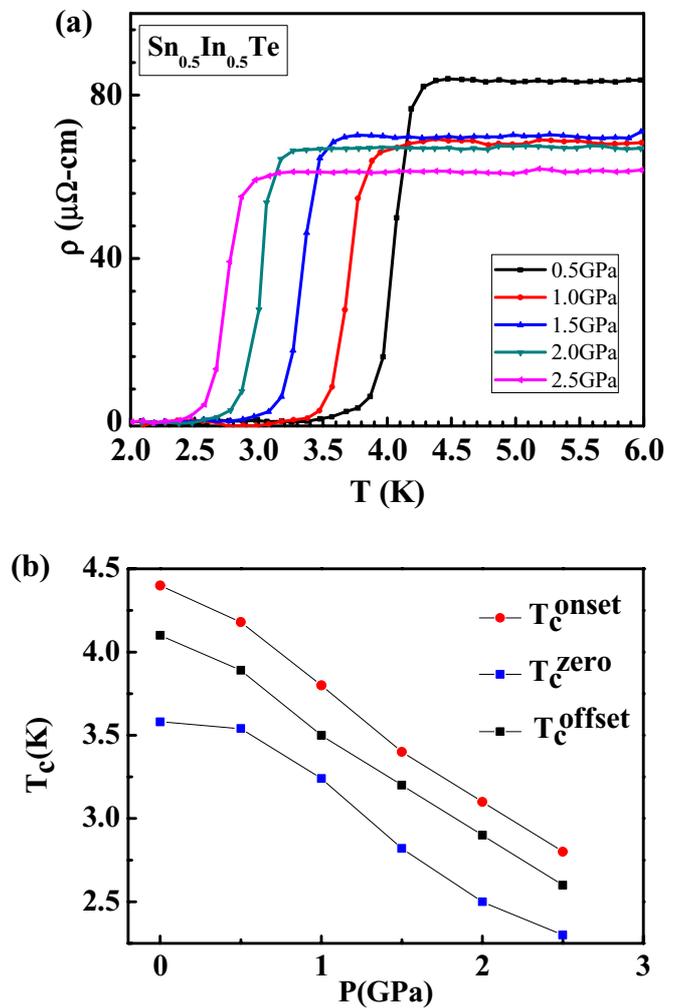
Results and discussion

The powder XRD pattern of $Sn_{0.5}In_{0.5}Te$ is shown in figure 1. It confirms pure phase synthesis in agreement with reference data from JCPDF (No. 089-3974). The specimen crystallizes in a rock-salt structure with space group Fm-3m. The calculated lattice parameter is **a** = 6.265 Å and the cell volume is 245.65 Å^3. We note that previous data have reported continuance of the rock salt structure of SnTe up to ($x = 0.5$) [6, 7]. Further band structure calculations indicate that structural changes are expected only above 5 GPa which is
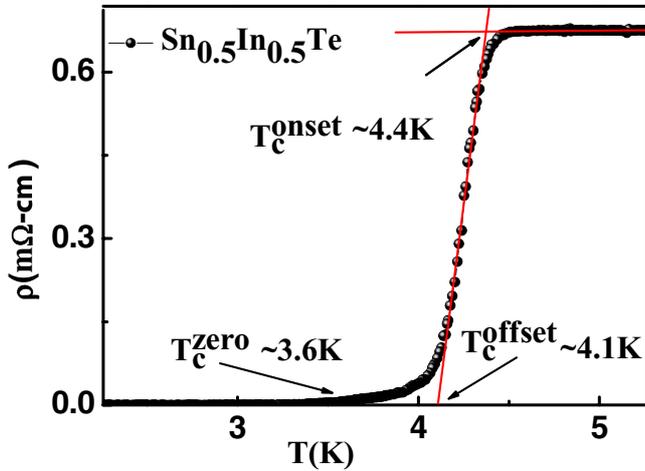
Figure 2. Resistive superconducting transition of $Sn_{0.5}In_{0.5}Te$ at ambient pressure. T_c^{onset}, T_c^{offset} and T_c^{zero} are indicated by arrows.

Figure 3. (a) Resistive superconducting transition of $Sn_{0.5}In_{0.5}Te$ at different pressures from 0.5 GPa to 2.5 GPa. An unambiguous decrease in superconducting transition temperature is seen. (b) Variation in superconducting T_c at various pressures for $Sn_{0.5}In_{0.5}Te$. We can see a negative pressure coefficient (dT_c/dp) for all transitions T_c^{onset}, T_c^{offset} and T_c^{zero}.

considerably above our experimental range [18]. From these accounts we infer that $x = 0.5$ does not go into a new phase due either to pressure (less that 2.5 GPa) or dopant concentration. In the inset, as-grown crystal flakes are shown. The electrical resistivity as a function of temperature $(\rho - T)$ for $Sn_{0.5}In_{0.5}Te$ at ambient pressure is shown in figure 2. The inset shows resistivity up to room temperature. We mark T_c^{onset} by the intersection of the two extrapolated lines, one corresponding to the superconducting transition line and the other being an extended normal state resistivity line. Similarly, T_c^{offset} is indicated by the intersection of the transition line and a zero resistivity line. We define T_c^{zero} as the temperature where a zero resistivity state was achieved. This is schematically shown in figure 2. From figure 2, The values of T_c^{onset}, T_c^{offset} and T_c^{zero} for single-crystal $Sn_{0.5}In_{0.5}Te$ are found to be 4.4 K, 4.1 K and 3.6 K respectively. The superconducting transition is sharp with a transition width of ~0.3 K.

The resistivity versus temperature behaviour near the superconducting transition (T_c) for varying pressure is shown in figure 3(a). For clarity, the data for ambient pressure are not included as they are an order of magnitude higher. It is seen that both the superconducting transition temperature and normal state resistivity decrease with increasing pressure. It can be seen that at a maximum pressure of 2.5 GPa, the T_c^{onset} decreases to 2.8 K from 4.4 K (ambient pressure), while the T_c^{offset} and T_c^{zero} decrease to 2.6 K and 2.3 K from 4.1 K and 3.6 K respectively. The T_c^{onset}, T_c^{offset} and T_c^{zero} for $Sn_{0.5}In_{0.5}Te$ superconductor at intermediate pressure are summarized in figure 3(b). It appears that the decrease in the T_c is approximately linear for two markers (onset, offset) of the superconducting transition. The negative coefficients of T_c suppression with pressure for three markers (T_c^{onset}, T_c^{offset} and T_c^{zero}) are estimated to be $-0.66\,K\,GPa^{-1}$, $-0.61\,K\,GPa^{-1}$ and $-0.57\,K\,GPa^{-1}$ respectively, and yield an average dT_c/dP of $-0.6\,K\,GPa^{-1}$ for the $Sn_{0.5}In_{0.5}Te$ superconductor. In fact, in conventional superconductors, it is very common to see the decreasing T_c trend as a function of increasing pressure. Sn, In and Pb all have negative dT_c/dP of $\sim0.4\,K\,GPa^{-1}$ [20]. The principal reason for this decrease in simple metals is stiffening of the lattice and the consequent decrease in electron–phonon
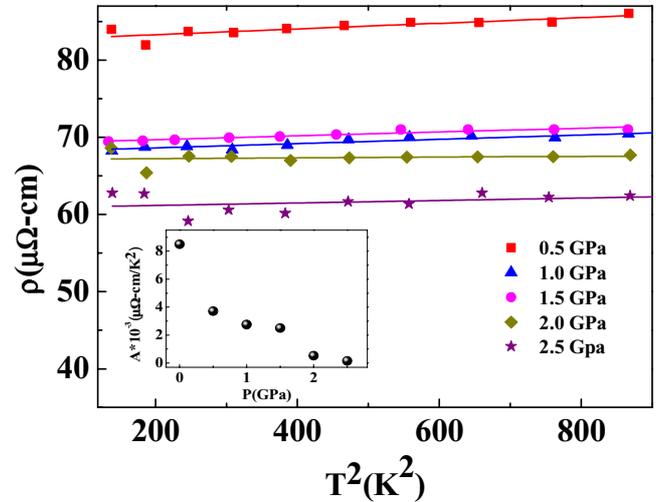
coupling rather than electronic effects. On the other hand, for high T_c cuprates generally $T_c(P)$ first increases with pressure and beyond a critical pressure it starts to decrease, exhibiting a dome structure in accordance with the T_c dependence on carrier concentration [21].

In figure 4(a) we compare resistivity versus temperature $(\rho - T)$ for $Sn_{0.5}In_{0.5}Te$ taken at ambient pressure and applied pressures in the extended temperature range up to 250 K. This is done to visualize the impact of hydrostatic pressure on the normal state conduction of $Sn_{0.5}In_{0.5}Te$. We see that at applied pressure 0.5 GPa the normal resistivity (resistivity just above the transition) decreases abruptly by nearly an order of magnitude. Quantitatively, a decrease of about 7.8 times $(659.76\,\mu\Omega\,cm$–$84.09\,\mu\Omega\,cm)$ is observed. With further increase in pressure, while the normal state resistivity continues to decrease, the rate of change with pressure decreases substantially. Overall, the change in resistivity with temperature shows a metallic behaviour at both ambient and applied pressures up to 2.5 GPa and clearly the
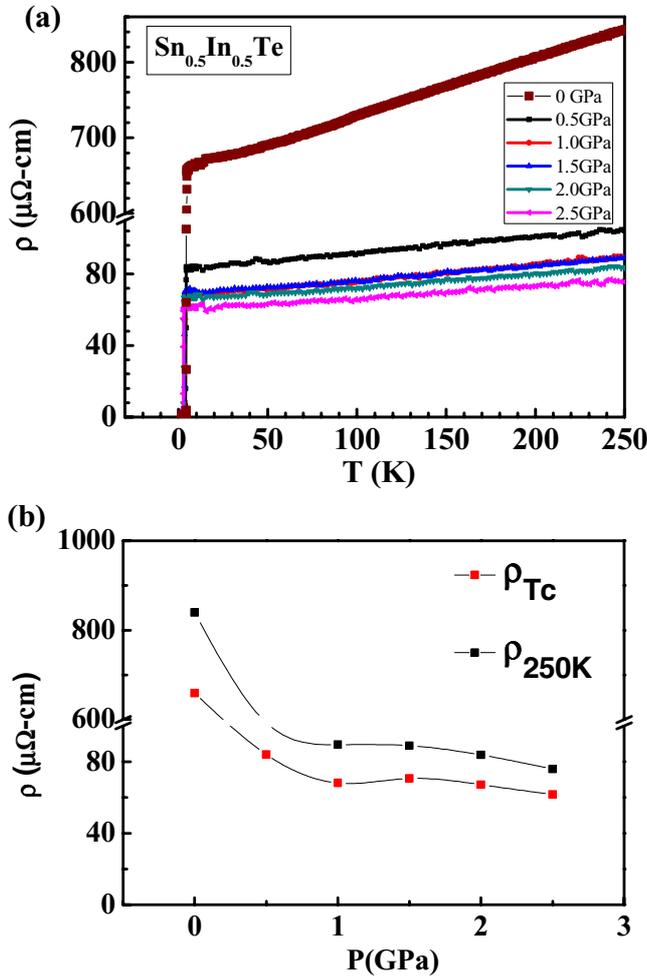
Figure 4. (a) Resistivity up to 250 K for $Sn_{0.5}In_{0.5}Te$. A large decrease in resistivity is seen on application of 0.5 GPa. For higher pressure, resistivity decreases at a smaller rate. No significant variation in residual resistivity ratio is observed. (b) Magnitude of electrical resistivity at 250 K and at temperatures close to T_c plotted as a function of pressure. A sharp decrease in magnitude is observed at 0.5 GPa followed by an almost linear decrease in the resistivity.

Figure 5. Resistivity versus T^2 for pressure 0, 0.5, 1, 1.5, 2, 2.5 GPa. The solid lines are linear fits to the equation $\rho = \rho_0 + AT^2$.

metallic behaviour increases with higher pressure. This is in contrast to $Cu_xBi_2Se_3$ where $\rho(T_c)$ increases by 7 times at 2.31 GPa. For low carrier density superconductors the BCS equation relates superconducting transition temperature with carrier concentration; $T_c \sim \theta_D \exp(-1/N(E_F)V_0)$ where θ_D is the Debye temperature, V_0 is the electron–phonon coupling calliper and density of state $N(E_F) \sim m^*n^{1/3}$ which is a product of effective mass m^* and carrier concentration n. A decreasing carrier concentration (increasing normal state resistivity) with pressure can explain decreasing superconducting T_c. But we find that normal state resistivity for $Sn_{0.5}In_{0.5}Te$ decreases many-fold with pressure. Thus the controlling parameter for the T_c suppression mechanism seems to be decreasing effective mass m^* in $Sn_{0.5}In_{0.5}Te$ under a simplistic s-wave correlation [7]. In figure 4(b) we show the change in the resistivity just above the transition temperature and at 250 K for ambient and various applied pressures. We can see that both decrease very rapidly on application of 0.5 GPa but for higher pressure, the change

is relatively much smaller. The RRR (residual resistivity ratio) between resistivity at 250 K temperature and temperature just above the transition remains ~1.25 from ambient pressure to 2.5 GPa. This indicates that within experimental error, the impurity band contribution to the normal state conduction mechanism in $Sn_{0.5}In_{0.5}Te$ remains mainly unaffected by applied pressure.

We note that both in high T_c cuprates and in pnictide superconductors, the transition temperature varies in a so-called dome structure as a function of doping concentration. In the overdoped region, the normal state resistivity is well characterized by Fermi liquid behaviour. To study the appropriateness of Fermi liquid theory (negligible electronic correlations), in figure 5 we plot the resistivity versus T^2 curves in the temperature range 11–30 K for $Sn_{0.5}In_{0.5}Te$. In this theory, particularly with regard to heavy fermion systems, the strong interaction between charge carriers is replaced by weakly correlated quasi-particles with high effective mass. In figure 5, the resistivity data taken at various pressure are fitted to the equation $\rho = \rho_0 + AT^2$ where ρ_0 relates to impurity scattering and the coefficient A relates to square of the effective mass of the quasi-particles. The calculated values for ρ_0 and A are listed in table 1. We find that for the pressure 0, 0.5, 1, 1.5, 2, 2.5 GPa the data follow a T^2 behaviour in the temperature range 11–30 K and the curves deviate from linearity above 30 K. In the inset of figure 5 we plot the pressure dependence of coefficient A which indicates a decreasing trend with increasing pressure. This is suggestive of a weakening correlation between charge carriers in the overdoped region. But the origin of this correlation phenomenon in the semiconducting parent SnTe needs to be ascertained.

Conclusion

We have prepared single crystals of $Sn_{0.5}In_{0.5}Te$ and applied pressures up to 2.5 GPa to check their superconducting properties under pressure. This is an optimally doped

Table 1. The values for ρ_0 and A at various pressures for the equation $\rho = \rho_0 + AT^2$.

Pressure (GPa)	0	0.5	1	1.5	2	2.5
$\rho_0(\mu\Omega\,\text{cm})$	669.96	82.5	68.07	69.1	67.2	60.8
$A(\mu\Omega\,\text{cm}\,\text{K}^{-2})$	8.49×10^{-3}	3.71×10^{-3}	2.75×10^{-3}	2.49×10^{-3}	5.25×10^{-4}	1.58×10^{-4}

specimen derived from a topological crystalline insulating phase. We found that superconducting T_c (onset) decreased with pressure from 4.4 K (ambient) to 3.8 K (2.5 GPa). This suppression of superconducting transition temperatures was found to be almost monotonic with pressure and the overall dT_c/dP was estimated to be $-0.6\,\text{K}\,\text{GPa}^{-1}$. Fermi liquid behaviour was indicated in the temperature range 11–30 K and we found that the normal state resistivity of the sample varied as a function of T^2 with increasing pressure. Such a systematic decrease with T_c with increasing metallicity and normal state T^2 behaviour is reminiscent of overdoped high T_c cuprates.

Acknowledgments

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