Quantum magnetotransport oscillations in the pure and robust 3D topological Dirac semimetal $\alpha$-Sn

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We report experimental evidence for the existence of topologically protected charge carriers in the pure and robust 3D topological Dirac semimetal $\alpha$-Sn(100) grown on InSb(100) substrate by employing a simple macroscopic four-point resistance measurement. We analyzed and compared electrical characteristics of the constituting components of the sample, and proposed a k-band drift velocity model accordingly. In consequence, a topological band, with low carrier density and high mobility, was identified as the origin of the observed Shubnikov de Haas oscillations. The analysis of these quantum oscillations resulted in a non-trivial value of the phase shift $\gamma = 0$, which is characteristic for massless Dirac fermions. This behavior was detected in the grown $\alpha$-Sn(100) films for both in plane and out of plane field orientation, suggesting 3D Topological Dirac semimetal behaviour of this material.

Topological Dirac semimetals (TDS) were theoretically predicted [1–5] as a class of quantum materials with a suitable crystalline symmetries and consequent bulk Dirac cones emerging. Such bulk topological bands embody 3D massless Dirac fermions, meaning that TDSs could be understood through a ‘3D graphene’ paradigm [6], opening the possibility to use properties of graphene and topological insulator (TI) in 3D. Due to 3D topological electronic carriers, TDSs exhibit a linear field dependence of the bulk quantum magnetoresistance [7–10]. Moreover this exotic topological state brings along effects of giant diamagnetism [5, 11, 12] and oscillating quantum spin Hall effect [13].

The 3D Dirac fermion in TDSs is composed of two massless Weyl fermions of opposite chirality [14, 15] occupying so called Weyl points which if separated in the momentum space (by braking time-reversal or inversion symmetry) would result in a topological Weyl semimetal (WSM). WSM is a topological quantum state of matter that exhibits a well-defined Fermi-arcs geometry [11, 16], which leads to the manifestation of an exotic property of quantized anomalous Hall effect [16].

In order to physically fabricate a 3D TDS one could, in principle, realize a fragile quantum critical point (TDS state) between normal insulator and TS by tuning chemical composition or spin-orbital coupling. On the other hand, much more robust TDS state can be realized in some stoichiometric compounds with suitable crystal symmetries which preserve time reversal symmetry and protect 3D Dirac points. Signatures of a TDSs were indeed experimentally reported to exist in Na3Bi [6, 17] and Cd3As2 [18, 20] by angle-resolved photoemission spectroscopy (ARPES).

Alpha phase of Sn is a diamond structured crystal which, if not subjected to strain, is a zero band gap semiconductor, with an evident covalent nature. Due to a suitable band structure $\alpha$-Sn was marked as a material capable to acquire several topological phases [21–23]. Interestingly an in-plane strain (band-gap opening), brings about the topologically protected states in $\alpha$-Sn, more specifically: in-plane tensile strain turns it into a 3D TI, while in-plane compressive strain converts it into a 3D TDS. The topological character of $\alpha$-Sn crystal thin films was fully established by experiments during the last decade. This was particularly done using angle-resolved photoemission spectroscopy (ARPES) which revealed a Dirac cone near the Fermi level ($E_F$) in the surface electronic structure of Te and/or Bi doped $\alpha$-Sn thin films on InSb substrate [21, 24, 29]. More importantly, the most recent soft X-ray ARPES investigations have shown a clear TDS characteristic in strained $\alpha$-Sn films [21, 26, 29]. Nevertheless, considering the scientific attention given to Na3Bi and Cd3As2, currently seen as prototypical TDSs, the number of $\alpha$-Sn 3D TDS studies is considerably limited. Further more, very well structurally defined (cubic), non-toxic elemental $\alpha$-Sn could be more favorable for applications than the TDS binary compounds whose crystal structure is often a matter of debate [30].

Strain has proven to be a useful tool for engineering topological phases in $\alpha$-Sn, and having that well defined Weyl points were predicted in this material [23], pursuing further TDS investigation in this elemental quantum material seems natural. Encouragingly, there are very recent reports about topological magnetotransport in $\alpha$-Sn. In the case of $\alpha$-Sn on InSb there is, until now, one report [28] on detected topologically protected carriers in magnetotransport in a relatively complicated sample configuration (Bi-doped $\alpha$-Sn/InSb/GaAs) and also one report for $\alpha$-Sn grown on CdTe [31].

Very recently we presented [22] a more straightforward method to grow a pure (no dopants) strained $\alpha$-Sn(100) 3D TDS films (with an accent on the importance of the InSb(100) substrate preparation). These films demonstrated an enhanced surface quality and a notable robustness against ambient conditions. Materials of such quality could become favorable for spintronics devices ap-
plications which are, in general, very sensitive to the noise that originates from crystal imperfections.

In this letter we present the evidence for the existence of topologically protected charge carriers in 30 nm thick α-Sn(100) films, grown using the above mentioned recently reported method [32], by analysing Shubnikov-de Haas (SdH) quantum oscillations extracted from the magnetoresistance measurements. In order to fully understand this finding, we analyze and compare electrical characteristics (sheet resistivity, carrier type, carrier concentration and mobility) of the grown 3D TDS α-Sn with the one of the InSb(100) substrate alone, and propose a k-band drift velocity model accordingly.

In the case of the electronic transport properties measurement of the composite systems (film + substrate) it is generally needed to investigate the transport properties of some of the constituting components separately. Therefore, along with the transport measurements on the α-Sn(100)/InSb(100) system we conducted the same measurements on InSb(100) alone (prepared using the same procedure [32] as in the case of grown α-Sn(100) films). Experiments were conducted at the temperature of 1.5 K, with contacts in the four-point geometry using the Van der Pauw technique (VdP) [33], at constant current (50–100 µA), up to magnetic field (B) of 8 T. Figure 1 depicts the conducted Hall effect measurements showing a clear qualitative difference between the off-diagonal resistance ($R_{xy}$) of the InSb(100) substrate and the deposited α-Sn film. While the substrate behaves linearly (n-type), the whole sample clearly displays a non-linear Hall effect indicating the presence of more than one band contributing to its conduction. At low fields ($<2$ T), the substrate is shunting the α-Sn film and the behaviour of $R_{xy}$ is somewhat non-linear. From the results of our extensive transport properties study of the substrate there are some indications that a prepared InSb(100), using Ar$^+$ plasma etching and thermal annealing, is electronically heterogenous. However, it is unclear what is exactly causing the low-field non-linear behaviour. Since it can be attributed to the substrate and its n-type behaviour is dominant, we omit this low field region from further analysis.

A useful method to identify different contributions leading to the observed Hall effect is to fit a k-band drift velocity model to the measured $R_{xy}$ behavior. $R_{xy}$ can, in general, be expressed using the following equations:

$$R_{xy} = \frac{\sigma_{xy}}{\sigma_{xx}^2 + \sigma_{xx}^{-2}},$$  \hspace{1cm} (1)

with the appropriate conductance values ($\sigma$):

$$\sigma = \sum_{i=1}^{k} \sigma^i,$$  \hspace{1cm} (2)

$$\sigma_{xx}^i = \frac{n_{is}^i \cdot e \cdot \mu^i}{1 + (\mu^i \cdot B)^2},$$  \hspace{1cm} (3)

$$\sigma_{xy}^i = \pm \frac{n_{is}^i \cdot e \cdot (\mu^i)^2 \cdot B}{1 + (\mu^i \cdot B)^2},$$  \hspace{1cm} (4)

under the constraint of the known sheet resistance at zero field:

$$R_s = \left( \sum_{i=1}^{k} n_{is}^i \cdot e \cdot \mu^i \right)^{-1}.$$  \hspace{1cm} (5)

Fitting the measured $R_{xy}$ behavior of the deposited α-Sn film to a two-band ($k = 2$) drift velocity model results in one p-type band ($i = p$) and one n-type band ($i = n$) with the sheet carrier concentration values of $n_{ps} = 2.1 \cdot 10^{13}$ cm$^{-2}$ and $n_{ns} = 1.7 \cdot 10^{13}$ cm$^{-2}$,
and the mobility values of $\mu^p = 1470 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$ and $\mu^n = 14.4 \cdot 10^4 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$. In this case the reduced chi-square value ($\chi^2$) for the fit equals 2.45. On the other hand, repeating the procedure for a three-band ($k = 3$) drift velocity model (Fig. 2) results in two $p$-type bands ($i = p_1, p_2$) and one $n$-type band ($i = n$) giving values of: $n_{p_1} = 3.3 \cdot 10^{13} \text{cm}^{-2}$, $\mu_{p_1} = 168 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$, $n_{p_2} = 1.5 \cdot 10^{12} \text{cm}^{-2}$, $\mu_{p_2} = 3470 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$, $n_n = 1.6 \cdot 10^{13} \text{cm}^{-2}$, $\mu_n = 14.3 \cdot 10^4 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$ and $\chi^2 = 1.47$. Considering these values, it is tempting to choose the latter model over the first one. Indeed, the bands with the large number of carriers ($n$ and $p_1$) are typical bulk bands expected for InSb and $\alpha$-Sn (considering the film/substrate thickness [32]), while the high mobility and low carrier density band ($p_2$) could be attributed to the topological carriers [34, 35]. Although the second model fits the data better, that does not prove its correctness so far.

To exclude one of the two models, we extracted the SdH oscillations from the magnetoresistance measurement of the deposited $\alpha$-Sn(100) film (Fig. 3). The experiments were conducted with $B$ applied perpendicular to the surface of the film (up to 8 T), at the temperature of 1.5 K, with macroscopic contacts in the linear four-point geometry. Figure 4 (inlay) depicts resistance oscillations ($\Delta R$) of the grown sample extracted by removing the background using the subtraction of a low order polynomial fit. The maximum of the fast Fourier transform (FFT) of $\Delta R$ gives an estimate on the characteristic frequency $f = 14 \pm 6$ T.

Assuming the oscillations come from a trivial bulk band, the estimated value for $f$ would correspond to a carrier density of $n = (2\pi f/\Phi_0)^{3/2}/3\pi^2 = (3 \pm 2) \cdot 10^{21} \text{m}^{-3}$, where $\Phi_0$ is the elementary flux quantum. If we convert this to the hypothetical sheet carrier densities for such bulk bands of the $\alpha$-Sn(100) film and InSb substrate, by multiplying with their thicknesses [32], this would give $(9 \pm 6) \cdot 10^{11} \text{cm}^{-2}$ and $(2 \pm 1) \cdot 10^{16} \text{cm}^{-2}$ respectively. From these values, which are orders of magnitude off from the measured values, it becomes clear that neither of the two models would be compatible with the observed SdH oscillations if assumed that they originate from a bulk band.

Instead, if we assume that the experimentally observed quantum oscillations arise from massless Dirac fermions, as it is expected for the topologically protected surface states, the sheet carrier density corresponding to the observed SdH frequency is given by $n_s = 2 \cdot n = 2 \cdot f/\Phi_0 = (1.4 \pm 0.6) \cdot 10^{12} \text{cm}^{-2}$. This carrier density value is in a very good correspondence with the second $p$ band ($i = p_2$) in the above applied three-band drift velocity model, but in contradiction with the two-band one. Note as well that the SdH oscillations are only observable if full orbits are possible in real space [36], i.e. when their cyclotron frequency is faster than their relaxation time. In our case this is equivalent with $\mu > 1/B \approx 3390 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$ for oscillations starting at $<3$ T. This condition as well is perfectly satisfied for the second $p$ band ($i = p_2$) in the three-band drift velocity model, but again in contradiction with the two-band one. Therefore, it can be concluded that the three-band drift velocity model is the one explaining the observations, while the two-band drift velocity model is insufficient.

The second $p$ band of the three-band model ($i = p_2$) is thus identified as a possible topological band of the $\alpha$-Sn(100) film. The first $p$ band ($i = p$) we identify as the bulk $\alpha$-Sn and the $n$ band as the bulk of the InSb substrate.

Consequently, using Eq. 5 we can now estimate the sheet resistance contribution of the bands separately: $R^p_{\text{res}} = 1.20 \text{k}^\circ/\square$ (possible topological band), $R^n_{\text{res}} = 1.13 \text{k}^\circ/\square$ ($\alpha$-Sn bulk band) and $R^n_{\text{res}} = 2.7 \Omega/\square$ (substrate band).

So far we did not supply direct proof of the topological
nature of the carriers in the second p band \((i = p2)\). However, the SdH oscillations contain the information on the topology of the corresponding carriers within their phase offset \(\gamma\) \cite{37}. The SdH oscillations can be expressed as:

\[
\Delta R \propto \cos \left(2\pi \left(\frac{f}{B} - \gamma\right)\right),
\]

where \(\gamma\) can be related to Berry’s phase acquired by the carriers in a cyclotron orbit \cite{38}. When the zero-field electronic dispersion is followed, then \(\gamma = 0\) (non-trivial Berry’s phase of \(\pi\)) for the Dirac fermions, while \(\gamma = 1/2\) (trivial Berry’s phase of 0) for ‘normal’ fermions \cite{9}. The intercept of a linear fit of the Landau level position \(N\) with respect to \(1/B\) then gives the \(\gamma\) value. However, for finite fields, as proposed by Wright and McKenzie, the fermions can no longer be expected to follow the zero-field dispersion, and a deviation of the perfect quantization of \(\gamma = \gamma(B)\) is to be expected \cite{9}. For that reason, a more accurate low-field fit procedure was proposed to extract the zero-field phase offset:

\[
N = \frac{f}{B} + A_1 + A_2 \cdot B,
\]

where \(f\) (characteristic frequency), \(A_1\) and \(A_2\) are the fit-parameters. The zero-field limit of the equation is equivalent with \(\gamma(0) = A_1\), therefore the parameter \(A_1\) is the quantized offset related to the zero-field Berry’s phase. Figure 4 displays the fit of the Eq. (7) to the observed SdH oscillations. Here, due to the dominance of bulk contributions to the conductance \cite{10}, minima of the SdH oscillations correspond to an integer Landau level index \((N)\), while maxima corresponds to a half-integer Landau level index \((N/2)\). The found fit-parameters are \(A_1 = 0.02 \pm 0.15\) and \(A_2 = 0.00 \pm 0.06\) and \(f = 13.3 \pm 1.7\) T. The characteristic frequency was determined by an initial linear fit and held fixed as the linear fit yields more accurate results on the frequency compared to the estimation of the FFT maximum of \(\Delta R\) which is usually used to reduce the number of fit parameters. As the analysis gives \(A_1 \approx 0\), it can be thus concluded that indeed the second p-type band \((i = p2)\) has a Dirac signature as it is expected for the topologically protected surface states in \(\alpha\)-Sn.

Having this conclusion, we can now estimate the values of the characteristic Fermi wavevector \cite{35,37}

\[
k_F = \sqrt{2\pi f/\Phi_0} = 0.21 \pm 0.04 \text{ nm}^{-1}
\]

and Fermi energy \(E_F = \hbar k_F v_F = 81 \pm 17\) meV. These values, estimated from transport measurements, are in close agreement with the ones estimated from our recent ARPES \((k_F^{\text{ARPES}} \sim 0.18 \text{ nm}^{-1}, E_F^{\text{ARPES}} \sim 60\) meV) and scanning tunneling spectroscopy study \((E_F^{\text{STS}} \sim 70\) meV) \cite{32}. Furthermore, from the calculated \(R_s^{2}\) value, using the resistance expression for nondegenerate 2D bands with a linear dispersion \cite{37,39}:

\[
R_s^{2} = \frac{4\hbar^2 \pi}{c^2 E_F^2} \frac{1}{\tau}
\]

we estimate the momentum relaxation time \(\tau \approx 350\) fs. This value is around six time larger than the one estimated for \(\alpha\)-Sn films with AlO\(_x\) capping layer \cite{28}, and almost two orders of magnitude larger compared with \(\alpha\)-Sn films covered with Ag \cite{40}, which indicates that there are fewer parallel momentum relaxation channels in our samples.

It is important to be noted that SdH oscillations were observed for \(\vec{B}\) applied both perpendicular and parallel (perpendicular to (010)) to the surface of the film, which could not be attributed to InSb or \(\alpha\)-Sn bulk bands. In the case where the topological phase resides only on the surface, one expects the oscillations to vanish for parallel field alignment. Moreover, phase offset analysis of these oscillations also gave a value close to the one of non-trivial Berry’s phase \((A_1 = -0.11)\). The deviation from zero value of the \(A_1\) parameter matches the dimensionality phase shift for 3D corrugated Fermi surface \(\delta = \pm 1/8\) in the analogous Lifshitz-Kosevich theory \cite{22}. Since in the case of \(\alpha\)-Sn(100) topologically protected bands reside also in the bulk \cite{21,23,29}, these results could be interpreted as a magnetotransport signature of a quasi 3D Dirac charge carriers. One should be aware that the phase offset of \(\pi\) in the SdH oscillation can not always be unquestionably seen as a direct proof of a 3D topological material \cite{41}. However, in this particular case, having all the previous spectroscopic reports in mind and the same SdH oscillations non-trivial behaviour for \(B\) applied in-plane and out of plane (opposite to the trivial behaviour of InSb alone), it is clear that the value of the phase offset strongly supports the 3D TDS picture of compressively strained \(\alpha\)-Sn on InSb substrate.

We can conclude that we found an evidence for the existence of 3D topological states in \(\alpha\)-Sn(100) films using a macroscopic transport measurement technique. A p-type band with low carrier density and high mobility was found and identified to be the origin of the observed SdH oscillations. To analyze these quantum oscillations a Landau index plots fitting was used, which gave the non-trivial value of the phase shift, characteristic for 3D massless Dirac fermions (Berry’s phase of \(\pi\)) for magnetic field applied both perpendicular and parallel to the (100) surface. This is an additional and independent evidence of the 3D TDS character of the pure and robust \(\alpha\)-Sn(100) films grown recently \cite{32}. Our findings suggest prominent accessibility of the topological properties in \(\alpha\)-Sn(100) grown on InSb(100) even without use of the capping layer. A remarkable fact that it is possible to detect topological carriers in our samples using a simple macroscopic four-point resistance measurement opens a great deal of opportunity to further investigate and apply this elemental quantum material.

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