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### High Fermi level spin polarisation in the $(Bi_{1-x}Sb_x)_2Te_3$ family of topological insulators - A Point Contact Andreev Reflection study

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Point Contact Andreev Reflection spectroscopy is employed to extract the effective Fermi level spin polarisation of three distinct compositions from the  $(Bi_{1-x}Sb_x)_2Te_3$  topological insulator family. The end members,  $Bi_2Te_3$  and  $Sb_2Te_3$ , exhibit high polarisation of 70(4) % and 57(3) %, respectively. High-field ( $\mu_0 H = 14 \text{ T}$ ) point-contact spectroscopy shows the carrier depletion close to the Fermi level for these two compositions with small bandgaps of 0.40(4) meVand 0.28(2) meV, respectively. The almost fully suppressed bulk conductivity in the  $(Bi_{0.18}Sb_{0.82})_2Te_3$  results in even higher spin polarisation of 83(9) %. Further, it is demonstrated that magnetic doping with Cr and V tends to reduce the spin polarisation values with respect to the ones of the pure compositions.  $Bi_{1.97}Cr_{0.03}Te_3$ ,  $Sb_{1.975}Cr_{0.025}Te_3$ ,  $Bi_{1.975}V_{0.025}Te_3$ , and  $Sb_{1.97}V_{0.03}Te_3$  exhibit spin polarisation of 52 %, 52 %, 58 %, and 50 %, respectively. In view of the rather high effective polarisation, non-magnetic topological insulators close to ( $Bi_{0.18}Sb_{0.82}$ )<sub>2</sub>Te<sub>3</sub> may provide a path towards the characterization of pair-breaking mechanisms in spin triplet superconductors.

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Keywords: Topological Insulators, Andreev Reflection, Point Contact Spectroscopy

#### 1

#### I. INTRODUCTION

36 The generation and detection of highly spin-polarised 37 2 currents is of crucial importance to the field of spin 38 3 electronics. Spin generators with high spin polarisation <sup>39</sup> 4 and low magnetisation are highly sought after. High 40 5 spin polarisation improves the tunnelling magnetoresis- 41 6 tance (TMR) effect in magnetic tunnel junctions (MTJs), 42 7 whereas decreased magnetisation generally lowers the 43 8 switching or precession currents in spin-transfer torque 44 9 based devices. Topological insulators<sup>1</sup>, spin-polarised 45 10 antiferromagnets<sup>2</sup> and compensated ferrimagnets<sup>3</sup> are 46 11 some classes of materials that can satisfy these seem- 47 12 ingly contradicting requirements. Indeed, magneti- 48 13 zation switching through giant spin-orbit torque has 14 been demonstrated in topological insulators (TIs) bi-15 layer structure<sup>4</sup>, and voltage control of the spin-orbit 16 torque of magnetic topological insulators was recently 49 17 achieved<sup>5</sup>. TIs possess helically spin-polarized Dirac cone 18 for their surface states. The spin momentum locking 50 19 is extremely beneficial as a mere switching of a ballis- 51 20 tic current direction provides the opposite spin polariza- 52 21 tion of the current. Although the spin polarized Dirac 53 22 cone have been studied in various TIs by angle-resolved 54 23 and spin-resolved photoemission spectroscopy (ARPES 55 24 and SRPES)<sup>6</sup>, the evidence for direct electrical detec- 56 25 tion is scarce. Direct electric current detection of the TI 57 26 spin polarized surface states have recently been reported 58 27 by magnetoresistance measurements<sup>7</sup> and non-local spin <sup>59</sup> 28 injection<sup>8-10</sup>. Here we report on the direct spin polar-  $_{60}$ 29 ized current detection by Point Contact Andreev Re- 61 30 flection (PCAR) spectroscopy. Among the three well- 62 31 established direct spin polarisation measurement tech-63 32 niques (SRPES, spin-polarized tunnelling<sup>11</sup> and Andreev 64 33

reflection<sup>12</sup>), PCAR has the distinct advantage that it can be used to evaluate the ballistic spin polarisation in a very narrow energy window centered at the Fermi level<sup>13</sup>, P. While the primary focus of this work is PCAR spin polarization measurements on high-quality thin films of the compositions Bi<sub>2</sub>Te<sub>3</sub>, Sb<sub>2</sub>Te<sub>3</sub>, and (Bi<sub>0.18</sub>Sb<sub>0.82</sub>)<sub>2</sub>Te<sub>3</sub>, high-field point contact spectroscopy is also studied, in order to demonstrate the low-temperature semiconducting behavior of the two end compositions and allows for the extraction of their gap values. Finally, the spin polarization in four magneticallydoped TIs (Bi<sub>1.97</sub>Cr<sub>0.03</sub>Te<sub>3</sub>, Sb<sub>1.975</sub>Cr<sub>0.025</sub>Te<sub>3</sub>, and Bi<sub>1.975</sub>V<sub>0.025</sub>Te<sub>3</sub>, and Sb<sub>1.977</sub>V<sub>0.03</sub>Te<sub>3</sub>) is probed to find substantially reduced values when compared to the same in the original compositions.

#### **II. EXPERIMENTAL DETAILS**

PCAR<sup>12</sup>, spin-polarised tunnelling, spin-resolved photoemission spectroscopy and spin-resolved field emission spectroscopy are just some of the techniques applicable towards the measurement of spin polarisation. PCAR is one of the most versatile ones, among these, for it has the ability to probe not only the diffusive, but also the ballistic definition of the spin polarisation in the meV vicinity of the Fermi level<sup>13</sup>. The experimental configuration utilized here is known as the needle-anvil approach, where a shear-cut superconducting niobium tip (Nb) lands on the sample. The "landing" procedure in our experimental setup is fully automatic and performed using a vertical attocube<sup>TM</sup> piezo stepper. Negative feedback based on zero-bias differential conductance is used to control the effective size of the contact. Whenever needed, hor-

izontal stepping is performed as well in order to ensure<sub>123</sub> 65 that a pristine (unscratched) area of the sample is al-124 66 ways probed. Direct bias-dependent differential conduc-125 67 tance measurements are taken instead of sampling the126 68 DC current-voltage characteristics and computing their<sub>127</sub> 69 numerical derivatives. This preferred approach has the128 70 advantage of better signal-to-noise ratio, due primarily to129 71 the use of lock-in amplifiers (LIA) and the corresponding<sub>130</sub> 72 narrowbanding and optimal pre-amplifier use. Further-131 73 more, the differential spectra are observed and recorded<sub>132</sub> 74 in real time, at a rate of 1 Hz. This permits more sophis-134 75 ticated post-processing, where the contact drifts can be  $_{135}$ 76 essentially eliminated and their influence on the further<sub>136</sub> 77 data analysis minimized. Low-frequency (0.5 Hz) analog $_{\scriptscriptstyle 137}$ 78 triangular waveform is modulated by the internal syn-79 chronous oscillator of a LIA (1.23 kHz). The modulation,  $_{130}$ 80 frequency is chosen high-enough to minimize the  $1/f_{-140}$ 81 noise contributions of the JFET pre-amplifiers, but at  $_{\scriptscriptstyle 141}$ 82 the same time low-enough, in order to keep to a minimum  $_{_{142}}$ 83 the signal dephasing due to the spurious cable inductance 84 and capacitance. The modulated voltage waveform is  $fed_{144}^{144}$ 85 in the contact and the current is pre-amplified at vari- $_{\scriptscriptstyle 145}$ 86 able gain and band filtered, at 6 dB/octave, before being  $_{146}$ 87 recorded by the LIAs. Two synchronized LIAs  $\mathrm{measure}_{_{147}}$ 88 and transfer data concurrently on the upward and down-  $_{\scriptscriptstyle 148}$ 89 ward trend of the triangular waveform, so that the dead 90 time of the measurement is practically zero. Further 91 details of the experimental setup, data acquisition and data preprocessing software can be found elsewhere<sup>14</sup>. 92 93 All measurements are taken in the variable tempera-94 ture insert of the commercially available Quantum De-95 sign Physical Property Measurement System (PPMS). 96 PCAR spectroscopy is a performed in the temperature 97 range 2.0 - 10.0 K. Measurements above the critical tem-98 perature of Nb ( $T_c = 9.2 \,\mathrm{K}$ ) are used as normalization 99 spectra and for corrections. PCS in high magnetic field 100  $(\mu_0 H = 14 \text{ T})$  is measured in order to study the density 101 of states of the compositions. The temperature evolu-157 102 tion of the zero-bias anomaly in the high-field PCS data  $^{^{158}}$ 103 is indicative of the insulating bulk behavior of the  $\operatorname{com^{-159}}$ 104 160 positions at low temperature. 105 161

High-purity Bi(99.999%), Sb(99.9999%) and Te<sub>162</sub> 106 (99.9999%) were evaporated from Knudsen effusion cells. 163 107 and Cr(99.999%) and V(99.995%) were deposited by 164 108 electron guns on heat-treated  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>(0001) substrates<sup>165</sup> 109 in a custom built molecular beam epitaxy system with 166 110 a base pressure better than  $5 \times 10^{-10}$  torr. The com-167 111 positions are determined by elements ratios obtained  $in_{-168}$ 112 situ during growth using separate quartz crystal moni-169 113 tors. The TI thickness of each sample is 20 quintuple<sub>170</sub> 114 layerd (QL) and the surface is protected with 2 nm insu-171 115 lating Te capping layer<sup>15</sup>. This particular ternary topo-172 116 logical insulator family  $(Bi_{1-x}Sb_x)_2Te_3$  has been previ-173 117 ously extensively studies by Zhang et al.<sup>15</sup>. The varia-174 118 tion of Bi and Sb concentration drives the Fermi level<sub>175</sub> 119 from the bulk conduction band  $(Bi_2Te_3)$  into the bulk<sub>176</sub> 120 valence band  $(Sb_2Te_3)$ . Respectively, the carriers are<sub>177</sub> 121 electrons and holes. At the optimized Bi:Sb ratio win-178 122

dow, the Fermi level can be tuned into the bulk gap and approached to the Dirac point. The compensated sample exhibits semiconductor-like activation behavior upon cooling. Due to the high sheet resistance, the  $Sb_2Te_3$ and  $(Bi_{0.18}Sb_{0.82})_2Te_3$  samples are shadow masked with Al(40 nm)/Ag(30 nm). Bottom shunting layer<sup>16</sup> would not significantly reduce the sheet resistance in this case due to the significantly reduced bulk conductivity. Spin polarization in topological insulators is in plane and locked to the direction of the in-plane wave-vectors  $(k_x)$ and  $\vec{k}_{y}$ ). Statistically each electron of a Cooper pair has finite vertical projection of its spin. Furthermore, in the ballistic regime of PCAR (with small axial magnetic field) or alternatively in cases where there is a significant tunneling barrier and the injection is performed from a superconductor with finite spin-orbit coupling, Cooper pairs spins are aligned along the perpendicular wave-vector( $\vec{k}_z$ ). Cooper pairs are not allowed to be injected in topological insulators (due to momentum conservation within the elastic limit and at small bias) with spins along z, hence the Andreev reflection is suppressed and the contact shows high spin polarization. The measurement is a direct indication that the transport spin polarization is in plane. Since the PCAR probes contributions from both the surface and bulk states, an increase in spin polarization is to be expected, related to carrier depletion in the bulk, *i.e.* pronounced semi-metallic or semiconducting transport properties at low temperature. We observe exactly that, the spin polarization is higher for the compositions with larger bulk bandgap.

#### III. SPIN POLARISATION OF $(BI_{1-x}SB_x)_2TE_3$ FAMILY

PCAR spectra are measured in the temperature range from  $2.0\,\mathrm{K}$  to  $10.0\,\mathrm{K}$ . The conductances of most contacts are in the range 5.0 - 20.0  $G_0$ . The high resistance of the point contacts is sought after because of two reasons: the transport current through the contact is kept in a quasiballistic regime, and the relative contribution of the TI sheet resistance is low. It has to be explicitly noted that reducing the point-contact resistance (increasing the effective contact area) leads to broadening of the spectral features and decreased value of the spin polarization. The analysis routine is based on Strijkers *et al.*<sup>17</sup> approach, although no significant proximity effect is observed in any of the contacts. The important extracted parameters from each fit are spin polarization P, barrier height Z, effective electronic temperature  $T_{\rm e}$ , and the proximity gap  $\Delta_1$ . The bulk superconducting gap of Nb,  $\Delta_2$ , is kept constant. No significant proximity effect is present as the values of  $\Delta_1$  and  $\Delta_2$  are similar. Furthermore, additional five fitting parameters might be used : zero bias offset  $(x_0)$ , conduction normalization offset $(y_0)$ , rescale of the conduction axis  $(y_s)$ , due to pre-amplifier gain and sheet resistance), rescale of the voltage axis  $(x_s, due to$ sheet resistance), and quadratic background component

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FIG. 1. PCAR spectra, fitting and extracted parameter for the three different compositions. (a) - Bi<sub>2</sub>Te<sub>3</sub> with P = 70(4) %, (b) - Sb<sub>2</sub>Te<sub>3</sub> with P = 57(3) %, and (c) - (Bi<sub>0.18</sub>S<sub>0.82</sub>)<sub>2</sub>Te<sub>3</sub> with P = 83(9) %. The model extracted parameters are indicated with asterisks. Panel (d) represents the electron injection process in PCAR. The electron spins of the TI surface states are in-plane locked perpendicular to the momentum. Cooper pairs (CP) with out of plane spin components are not allowed to be injected elastically in the TI surface states due to momentum conservation.

 $(y_{q}, \text{ due to tunneling contributions})$ . A multiparame-197 179 ter fit is susceptible to converging in local minima rather198 180 than the global solution and the uniqueness of the PCAR<sub>199</sub> 181 fit has been discussed before  $^{18,19}$ . Our fitting procedure<sub>200</sub> 182 includes close-interval minimization and calculates the<sub>201</sub> 183 correlation and covariant matrices of the obtained fit in<sub>202</sub> 184 order to uncover interdependence between the parame-203 185 ters and the errors. An example is given on Fig. 4 (f).204 186 Three representative spectra along with the extracted fit-205 187 ting parameters for the three topological compositions<sup>206</sup> 188  $(Bi_2Te_3, Sb_2Te_3, and (Bi_{0.18}Sb_{0.82})_2Te_3)$  are shown in<sup>207</sup> 189 Fig.(1). It has to be pointed out that all spectra exhibit  $_{208}$ 190 small quadratic high bias background. The background<sup>209</sup> 191 is attributed to the DOS structure of the topological in-210 192 sulators and small tunneling current contribution. Pre-211 193 viously extensive studies of the DOS bias dependence in212 194 topological insulators have been performed by scanning213 195 tunnelling microscopy $^{20,21}$ . Here, the energy scale is  $\lim_{214}$ 196

ited to the immediate (few meV) vicinity of the Fermi level. The PCAR spectra are normalized with spectra above the  $T_{\rm c}$  of Nb. It has to be stated that the effective electronic temperature  $T_{\rm e}$  is found to always be above the bath temperature of the setup T. The typical value for  $T_{\rm e}$  is from 4.0 to 6.0 K, whereas the typical bath temperature T is from 2.0 to 2.4 K. Such electron heating and additional thermal broadening is often observed in PCAR of thin films<sup>22</sup>, exception being only cases of cooling in low transparent superconducting tunnel junctions $^{23}$ . In this set of measurements, the elevated electronic temperature is mainly attributed to the formation of a narrow (tunneling-transparent) Schottky barrier at the tip-TI contact<sup>24</sup>, which will be demonstrated later on Fig. 2 (c) and (d). The extracted spin polarization is 70(4), 57(3) and 83(9)%, respectively for the three compositions. In order to confirm that the observed spectral features are due to Andreev reflection, tempera-



FIG. 2. Temperature scans of Bi<sub>2</sub>Te<sub>3</sub> and Sb<sub>2</sub>Te<sub>3</sub>. (a) - PCAR spectra of Bi<sub>2</sub>Te<sub>3</sub> between 2.0 to 10.0 K, (b) - PCAR spectra of Sb<sub>2</sub>Te<sub>3</sub> between 2.0 to 10.0 K, (c) - PCS of Bi<sub>2</sub>Te<sub>3</sub> in magnetic field of  $\mu_0 H = 14$  T, and (d) - PCS of Sb<sub>2</sub>Te<sub>3</sub> in magnetic field of  $\mu_0 H = 14$  T, where  $\Delta_2 = 1.5$  meV is the bulk superconducting gap of Nb and q is the elementary charge.

ture scans of some of the contacts are recorded (Fig.(2)).241 215 The spectra are normalized with a background curve ac-242 216 quired at 10.0 K (above the critical temperature of Nb  $_{-243}$ 217  $T_{\rm c} = 9.2 \,\mathrm{K}$ ). PCAR temperature scans indicate the<sup>244</sup> 218 usual evolution of the spectra where the features be-245 219 come narrower and lower due to the reduction in the246 220 Nb superconducting gap combined with the temperature<sup>247</sup> 221 smearing; spectra are essentially flat above  $T_{\rm c}$ . The last<sub>248</sub> 222 observation is a clear confirmation that the investigated<sup>249</sup> 223 samples demonstrate the expected high effective spin po-250 224 larisation. Furthermore, the fact that Andreev signal<sub>251</sub> 225 is measured all the way to the critical temperature of<sub>252</sub> 226 Nb (see Fig. (3) (a) and (b)) is a clear demonstration<sub>253</sub> 227 that the transport is elastic and there is no appreciable<sub>254</sub> 228 Joule heating in the contact area. Once the supercon-255 229 ducting tip is quenched (either in high magnetic field or<sub>256</sub> 230 at high temperature) and provided that the density of<sub>257</sub> 231 states around the Fermi level ( $\approx \pm 15 \,\mathrm{meV}$ ) is feature-258 232 less, the point-contact spectroscopy (PCS) must exhibit<sub>259</sub> 233 a flat line. Interestingly, spectra exhibit really structure<sub>260</sub> 234 in magnetic field above the upper critical field of Nb. As<sub>261</sub> 235 the magnetic field is scanned from 0 to 14 T, the spectral<sub>262</sub> 236 features change from the well-established Andreev reflec-263 237 tion structure to a Lorentzian-like shape. The latter is an<sub>264</sub> 238 indication of carrier depletion and existence of small band<sub>265</sub> 239 gap in the bulk density of states of the topological insula-266 240

tors. The conductance features of the PCS are an order of magnitude smaller than the features of the PCAR. Hence, contribution from the carrier depletion towards the overall PCAR signal is considered insignificant for the analysis of the spin polarization. The high-field PCS demonstrates essentially the differential conductance of the tunneling barrier between the Nb tip and the TI sample. The zero bias anomaly (ZBA) of the PCS temperature scans (Fig. (2) c and d) decays much faster than the ZBA of the PCAR temperature scans (Fig. (2) a and b) as temperature is increased. The temperature evolution of the ZBA in PCS decays exponentially following Arrhenius law. On the other hand, the ZBA of PCAR should follow the Bardeen-Cooper-Schrieffer  $(BCS)^{25,26}$ evolution of the superconducting gap, provided that the density of states (DOS) of the studied material are flat within the DC bias range. The critical exponent of the superconducting transition in the weak-coupling regime following the BCS-theory is  $\gamma = 0.5^{25,26}$ . The extracted critical exponents for the transitions of the Bi<sub>2</sub>Te<sub>3</sub>/Nb and  $Sb_2Te_3/Nb$  contacts are significantly higher : 0.91(2)and 0.71(2), respectively. The deviation from the BCS bahaviour is due to formation of a narrow Schottky barrier and that the spin polarisation is efficient Cooper pair breaker<sup>27</sup>. A consequence of this is that the effective electronic temperature is significantly above the bath tem-



FIG. 3. Temperature evolution of the ZBA of PCAR (top panels) and PCS (bottom panels) along with extracted critical exponents ( $\gamma$ ) of the superconducting transition and the bulk band gap ( $E_{\rm g}$ ). (a) - ZBA of the PCAR temperature scan in zero magnetic field of Bi<sub>2</sub>Te<sub>3</sub> with  $\gamma = 0.91(2)$ , (b) - ZBA of the PCAR temperature scan of Sb<sub>2</sub>Te<sub>3</sub> in zero magnetic field with  $\gamma = 0.71(2)$ , (c) - ZBA of the PCS temperature scan of Bi<sub>2</sub>Te<sub>3</sub> in  $\mu_0 H = 14$  T with  $E_{\rm g} = 0.40(4)$  meV, and (d) - ZBA of the PCS temperature scan of Sb<sub>2</sub>Te<sub>3</sub> in  $\mu_0 H = 14$  T with  $E_{\rm g} = 0.28(2)$  meV. The presented ZBA plots are extracted from the ZBA temperature dependence in Fig. (2).

<sup>267</sup> perature (injection occurs above the Fermi level). Fur-<sup>290</sup> thermore, as these materials exhibit very small gap at<sup>291</sup> low temperatures, the observed energy gap evolves sig-<sup>292</sup> nificantly from 2 to 10 K, and the  $T_{\rm e}$  does not follow<sup>293</sup> linearly the increase in T.<sup>294</sup>

The temperature evolution of ZBA of PCS in 272  $\mu_0 H = 14 \text{ T}$  is presented in Fig. (3). The ZBA temper-273 ature evolutions in high field (bottom panels on Fig. (3)) 274 is in contrast with the ZBA temperature scans in zero<sup>296</sup> field (top panels on Fig. (3)). Our very high field PCS<sup>299</sup> 275 276  $(\mu_0 H = 14 \text{ T})$  demonstrates small bulk bandgaps for 301 277 both compositions. The Arrhenius fitting of the data gives  $E_{\rm g} = 0.28(2) \,\mathrm{meV}$  and  $0.40(4) \,\mathrm{meV}$ , respectively<sup>302</sup> 278 279 for Sb<sub>2</sub>Te<sub>3</sub> and Bi<sub>2</sub>Te<sub>3</sub>. The larger gap of Bi<sub>2</sub>Te<sub>3</sub> cor-303 280 relates with the measured higher spin polarization. In<sub>304</sub> 281 fact, resistance upturn at very low temperatures has al-305 282 ready been reported for these compositions<sup>15</sup>. It is im-<sub>306</sub> 283 portant to note that the observed gap should not be di-307 284 rectly linked to the gap in the TI electronic structure.<sup>308</sup> 285 The Nb tip might introduce disorder (vacancies, disloca-309 286 tions etc.) in the crystal structure, and, hence, alter the<sub>310</sub> 287 density of states. The work of Jiang et al.<sup>28</sup> has demon-311 288 strated that the bulk gap in  $Sb_2Te_3$  decreases with in-312 289

creasing the thickness of the TI sample. Furthermore, these compositions exhibit close to degenerate semiconducting behavior with the Fermi level positioned in the vicinity of the conduction or the valence band edges, respectively. Therefore, the extracted gaps demonstrate the energy spacing between the Fermi level and the corresponding band edge rather than the full band gap. The determined bulk band gap in these samples is 100 meV-200 meV<sup>15</sup>. The larger extracted gap of Bi<sub>2</sub>Te<sub>3</sub> correlates with the higher value of critical exponent extracted from the PCAR temperature dependence (see Fig. (3)c). This observation of high-field PCS is an explicit demonstration of carrier depletion in these topological insulators.

It is important to comment on the full field scan of a  $\text{Bi}_2\text{Te}_3/\text{Nb}$  point contact(Fig. (5)). The low field scan indicates that the Andreev-reflection features disappear in small field  $\mu_0 H \approx 0.5 \text{ T}$ . The contact quench field is on par with the bulk upper critical field  $H_{c3}$  of  $\text{Nb}^{29}$  and within the error expected for the lack of proper polycrystalline average within the point contact  $(\pm 36\%)^{30}$ . As was the case with the temperature evolution, the field evolution of the superconducting gap does not follow the expected BCS evolution. The reasons are likely

related to the highly spin polarized current through the<sub>368</sub> 313 contact<sup>27</sup>. Furthermore, the full field scan indicates that<sub>369</sub> 314 the ZBA is relatively field insensitive from the guench<sub>370</sub> 315 field  $(\mu_0 H = 0.5 \text{ T})$  up to the maximum available<sub>371</sub> 316 field  $\mu_0 H = 14 \,\mathrm{T}$ . The effective field scale over which<sub>372</sub> 317 the conductance dip diminishes is about  $\mu_0 H = 10 \text{ T}_{,373}$ 318 which corresponds to Zeeman splitting of approximately<sub>374</sub> 319 0.92 meV or narrowing of the gap, very close to the one375 320 observed by thermal activation in Fig. (3). This could be<sub>376</sub> 321 interpreted as either the inelastic tunneling between two377 322 states (one in the valence part and one in the conduc-378 323 tion part of the Dirac cone) at constant momentum, 01379 324 potentially the spin-orbit splitting of a bulk conduction<sub>380</sub> 325 state at the Fermi level. 381 326

## IV. SPIN POLARISATION OF MAGNETICALLY-DOPED TOPOLOGICAL INSULATORS

We further investigate the effect of magnetic ions dop-330 ing on the spin polarization of Bi<sub>2</sub>Te<sub>3</sub>, and Sb<sub>2</sub>Te<sub>3</sub>. Mag-331 netic topological insulators have been realized by Cr- and 332 V-doping<sup>31,32</sup>. Recently, high Curie temperature and ro-<sup>389</sup> 333 bust Quantum Anomalous Hall Effect (QAHE) were ob-334 served in optimally V-doped TIs<sup>33</sup>. Both Cr and V are<sub>390</sub> 335 substitutional on the Bi(Sb) sublattice. While V acts as<sub>391</sub> 336 electron donor<sup>33</sup> in  $Sb_2Te_3$ , the contribution of carriers<sup>392</sup> 337 by Cr doping is smaller but hole type<sup>31,34</sup> in the same<sub>393</sub> 338 parent compound. The carrier concentration in the Cr-394 339 Bi<sub>2</sub>Te<sub>3</sub> should be reduced and the spin polarization in-395 340 creased. Contrarily, Cr-doping was shown to decrease the396 341 spin polarization of the Bi<sub>2</sub>Te<sub>3</sub> composition which is at-397 342 tributed to spin-flip scattering effect from paramagnetic<sub>398</sub> 343 impurities<sup>35</sup>. On the other hand, Cr-doping of Sb<sub>2</sub>Te<sub>3399</sub> 344 is expected to affect very little or to slightly increase400 345 the bulk hole carrier concentration, which should lead to<sub>401</sub> 346 lower spin polarization compared with the pure  $Sb_2Te_{3402}$ 347 sample. Decreased spin polarization is indeed measured<sub>403</sub> 348 in this composition. The extracted spin polarization<sub>404</sub> 349 of  $Bi_{1.97}Cr_{0.03}Te_3$  and  $Sb_{1.975}Cr_{0.025}Te_3$  is 52(1)% and 405 350 52(1)%. Vanadium doping has been shown to induce<sub>406</sub> 351 harder magnetism and higher Curie temperature than<sub>407</sub> 352 chromium<sup>33</sup>. V-doped Bi<sub>2</sub>Te<sub>3</sub> demonstrated a lower spin<sub>408</sub> 353 polarization value of P = 58(5)% than the pris-409 354 tine composition. Three of the measured compositions<sup>410</sup> 355 do not demonstrate magnetic ordering : Bi<sub>1.97</sub>Cr<sub>0.03</sub>Te<sub>3,411</sub> 356  $Sb_{1.975}Cr_{0.025}Te_3$ , and  $Bi_{1.975}V_{0.025}Te_3$ . The decreased<sup>412</sup> 357 values of the spin polarization is due to spin-flip scat-413 358 tering by paramagnetic impurities<sup>36</sup>. Non-magnetic im-414 359 purities and disorder do not cause backscattering of<sub>415</sub> 360 electrons from the TI surface states<sup>37</sup>, however, doping<sub>416</sub> 361 with magnetic ions must result in spin-flip scattering<sup>35</sup>.417 362  $Sb_{1.97}V_{0.03}Te_3$  is the only composition which demon-418 363 strates magnetic behaviour in the measurement temper-419 364 ature range ( $T > 2.0 \,\mathrm{K}$ ). The extra free electrons, pro-420 365 vided by the vanadium, reduce the natural p-type char-421 366 acter of the original composition and this should result<sub>422</sub> 367

in a lower bulk conduction and higher contribution of the surface carriers towards the measured spin polarization. However, it exhibits the lowest value of spin polarization (P = 50%). The decreased spin polarization in the magnetically-ordered sample is attributed to the fact that the magnetic easy axis is perpendicular to plane (as evidenced by AHE) whereas the spins of the electrons in the TIs surface state are usually locked in the plane. This must increase the electron-electron spin scattering and via the Kramers-Kronig relations reduce the density of surface states and their bulk penetration thus impacting the overall effective spin polarization as perceived by PCAR. No features similar to the PCS in Fig. (1 (c) and (d)) have been observed in the magnetically doped samples<sup>38,39</sup>. The ferromagnetic ordering of all compositions is probed by Anomalous Hall Effect (AHE) in van der Pauw configuration. Only Sb<sub>1.97</sub>V<sub>0.03</sub>Te<sub>3</sub> demonstrated AHE signal at temperature of 2.0 K or higher. The AHE amplitude of the latter composition is evaluated as a function of temperature and the Curie temperature of  $T_{\rm C} = 11.6(6)$  K determined (see Fig. (4)e).

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#### V. CONCLUSIONS

The high in-plane spin polarization in the topological insulator family  $(Bi_{1-x}Sb_x)_2Te_3$  is verified by a direct electrical measurement. The measured spin polarization is in the range of 60 to 85% and represents a low limit for the intrinsic spin polarization in these topological insulators. Various artefacts stay in the way of a more accurate determination: tip induced damage, directionality of the current, not-fully ballistic transport and interfacial spin-flip events at the topological insulator/superconductor interface. It has to be emphasized that the value is obtained at 2K, and no temperature dependence of the spin polarization is studied due to the limitations of PCAR imposed by the low-critical temperature of Nb. Carrier depletion by direct point contact spectroscopy was demonstrated for first time. The extracted bulk bandgaps of Bi<sub>2</sub>Te<sub>3</sub> and Sb<sub>2</sub>Te<sub>3</sub> correlate with the expectation that higher bulk carrier depletion results in higher effective spin polarisation. Furthermore, we investigated the influence of magnetic ions on the spin polarization of four topological insulator compositions :  $Bi_{1.97}Cr_{0.03}Te_3$ ,  $Sb_{1.975}Cr_{0.025}Te_3$ ,  $Bi_{1.975}V_{0.025}Te_3$ , and  $Sb_{1.97}V_{0.03}Te_3$ . Low doping concentration, insufficient to induce ferromagnetic behaviour, was shown to reduce the value of the spin polarization. The latter demonstrates that paramagnetic ions act as spin-flip scattering centers and decrease the TI surface states spin polarization<sup>35</sup>. The only ferromagnetic composition among the four,  $Sb_{1.97}V_{0.03}Te_3$ , had the lowest spin polarization which is attributed to the competition between in-plane spin locking and perpendicular magnetic anisotropy. Extensions of the current investigation should permit the measurement of the spin polarization of optimally V-doped  $Sb_2Te_3$ , the tracking of the spin polarization as a func-



FIG. 4. Fitted PCAR data along with the extracted parameters of four magnetically doped TI compositions. Panel (a) shows  $Bi_{1.97}Cr_{0.03}Te_3$ , Panel (b) -  $Sb_{1.975}Cr_{0.025}Te_3$ , Panel (c) -  $Bi_{1.975}V_{0.025}Te_3$ , Panel (d) -  $Sb_{1.97}V_{0.03}Te_3$ , Panel (e) - AHE temperature scan of  $Sb_{1.97}V_{0.03}Te_3$  with the extracted Curie temperature of  $T_C = 11.6(6)$  K. Panel (f) - example of a covariant matrix on the fit on Panel (a) and the vector of the parameters errors.

tion of the applied gate bias, and the evaluation of the
spin polarization in the QAHE regime.

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FIG. 5. Magnetic field scan of a Bi<sub>2</sub>Te<sub>3</sub>/Nb point contact at 2.0 K. Left panel represents the low-field scan up to  $\mu_0 H = 1.0$  T and the right panel represents the high-field scan from 1 T up to  $\mu_0 H = 14.0$  T. Where  $\Delta_2 = 1.5$  meV is the bulk superconducting gap of Nb and and q is the elementary charge.

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