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Logarithmic singularities and quantum oscillations in magnetically doped topological insulators

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We report magnetotransport measurements on magnetically doped $(Bi,Sb)_2Te_3$ films grown by molecular beam epitaxy. In Hallbar devices, we observe logarithmic dependence of transport coefficients in temperature and bias voltage which can be understood to arise from electron - electron interaction corrections to the conductivity and self-heating. Submicron scale devices exhibit intriguing quantum oscillations at high magnetic fields with dependence on bias voltage. The observed quantum oscillations can be attributed to bulk and surface transport.

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I. INTRODUCTION

Breaking of time reversal symmetry in topological insulators can unlock exotic phenomenon such as the quantized anomalous Hall effect[1–5], giant magneto-optical Kerr and Faraday effects[6], the inverse spin-galvanic effect[7], image magnetic monopole effect[8] and chiral Majorana modes[9,10]. Angle resolved photoemission spectroscopy measurements have revealed presence of a magnetic gap at the Dirac point as well as hegdehog spin texture in magnetic topological insulators[11,12]. Proximity coupling to a magnetic insulator such as EuS, YIG and TIG[13–15] or introducing magnetic dopants like Mn, Cr and V[2,3,16] can remove time reversal symmetry. Such efforts have induced long range ferromagnetic order in topological insulators.

In this paper we explore magnetotransport in magnetically doped ultrathin films of $(Bi,Sb)_2Te_3$ to understand the role of different scattering mechanisms. By studying the effect of temperature and voltage bias on the longitudinal and anomalous Hall resistances, we observe logarithmic dependences on temperature and voltage bias. Joule heating due to voltage bias increases the effective temperature of the hot electrons[17]. The logarithmic singularities are originating from interplay of electron-electron interaction and disorder. We find that our observed logarithmic corrections quantitatively agree with the Alshuler-Aranov theory of electron-electron interactions. Furthermore, in submicron sized mesocale devices we observe quantum oscillations that depend on voltage bias and weaken with increasing sample width.

II. MATERIALS AND METHODS

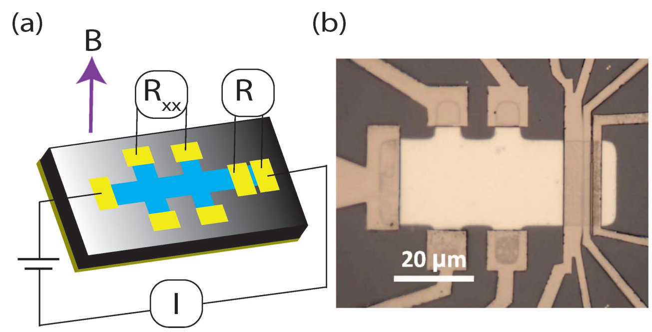

FIG. 1. (color online) (a) Schematics of a magnetic topological insulator device. (b) Optical image of a Hallbar device H_1 and a two terminal device D_1 simultaneously patterned on magnetically (Vanadium) doped topological insulators.

The 4 quintuple layers (QLs) thick pristine and V-doped $(Bi,Sb)_2Te_3$ films are grown on $SrTiO_3$ (111) substrate by molecular beam epitaxy (MBE). The growth process was monitored in-situ by refection high-energy electron diffraction (RHEED) to ensure high quality films[3,18,19]. To prevent oxidation, a capping layer of 10 nm tellurium was deposited. The devices were fabricated employing standard photolithography and electron-beam lithography techniques. The device schematic and optical image of a Ti/Nb/NbN contacted magnetic topological insulator film is shown in Figs. 1(a)-1(b). The transport measurements were done in a ^3He-^4He dilution refrigerator using standard ac lock-in measurement techniques. Here we summarize results from a pristine Hallbar device H_0, a V-doped Hallbar device H_1 and a V-doped submicron scale device D_1.

III. RESULTS AND DISCUSSION

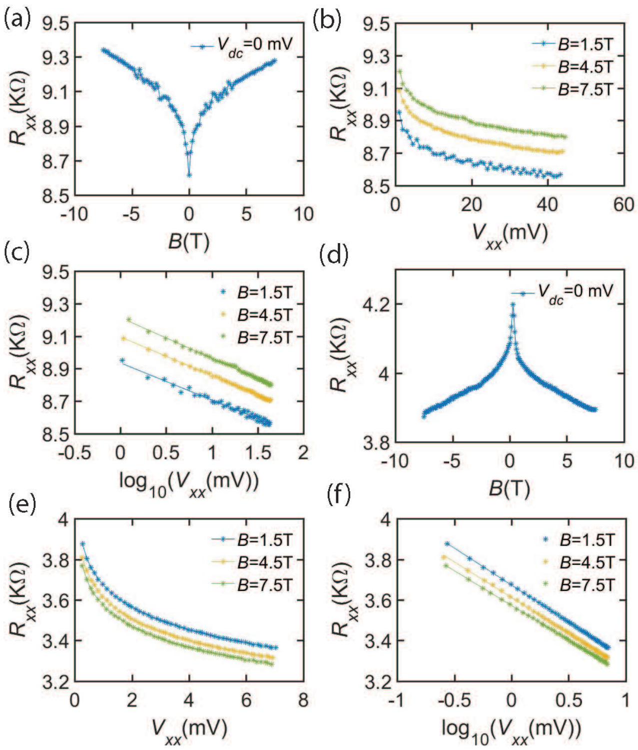

FIG. 2. Comparison of magnetotransport in pristine and V-doped 4 QL thick $(Bi,Sb)_2Te_3$ films. (a) Magnetoresistance dip at B=0 T in the sweep range B = -7.5 to +7.5 T for pristine $(Bi,Sb)_2Te_3$ samples. (b) Bias dependence of pristine $(Bi,Sb)_2Te_3$ film at B = 1.5, 4.5 and 7.5 T. (c) Logarithmic plot for bias dependence of 4 QL pristine $(Bi,Sb)_2Te_3$ film at B = 1.5, 4.5 and 7.5 T. (d) Magnetoresistance peak at B=0 T in the sweep range B = -7.5 to +7.5 T for V-doped $(Bi,Sb)_2Te_3$. (e) Bias dependence of V-doped $(Bi,Sb)_2Te_3$ film at B = 1.5, 4.5 and 7.5 T. (f) Logarithmic plot for bias dependence of V-doped $(Bi,Sb)_2Te_3$ film at B = 1.5, 4.5 and 7.5 T.

Magnetotransport measurements in a Hallbar device H_0 on pristine 4QL $(Bi,Sb)_2Te_3$ films presented in Fig. 2(a) exhibit a dip in the longitudinal resistance R_{xx} at B = 0 T which is attributed to weak antilocalization effect[20,21]. This is because in the presence of strong spin-orbit coupling time reversed trajectories have opposite spin orientations which lead to a destructive interference and a resistance minimum[22].

We measured the bias dependence of longitudinal resistance R_{xx} in the same device. The results are shown in Fig. 2(b) - Fig. 2(c) for a few different magnetic fields and exhibit a logarithmic dependence on voltage bias. Weak antilocalization is in itself a possible cause of logarithmic correction. However lowering temperature or voltage bias is expected to make weak antilocalization effect more pronounced thereby decreasing resistivity which is inconsistent with Fig. 2(b) - Fig. 2(c).

Further by introducing magnetic impurities, the weak antilocalization effects can be heavily suppressed as has been reported in Fe-doped Bi_2Te_3 and Cr-doped Bi_2Se_3 films[23–25]. Fig. 2(d) shows the magnetoresistance in a V-doped $(Bi,Sb)_2Te_3$ has a peak instead of a dip at B = 0 T seen in pristine samples. Even when the weak antilocalization effects are suppressed, the longitudinal resistance R_{xx} in V-doped $(Bi,Sb)_2Te_3$ has a logarithmic dependence on voltage bias as shown in Fig. 2(e) - Fig. 2(f) at different magnetic fields.

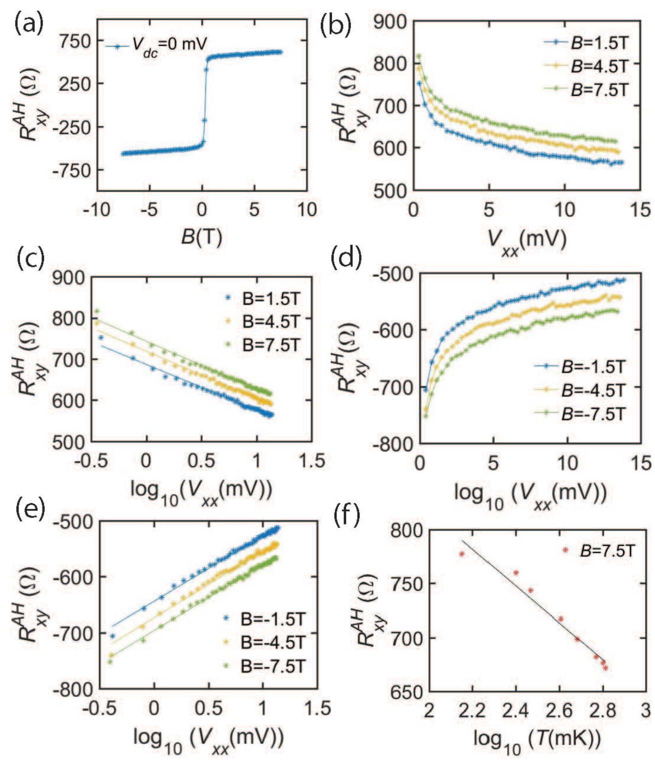

FIG. 3. (color online) Voltage bias and temperature dependence of anomalous Hall effect of V-doped 4 QL thick $(Bi,Sb)_2Te_3$ films. (a) Magnetic field dependence of anomalous hall resistance R^{AH}_{xy} measured at V_{dc} = 0. (b) Bias dependence of R^{AH}_{xy} at B = 1.5, 4.5 and 7.5 T. (c) Logarithmic plot for bias dependence of R^{AH}_{xy} at B = 1.5, 4.5 and 7.5 T. (d) Bias dependence of R^{AH}_{xy} at B = -1.5,- 4.5 and -7.5 T. (e) Logarithmic plot for bias dependence of R^{AH}_{xy} at B = -1.5, -4.5 and -7.5 T. (f) R^{AH}_{xy} exhibits a logarithmic dependence on temperature at B = 7.5 T in the temperature range of 140 mK to 650 mK .

Weak localization in disordered two dimensional (2D) systems is also a potential explanation for logarithmic increase in resistance at low temperatures. The existence of weak localization relies on the existence of coherent constructive interference of time reversed trajectories for an electron to return to the origin[26]. Moderate external magnetic fields as well as magnetic impurities, that break time reversal symmetry, are typically enough to suppress logarithmic corrections arising from weak localization[27–29]. However, the logarithmic corrections observed in our experiments persist even at fields of 7.5 T.

A way to identify logarithmic corrections due to weak localization is by the absence of logarithmic corrections to R_{xy} [27,28,30]. In Fig. 3(a) anomalous Hall measurements are shown without an applied bias. The R_{xy}^{AH} jumps at the coercive field when the magnetization switches its direction. Fig. 3(b)- Fig. 3(c) shows that the logarithmic dependence on voltage bias are present in anomalous Hall resistance R_{xy}^{AH} as well. The data is antisymmetric in magnetic field as shown in Fig. 3(d)- Fig. 3(e) because of which we can rule out spurious R_{xx} contributions. Interestingly, if instead of bias voltage temperature of the sample is changed, similar decrease in resistance R_{xy}^{AH} is observed as shown in Fig. 3(f). Therefore, weak localization cannot explain the transport behavior that we observe.

Logarithmic corrections to conductance could also arise from scattering off magnetic impurities as in the Kondo effect[31–33]. However, in the ferromagnetic state magnetic spin-flips should become increasingly energetically unfavorable at low temperatures and at large external magnetic fields. More importantly, the fact that logarithmic dependences are also observed in topological insulator thin films in the absence of magnetic dopants[34,35] makes this scenario an unlikely explanation of our findings.

Magneto-transport studies in pristine topological insulator Bi_2Se_3 films found it crucial to include electron-electron interactions[34,35]. We explain why the observed logarithmic singularities are due to electron-electron interactions in the 2D surface states. As first realized by Altshuler and Aronov[36] (AA) disordered 2D electron systems exhibit a breakdown of the Fermi-Liquid theory due to reduced ability of the disordered electron gas to screen the Coulomb interaction. The logarithmic corrections in the AA theory are pervasive and are expected to arise not only in transport properties but also in equilibrium thermodynamic quantities such as specific heat[28]. One of the key differences of the AA corrections with those in the localization theory is that both the longitudinal and Hall resistivities are expected to acquire logarithmic corrections[27,28]. In fact the logarithmic corrections are most easily expressed in terms of conductivities rather than resistivities in the AA theory, because the Hall conductivity is expected to remain unchanged. Specifically one expects the logarithmic corrections in the AA model to be given by[27–29]:

$$\delta\sigma_{xx}(\varepsilon) = \kappa\frac{e^2}{h}\log(\frac{\varepsilon\tau}{\hbar}),\ \delta\sigma_{xy} = 0, \quad (1)$$

where τ is the elastic scattering time, and ε is an appropriate energy scale that can be chosen to be the largest among the temperature k_BT or the frequency $\hbar\omega$, at which the conductivity is probed. κ is a dimensionless number that takes different values for spinless and spinful electrons, and depends on a dimensionless parameter F that characterizes a Hartree contribution to the conductivity corrections[29,37,38]. This parameter takes the following forms for spinless and spinful electrons:

$$\kappa^{\text{spinless}} = \frac{1}{2\pi}, \quad (2)$$

$$\kappa^{\text{spinfull}} = \frac{1}{2\pi}(2-2F), \quad (3)$$

For short range interaction, F=1 and long range interaction F=0[39]. For spin-split bands one expects that for a spin splitting $\Delta \gg k_BT$, the only singular logarithmic terms arise from exchange and $S_z = 0$ Hartree contributions, and the expression for κ is[38]:

$$\kappa^{\text{spin}-\text{split}} = \frac{1}{2\pi}(2-F) \approx 0.32\left(1-\frac{1}{2}F\right). \quad (4)$$

Our magnetic samples are expected to be spin-split, whereas the precise level of spin polarization is unknown to us [28,29].

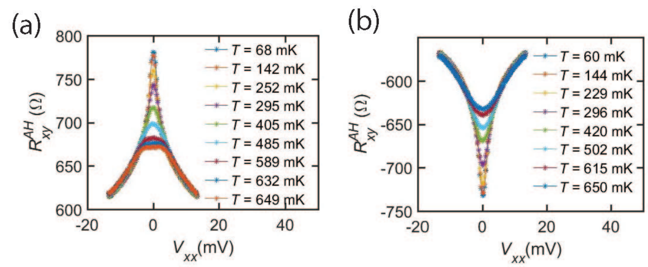

FIG. 4. (color online) Voltage bias dependence of the anomalous Hall effect at several fixed temperatures in V-doped 4 QL thick $(Bi,Sb)_2Te_3$ films for (a) B= 7.5T and (b) B= -7.5T.

Fig. 4(a)-(b) further explores the dependence of anomalous Hall effect on both voltage bias and temperature. Increasing either voltage or temperature lowers the anomalous Hall resistance which supports a self-heating mechanism due to applied bias.

We believe that the origin of the non-Ohmic behavior we observe, namely the logarithmic dependence of the conductivity on voltage bias, is fundamentally no different than the logarithmic dependence on temperature and can be understood simply as a consequence of Joule heating. In other words, as the electrons are accelerated by the electric field they inevitably gain energy, and, once they reach a steady state of current flow, this inevitably implies that the electrons posses a larger effective temperature compared to that of the lattice or other reservoirs that serve as heat sinks. By appealing to a simple model of Joule heating[40] one can effectively replace the argument of the logarithm in Eq. (1) by $\varepsilon \sim \max(AV^{2/(2+p)}, K_BT, \hbar\omega)$, where V is the voltage bias that drives the transport, A is a constant, and p is the

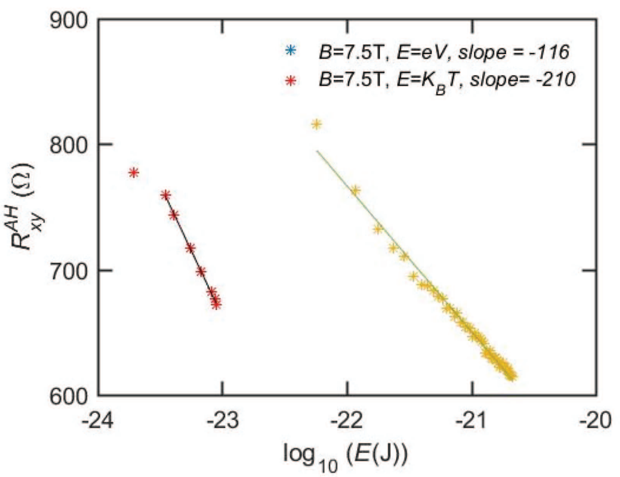

FIG. 5. (color online) Comparison of the dependence of anomalous Hall resistance R_{xy}^{AH} on logarithm of $K_B T$ and eV in V-doped 4 QL thick $(Bi,Sb)_2Te_3$ films at B= 7.5 T.

power that controls the temperature dependence of the electron's inelastic scattering rate, $\tau_{in} \propto T^{-p}$. The logarithmic fits of the Hall resistivity vs temperature have approximately twice the slope of those Hall resistivity vs the bias voltage indicating that $p \sim 2$, as shown in Fig. 5 .

The Joule heating induced by the bias voltage results to be a more efficient way to tune the electron temperature than the direct control of the temperature of the sample, and, hence we will focus on this dependence for the remainder of the discussion. The expected behavior of the correction to the conductivity for low temperatures dc measurements from the AA theory as a function of voltage is:

$$\delta\sigma_{xx}(V) = \frac{2\kappa}{(2+p)}\frac{e^2}{h}\log(V),\ \delta\sigma_{xy}(V) = 0. \quad (5)$$

From the data, σ_{xx} and σ_{xy} is calculated using the relations

$$\sigma_{xx} = \frac{\rho_{xx}}{\rho_{xy}^2 + \rho_{xx}^2}, \sigma_{xy} = \frac{\rho_{yx}}{\rho_{xy}^2 + \rho_{xx}^2} \quad (6)$$

where ρ_{xx} and ρ_{xy} are the resistivities considering the square shaped sample geometry. As illustrated in Fig. 6(a) our data is consistent with logarithmic corrections in σ_{xx} while no apparent logarithmic corrections in σ_{xy}, as expected from the AA theory. The anomalous Hall conductivity is nearly quantized at +/- e^2/h as shown in Fig. 6(b). Anecdotally, we argue that quantization of σ_{xy}^{AH} is less sensitive to finite bulk carriers than R_{xy}^{AH}.

To quantify the logarithmic behavior we fit the voltage-dependent nonlinear conductivity as (expressing bias in volts):

$$\sigma_J(V) = \sigma_J^0 + \delta\sigma_J \log(V),\ J = \{xx, xy\} \quad (7)$$

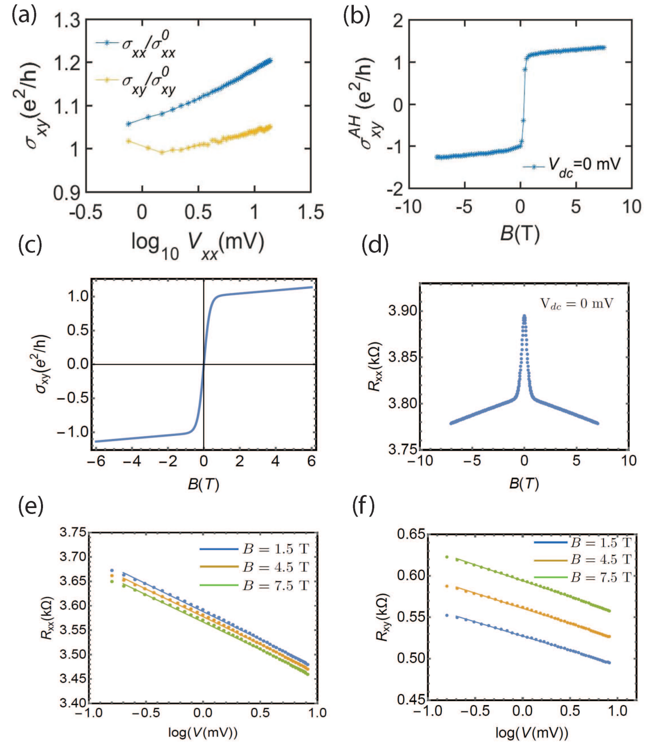

FIG. 6. (color online) Transport coefficients in the V-doped Hall bar (device H_1) as a function of bias. (a) Logarithmic plot for σ_{xx} (blue) and σ_{xy} (orange) at $B = 1$ T. (b) Magnetic field dependence of σ_{xy} at V_{dc}=0. (c),(d) Theoretical model of the magnetic field dependence of σ_{xy} and R_{xx}. (e),(f) Simulation of dependence of R_{xx} and R_{xy} on bias at B=1.5 T, 4.5 T and 7.5 T as expected from interaction corrections in disordered 2D films.

we obtain $\sigma_{xx}^0 \approx 9.17e^2/h$, $\delta\sigma_{xx} \approx 0.33e^2/h$, and $\sigma_{xy}^0 \approx 1.58e^2/h$, $\delta\sigma_{xy} \approx 0.02e^2/h$. Notice the smallness of the bias dependence of σ_{xy} compared to σ_{xx}. Therefore, considering that it is possible that small systematic errors can arise from mixing of R_{xx} and R_{xy} (e.g. if contacts are sligthly misaligned R_{xy} picks a small contribution from R_{xx}), we conclude that our data is consistent with σ_{xy} with negligible logarithmic bias dependence and while having significant logarithmic bias dependence on σ_{xx}, as expected from the AA theory. We observe, however, an interesting quantitative deviation from the expectation of the AA theory. Using the approximate value of $p \sim 2$, obtained by comparing the temperature and the voltage fits (see Fig. 5), the fitted parameter κ reads as:

$$\pi\kappa_{\rm fit} \sim 2. \quad (8)$$

However, from Eqs.(2)-(4), we expect $\pi\kappa \leq 1$, under the natural assumption of repulsive interactions $F > 0$. The origin of this discrepancy is at present unknown to us, but we wish to remind the reader that the equations of the AA we have employed were derived for parabolic electrons without Berry phase effects, and, it remains to be determined whether nontrivial orbital coherence, such as those giving rise to Berry curvatures for the bands of interest here, affect in any way the classic results of the AA theory.

A simple modeling of the resistivity can be done by using the expected conductivity behavior from the AA theory. The resistivity is taken to be of the form: $\sigma_{xx} = \sigma^0_{xx} + \delta\sigma^0_{xx}\log(|V| + V_0)$, where $V_0 \sim 0.4$ mV is essentially a cutoff of the logarithm at small bias (which is controlled by the temperature scale T_0 and the constant A in the Joule heating model), and σ^0_{xx} and $\delta\sigma^0_{xx}$ are field and bias independent quantities obtained by linear fitting of the logarithmic plots of the conductivity[41]. We add a simple description of the AHE in which the Hall conductivity has a jump of e^2/h near zero applied magnetic field in addition to the usual linear term reflecting the classical Hall effect. σ_{xy} in the model is presented in Fig. 6(c) and has the form: $\sigma_{xy} = \frac{e^2}{h}\tanh(B/B_0) + \delta\sigma^0_{xy}B$, where $B_0 \sim 0.3$ T reflects broadening of the jump of the magnetization as a function of field and $\delta\sigma^0_{xy}$ is field and bias independent. The model is able to reproduce the essential behavior of the resistivities and it is shown in Fig. 6(d) - Fig. 6(f).

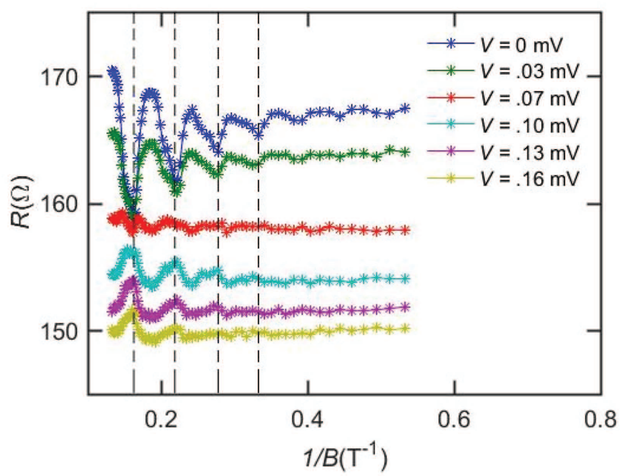

FIG. 7. (color online) Voltage bias dependence of quantum oscillations from magnetoresistance in V-doped $(Bi,Sb)_2Te_3$ device D_1 of width W=0.2 μm.

The transport results discussed above are for larger Hall bar (~ 20 μm) samples. Interestingly, when the device dimension was reduced to submicron range, prominent Shubnikov-de Haas (SdH) oscillations were observed. For example, in a 0.2 μm wide device (device D_1) measured by two terminal the oscillations were periodic in 1/B and have a non-trivial dependence on bias voltage. These quantum oscillations were seen in multiple samples with Ti/Nb/NbN and Ti/Al contacts. In particular, the zero bias minima turn into maxima in resistance at large voltage bias as shown in Fig. 7.

We also studied the effect of temperature on the magneto-oscillations as shown in Fig. 8. The ampli-

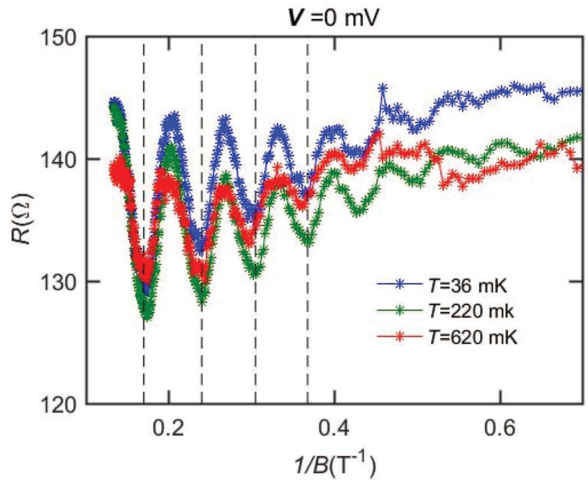

FIG. 8. (color online) Temperature dependence of quantum oscillations from magnetoresistance in V-doped $(Bi,Sb)_2Te_3$ device D_1 of width W=0.2 μm.

tude of the quantum oscillations is found to decrease with increase in temperature. However, the transition from maxima to minima could not be observed at temperatures accessible in the dilution fridge. The inferred electron density is 9×10^{11} cm^{-2} (4.5×10^{11} cm^{-2}) for spinful (spinless) Fermions. The period of the SdH could not be changed by applying a backgate or topgate. The screening of the top and bottom gates by the surface states results in inability to change the Fermi energy of the bulk states as has been observed in other topological materials[42]. An estimate of the electron gas mobility is made from the onset magnetic field of the SdH oscillations $\mu_q \approx \frac{1}{B_q} \approx 6,000$ cm^2 V^{-1}s^{-1} for the 200 nm wide device[43]. This mobility is intriguingly large compared to macroscopic samples.

We describe a transport model for the non-trivial dependence of the quantum oscillations on voltage bias that we have observed in the narrow junctions. We assume the magneto-conductance to be given by

$$g(B, V) = g_0 + \alpha\rho(\epsilon_F, B)\ln(\varepsilon\tau/\hbar) \quad (9)$$

where g_0 is assumed to be a constant background conduction and $\rho(\epsilon_F, B)$ is the SdH density of states given by

$$\rho(\epsilon_F, B) = \frac{2eB}{h}\sum_{j=0}^{j=\infty}\frac{1}{(2\pi)^{1/2}\Gamma}\exp(-\frac{(\epsilon_F - \epsilon_j)^2}{2\Gamma^2}) \quad (10)$$

where Γ is the half-width of Landau level broadening and ϵ_j is the single particle Landau level energy[44]. Presence of both bulk and surface conduction mechanisms in topological insulators has been found previously[45–50]. Fig. 9(a) and Fig. 9(b) display the magneto-transport data normalized to zero bias and zero magnetic field. Similarly, in the model, the resistance $r(B, V) = 1/g(B, V)$ is normalized to its value at zero bias voltage and magnetic field as is shown in Fig. 9(c) and Fig. 9(d) respectively.

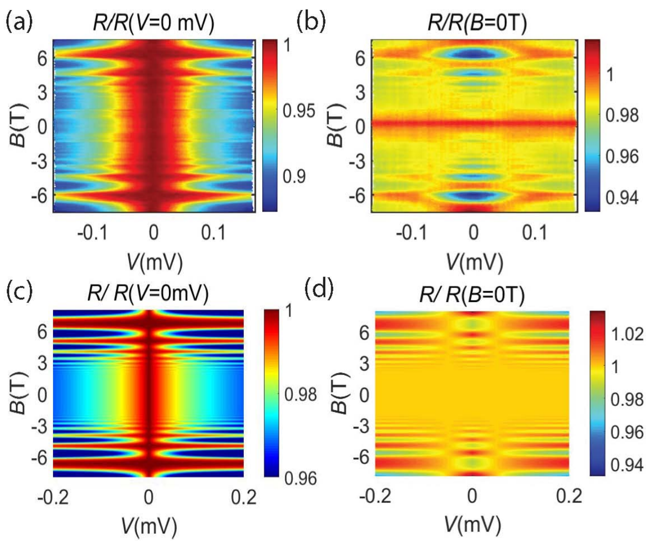

FIG. 9. (color online) (a),(b) The resistance in V-doped $(Bi,Sb)_2Te_3$ device D_1 is normalized to its zero bias value and normalized to its zero magnetic field value respectively. The evolution is studied with magnetic field and applied bias voltage. The SdH oscillations are present both at small and large bias voltages. However, the zero bias maxima become minima at large bias voltages at a fixed magnetic field and vice versa. (c) A model with two conduction mechanisms in parallel that incorporates logarithmic decay with applied bias of the SdH oscillations on top of a constant background conduction. The resistance is normalized to the zero bias value for comparison to the experimental data. (d) The two conduction model captures the evolution from maxima to minima of the resistance normalized to the zero magnetic field value with voltage bias.

While we do have an understanding of the voltage dependence of the oscillations, there are properties that are less well understood. The contrast of the quantum oscillations is found to decrease systematically with increasing width. Such dependence of visibility of quantum oscillations on channel width is unusual. The quantum oscillations are discussed in further detail in supplementary materials[41].

IV. CONCLUSION

We have studied magnetotransport in V-doped $(Bi,Sb)_2Te_3$ and find logarithmic singularities in longitudinal resistance R_{xx} and anomalous Hall resistance R_{xy}^{AH} which is well explained quantitatively by quantum corrections due to electron-electron interactions. In submicron scale devices, SdH oscillations are observed where the maxima transition to minima with voltage bias. A simple transport model explains these observations.

V. ACKNOWLEDGEMENTS

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