Coexistence of Surface and Bulk Ferromagnetism Mimics Skyrmion Hall Effect in a Topological Insulator

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Here, we report the investigation of the anomalous Hall effect in the magnetically doped topological insulator $(\text{V}, \text{Bi}, \text{Sb})_2\text{Te}_3$. We find it contains two contributions of opposite sign. Both components are found to depend differently on carrier density, leading to a sign inversion of the total anomalous Hall effect as a function of applied gate voltage. The two contributions are found to have different magnetization reversal fields, which in combination with a temperature dependent study points towards the coexistence of two ferromagnetic orders in the system. Moreover, we find that the sign of total anomalous Hall response of the system depends on the thickness and magnetic doping density of the magnetic layer. The thickness dependence suggests that the two ferromagnetic components originate from the surface and bulk of the magnetic topological insulator film. We believe that our observations provide insight into the magnetic behavior, and thus will contribute to an eventual understanding of the origin of magnetism in this material class. In addition, our data bear a striking resemblance to anomalous Hall signals often associated with skyrmion contributions. Our analysis provides a straightforward explanation for both the magnetic field dependence of the Hall signal and the observed change in sign without needing to invoke skyrmions, and thus suggest that caution is needed when making claims of effects from skyrmion phases.

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The anomalous Hall effect (AHE), despite being first reported over a hundred years ago, [1] remains of significant modern research interest as its investigation in novel magnetic materials continues to yield rich physics. Recent examples of this are the quantum anomalous Hall effect [2–12], which offers both potential for metrological applications [13,14] and for the academic study of axion electrodynamics [5,12,15,16], as well as reports on alleged contribution to the Hall effect [17–24] associated with skyrmion magnetic textures.

Our current investigation pertains to both of these observations, as it aims to both deepen our understanding of the nature of the magnetic state in materials where the quantum anomalous Hall effect has been reported, and shows that signals currently identified as a contribution from skyrmions are potentially being misinterpreted since, as we show here, the same signal can be more easily interpreted as simply resulting from the interplay between

two ferromagnetic contributions in a material which does not host skyrmions.

Our studies are carried out on layers of the magnetic topological insulator V-doped $(\text{Bi}, \text{Sb})_2\text{Te}_3$. Most works superficially describe its magnetic state as simply ferromagnetic, but some show indications of superparamagnetism [11], magnetically fluid behavior, or even claims of adiabatic cooling [10]. Moreover, a clear explanation of the origin of the strong perpendicular-to-plane magnetic anisotropy, and of the relative difference in the strength of this anisotropy for V vs Cr doping [9] is still lacking.

In this paper, we aim to shed more light on the nature of the magnetic state through a study of the AHE [25–30] in these materials. We find that it shows indications of a rich magnetic phase resulting from the interplay between two individual magnetic components.

All investigations are performed on samples with a standard Hall bar geometry prepared by optical lithography, and are equipped with an electrostatic gate that allows tuning of the Fermi level in the layers. The topological insulator heterostructures are $(\text{Bi}, \text{Sb})_2\text{Te}_3$ based, with the magnetic layers doped by V. They are grown using molecular beam epitaxy (MBE) on insulating, hydrogen passivated Si (111) substrates, and are covered *in situ* with

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10 nm of Te to protect the surface from aging [12,31]. The Bi/Sb ratio is 0.21/0.79 for all samples. More details on the growth and fabrication can be found in the Supplemental Material [32]. Measurements are done in basic ^4He magnetocryostats at temperatures ranging from 2.4 to 13 K and unless otherwise specified, at the applied gate voltage yielding the highest value of ρ_{xx} .

Our main investigations are focused on device *D1*, which was selected because it most clearly illustrates the rich nature of the AHE behavior in these materials, while at the same time showing a total anomalous Hall signal which is very reminiscent of the one associated with the topological skyrmion term. It comprises a 9-nm-thick TI layer, the bottom 2 nm of which contain 6.5% V doping. Figure 1(a) shows the AHE response of this device at a temperature of 2.4 K and with zero gate voltage applied. Its shape has significantly more features than expected from a simple ferromagnet, and as we will demonstrate, is the result of the coexistence of two ferromagnetic contributions in the sample.

Despite being produced by high-precision lithography, in any sample where the longitudinal resistance is larger than the Hall resistance, a small admixing of R_{xx} into the Hall signal is inevitable. This explains why the hysteresis loop in

FIG. 1. (a) Measurement of the Hall resistance as a function of H field for device *D1* (layer schematic on the right). The red arrow indicates the offset resulting from mixing in of the longitudinal resistance (see text for details). (b) Solid line: Same data after removing offset and classical Hall contributions as described in the text. Dashed line: Result of the two-component model used to describe the data. (c) Each of the two anomalous Hall contributions extracted from fitting the data. To the right of the figure are a picture of one of the devices, and a schematic describing how we model imperfections in the device (see text).

Fig. 1(a) is not vertically symmetric around the origin. To carefully analyze the Hall signal, we first remove this small additional contribution by treating admixing (which always results from the fact that the Hall voltage is not being picked up strictly orthogonal to the current flow) as an effective misalignment of the Hall leads, as illustrated (strongly exaggerated) in the sketch of Fig. 1. The pure Hall signal can then be obtained by subtracting away the correct fraction of the longitudinal voltage (conceptually $R_{xx} \Delta x/L$), where in our case $\Delta x/L$ is always smaller than 0.005. The resulting total Hall data contain not only the AHE, but also the classical Hall effect, which can be removed by subtracting the appropriate linear slope (a positive slope corresponds to n -type transport). The result of removing both the admixing and the classical Hall effect leaves us with the pure AHE contribution, which we label as R'_{xy} to distinguish from the measured values, and which is presented as the pale solid line in Fig. 1(b).

These corrections obviously change the quantitative details of the curve, but do not modify its overall qualitative appearance, which remains quite different from that expected from a simple ferromagnet. In fact, the curve can be decomposed into two contributions, each having the appearance of a simple AHE signal and having anomalous Hall coefficients of opposite signs. This decomposition is displayed in Fig. 1(c), where the blue and red lines show the two individual contributions, one positive (AH1) and the other negative (AH2). The sum of these two is shown as the dashed line overlaid to the data in Fig. 1(b). We adopt the convention that the negative anomalous Hall contribution is the one which has the same sign as the quantum anomalous Hall effect displayed by our samples when cooled to below 100 mK (see Supplemental Material [32]). We therefore refer to the contribution described by the red line in Fig. 1(c) as negative.

Because this device is gated, we can now study the evolution of these two components as we change the carrier concentration in the layer. In Fig. 2(a) we plot the $H = 0$, ρ_{xx} , and R'_{xy} value for the device as a function of gate voltage, showing that the device can be gated through its minimum conductivity point. In Fig. 2(b), we show R'_{xy} (the measured R_{xy} curves are given in Fig. S6 of the Supplemental Material [32]) as a function of magnetic field for different gate voltages. Interestingly, not only does the shape of the curve change, but even the overall sign of the AHE reverses as we go from large negative to large positive gate voltages. In all cases, the data can be modeled by decomposing it into two contributions as in Fig. 1(c). The agreement between the resulting fits and the data is illustrated in Fig. 2(e), where the upper panel is a color scale of the experimental R'_{xy} as a function of magnetic field and gate voltage, and the lower panel is the same plot, as generated by the model.

The strength of the two contributions determined by this fit is presented as the red and blue points in Fig. 2(c), with

FIG. 2. (a) Zero-field ρ_{xx} and R'_{xy} of our device as a function of gate voltage. (b) Magnetic field dependence of the offset corrected Hall resistance (R'_{xy}) as a function of magnetic field for various gate voltages. (c) Strength of each of the contributions extracted from the data. (d) The two normalized anomalous Hall conductivity components as a function of gate voltage compared to the ratio between electron and hole Drude conductivity extracted from the high field data (see Supplemental Material [32]). (e) Color plot confirming the agreement between the experimental data (top) and the description provided by our model (bottom).

the black points giving the sum of the two, and reproducing the fact that the sign of the overall AHE changes at around 6 V of applied gate voltage. The coercive fields of the positive and negative contributions, defined as the magnetic field where the contribution crosses zero, are 390 and 206 mT, respectively, and have no dependence on gate voltage. Using standard matrix inversion, these results can be converted to anomalous Hall conductivity contributions, yielding the red and blue points of Fig. 2(d). We compare these to the black points in the figure, which we get from fits of the high field classical Hall signal (see Supplemental Material [32] for details). It is noteworthy that while the negative anomalous Hall contribution is relatively independent of gate voltage, the positive one decreases quickly as electron conductivity begins to turn on.

Further evidence of the relative independence of these two contributions is provided by a temperature dependence study. The same analysis as in Fig. 2 is now conducted on the sample at temperatures ranging from 2.4 to 13 K, and the results are summarized in Fig. 3. In Figs. 3(a) and 3(b), we plot the amplitude of each contribution as a function of gate voltage, and for several temperatures. From this data, we plot in Fig. 3(c) the normalized amplitude vs T for a few

FIG. 3. (a),(b) Gate voltage dependence of the positive and negative contribution for measurements at various temperatures. (c) Temperature dependence of each of the (normalized) components, and (d) temperature dependence of the coercive field associated with each of the component. (The lines are guides to the eye.)

characteristic gate voltage values, and in Fig. 3(d), the coercive field of each component at a few temperatures, again showing the clearly different behavior of the two components.

Device *D1* was ideal for this detailed study because it has both negative and positive contributions to its AHE which are similar enough in amplitude yet different enough in coercive fields that both can be reliably extracted for the analysis, and also because it clearly highlights the similarities between our results and those attributed to skyrmions. A study of more and varied samples allows us additional insights, and indeed suggests that in our samples, the two ferromagnetic states can be associated with surface and bulk contributions. To verify this, we have examined a large set of samples where the thickness and composition of the magnetic layer is varied. A sampling of these results is presented in Fig. 4 (with the results for still more samples provided in the Supplemental Material [32]).

As seen in Fig. 4(a), while some devices (*D4*) show both contributions as did *D1*, other samples have anomalous Hall signal which are dominated by a single contribution. Even there, however, the situation is nontrivial, as the AHE can be either positive (*D2*) or negative (*D3*) depending on the properties of the individual layer. As is evident from the summary of the results given in Figs. 4(b)–4(d), it appears that the positive contribution is driven from very thin and highly magnetically doped layers.

For a constant high magnetic doping level, positive total AHE is observed only when the magnetically doped film thickness is sufficiently small. When thickness is manipulated under a constant magnetic doping concentration [see Fig. 4(d)], the amount of magnetic dopants on the film

FIG. 4. (a) Magnetic field dependence of the offset corrected Hall resistance (R'_{xy}) for devices D2–D4. (b) Magnetic doping per thickness phase space with position of the devices D2–D8, including the position where the quantum anomalous Hall effect was observed in this material system (device D8, see Supplemental Material [32]). (c), (d) Offset corrected zero field value of the Hall resistance as a function of magnetic doping level (c) and magnetic layer thickness (d) for the positively magnetized state. A schematic layer layout for devices D2–D8 is presented at the top of the figure.

surface remains unaffected (the two main surfaces are only shifted with respect to each other), whereas the amount of magnetic dopants in the bulk grows as the two surfaces shift away from each other. Based on that we suggest that the positive contribution originates on the surface of the magnetically doped layer, and the negative one in the bulk. This assignment is also supported by the fact that the positive contribution is much more sensitive to gate voltage than the negative one. Moreover, the different coercive fields and temperature dependence of each component indicate separate ferromagnetic origin, pointing towards the coexistence of surface and bulk ferromagnetism. It is not surprising that different magnetic and charge environments (especially with regard to a topological surface state) for the magnetic dopants on the surface and in the bulk would result in different magnetic interactions leading to the observed coexistence of two magnetic orders. The exact microscopic origin of the distinct surface and bulk magnetism remains an open question, and one that, when answered, may provide deep insight as to the underlying nature of the, still badly understood, magnetism in this material.

We note that all published transport results comprising similar phenomenology [17–24] could be well explained using our model introducing two coexisting ferromagnetic orders. Moreover, our explanation successfully describes every electrical transport feature, including a dependence on magnetic field, gate voltage, temperature, and thin film parameters such as doping level and film thickness, without

the need to speculate about new topological physics. While our analysis cannot explicitly rule out a skyrmion origin to the observed transport features in some cases, it certainly provides the simplest explanation yet, and implies that extreme caution should be used when making claims of effects from skyrmion phases.

Conclusions.—We have investigated the magnetotransport properties of magnetically doped (V,Bi,Sb)₂Te₃ topological insulator structures. A rich anomalous Hall response exhibiting sign reversal as a function of magnetic layer thickness, magnetic doping density, and applied gate voltage was observed. All transport features can be well explained by the coexistence of two ferromagnetic orders in the system. Moreover, the analysis suggests that one of the ferromagnetic orders originates on the surface while the other in the bulk of the magnetic topological insulator film. Our work provides yet another valuable piece to the puzzle of the origin of magnetism in this material system, and we believe will contribute to a final understanding of physics underlying it. Moreover, the characteristic shape of the anomalous Hall signal in the case where the two contributions are comparable in amplitude, as well as the observed change of overall sign of the signal as a function of parameters such as gate voltage and temperature comprise an identical phenomenology to that commonly attributed to skyrmion states. Our findings therefore suggest the necessity for reexamination of these earlier claims.

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