

Investigating “kagome-derived” phenomena in the RT_6X_6 family

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Abstract

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Materials containing kagome lattices have gained substantial attention in part since their band structures are predicted to simultaneously host Dirac points, van Hove singularities, and flat bands. In this thesis I investigate the consequences (or lack thereof) of the first two of these features on a couple of materials in the RT_6X_6 kagome family. First, I will discuss the ferrimagnet $TbMn_6Sn_6$ which displays a large intrinsic anomalous Hall effect (AHE) previously attributed to gapped Dirac points in the band structure. Through tuning this material with Cr doping, we reveal key issues with this interpretation, finding instead that the large intrinsic AHE can be attributed to multiple Berry curvature “hot spots” that exist throughout the Brillouin zone. Coincidentally, this doping series leads to magnetic compensation that is highlighted by record-breaking coercive fields, offering a system with a large intrinsic AHE coupled with nearly vanishing magnetization. After this, I will focus for the rest of the thesis on my work on the charge density wave (CDW) system ScV_6Sn_6 . The first result comes from careful measurements of the magnetotransport of this system; these data and accompanying analysis provide strong evidence for pseudogap behavior in the “normal” state of this compound in which short-range CDW fluctuations open up a partial electronic gap. Then I will discuss my work carefully optimizing the crystal growth of ScV_6Sn_6 which allowed me to obtain crystals with a far better quality than those reported in literature. Magnetotransport measurements of these crystals reveal a lack of evidence of any AHE (which was originally reported) and a complicated series of apparent phase transitions near the nominal T_{CDW} . Lastly, I will present recent angle-dependent magnetoresistance measurements these high quality samples of ScV_6Sn_6 . While electrical transport on RT_6X_6

materials often is dominated by small, light 3D Dirac-like pockets, our high-quality single crystals allow us to detect the transport contribution of a large quasi-2D vHs pocket near the Fermi energy at low temperatures. These results indicate the transport properties of this nominally 3D stannide maintains some connection to the 2D kagome layers it contains.

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My journey to and through this PhD has been extremely rewarding, but also very challenging and I have many people to thank for helping me accomplish this goal. Through high school I had always been broadly interested in science. As an undergrad at the University of Florida I started as an “undeclared engineering” major with the intention of getting a PhD but having the option to get a “real” job upon graduating. However, in my physics 1 course Prof. James Hamlin discussed his research on pressure induced superconductivity and I was hooked on the beauty (and lack thereof) of experimental condensed matter physics. My sophomore year I joined Prof. Amlan Biswas’ lab where I studied thin film manganites via magnetotransport measurements. Amlan was a wonderful boss; he was constantly encouraging and helping me despite my youth and inexperience. After a year in this lab, I joined James’ lab where I fell in love with materials synthesis. James’ careful approach to science (extraordinarily well documented due to his feud with the now disgraced Ranga Dias) continues to be a major influence on my research. Lastly, during the peak of Covid in summer 2020 I worked remotely with Dr. Lin-Lin Wang at Ames National Laboratory. Despite never mentoring a student before (and me having only taken 1 semester of quantum mechanics), Lin-Lin somehow had me (kind of) understanding and performing basic density functional theory calculations by the end of the 10-week program. Honestly speaking, this is what truly convinced me that I could “do science” and reinforced my notion that I should never do research that requires a lot of knowledge about computers.

This broad experience with condensed matter physics made me realize that I wanted to get a PhD in this field that focused on materials synthesis and some sort of “non-traditional” technique. After speaking with Prof. Jiun-Haw Chu while searching for a graduate school, I was convinced to come to UW to work for him due to his very clear passion and huge breadth of knowledge. He has been incredibly supportive throughout my PhD and has given me far too much freedom to “hack through the jungle” in a much less structured PhD than many of his other students who have focused on one or two main ideas/topics. As one example, I will always be thankful for him allowing me to move across the country to work at Penn State on nanodevices for an entire summer. His clear vision of what needs to be done next (both experimentally and in terms of analysis) is always inspiring. I hope we can continue to work together in the future.

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them, and more recently each of these couples has had their first child (CJ and Macy). It is very cool (and more than a little strange) to see the DeStefano family grow, and I am very excited for the future.

Lastly, I'm extremely thankful for my wonderful partner, Emma Steinebronn. She has been my rock throughout our nearly 5 years together while she was getting her PhD at Penn State. She is one of the kindest and most caring people I have ever met. Our trips across the country to visit each other (and the summer I spent at Penn State) were always filled with joy whether we were hiking or going to trivia nights together. Despite being long distance most of our relationship, we somehow always managed to have fun just playing video games or watching movies together, and I felt an incredible bond to her the entire time. I'm over the moon that we will soon be living together in upstate New York; I genuinely cannot wait to be together.

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1 Introduction

This chapter presents a broad overview of band theory in order to introduce the modern interest in materials containing kagome lattices. Namely, there are three main features that arise in the ideal band structure of these materials that have caught the attention of physicists: flat bands, van Hove singularities (vHs), and Dirac points. This thesis investigates the impacts (or lack thereof) of Dirac points and van Hove singularities on the emergent properties in several members of the RT_6X_6 family which will also be discussed in this chapter. Due to the variety of motivations for the different chapters in this thesis, each subsequent chapter will have an introduction that builds upon this initial, brief introduction.

1.1 Band Theory

Materials have been essential to the development of human society to the point that different times in history are sometimes referred to by the material humans were primarily interested in at that time (ex. the Bronze Age). Despite this long-standing tradition, we did not have an accurate microscopic picture for why materials act the way they do until the advent of quantum mechanics in the early 20th century. Quantum mechanics allowed for an understanding of systems on the scale of atoms. One of the primary realizations comes from the fact that quantum systems cannot take on arbitrary values for measured quantities; for example, the electron energy levels of a free atom is quantized to discrete values. Upon bringing atoms close together, these energy levels will shift as the atoms interact with one another. In the specific case of crystalline solids, the periodic potential of the lattice provides a term to the Hamiltonian of the system; thus, there are only specific allowed electron energies for a given crystalline compound. These allowed energies (plotted as a function of momentum as with any dispersion) constitute a band structure.

In the independent electron approximation, the band structure of a material starts from just the Hamiltonian for a single electron:

$$\hat{H} = \frac{\hat{p}^2}{2m} + V(\mathbf{r}) \quad (1.1)$$

where $V(\mathbf{r} + \mathbf{a}) = V(\mathbf{r})$ is a periodic potential with $\mathbf{a}$ corresponding to a Bravais lattice vector. It was shown by Felix Bloch [1] that solutions of the Schrödinger equation for a periodic potential like this must be of the special form

$$\psi_{n\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{n\mathbf{k}} \quad (1.2)$$

where $\mathbf{k}$ is referred to as the wave vector. For each value of $\mathbf{k}$, there are multiple solutions of the Schrödinger equation. n (the band number) refers to each of these solutions, giving an $E_n(\mathbf{k})$ which gives the dispersion relation for electrons in that band. Note that in principle $\mathbf{k}$ can take on any value, but there is only a subset of $\mathbf{k}$ that is relevant here which exist in the Brillouin zone. Any other $\mathbf{k}$ can be mapped back into the Brillouin zone using reciprocal lattice vectors, meaning that they are not physically different states (the reciprocal lattice is periodic just as the real space lattice is periodic). In practice, calculating the band structure for a real material is reasonably computationally intensive and is nowadays often performed using density functional theory (DFT) calculations. However, some basic techniques can be used to get a qualitative feel for a band structure in simple cases.

One such technique is known as the tight-binding model. A basic understanding of this toy model can be gained by starting from the basic case of bringing two hydrogen atoms together. When they are infinitely far away from one another, they will each have an electron in the $1s$ ground state with wave functions ψ_A and ψ_B . When these atoms are brought together, their wave functions will overlap and split into two separated energy levels for each level of a single isolated atom (namely $\psi_A + \psi_B$ and $\psi_A - \psi_B$). This idea holds in general for groups of N atoms where N orbitals are formed for each orbital of the isolated atoms. As these atoms are brought together, the orbitals from different atoms will overlap and spread into energy bands.

In order to get a deeper understanding of this toy model, I will delve briefly into some of the formalism here following the notation of Kittel [2]. We will assume that the ground state of an s electron in the potential $U(\mathbf{r})$ of an isolated atom is $\phi(\mathbf{r})$ and that the influence of the atoms as we bring them close together will be small. We can then write the wavefunction for one electron in the crystal in the basis of these atomic orbitals, namely

$$\psi_{\mathbf{k}}(\mathbf{r}) = \sum_j C_{\mathbf{k}j} \phi(\mathbf{r} - \mathbf{r}_j) \quad (1.3)$$

with j running over all of the lattice points. In order for this to be a Bloch wave function, it ends up that $C_{\mathbf{k}j} = N^{-1/2} e^{i\mathbf{k}\cdot\mathbf{r}_j}$. To find the first-order energy of this system, we will take the expectation value of the Hamiltonian:

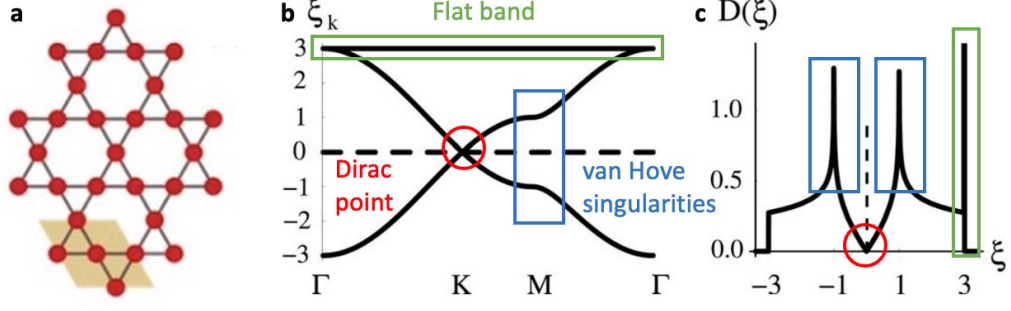


Figure 1.1: **Kagome lattice and tight-binding model band structure [3].** **a**, The kagome lattice with the unit cell shaded. **b**, Tight-binding model band structure of a two-dimensional kagome lattice. **c**, Corresponding tight-binding model density of states.

$$\varepsilon_{\mathbf{k}} = \langle \mathbf{k} | H | \mathbf{k} \rangle = \sum_m e^{-i\mathbf{k} \cdot \boldsymbol{\rho}_m} \int dV \phi^*(\mathbf{r} - \boldsymbol{\rho}_m) H \phi(\mathbf{r}) \quad (1.4)$$

with $\phi_i \equiv \phi(\mathbf{r} - \mathbf{r}_i)$ for all i and $\boldsymbol{\rho}_m \equiv \mathbf{r}_m - \mathbf{r}_j$. We will consider only on-site ($\boldsymbol{\rho}_m = 0$) and nearest neighbor (connected by $\boldsymbol{\rho}$) terms. In turn, this equation can be written in two parts using the following shorthand notation:

$$\int dV \phi^*(\mathbf{r}) H \phi(\mathbf{r}) = -\alpha; \int dV \phi^*(\mathbf{r} - \boldsymbol{\rho}) H \phi(\mathbf{r}) = -\gamma \quad (1.5)$$

leading to

$$\varepsilon_{\mathbf{k}} = -\alpha - \gamma \sum_m e^{-i\mathbf{k} \cdot \boldsymbol{\rho}_m} \quad (1.6)$$

with the α term corresponding to an atomic energy shift due to the neighboring atoms and the γ term describing the dispersion of the bands coming from the interaction of the nearest neighbor atoms. With this formalism, a qualitative picture of a band structure can be obtained without any need for caring about specific details of the system. While this makes this technique useful, it also has significant downsides. For example, note that here we only considered s orbitals; higher angular momentum orbitals are not as simple to deal with. This issue (as well as several others) and their applications to real materials will be discussed in the context of kagome materials below.

1.2 Kagome Lattice

The kagome lattice (shown in Fig. 1.1a) has gained significant attention in the past decade in large part due to the extremely rich tight-binding model band structure calculated for a two

monolayer with this crystal structure (shown along with the corresponding density of states in Fig. 1.1b and c) [3]. As marked in the band structure, this calculation reveals a Dirac point at the K point, vHs (saddle points in the band structure) at the M point, and a flat band that stretches across the entirety of the Brillouin zone. As outlined below, the coexistence of all three of these features indicates that kagome materials may be able to host both correlated and topological physics. Notably, this calculation does not rely on the microscopics of a given system beyond the fact that it contains a kagome lattice; thus, the band filling of a real material could in principle be tuned to bring any of these features to the Fermi energy (E_F) where they should dominate transport and thermodynamic responses.

Flat Bands and vHs

Flat bands and vHs are (at a basic level) somewhat similar — while a flat band is just that (flat), a vHs is a local “flattish” part of the band structure (more specifically it is a saddle point in the band structure). This flatness is related directly to the density of states; a very flat band means that there are many states to fill at a particular energy. On a grand scale, this is very interesting to physicists because if this large density of states happens to lie near E_F the system is primed to undergo a symmetry-breaking phase transition (ex. superconductivity, magnetism, charge density wave, etc.). This transition will break this large degeneracy of states at E_F (shifting some of the states down in energy), lowering the total energy of the system. As a specific example of this phenomenon, the Bardeen-Cooper-Schrieffer theory of superconductivity predicts that for a given conventional superconductor, the critical temperature (T_c) should be related to the density of states at E_F ($N(0)$) through the relationship $T_c \propto e^{-1/N(0)}$ [4]. Importantly in the context of this thesis, a flat band or vHs indicates a real space confinement of carriers (through the relation $v \propto \frac{dE}{dk}$) and high effective mass of the carriers (through $m^* \propto \frac{1}{d^2E/dk^2}$) [2]. This high effective mass will be important to the contents of Chapter 5.

Dirac Points

Dirac points are in some ways the antithesis to flat bands. This band structure feature consists of two (two-fold degenerate) bands crossing at some point in momentum space in a linear fashion. This linear dispersion is analogous to the dispersion relation of light, and it leads to extremely low effective masses and high mobilities. The poster child of Dirac

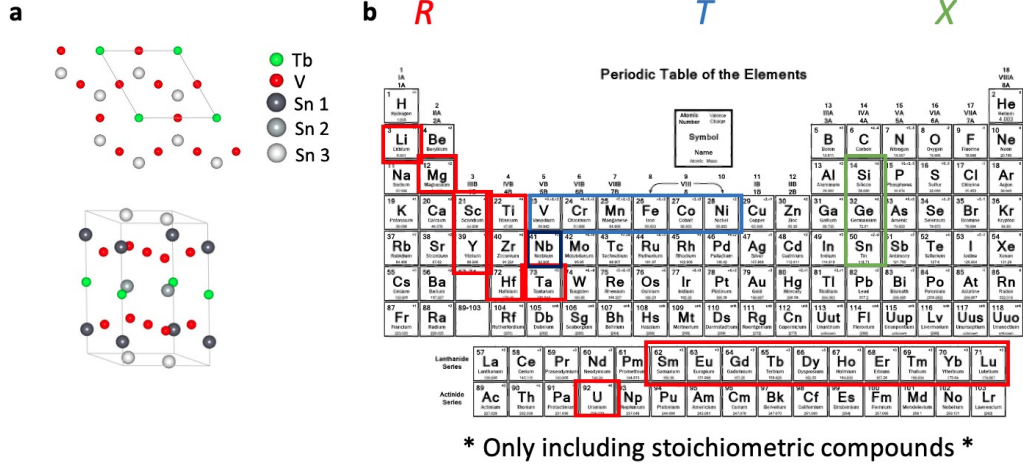


Figure 1.2: **RT_6X_6 crystal structure and compounds.** **a**, The RT_6X_6 crystal structure using TbV_6Sn_6 as an example [6]. **b**, A periodic table color coded by the different R , T , and X elements that have been used to form stoichiometric RT_6X_6 compounds.

semimetals is monolayer graphene in which the band structure hosts a very prominent isolated Dirac point [5]. However, in bulk metals (like the RT_6X_6 materials that are the focus of this thesis as discussed in the next section) typically many other (often times trivial) bands coexist with these Dirac bands. Due to the extremely low density of states coming from the Dirac point, these trivial bands will often dominate the thermodynamical and magnetotransport responses of these materials. This idea will be vital in the understanding of Chapter 3.

1.3 The RT_6X_6 Family

With these ideas in mind, we can now turn to the realization of real kagome materials. While the zoo of kagome materials has expanded immensely over the past few years (see Ref. [7] for a reasonably comprehensive list), I will focus here on the RT_6X_6 family (R = rare earth, T = transition metal, X = Si, Ge, or Sn). These materials form with kagome layers comprised solely of the T ion in the ab -plane which are spaced along the c -axis by R and X layers as can be seen in Fig. 1.2a using the example of TbV_6Sn_6 [6]. The chemical variety available for this family is massive as shown in panel b of this figure which shows a periodic table which is color coded by the different R , T , and X elements that have been used to form stoichiometric RT_6X_6 compounds. Interestingly, many of these compounds were synthesized and characterized decades ago (without the modern context of the kagome band structure) due to the idea of magnetic geometric frustration that often arises in crystal

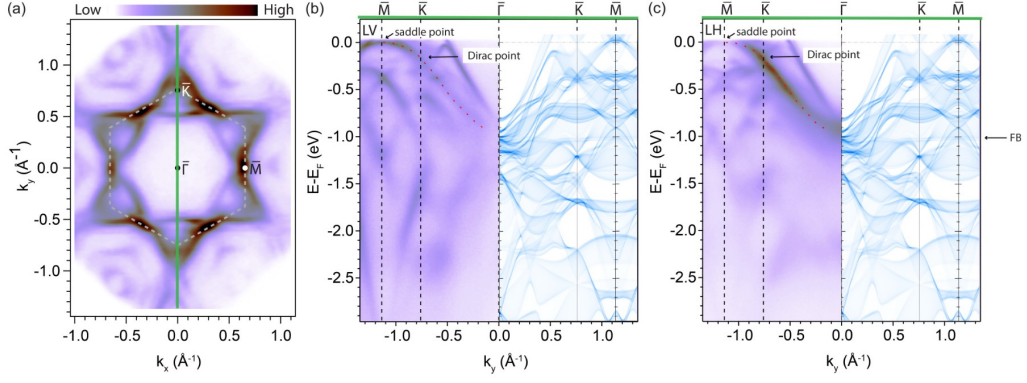


Figure 1.3: **Electronic structure of the kagome termination of TbV_6Sn_6 [6].** **a**, The Fermi surface of TbV_6Sn_6 measured with ARPES. The white dashed line denotes the first Brillouin zone. **b and c**, $\bar{\Gamma} - \bar{K} - \bar{M}$ cut measured with ARPES (left hand side of panels) compared with DFT calculations (right hand side of panels). **b** is measured with linear vertical polarization, **c** is measured with linear horizontal polarization.

structures with triangular motifs (see, for example, Refs. [8, 9, 10, 11, 12]). Due to the f and d electrons coming from the R and T sites, many different (and often complex) magnetic states were found in these RT_6X_6 materials (a brief review on magnetism will be presented in Chapter 3). These materials came back into the spotlight in the 2020s in large part due to Ref. [13] which pointed out that these varied magnetic states could be used to tune the interesting band structure features arising from the kagome lattice. Furthermore, the chemical variety available in this family allows for careful tuning of E_F . Thus, the RT_6X_6 family seems to be an ideal set of systems to study kagome physics.

However, as with any real material, the story is never as simple as it seems. While DFT calculations and angle-resolved photoemission spectroscopy (ARPES) experiments often reveal the expected kagome features in the band structure of these materials (for an example, see Fig. 1.3 which was adapted from Ref. [6]. Note the Dirac points near K and the saddle points (νHs) near M as well as a flattish band.), many other bands complicate the situation. This is not to be unexpected; remember that the ideal kagome band dispersion shown above was calculated with a nearest-neighbor tight-binding model calculation for the specific situation of a monolayer kagome layer with only s orbitals. The situation here is obviously different. First, the RT_6X_6 family has the R and X spacer layers in between T layers. While the T layers are physically separated by these layers, there are still two of them in the unit cell, leading to copies of the kagome band structure features. Furthermore, the

chemical bonding in these materials is quite three dimensional, causing each layer to couple and hybridize with each other. As a result, the expected kagome features are substantially distorted and smeared, and many trivial bands complicate the band structure. These bands play a significant role in resultant properties of these materials, making it difficult to parse which parts of the responses are really due to the kagome band structure and which are not.

This thesis is precisely focused on this topic. In Chapter 3 I will critically analyze anomalous Hall measurements initially attributed to a gapped Dirac point in TbMn_6Sn_6 , proving conclusively through Cr doping this material that these results are actually due to trivial bands located near E_F . The Cr-doped materials also coincidentally show a number of other interesting features including (to our knowledge) the highest coercive field ever recorded for a single crystal. In Chapter 4 I will present magnetotransport measurements on the charge density wave material ScV_6Sn_6 revealing pseudogap-like behavior stemming from charge density wave fluctuations in the normal state. These results are the backbone of Chapter 5 in which I will show how to greatly increase the crystal quality of ScV_6Sn_6 compared to literature. Using these improved quality crystals I was able to measure the angle-dependent magnetoresistance response of ScV_6Sn_6 revealing a distinct switching from magnetotransport dominated by small, Dirac-like pockets at high temperatures to magnetotransport dominated by a large pocket associated with a vHs near the E_F at low temperatures.

2 Experimental Methods

2.1 Crystal Growth

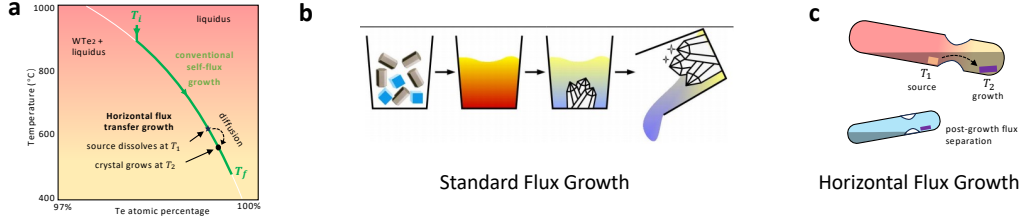


Figure 2.1: **Growth methods.** **a**, A small section of the W-Te binary phase diagram with growth paths of the standard flux growth and horizontal flux growth methods. **b**, A schematic showing the basic steps of a standard flux growth. Image credit to Paul Malinowski. **c**, A schematic showing the basic steps of a horizontal flux growth. Image credit to Chaowei Hu.

As a member of a crystal growth group, the first step in measuring a sample is making the sample. The single crystals of $RT_6\text{Sn}_6$ used for the results in this thesis were all grown using a standard flux method as has been previously described in literature [14, 13, 15]. In these cases, an excess of Sn was mixed with the other constituent elements (often in Canfield crucible sets [16]), then sealed in quartz tubes under vacuum. Upon warming to high temperatures, the molten Sn dissolves the other elements. If the growth is in the proper part of composition/temperature phase space, upon slow cooling a supersaturated solution will form, and single crystals will slowly precipitate out of melt. Fig. 2.1a shows a small section of the W-Te binary phase diagram. The green line on the liquidus shows the path that the liquid will take through phase space as crystals form (this process makes the remaining liquid increasingly Sn rich). Upon reaching a final temperature that is still above the melting point of Sn, the growth is removed from the furnace and centrifuged. This removes the excess molten metal from the single crystals, allowing them to be harvested. This process is shown schematically in Fig. 2.1b. The specifics of how the crystals used in each chapter were grown can be found in the introductions of the chapters. Notably, Chapter 5 describes my work (along with the assistance of Chaowei Hu) to grow high quality single crystals of ScV_6Sn_6 and it includes some tips and tricks we found useful in growing high quality intermetallics.

A small part of my time in Jiun-Haw's lab was spent working with Chaowei Hu on

the horizontal flux growth method he was refining to grow extremely pure transition metal dichalcogenides (TMDs). Note that this work was primarily headed by him. The original description of this technique can be found in Ref. [17] and our updated technique is outlined in Ref. [18]. This technique is a play on the traditional flux growth method described above, but instead of relying on a changing temperature to run the reaction, a clever trick allows the growth to occur at the same temperature the entire time. In short, a temperature gradient is applied to a tilted sealed quartz ampoule with a neck in it. The low (cold) side of the ampoule begins full of the solvent (the chalcogenide), and the high (hot) side of the ampoule contains more solvent and a chunk of the metal (in the case of WTe_2 we usually use mass ratios of W:Te of about 1:50 with about 50 grams total of reagents). Upon heating to a modest temperature, the chalcogenide melts and gradually incorporates some of the metal. Note that not all the metal can be incorporated at once at the selected position in composition-temperature phase space. This dissolved metal diffuses to the cold side of the ampoule where single crystals of the desired TMD begin to form due to the smaller amount of metal that can be incorporated in this colder side of the tube. Now more metal can be dissolved on the hot side of the ampoule, leading to a continuous process of the metal source dissolving on the hot side and single crystals forming on the cold side. This type of growth can lead to extremely clean crystals as it is performed at constant “low” temperatures. Upon growth completion, the tilt angle and temperature profile are reversed to remove the flux from the crystals. This process is shown schematically in phase space in Fig. 2.1a and a sketch of the growth process is shown in panel c. As of now we have been unable to grow anything except binary compounds with this method. This is due to the extremely complex composition-temperature phase spaces that begin to form when more than two reagents are added. For example, multiple attempts to grow ScV_6Sn_6 with this method led only to crystals of VSn_2 . While it is possible to grow more complex materials with this method (for example, YFe_2Ge_2 single crystals have been grown in this way [19]), in order to grow new materials this process requires significant trial and error that is hindered by the long growth times this growth needs (often > 1 month).

Once crystals have been grown, they are characterized by X-ray diffraction (revealing the lattice constants via Bragg’s law) using a Rigaku MiniFlex 600 system with a Cu source and Hy-Pix 400MF 2D detector and energy dispersive X-ray spectroscopy (revealing the chemical constituents) using a Sirion XL30 scanning electron microscope. When determin-

ing the doping levels of crystals, each crystal is polished before the measurement to ensure a reasonably flat surface and at least eight data points were taken in different parts of the sample to ensure chemical homogeneity.

2.2 Electrical Transport Measurements

Samples were polished and cut by a wire saw to be bar-shaped with dimensions typically about $1\text{ mm} \times 0.5\text{ mm} \times 0.1\text{ mm}$. Silver paste and gold wires were used to 5-point (Hall pattern) measurements. The measurements were performed in a Quantum Design Dynacool Physical Property Measurement System (PPMS) with standard lock-in techniques in temperatures ranging from 1.7 to 400 K and in magnetic fields up to 14 T. To eliminate any contributions from contact misalignment, the in-line and Hall resistivities were symmetrized and anti-symmetrized, respectively. For some of the measurements, the samples were rotated in situ using a Quantum Design in-plane rotator. When measuring small resistance samples like the ones presented here (typically $< 0.1\Omega$), it was found that special care should be taken when doing these measurements as they can be quite noisy. The length of wires connecting the puck to the sample should be minimized, and any wire loops should be minimized in size. Measurements taken while actively rotating were often found to be very noisy even at quite low rotation speeds (ex. $0.5^\circ/\text{s}$). I found that rotating a small amount, then waiting and collecting data for a few seconds at the next position before rotating a small amount to the next target angle got much higher quality data (although it takes a lot of time!). Often, measuring only every 2 degrees or so gave enough data points to reveal if more data points are needed in a certain angle range.

2.3 Magnetization Measurements

Magnetization measurements presented in this thesis were performed with the vibrating sample magnetometer option of the PPMS. Samples of the same size as the transport samples were attached to a quartz paddle with GE varnish such that the magnetic field was either out-of-plane or in-plane. UW recently purchased a Quantum Design Magnetic Property Measurement System 3 (MPMS) through MEM-C which has a much higher signal to noise ratio than the PPMS. I highly recommend using the MPMS when measuring samples with very small magnetic moments.

3 Anomalous Hall Effect in $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$

3.1 Background and Motivation

The ferrimagnet TbMn_6Sn_6 displays a large intrinsic anomalous Hall effect (AHE) that was originally attributed to a gapped Dirac point at the K point stemming from the kagome band lattice. Through tuning this material with Cr doping, we reveal key issues with this interpretation, finding instead that the large intrinsic AHE is associated with multiple Berry curvature “hot spots” that exist through the entire Brillouin zone. Coincidentally, this doping series leads to magnetic compensation that is highlighted by record-breaking coercive fields, offering a unique system with a large intrinsic AHE coupled with nearly vanishing magnetization. This chapter is based on work previously published in Ref. [20].

Magnetic Order

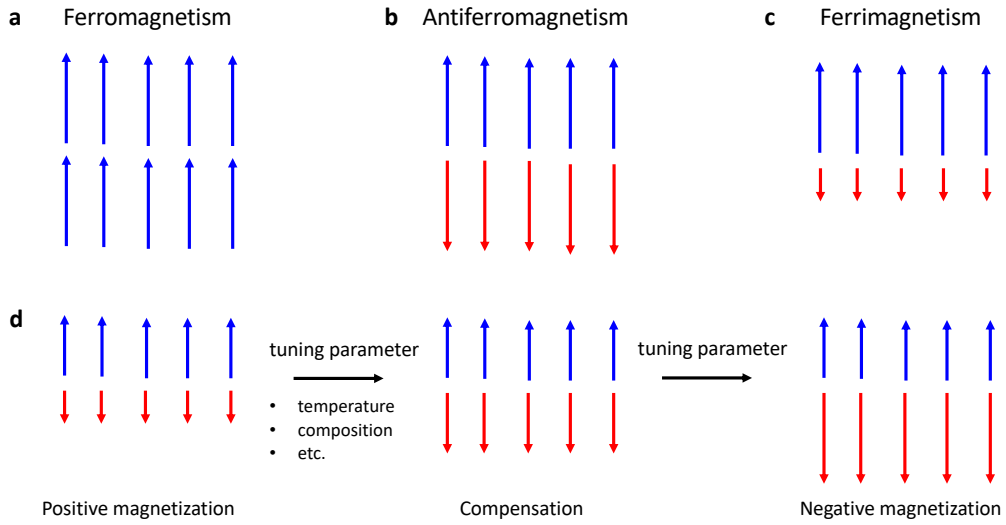


Figure 3.1: **Magnetism basics.** **a, b, and c,** Schematics showing the spin arrangements of simple ferromagnetism, antiferromagnetism, and ferrimagnetism. **d,** Schematic showing the evolution of spin arrangements of a ferrimagnet with a tuning parameter. Depending on the size of the tuning parameter, a positive, compensated, or negative magnetization state can be observed.

Much of this introduction on basic magnetic orders is based on Ref. [21]. At sufficiently high temperatures, materials with unpaired electrons are paramagnets. In the paramagnetic state these unpaired electrons act independently of each other, randomly fluctuating due

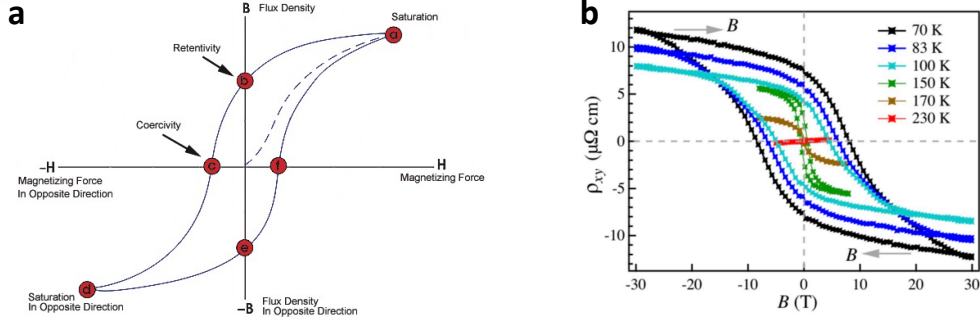


Figure 3.2: **Hysteresis loops for ferromagnets.** **a**, Schematic hysteresis loop for a ferromagnet when measuring $M(\mu_0 H)$. **b**, Example hysteresis loop for a ferromagnet when measuring $\rho_{xy}(\mu_0 H)$.

to the thermal energy of the system. Upon cooling, this thermal energy will decrease and other energy scales (here the magnetic energy scale) will become relevant. In some cases a magnetically ordered state can emerge where the angular momentum of the electrons in the system work together to form a long range symmetry breaking state.

To look at this slightly more rigorously, we can consider the simple case of a ferromagnet, a state in which there is a spontaneous parallel alignment of magnetic moments even in the absence of a magnetic field (see Fig. 3.1a). The magnetic moment of an atom is proportional to its total angular momentum $\mathbf{J}$ where

$$\mathbf{J} = \mathbf{L} + \mathbf{S} \quad (3.1)$$

with $\mathbf{L}$ and $\mathbf{S}$ referring to the orbital angular momentum and the spin angular momentum. An increase in applied magnetic field will align the moments while increasing temperature disorders them. For simplicity, we will ignore $\mathbf{L}$. In the presence of an applied field $\mathbf{B}$, the Hamiltonian of the magnetic part of the system will be of the form

$$\hat{H} = -\sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + g\mu_B \sum_j \mathbf{S}_j \cdot \mathbf{B} \quad (3.2)$$

where J_{ij} is the exchange constant for nearest neighbors (positive for ferromagnets), g is the Landé factor and μ_B is the Bohr magneton. The first term on the right is the Heisenberg exchange energy and relates how neighboring atoms influence each other, and the second term on the right is the Zeeman energy. Note that the actual magnetization is in the opposite direction of $\mathbf{S}$, hence the Zeeman energy term tends to align moments with the field. In a

ferromagnet the moments stay ordered even at zero field because of the coupling between nearby ions. In fact, if one is to measure $M(\mu_0 H)$, a finite hysteresis loop will emerge because a finite magnetic field in the opposite direction as the spins is necessary to force them to flip as shown in Fig. 3.2a. Furthermore, for some ferromagnets, an anomalous Hall effect (AHE) can be measured which will show up as a similar hysteresis loop when measuring $\rho_{xy}(\mu_0 H)$ as seen in Fig. 3.2b. This will be discussed more in depth in the next subsection.

Beyond ferromagnetism, the next simplest magnetically ordered state is antiferromagnetism. Here neighboring spins are instead arranged in an antiparallel moment (shown schematically in Fig. 3.1b) leading to no net magnetic moment at zero field. These materials can be described by the same Hamiltonian as shown in Eqn. 3.2 although in this case J_{ij} is negative. Note that with a large enough magnetic field it will eventually become energetically favorable for the spins pointing anti-parallel to the field to flip. Depending on the exact microscopic details, these metamagnetic transitions can be classified in various ways including spin-flop and spin-flip transitions.

The relevant magnetic ordering for this chapter is ferrimagnetism which acts in some way as a bridge between ferromagnetism and antiferromagnetism. In the simplest case of this ordered state, half of the spins point antiparallel to the other half of the spins like an antiferromagnet. However, the two spin sublattices have different moment sizes (shown schematically in Fig. 3.1c), leading to a net magnetic moment like a ferromagnet. Thus, the macroscopic ferromagnetic behaviors discussed above can still be observed despite the difference in microscopic details between these two cases. An interesting situation emerges when you begin to consider being able to independently tune the two magnetic sublattices of a ferrimagnet which is shown in Fig. 3.1d. In the initial case the system has a positive magnetization. Tuning one of the sublattices with a given parameter (temperature, composition, etc.) a case can arise where the system reaches magnetic compensation. Here the two sublattices have equal moments, leading to zero net magnetic moment. On the surface, this looks just like an antiferromagnet. However, it is important to note that this zero magnetization state is “accidental” whereas in an antiferromagnet the zero magnetization state is symmetry enforced. These compensated ferrimagnets have gained significant attention in recent years in large part because they can still show some ferromagnetic behaviors (ex. an AHE) without a significant magnetic moment [22, 23, 24]. In principle these materials may

allow for spintronics devices that can be packed closely together without needing to worry about stray magnetic fields coming from the samples. Upon further increasing the tuning parameter, a negative magnetization state can be reached where the sublattice that initially had a smaller moment size now has a larger moment size relative to the magnetic sublattice that was unchanged.

As one final comment in this section, it is important to point out the difference between localized magnetic moments and itinerant magnetic moments. Localized moments are physically essentially what I have used to sketch these different magnetic configurations; they are well-defined moments coming from a given ion. Many rare earth magnets containing f electrons are local moment magnets. On the other hand, itinerant magnets require a band structure picture to understand. For some materials the Zeeman effect will split the originally two-fold degenerate energy bands into separate up-spin and down-spin bands. Depending on the location of these bands relative to E_F , there are situations where these up- and down-spin bands will be filled unequally, leading to a total moment of the system. Many transition metal magnets containing d electrons are itinerant magnets. The distinction between these two types of magnets will become important when considering the AHE.

Anomalous Hall Effect

In order to begin understanding the origin of the AHE, we must begin with the idea of Berry phase, a geometric phase a quantum mechanical state picks up while undergoing adiabatic evolution in the parameter space of the system's Hamiltonian [25, 26]. In solid-state systems, Bloch wavefunctions acquire this phase as they evolve in k space through the equation

$$\gamma_n = \oint_C d\mathbf{k} \cdot \langle u_n(\mathbf{k}) | i\nabla | u_n(\mathbf{k}) \rangle \quad (3.3)$$

where $\langle u_n(\mathbf{k}) | i\nabla | u_n(\mathbf{k}) \rangle$ is known as the Berry connection (A_n). This Berry connection is gauge-dependent, so in analogy to the classical electrodynamics equation $\mathbf{B} = \nabla \times \mathbf{A}$ (which transforms the gauge-dependent vector potential $\mathbf{A}$ into a gauge-independent magnetic field), a similar gauge-independent field can be defined:

$$\Omega_n = \nabla \times \mathbf{A}_n \quad (3.4)$$

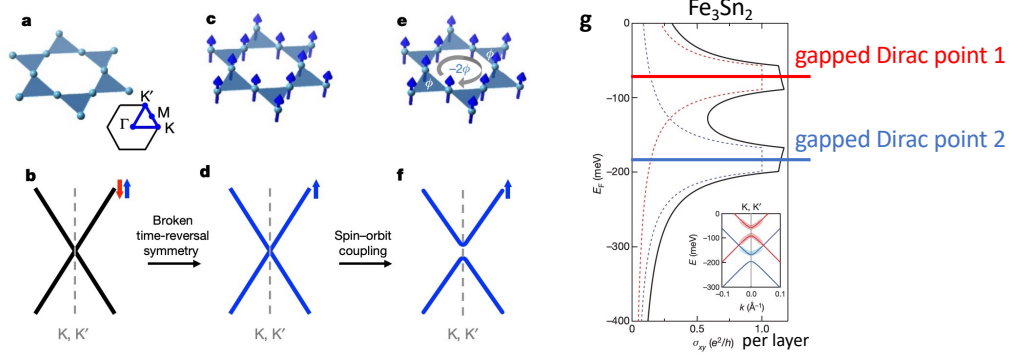


Figure 3.3: **Ferromagnetic kagome lattice** [27]. **a, b**, Original Dirac point from a kagome lattice. **c, d**, Spin-split Dirac point from a ferromagnetic kagome lattice. **e, f**, Gapped Dirac point upon allowing spin-orbit coupling which brings about Berry curvature (here denoted ϕ). **g**, An example of the contributions of gapped Dirac points to the intrinsic part of the AHE using Fe_3Sn_2 as an example.

with $\Omega_{\mathbf{n}}$ referring to the Berry curvature. Just like $\mathbf{B}$, the Berry curvature is a pseudovector, meaning it is only non-zero when inversion or time-reversal symmetry is broken (note that the breaking of either of these symmetries breaks Kramer's degeneracy, allowing two-fold degenerate energy bands to be split in energy into two separate bands). The Berry curvature is usually small in most of momentum space, but near band crossings or avoiding crossings this quantity can become large. It should be noted that TbMn_6Sn_6 has many band crossings near E_F meaning that there are many Berry curvature “hot spots” that should impact electronic transport properties, a fact that will become important in this chapter.

This Berry curvature becomes important in the context of magnetotransport because it can be directly linked to the intrinsic part of the AHE (the extrinsic part will be discussed soon). Namely,

$$\sigma_{ij}^{AH-int} = -\epsilon_{ijl} \frac{e^2}{h} \sum_n \int \frac{d\mathbf{k}}{(2\pi)^d} f(\epsilon_n(\mathbf{k})) \Omega_n^l(\mathbf{k}) \quad (3.5)$$

where $f(\epsilon_n(\mathbf{k}))$ describes the Fermi-Dirac distribution over the energy bands. A simple way to break down this equation comes from considering that $f(\epsilon_n(\mathbf{k})) = 0$ for bands that are unfilled (ie. bands that are above E_F). Furthermore, for spin-split bands that are buried at energies far below E_F , the integration over both the up- and down-spin bands will cancel. Thus, the only bands that will contribute to the intrinsic part of the AHE are bands near E_F with non-zero Berry curvature.

This idea becomes relevant in the context of a ferromagnetic kagome lattice as shown in Fig. 3.3 which was adapted from Ref. [27]. Panels a and b show the original Dirac point from a kagome lattice. Panels c and d show that when the kagome lattice orders ferromagnetically, the Dirac point will spin-split. Panels e and f show that upon introducing spin-orbit coupling, the Dirac point will become gapped. This gapped Dirac point induces significant Berry curvature which is shown schematically with ϕ . This situation is very similar to the idea of a two dimensional magnetic topological insulator in which the surface Dirac point is spin-split and gapped out, leading to a quantum anomalous Hall effect in which the Hall conductance is quantized [28]. While in three dimensional samples the Hall conductance will not be quantized, if E_F is positioned in the gap of the Dirac point it is expected that there will be a quantized intrinsic Hall conductance (to e^2/h) per layer. As E_F is moved away from the gap, the expected σ_{xy}^{AH-int} will correspondingly fall off. This is shown in panel g using Fe_3Sn_2 as an example where the dashed red and blue lines correspond to σ_{xy}^{AH-int} from each of the two gapped Dirac points in the band structure this material. Note that the contributions of each of these Dirac points (and any other bands near E_F) needs to be summed according to Eqn. 3.5 which is shown with the black solid line. Because of this, σ_{xy}^{AH-int} per layer is not actually quantized except by coincidence. In systems with many bands near E_F this complication makes interpretation of the AHE difficult.

Beyond the above issues, accurate analysis of the AHE is difficult because there are both intrinsic and extrinsic contributions to this effect. While parsing these two parts is non-trivial, it has become more-or-less generally accepted that a scaling relation can accomplish this feat. This relation is of the form

$$\sigma_{AH} = a\sigma_{xx}^2 + c \quad (3.6)$$

where $a\sigma_{xx}^2$ is the extrinsic part of the anomalous Hall conductivity (AHC) and $c \equiv \sigma_{AH}^{int}$ is the intrinsic part of the AHE [29]. Without getting too far into the details, the extrinsic part of the AHE is largely due to the side jump and skew scattering mechanisms in which up- and down-spin charge carriers scatter asymmetrically in the crystal. This is the underlying physics of Eqn. 3.6: the extrinsic part of the AHC has a dependence on σ_{xx} because the amount of scattering in the crystal should correspondingly change σ_{xx} whereas the intrinsic part of the AHC should be agnostic to scattering and hence is constant as a function of σ_{xx} . Thus, by changing σ_{xx} by significant margins (ex. with temperature) σ_{AH}^{int} can in principle

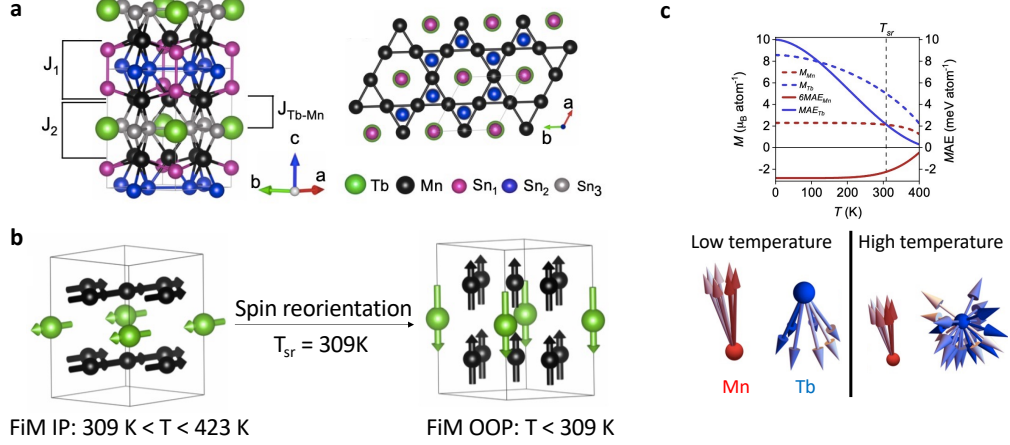


Figure 3.4: **Magnetism of TbMn_6Sn_6 .** **a**, Crystal structure of TbMn_6Sn_6 with couplings (J terms) noted [30]. The kagome lattice of Mn is seen. **b**, A schematic of the phase transitions in TbMn_6Sn_6 [31]. **c**, A plot of the MAE and magnetization of the Tb and Mn ions as a function of temperature in TbMn_6Sn_6 [31]. A schematic of the Tb and Mn fluctuations at high and low temperatures is also shown.

be extracted. The careful experimental verification of this equation is presented in Ref. [29] in which thin films of Fe with varying thicknesses were measured. Fe is a reasonably well-behaved ferromagnet (meaning here that the spins do not fluctuate much below T_C); this equation may not hold in more complex materials with different scattering mechanisms.

The earlier mention of itinerant and localized magnetism becomes very relevant here. In itinerant electron magnets, the bands near E_F are (by definition) responsible for the magnetism. Since they are near E_F , the Berry curvature they produce can cause an AHE. On the other hand, in localized magnet systems, the magnetic bands are buried deep below E_F and thus an AHE will often not be seen. This is key to remember in the case of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ — the Mn and Cr moments will be responsible for any AHE and the Tb ions will not contribute.

TbMn_6Sn_6 Background

With the above ideas in mind, we can now move into the discussion of TbMn_6Sn_6 . Fig. 3.4a shows the crystal structure of this material; as expected for the RT_6X_6 family, the ab-plane kagome lattice is comprised entirely of Mn ions. Note that the various magnetic coupling terms (J) are also labeled. Interestingly, upon cooling from high temperatures this material

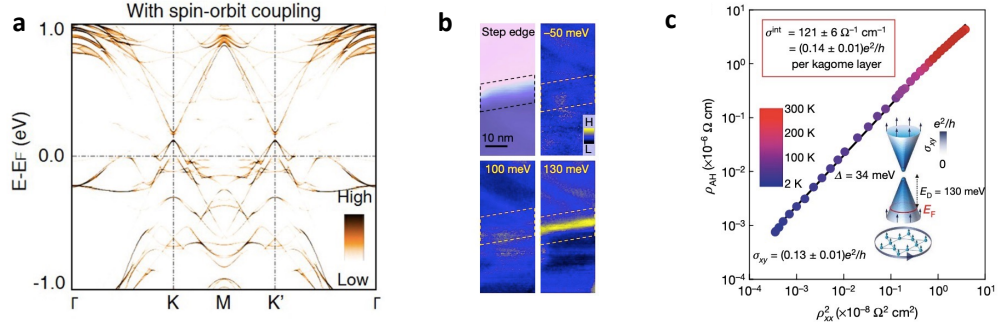


Figure 3.5: **Proposed quantum limit magnetism in TbMn₆Sn₆ [13].** **a**, Calculated band structure of TbMn₆Sn₆. **b**, Scanning tunneling spectroscopy data on TbMn₆Sn₆ using various bias voltages. **c**, AHE scaling of TbMn₆Sn₆.

orders as an in-plane ferrimagnet (FiM IP) at about 423 K (T_C) before undergoing a spin reorientation at 309 K (T_{sr}) and ordering as an out-of-plane ferrimagnet (FiM OOP) as shown in panel b [32, 31]. Note that the Mn and Tb sublattices both order ferromagnetically but they are arranged antiferromagnetically relative to one another. Fig. 3.4c shows a plot of the calculated magnetoanisotropy energy (MAE) and experimental magnetization of the Tb and Mn ions as a function of temperature in TbMn₆Sn₆ [31]. The MAE is essentially a measure of how strongly the magnetic moments of a given sublattice want to point in a particular direction. In this case, the Mn want to order in-plane (a negative MAE) and the Tb want to order out-of-plane (a positive MAE). This competition is what drives the spin reorientation in this compound. The element specific magnetization as a function of temperature reveals that the Mn moments are reasonably well-ordered at 400 K (note that cooling down further does not change M_{Mn} very much). On the other hand, the Tb moment size grows considerably upon cooling. The well-ordered Mn determine the ordering at T_C and thus TbMn₆Sn₆ orders as a FiM IP, but upon cooling the Tb moments becomes increasingly well behaved until finally the Tb moments are able to dictate the ordering along the c-axis at T_{sr} . At first glance this may be confusing considering that at base temperature $M_{Tb} \approx 9\mu_B$ and $M_{Mn} \approx 2.5\mu_B$, thus making the net moment per formula unit (F.U.) about $6\mu_B$ since there are 6 Mn ions per Tb ion. However, the large spin-orbit coupling of Tb as compared to Mn allows the Tb to have a very large MAE. It should be noted that these Tb fluctuations are quite significant; in fact, replacing the Mn with non-magnetic V drives T_C down from 423 K in TbMn₆Sn₆ to 4 K in TbV₆Sn₆ as reported in a publication headed by Elliott Rosenberg and me [6].

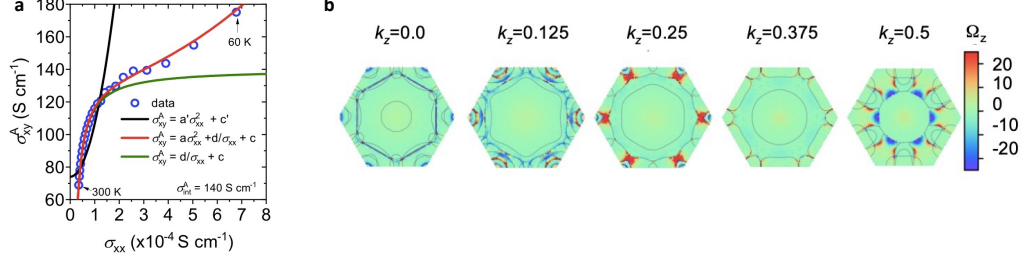


Figure 3.6: **New AHE scaling for TbMn₆Sn₆ [30].** **a**, Different AHE scalings applied to TbMn₆Sn₆. **b**, Calculated Berry curvature at different regions of the Brillouin zone.

As shown in Fig. 3.3, a ferromagnetic kagome lattice should be quite interesting. TbMn₆Sn₆ seems to be a nearly ideal case of this, and correspondingly a report on this compound proposed some very interesting findings [13] as shown in Fig. 3.5. Panel a shows their calculated band structure of TbMn₆Sn₆ revealing the expected gapped Dirac points from the kagome lattice about 130 meV above E_F . Panel b shows scanning tunneling spectroscopy data on TbMn₆Sn₆ using various bias voltages. When the bias voltage is tuned such that E_F is located within the gapped Dirac points, a high conductance edge state emerges which is expected for a topologically non-trivial state. Panel c shows the data we are most concerned with here, the scaling of the AHE in this compound to extract σ_{AH}^{int} . Here they are using the resistivity form of Eqn. 3.6; using the fact that $\rho = \sigma^{-1}$, the resistivity form of this scaling relation should be

$$\rho_{AH} = a + \sigma_{AH}^{int} \rho_{xx}^2 \quad (3.7)$$

which is (of course) equally valid to use. Upon applying this scaling, they recover a value for σ_{AH}^{int} that is consistent with a gapped Dirac point with the gap size and distance from E_F that they were expecting. However, there is a key physical reason this equation should not work here: no other bands near E_F were considered for this itinerant system. On a deeper level this is an even deeper reason for concern: here they have plotted $\rho_{AH}(\rho_{xx}^2)$ on a log-log scale and fit their data with a line. However, according to Eqn. 3.7 they should be fitting this data on a linear-linear scale with a line! The high profile nature of this paper along with some glaring issues such as this one quickly caught the attention of the condensed matter community.

One of the more serious attempts to understand this material came about in Ref. [30] as shown in Fig. 3.6. Panel a shows their attempt at scaling the AHE in TbMn₆Sn₆ using

equations similar to Eqn. 3.6. As they point out, in principle there is no physical reason to use more powers of σ_{xx} in this scaling relation, but (of course) it is better to use as few as possible. While Eqn. 3.6 (black) completely fails to fit the data, a nonstandard d/σ_{xx} term is able to capture the high temperature response quite well (green). Upon adding this term to Eqn. 3.6 to make an expression of the form

$$\sigma_{AH} = a\sigma_{xx}^2 + d/\sigma_{xx} + c \quad (3.8)$$

the entire data set is able to be fit quite well (red). Because the high temperature response is able to be fit so well by this new term, they tentatively propose that this term may be due to the substantial Tb fluctuations at high temperatures (as shown in Fig. 3.4c), a response that would be completely missed while studying Fe as Eqn. 3.6 was derived from [29]. This indicates that Ref. [13] did not properly fit their data. On top of this, upon doing a computational analysis of the Berry curvature in the Brillouin zone, the authors of Ref. [30] found that there are many Berry curvature “hot spots” all around the Brillouin zone as shown in Fig. 3.6b, not just at the K points (corners) where the gapped Dirac points were proposed. This is to be expected, there are many itinerant bands near E_F as shown in Fig. 3.5a.

While these results put a major hole in the story presented in Ref. [13], it did leave some open questions. Two main questions: how much (if at all) do the gapped Dirac points influence the large σ_{AH}^{int} that was extracted even in Ref. [30], and how important is this new non-standard AHE scaling term? In order to conclusively put this story to bed, we decided to Cr dope TbMn_6Sn_6 to form $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$. This hole dopes the compound, lowering E_F . If the gapped Dirac points are largely relevant to σ_{AH}^{int} , this quantity should decrease with increasing x . Furthermore, by using Eqn. 3.6 across a range of x , we should be able to determine how necessary the new AHE scaling term is.

With this intro, I now reproduce Ref. [20] with small changes below to describe our results.

3.2 Published Results

Introduction

Compensated ferrimagnets have recently emerged as a promising material platform for spintronics applications, combining advantageous properties of both ferromagnets and antifer-

romagnets [22, 23, 24]. In antiferromagnets, the net moment is zero by symmetry. This feature leads to faster switching times, reduced stray fields and resistance against external fields, but also leads to difficulty to read and write magnetic domains. A related class of Luttinger-compensated ferrimagnets [33] also features exact compensation, albeit not by symmetry but by virtue of Luttinger theorem. These can be insulators, or half-metals (metallic in one spin channel). A more promising possibility are chemically compensated ferrimagnets, which are metallic in both spin channels. There, the net magnetization can be tuned to zero by controlling the chemical composition of their inequivalent spin sublattices. A great advantage here is that the net magnetic moment can be tunable by chemical composition or temperature, so that it is small, but not zero, thus allowing domain control by external fields. Another important advantage is that despite near-zero magnetization, compensated ferrimagnets allow for large (on the order of hundreds meV) exchange splitting. Consequently, they can exhibit finite anomalous Hall effect (AHE), providing a relatively straightforward reading mechanism.

A well-known class of compensated ferrimagnets are the rare-earth transition-metal alloys [34]. In these materials the magnetic moments of rare-earth and transition-metal ions are antiferromagnetically coupled, and the net magnetic moment can be controlled by tuning their relative concentrations. The magnetization of the rare-earth and the transition-metal moments have different temperature dependencies. As a result, zero net magnetization occurs at a compensation temperature, denoted as T^* , which can be smoothly modulated by chemical composition. While amorphous materials have dominated the study of rare-earth transition-metal compensated ferrimagnets, crystalline compounds have received comparatively less attention. Unlike amorphous materials, crystalline materials host well-defined Bloch energy bands, characterized by Berry curvatures capable of generating large intrinsic anomalous Hall effects. Consequently, the search and design of materials with large Berry curvature has also emerged as a key focal point in spintronics research [35].

Here we report the realization of compensated ferrimagnetism in a rare-earth transition-metal intermetallic compound, $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$. $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ belongs to the RT_6X_6 (R = rare earth, T = transition metal, X = Si, Ge, Sn) family, known for its kagome lattice structure and intriguing band structure features such as flat bands, van Hove singularities, and Dirac points [36, 37, 38]. Notably, the ferrimagnet TbMn_6Sn_6 exhibits a large intrinsic AHE, which was initially attributed to Berry curvature arising from a gapped 2D Dirac point

above but near the Fermi level [13]. This picture, if correct, would preclude Cr doping (hole doping) as a knob to tune the net magnetization without decreasing the intrinsic AHE, since that would shift the Fermi level away from the Dirac point. Furthermore, the 2D nature of the Dirac point would limit the large AHE to one particular geometry. Fortunately, it was later shown [39, 40, 30] that the AHE is accumulated over the entire Brillouin zone and does not depend dramatically on the Fermi level; it was also shown that the AHE in similar compounds is, in fact, 3D [41], which is also favorable for applications.

In this work, we show that the substitution of Cr for Mn reduces the magnetic moment of the transition metal site, which eventually turns $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ into a compensated ferrimagnet. Near compensation, we observed a divergence of the coercive field at low temperatures. Furthermore, contrary to the initially advocated 2D Dirac point scenario, Cr substitution substantially enhances intrinsic AHE, likely due to the tuning of the Fermi level to various Berry curvature hot spots [30]. Our findings pave the way for the rational design of a compensated ferrimagnet with a large intrinsic AHE.

Results

Basic characterization

High-quality single crystals of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ with x up to 0.55 were grown using a Sn flux method. The magnetization and magneto-transport properties of these crystals were measured by the vibration sample magnetometry (VSM) option of a Quantum Design Physical Property Measurement System (PPMS) Dynacool and standard AC electrical transport techniques, respectively. Further details can be found in the methods section. Fig. 3.7A shows the crystal structure of TbMn_6Sn_6 , which consists of two layers of kagome lattices formed by Mn atoms and one layer of triangular lattice composed of Tb atoms, viewed along the ab-plane (top) and c-axis (bottom). This crystal structure is preserved with Cr doping, as exemplified in the Z-contrast atomic-resolution scanning transmission electron microscope (STEM) image shown in Fig. 3.7B where the ab-plane kagome motif can be observed in an $x = 0.55$ sample. Additionally, atomic-resolution electron energy-loss spectroscopy (EELS) was used to map out the elemental distribution in this sample which is presented in Fig. 3.7C. Notably, the Cr substitution for Mn is relatively homogeneous even at the nanometer scale.

TbMn_6Sn_6 orders ferrimagnetically at 423 K (T_c) [31]. Within the layers the Mn mo-

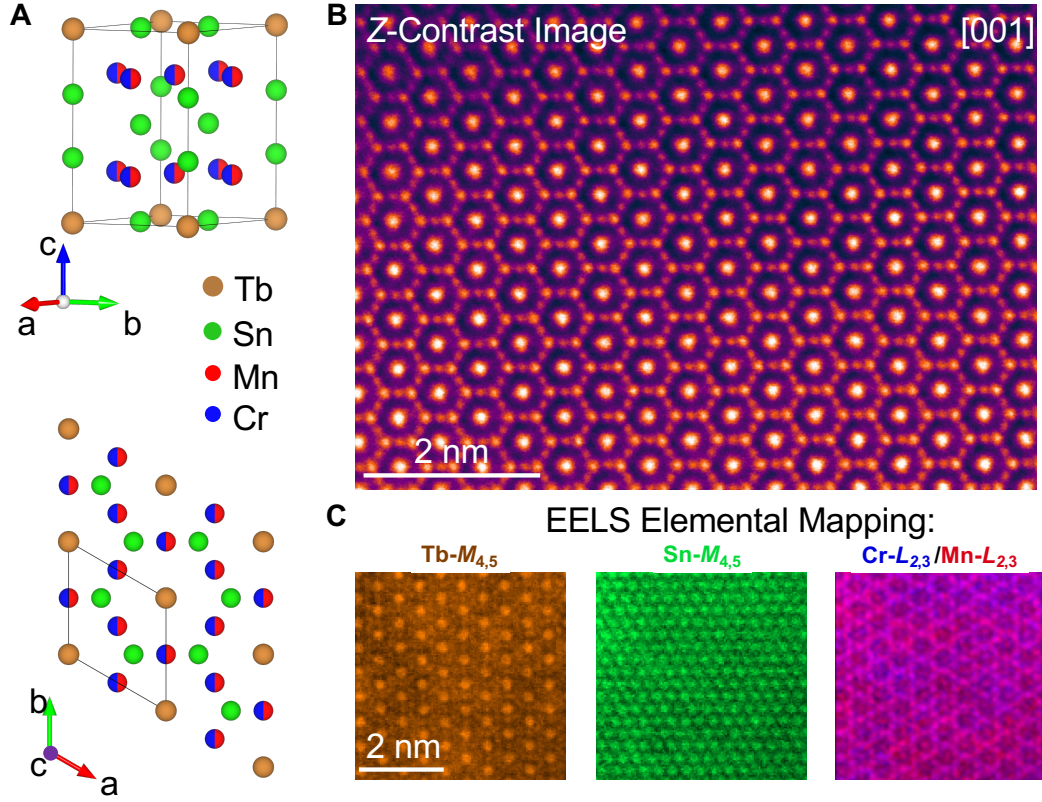


Figure 3.7: **Atomic-scale visualization of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ kagome lattice.** **A**, Atomic models of the $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ lattice viewed along the ab -plane in the upper panel and along the c -axis in the bottom panel. The kagome layers have a mixed occupancy of Cr-dopants and Mn (depicted in blue and red, respectively). **B**, Z-contrast atomic-resolution STEM image of an $x = 0.55$ crystal viewed along the c -axis. **C**, Color maps of elemental distribution by atomic-resolution EELS. Left: Tb ($M_{4,5}$ edge), middle: Sn ($M_{4,5}$ edge), right: mixed distribution of Cr and Mn ($L_{2,3}$ edges).

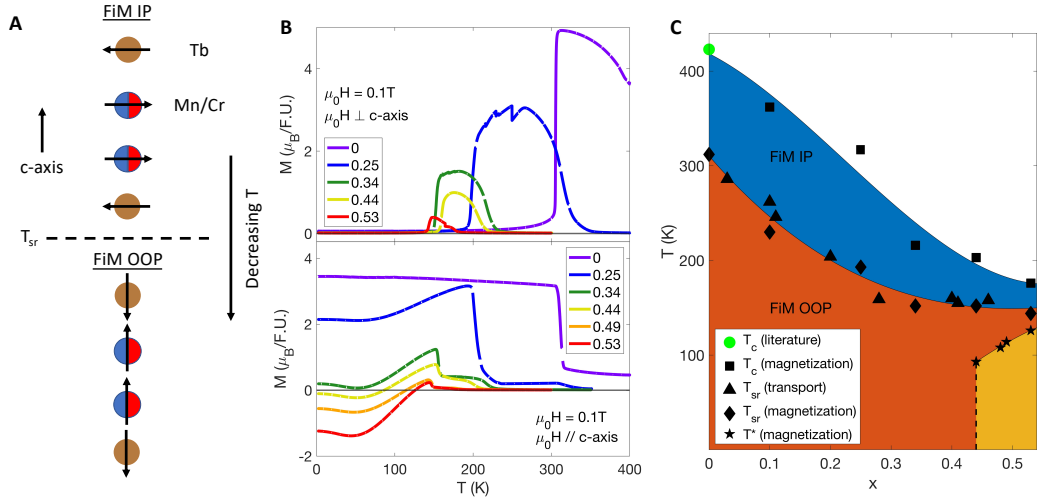


Figure 3.8: **Spin reorientation transition and the phase diagram of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$.**

A, Orientation of magnetic moments of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ in the high-temperature ferrimagnetic in-plane (FiM IP) (top) and low-temperature ferrimagnetic out-of-plane (FiM OOP) (bottom) phases. **B**, Low-field magnetization as a function of temperature of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ for a variety of x with an in-plane (top) or out-of-plane (bottom) magnetic field of 0.1 T applied. The sudden decrease/increase of in-plane/out-of-plane magnetization indicates the spin reorientation transition at T_{sr} . **C**, $x - T$ phase diagram of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ as determined from magnetization and transport measurements.

ments and the Tb moments order ferromagnetically, but the Tb and Mn layers couple antiferromagnetically, resulting in a ferrimagnetic structure where Tb and Mn moments are antiparallel (schematically shown in the top portion of Fig. 3.8A (FiM IP)). Neutron scattering studies on polycrystalline samples have revealed that at base temperature the size of Mn moment is $2.4 \mu_B$ and the size of Tb moment is $8.6 \mu_B$, leading to a net $5.8 \mu_B$ per formula unit along the direction Mn moments [31]. It should be noted that the saturated magnetization of single crystals of TbMn_6Sn_6 from bulk magnetization measurements [13, 32, 30] is $\approx 4 \mu_B$ per formula unit, which is consistent with our results but less than the value extracted from neutron scattering; this discrepancy has previously been noticed [32] and may be due to inherent differences between polycrystalline and single crystal samples. Fig. 3.8B shows the temperature dependence of the magnetization of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ with a small in-plane (top) and out-of-plane (bottom) magnetic field of 0.1 T. The ferrimagnetic ordering temperature of the $x = 0$ sample exceeds the instrument limit (400 K), thus it is not visible within this dataset. Nevertheless, a sudden decrease in in-plane magnetization and an increase in out-of-plane magnetization are clearly observed at 309 K (T_{sr}). The abrupt change in magnetization is associated with a spin reorientation transition [13], where the Tb and Mn moments switch from a high temperature in-plane orientation to a low temperature out-of-plane orientation (FiM OOP), as shown in the bottom portion of Fig. 3.8A. This transition can be attributed to the competition between the c-axis uniaxial anisotropy favored the Tb moments and the easy-plane anisotropy preferred by the Mn moments. At high temperatures, the Tb moments fluctuate more due to a weaker exchange interaction, allowing the easy-plane anisotropy of the Mn moments to dominate. At low temperatures, the scenario is reversed, and the c-axis uniaxial anisotropy of the Tb moments prevails [30, 42].

As shown in Fig. 3.8B, the temperature dependence of the in-plane magnetization for Cr doped samples closely resemble that of the parent compound except that T_c , T_{sr} and the magnitude of the magnetization steadily decrease with increasing x . This is consistent with a previous study on polycrystalline samples, where Cr doping reduced the average moment size of the transition metal, thereby suppressing the overall ferrimagnetic ordering temperature [43]. However, the out-of-plane magnetization exhibits a more intriguing behavior. While the features at T_c and T_{sr} are consistent with the in-plane magnetization data, the out-of-plane magnetization demonstrates a strong temperature dependence below T_{sr} , con-

trasting with the nearly constant magnetization observed in the parent TbMn_6Sn_6 over this temperature range. For the Cr doped samples, the out-of-plane magnetization decreases with temperature, reaching a local minimum near 60 K. For x greater than 0.4, the magnetization continues to decrease even after reaching zero at T^* , resulting in negative value for temperatures below T^* . This anti-alignment of the moment with the external magnetic field has been observed in other ferrimagnets driven to magnetic compensation by changing temperature [44].

Using the magnetization data presented here and the resistivity data presented in Supplementary Note 1, along with the literature value of T_c for $x = 0$ [32, 31], an $x - T$ phase diagram was constructed and is shown in Fig. 3.8C. It can be seen that both T_c and T_{sr} decrease rapidly with increasing x in the low x regime, but become relatively constant with further doping above approximately $x = 0.3$. For higher Cr concentration, T^* emerges, delineating the boundary of a region where the magnetization is anti-aligned with the external field during the cool-down process. In the following sections, we will examine the hysteresis loop of both the magnetization and the anomalous Hall resistivity as a function of doping and temperature. Our results reveal that this unusual behavior arises from magnetic compensation driven by Cr doping and thermal fluctuations of Tb moments.

Chemical doping induced magnetic compensation and giant coercivity

We begin by examining the doping dependence at the base temperature. Fig. 3.9A shows the magnetization (top row) and ρ_{xy} (bottom row) as functions of the magnetic field parallel to the c -axis at 2.5 K for five representative Cr concentrations. The magnetization of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ shows a clear hysteresis loop characteristic of a hard ferrimagnet with uniaxial anisotropy. As the Cr concentration increases, the saturated magnetization decreases, reaching a value near zero at a composition $x^* \approx 0.43$, before increasing again for $x > x^*$. The reduction in saturated magnetization is accompanied by a considerable increase in the coercive field. A previous neutron scattering study on low- x $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ revealed that Cr doping at the Mn site reduces the moment on that site without substantially changing the moment on the Tb site [43]. Extending this observation to higher dopings suggests that at x^* the total magnetization from the Mn/Cr and Tb moments becomes equal, resulting in a net magnetization of zero. As x increases beyond x^* , the total magnetization of the Tb moments surpasses that of the Mn/Cr moments, causing the net magnetization to

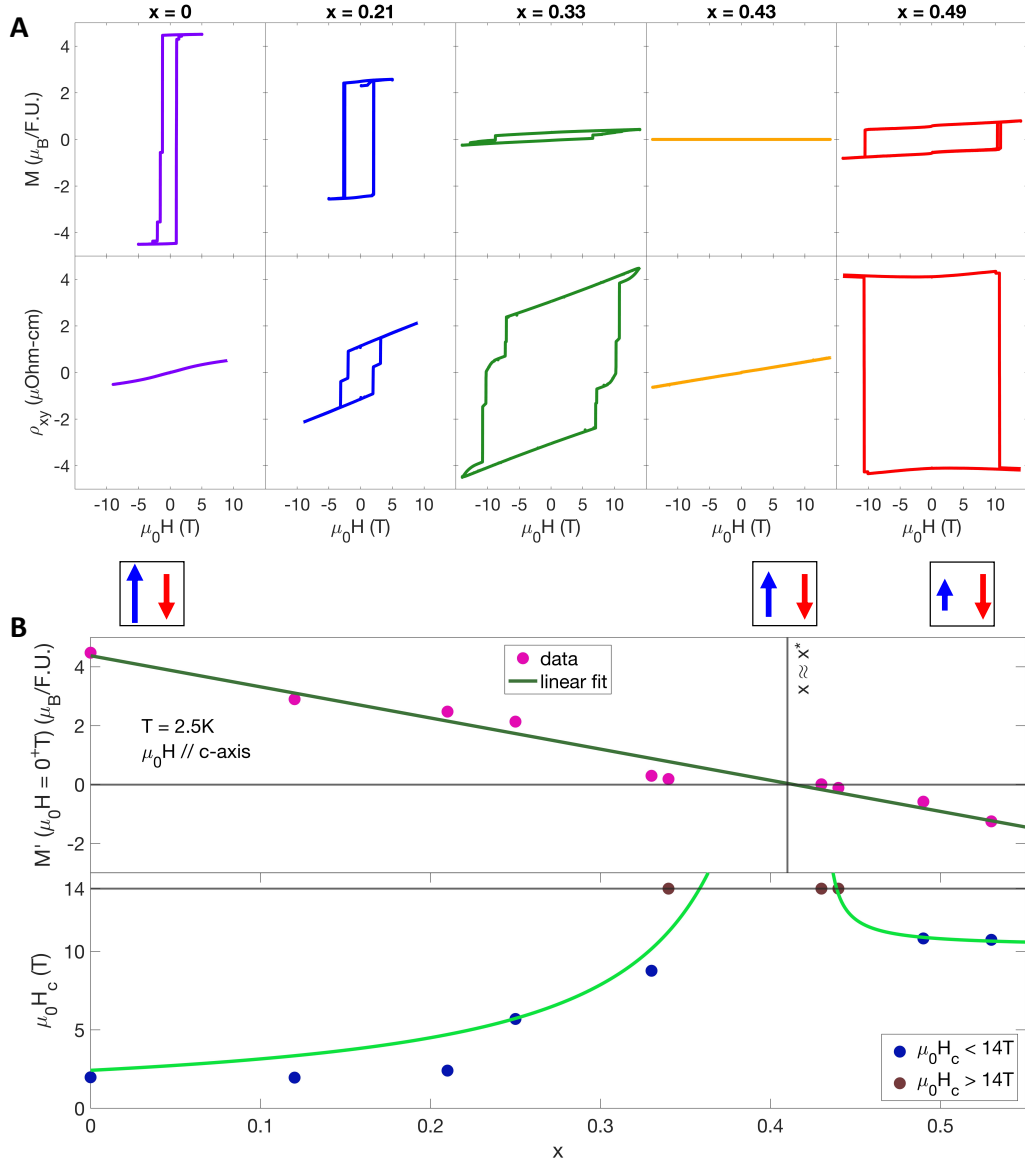


Figure 3.9: **Magnetic compensation and giant coercivity through chemical doping.** **A**, Magnetization (top row) and ρ_{xy} (bottom row) as a function of magnetic field for a variety of x at 2.5 K. Plots in the same column are from the same x which is indicated by the text on top of the column. **B**, Top: M' (described in the main text) as a function of x at 2.5 K with decreasing magnetic field. Bottom: $\mu_0 H_c$ as a function of x from magnetization measurements.

rise again, aligning with the Tb moment.

To confirm this hypothesis, we plot M' in the top panel of Fig. 3.9B, where $M' = M$ for $x < x^*$ and $M' = -M$ for $x > x^*$. Here M represents the experimentally measured zero field magnetization after the application of a large positive field to polarize the moments. The assumption is that M' corresponds to the net magnetization of the Mn/Cr moments minus the net magnetization of the Tb moments. Indeed, M' decreases roughly linearly as a function of x while (by construction) crossing zero at x^* . A linear fit of these data indicates that the average moment decreases by roughly $1.8\mu_B$ per Cr substitution, in good agreement with the previous neutron scattering study [43]. Since each Mn ion has a moment of $2.4\mu_B$ in TbMn_6Sn_6 , this implies that each Cr ion in $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ carries a moment of $0.6\mu_B$ if the Mn moments remain the same regardless of doping. This is quite similar to the value of $0.48\mu_B$ which was found to be the moment per Cr ion in TbCr_6Ge_6 via neutron scattering measurements [45]. Thus, Cr is likely in a low-spin state in these materials.

To gain further insight, we performed density functional theory (DFT) calculations to investigate the magnetic state of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$. Interestingly, we found that for $\text{TbCr}_2\text{Mn}_4\text{Sn}_6$ (with both DFT+U frozen core Tb^{3+}) the lowest energy states correspond to Cr forming one-dimensional in-plane antiferromagnetic chains. Thus, a potential major contribution to the magnetic moment reduction with Cr doping is due neither to dilution of the Mn sublattice by nonmagnetic ions, nor to suppression of Mn moments, but by the tendency of Cr appearing next to each other in the plane to form antiferromagnetic clusters. The results of these calculations are shown in Supplementary Note 2.

The bottom panel of Fig. 3.9B shows the coercive field, $\mu_0 H_c$, extracted from the magnetization measurement. The coercive field approximately follows the inverse of the zero-field magnetization, consistent with the flipping of the magnetization when the Zeeman energy $\mu_0 H_c M$ overcomes the domain depinning energy [46, 47]. Near x^* the coercive field exceeds 14 T, the maximum field achievable by the PPMS used. To our knowledge, this is the largest coercive field ever observed in a crystalline material [48]. This massive coercivity is a result of the small magnetic moment observed combined with the large magnetocrystalline anisotropy of the Tb ions at low temperatures [30, 6]. These observations are consistent with $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ behaving as a nearly compensated ferrimagnet, exhibiting a giant coercive field at low temperatures between $0.30 \leq x \leq 0.45$, where the net Tb moments and the net Mn/Cr moments nearly cancel each other.

In contrast to magnetization, ρ_{AH} increases with Cr concentration. For samples very close to x^* , the hysteresis in ρ_{AH} cannot be detected by magnetic field sweep at base temperature simply because the coercive field exceeds 14 T. However, when the sample is field-cooled from high temperature, a large ρ_{AH} can still be observed even as M approaches zero (see Fig. S12E in Supplementary Note 6). For $x > x^*$, the anomalous Hall effect switches sign, i.e. ρ_{AH} switches from positive to negative when the magnetization is positive. Given that the energy bands near the Fermi level are primarily composed of transition metal d-orbitals, the sign change of is consistent with the anomalous Hall effect being driven by the transition metal sites. In-depth analysis of the anomalous Hall effect will be presented in Sec. 3.2.

Temperature induced magnetic compensation

In addition to chemical substitution, temperature is another effective parameter for controlling magnetic compensation. Fig. 3.10A displays the previously shown out-of-plane magnetization as a function of temperature for a sample with $x = 0.49$. This data was collected as the sample is cooled under a 0.1 T c-axis magnetic field. At the spin reorientation transition temperature $T_{sr} = 150$ K, the magnetization abruptly increases as all moments reorient from in-plane to out-of-plane. Below T_{sr} , the magnetization reaches a maximum and then decreases with decreasing temperature. It crosses zero at $T^* = 114$ K and remains negative for temperatures below T^* .

To gain a deeper understanding of this behavior, we measured the field dependence of magnetization and Hall resistivity ρ_{xy} across a range of temperatures, as shown in the top and bottom rows of Fig. 3.10B, respectively. Each column represents data taken at the same temperature, which is noted above the column. In the 2.5 K data, in addition to the large hysteresis loop, a small additional paramagnetic contribution can be observed near $\mu_0 H = 0$ T in the magnetization versus field data. This contribution is highly sample dependent and only noticeable in samples with x greater than 0.4, suggesting it may result from either disordered spins or a minor impurity phase. As the temperature increases, $\mu_0 H_c$ decreases substantially (the temperature dependence of $\mu_0 H_c$ for multiple samples is detailed in Supplementary Note 3). Near T^* (≈ 120 K), the hysteresis loop in ρ_{xy} reverses direction, whereas the magnetization vanishes. As temperature increases above T^* , the magnetization increases again. The temperature dependence of the magnetization and the anomalous Hall effect mirrors

the doping dependence at base temperature, indicating that the magnetic compensation can also be induced by temperature sweep. Instead of a reduction in average Mn/Cr moments due to Cr substitution, the magnetic compensation at T^* is driven by the reduction of the average Tb moments at elevated temperatures due to thermal fluctuations, which is the same mechanism that drives the spin-reorientation transition [30]. Such temperature-driven compensation is commonly seen in other rare-earth transition metal alloy system, where sign switching of ρ_{AH} has been observed [49].

With these new insights, we can now explain the magnetization reversal observed in the field-cooled measurements. In samples with $x > \approx 0.4$, the Mn/Cr moments have decreased sufficiently that the Tb moments can match them in magnitude at T^* . As the temperature continues to decrease in these samples, the net Tb moment becomes larger in magnitude than the Mn/Cr moment, causing the net magnetization to become anti-aligned with the external field. Additionally, the coercive field rapidly increases with decreasing temperature and exceeds the external field below T^* . Consequently, the magnetization becomes trapped in the reverse configuration, leading to an overall negative value of magnetization.

Enhancement of anomalous Hall effect with doping

Table 1: Extracted parameters from conductivity fitting of AHE.

x	a (Ohm-cm)	c ([Ohm-cm] $^{-1}$)	d ([Ohm-cm] $^{-2}$)
0	$(7.21 \pm 0.64) \times 10^{-9}$	186 ± 2.8	$(-3.26 \pm 0.22) \times 10^5$
0.03	$(-7.48 \pm 2.45) \times 10^{-9}$	140 ± 3.5	$(-2.04 \pm 0.18) \times 10^5$
0.10	$(-1.16 \pm 0.73) \times 10^{-7}$	313 ± 20.4	$(-4.87 \pm 0.88) \times 10^5$
0.28	$(-1.03 \pm 0.41) \times 10^{-5}$	562 ± 177	$(-1.08 \pm 0.43) \times 10^6$
0.49	$(-7.86 \pm 2.72) \times 10^{-6}$	1107 ± 262	$(-3.90 \pm 0.97) \times 10^6$

We now turn to a more quantitative analysis of the anomalous Hall effect. Fig. 3.11A and B show the field dependence of Hall resistivity ρ_{xy} of samples with $x = 0.28$ and 0.43 measured at various temperatures. Similar plots of samples with other Cr concentrations can be found in Supplementary Note 4. All samples exhibit pronounced hysteresis and a well-defined anomalous Hall effect at zero field. For higher Cr concentrations, the anomalous

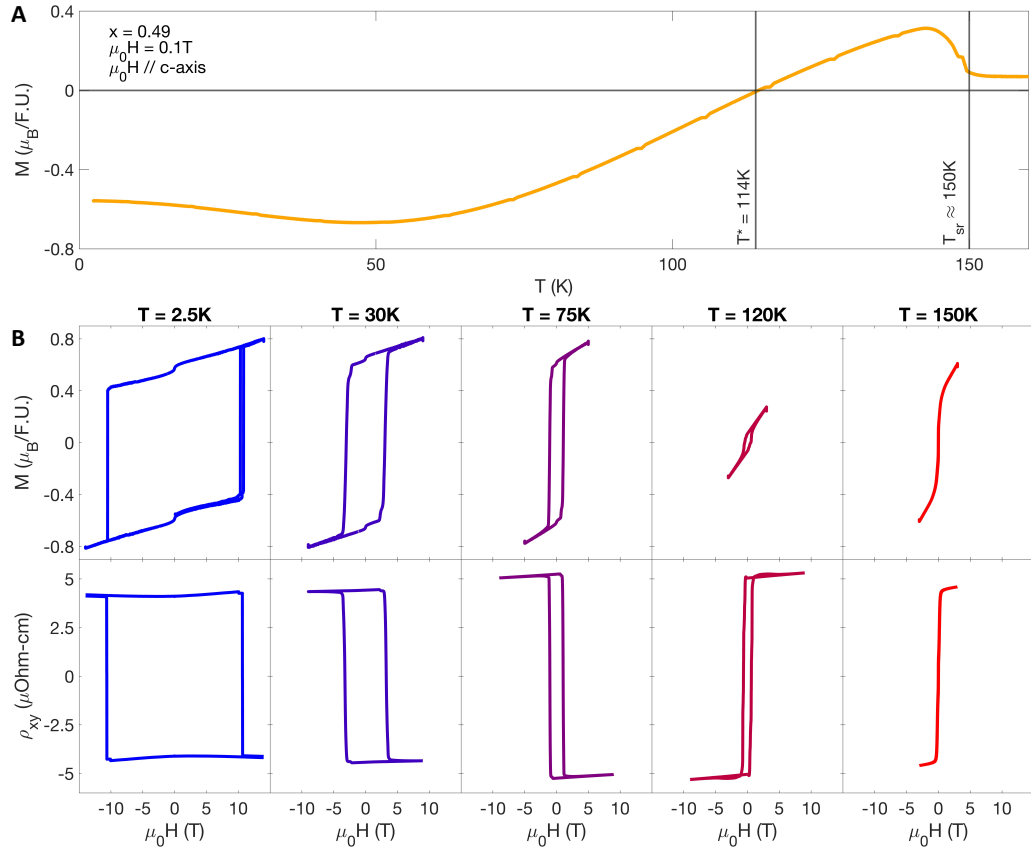


Figure 3.10: **Temperature dependence of magnetization and sign reversal of ρ_{AH} .** **A**, Low-field magnetization with an out-of-plane magnetic field for $x = 0.49$ as a function of temperature. **B**, Magnetization (top row) and ρ_{xy} (bottom row) as a function of magnetic field at a variety of temperatures for the same sample. Plots in the same column are taken at the same temperature which is indicated by the text on top of the column.

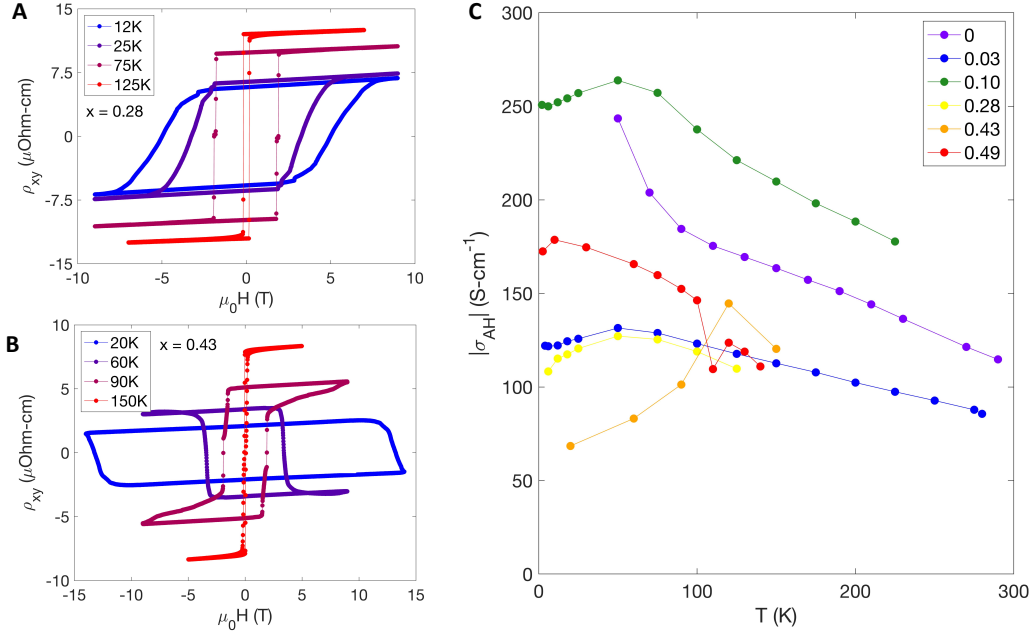


Figure 3.11: **Anomalous Hall effect of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$.** **A, B,** ρ_{xy} as a function of $\mu_0 H$ at several temperatures for $x = 0.28$ and 0.43 , respectively. **C,** Anomalous Hall conductivity $|\sigma_{AH}|$ as a function of temperature for a variety of dopings of $\text{Tb}(\text{Mn}_{1-x}\text{Cr}_x)_6\text{Sn}_6$.

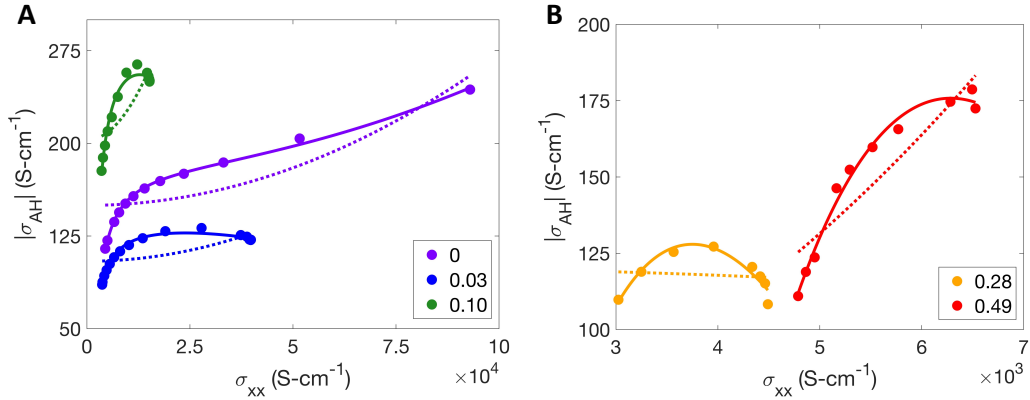


Figure 3.12: **Fitting of anomalous Hall conductivity.** **A, B,** $|\sigma_{AH}|$ as a function of σ_{xx} for low (A) and high (B) x . Dashed lines correspond to fits utilizing the standard anomalous Hall fitting function and solid lines show fits which include the extra term described in the main text.

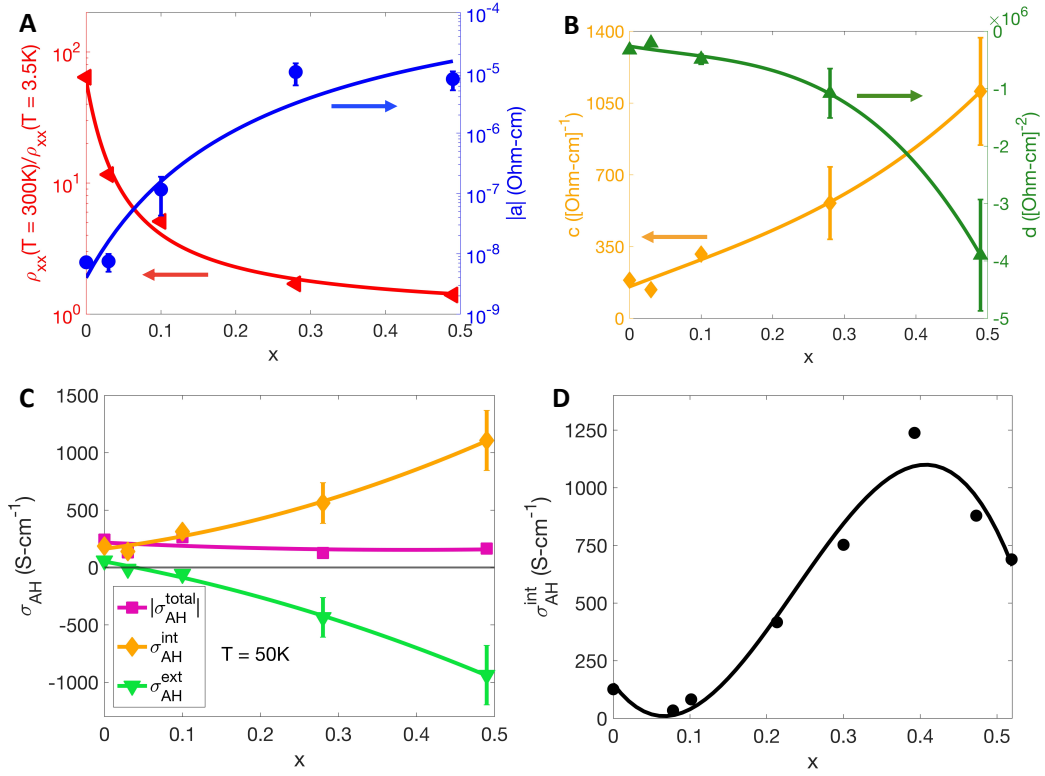


Figure 3.13: **Doping dependence of intrinsic and extrinsic anomalous Hall effect parameters.** **A**, Residual resistivity ratio and fit parameter a (extrinsic contribution) as a function of x . **B**, Fit parameters c (intrinsic contribution) and d (spin fluctuations contribution) as a function of x . **C**, $|\sigma_{AH}|$ and extracted σ_{AH}^{int} and σ_{AH}^{ext} at 50 K as a function of x . **D**, Calculated σ_{AH}^{int} as a function of x using a rigid band model.

Hall effect changes sign as a function of temperature as a result of magnetic compensation, as discussed in the previous section.

We calculated the anomalous Hall conductivity, σ_{AH} , using the relation $\sigma_{AH} = -\rho_{AH}/(\rho_{xx,0}^2 + \rho_{AH}^2)$, where $\rho_{xx,0}$ is the zero-field longitudinal resistivity. Fig. 3.11C presents the anomalous Hall conductivity as a function of temperature for various Cr concentrations. To facilitate comparison across different samples without the influence of sign switching, we plot only the absolute value $|\sigma_{AH}|$. The results for $x = 0$ are consistent with previous reports [30]. Cr doping does not substantially alter $|\sigma_{AH}|$, which changes by less than an order of magnitude across all measured Cr concentrations. Additionally, this quantity is not strongly dependent on temperature.

There are two contributions to the anomalous Hall effect: the intrinsic and extrinsic contributions. The former is originated from the Berry curvature of the band structure, while the latter is due to the skew scattering and side-jump mechanisms. Separating the intrinsic and extrinsic contributions of the anomalous Hall effect is a challenging task. Traditionally, this is done by fitting the anomalous Hall conductivity versus longitudinal conductivity to the function $\sigma_{AH} = a\sigma_{xx}^2 + c$, where $c = \sigma_{AH}^{int}$ is the intrinsic contribution, and the $a\sigma_{xx}^2$ term corresponds to extrinsic skew scattering and side jump contributions [29]. However, it has been shown that this function does not adequately fit the anomalous Hall conductivity in TbMn_6Sn_6 . An additional non-standard term, tentatively associated with spin fluctuations as discussed in Ref. [30], can be included to achieve a better fit, leading to an equation of the following form:

$$\sigma_{AH} = a\sigma_{xx}^2 + c + d\sigma_{xx}^{-1} \quad (3.9)$$

We fitted the σ_{AH} versus σ_{xx} data for each sample, as shown in Fig. 3.12A and B for low and high doping levels, respectively, to extract σ_{AH}^{int} and σ_{AH}^{ext} . The dashed lines represent fits to the standard anomalous Hall equation, while the solid lines include an additional term proportional to $\frac{1}{\sigma_{xx}}$ (Eqn. 3.9). This additional term decisively improves the fit, especially for higher dopings.

The doping dependence of the fit parameters are presented in Table 1 and plotted in Fig. 3.13. It can be seen that the a coefficient associated with the skew scattering changes sign and increases dramatically as Cr concentration increases. This can be explained by the increase of disorder scattering due to Cr doping, and it is consistent with the substantial

drop of residual resistance ratio (RRR), as shown in Fig. 3.13A. The extracted non-standard parameter (d) increases roughly an order of magnitude over the measured dopings as shown in Fig. 3.13B, which is consistent with the enhanced Tb fluctuations corresponding to reduction of T_{sr} with increasing x . Also shown in this panel is a substantial enhancement of the extracted parameter c ($= \sigma_{AH}^{int}$) in orange. To further validate these fits, analyses using the resistivity form of the AHE equations are detailed in Supplementary Note 4 and yield similar results.

This extracted σ_{AH}^{int} with increasing x is plotted again in orange in Fig. 3.13C. Also shown is a simultaneous development of a large negative σ_{AH}^{ext} with increasing x (results shown for data taken at 50 K). Due to the opposite signs of the intrinsic and extrinsic contributions, the overall $|\sigma_{AH}^{total}|$ does not vary substantially with doping, and thus can be completely missed barring a proper scaling analysis.

Discussion

TbMn₆Sn₆ has attracted appreciable attention due to reports of a large intrinsic anomalous Hall effect, initially attributed to quasi two-dimensional gapped Dirac points, a characteristic feature of the kagome lattice band structure [13]. However, this interpretation has been challenged by recent calculations, which suggest that the relevant Dirac points are far from the Fermi level and that the intrinsic anomalous Hall effect primarily arises from the anti-crossing of energy bands at other locations in momentum space [30, 46, 40]. These studies further indicate that intrinsic anomalous Hall conductivity could be substantially enhanced if the material is hole-doped, a condition that can be achieved through Cr substitution.

The pronounced doping dependence of σ_{AH}^{int} observed in this work sheds more light on this debate. According to the gapped Dirac point model proposed in Ref. [13], σ_{AH}^{int} should decrease with Cr doping, as Cr doping lowers the Fermi level away from the gapped Dirac points, where the Berry curvature is concentrated. In contrast, the observed increase in σ_{AH}^{int} with Cr doping (i.e., hole doping) aligns better with scenarios proposed in Refs. [30, 46, 40]. To verify this, we calculate σ_{AH}^{int} as a function of hole doping by rigid band shift the Fermi level of TbMn₆Sn₆, as shown Fig. 3.13D. The good agreement between the experiment (orange curve in Fig. 3.13C) and the calculation (Fig. 3.13D) unambiguously confirms that multiple anti-crossing features in the band structure contribute to the intrinsic anomalous

Hall effect.

In summary, we successfully grew Cr-doped single crystals of TbMn_6Sn_6 and studied their magnetic and magnetotransport properties. Cr doping tunes the ferrimagnetic state towards magnetic compensation with a giant coercive field. Additionally, with increased Cr doping, σ_{AH}^{int} becomes substantially larger, in strong agreement with first-principles calculations. This work offers a pathway for identifying and synthesizing compensated ferrimagnets with large intrinsic anomalous Hall conductivities.

4 Pseudogap Behavior in ScV_6Sn_6

4.1 Background and Motivation

The AV_3Sb_5 ($A = \text{K, Rb, Cs}$) family of compounds contain kagome layers comprised of V, and they gained attention after it was found that they host charge density waves (CDW) below ≈ 80 K and superconductivity below ≈ 3 K. Upon further investigation, rich phase diagrams were found (see Ref. [50] for a review) with potentially broken time-reversal and rotational symmetry, leading to speculation of orbital current loop states and electronic nematicity. ScV_6Sn_6 also contains kagome layers made only of V, and (unique to the RT_6X_6 family at the time of discovery) was found to host a CDW below $T_{\text{CDW}} = 90$ K. Interestingly, despite this CDW being a first-order phase transition, studies showed that short-range CDW fluctuations exist well above T_{CDW} which do not exist in the AV_3Sb_5 compounds. In this chapter I present a comprehensive magnetotransport study on this material that provide evidence of pseudogap behavior in the normal state. This chapter is based on work previously published in Ref. [51]. Note that this chapter has been written with the understanding the field had on these materials in 2023; an updated introduction as of 2026 has been written for Chapter 5.

Charge Density Waves

A charge density wave is an ordered state of matter in which the charge carriers form a standing wave pattern that is accompanied by a periodic distortion of the lattice. There are multiple possible driving factors that contribute to the formation of a CDW in a given material including electronic degrees of freedom (related to the topology of the Fermi surface) and lattice degrees of freedom (ex. unstable phonon modes). In general (of course) these factors conspire to differing degrees to realize the resultant ground state ordering.

The chronologically first attempt at describing charge density waves (and most basic example of this phenomenon) is known as a Peierls distortion [52] which is shown schematically in Fig. 4.1 (figure adapted from Prof. Crommie's group website from UC Berkeley). In this model, a 1D chain of ions with lattice spacing a is considered. The lowest energy Bloch band will simply be a parabola. If each ion contributes two electrons then this band will be completely filled and no distortion will occur. However, if each ion contributes only one electron then this band will only be half filled (to $k_F = \pm \frac{\pi}{2a}$). In this case the electronic

energy of the system can be lowered by opening up a gap at this wave vector, pushing unfilled states to higher energies and filled states to lower energies. This is accomplished by the system undergoing a distortion that moves every other ion closer to one neighbor and farther from the other neighbor, doubling the period of the lattice and thus halving the size of the Brillouin zone, opening up energy gaps at $k_F = \pm \frac{\pi}{2a}$. This raises the total elastic energy of the system, but if the electronic energy lowering outweighs this elastic energy gain then this transition is energetically favorable. Thus, this is an electronically driven CDW.

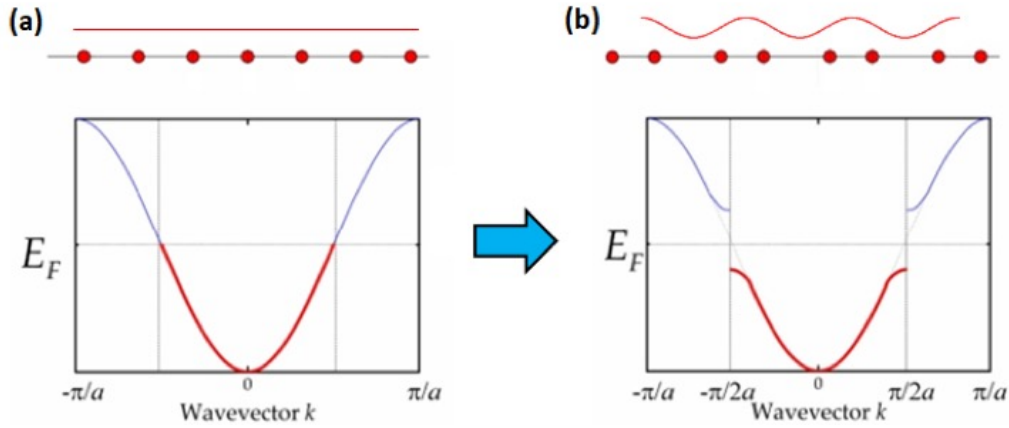


Figure 4.1: **Peirels distortion** (figure adapted from Prof. Crommie’s group website from UC Berkeley). **a**, A 1D chain of ions, each of which is contributing one electron to the Fermi sea. **b**, The same chain of ions after going through a Peirels distortion. Note that a gap has been opened in the electronic dispersion.

Note that in this case the parts of the Fermi surface that connect in order to complete this phase transition are connected by the reciprocal lattice vector $\frac{\pi}{a}$. This idea of “Fermi surface nesting” holds in higher dimensional systems. In general, if large parallel parts of a Fermi surface can be connected by a reciprocal lattice vector the material can be susceptible to a CDW distortion [53]. This is due to a divergence (technically only a divergence in one dimension, but still a sharp change in two dimensions) in the electronic susceptibility such that the system would be unstable even if the ions were somehow clamped to their original positions.

On the other hand, lattice degrees of freedom can also cause a CDW if there is strong electron-phonon coupling [54]. In these cases a lattice instability drives a structural phase transition and the electronic degrees of freedom then follow this new periodicity. Pieces of evidence for a structurally driven CDW like this often stem from a lack of possible Fermi

surface nesting and the hardening of a phonon mode below the charge density wave transition temperature.

To slightly editorialize here, I would like to note that categorizing charge density wave transitions as “structurally driven” or “electronically driven” is somewhat misleading. Both the electronic and elastic factors are key in determining whether a material will undergo such a transition (although the relative importance of each of these factors varies material to material). In fact, some argue that the concept of Fermi surface nesting driving phase transitions in real materials is likely almost never the case; perfect Fermi surface nesting is never realized and electron-phonon coupling likely plays a key role in nearly every charge density wave [55].

AV₃Sb₅ Family

The AV₃Sb₅ family first caught the condensed matter community’s attention when it was reported that these materials hosted CDWs with $T_{CDW} \approx 80$ K and superconductivity below about 3 K (shown in panels a and b of Fig. 4.2) and contained kagome layers comprised solely of V [50]. Interestingly, the electron band structure of these materials contain kagome-derived vHs almost exactly at E_F at the M point; it was quickly noted that Fermi surface nesting could describe the 2×2 in-plane CDW ordering in this family (panel c) [56]. Furthermore, a field-induced anomalous Hall effect was reported in these materials that onset at T_{CDW} (panel d), implying that time-reversal symmetry was broken by this phase transition [57]. With these realizations, significant interest came about as the field was interested to see how these interesting phases were related to the kagome-derived band structure.

However, as more and more experimental probes were applied to these systems, the story became increasingly muddled. The reported phase diagrams became increasingly complex (see Fig. 4.3 from Ref. [50]), but issues began to arise. For a variety of reasons (likely including widely varied sample quality and apparent extreme sensitivity to various external parameters [59]) different groups began to report very different results. Two concrete examples with different explanations include the out-of-plane charge density wave ordering vector and the existence of nematicity (rotational symmetry breaking order) in CsV₃Sb₅. The former of these can be reasonably easily explained by the quasi-2D nature of these crystals. As such, the interplane crystallinity is relatively poor, leading to a variety of different stacking faults. In turn, X-ray and neutron scattering data (typical ways to determine

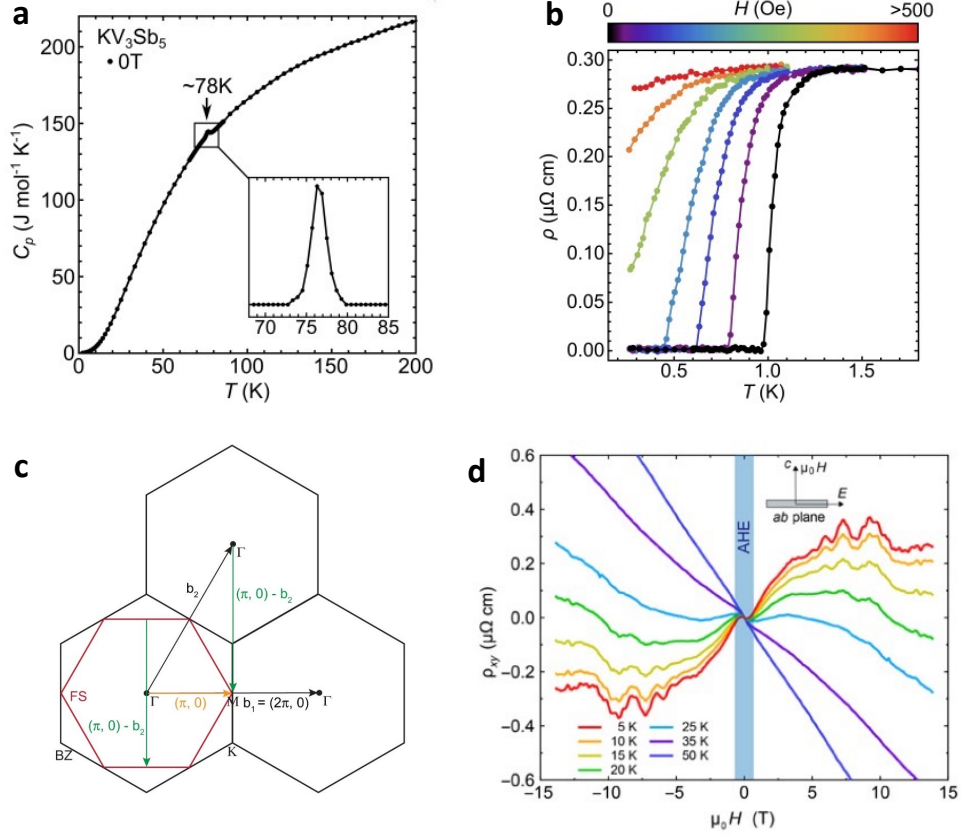


Figure 4.2: **AV₃Sb₅ basics.** **a**, Heat capacity anomalous of the CDW in KV₃Sb₅ [58]. **b**, Resistivity evidence of superconductivity in KV₃Sb₅ [58]. **c**, Fermi surface nesting in the AV₃Sb₅ family [56]. **d**, An apparent field-induced anomalous Hall effect in KV₃Sb₅ [57].

the ordering wave vector) from different studies led to a variety of different proposed wave vectors including $2 \times 2 \times 2$, and $2 \times 2 \times 4$ (for example, see Ref. [60]). Thus, this is an intrinsic issue with this system that (perhaps) very careful crystal growth efforts could solve. On the other hand, this idea of nematicity in CsV₃Sb₅ arose in part from the use of outdated (and improper) elastoresistance measurements [61]. When replication of these results were attempted in Jiun-Haw's lab led by Zhaoyu Liu, we quickly realized that proper measurements of the elastoresistance of this material reveal a distinct *lack* of nematicity [62]. It is likely that many of the contradicting results on these materials actually arise from a variety of these factors.

To again get on my soapbox, these issues of reproducibility in condensed matter physics are paramount to get under control. While some of these issues are out of the hands of the scientists doing the work, the push for exciting results (ex. a new symmetry breaking phase in an exciting materials that can be connected to a model band structure) is poisoning the

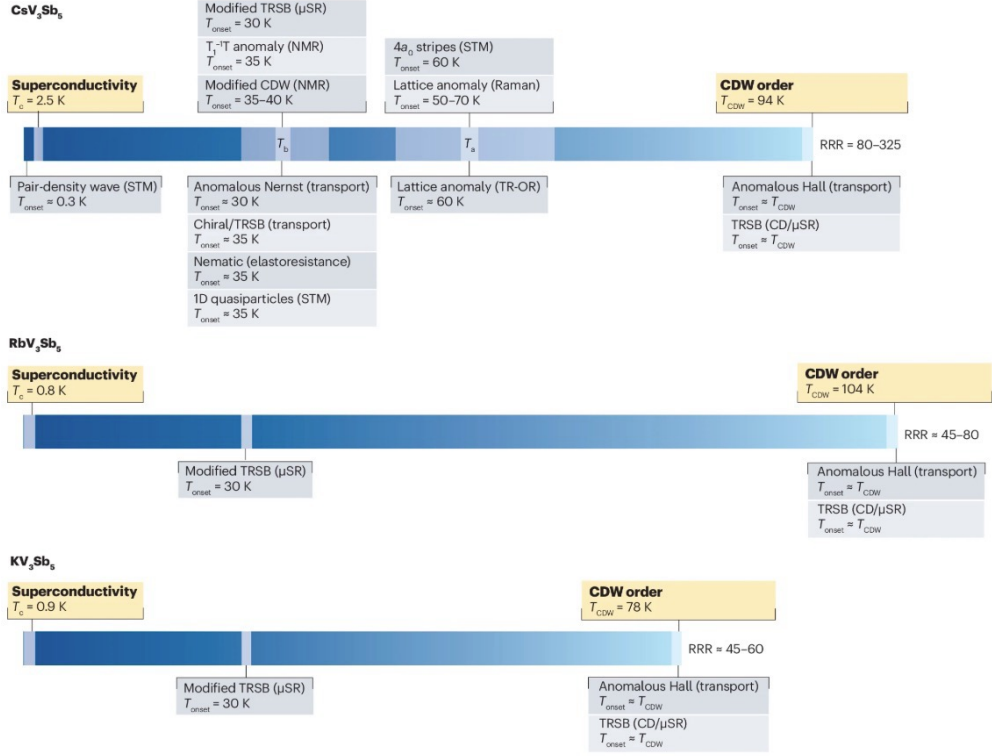


Figure 4.3: AV_3Sb_5 phase diagrams [50].

field. For example, Ref. [61] was published in Nature (one of the most prestigious scientific journals) and has received 523 citations in 4 years and 2 months whereas the *correct* null result was published in Physical Review X and has received 31 citations in 1 year and 9 months. This highlighting of exciting “results” overall harms the actual understanding of the science that is being investigated.

Regardless, at this time it seemed that kagome-derived charge density waves held promise for a wide variety of interesting phases of matter. As such, the search for kagome materials hosting CDWs became a hot topic in this field.

ScV₆Sn₆

The first CDW in the RT_6X_6 family was reported by David Mandrus’ group in ScV₆Sn₆ in 2022 [15]. At first glance this material actually seemed quite similar to the AV_3Sb_5 family: T_{CDW} is approximately 90 K, the kagome lattice is formed by V ions, and signatures of a field-induced anomalous Hall effect below T_{CDW} in both ρ_{xy} and ρ_{zx} (indicating, once again, potential time-reversal symmetry breaking) were quickly observed. However, there were also obvious differences: the crystals themselves are significantly more three-dimensional,

the ordering wave vector is $\sqrt{3} \times \sqrt{3} \times 3$, no superconductivity was observed, and the CDW itself was reported to be primarily structurally driven [63, 64, 65]. Nevertheless, one particularly intriguing unique feature related to this CDW did emerge: despite the CDW transition being first order (as evidenced the hysteresis loop in this transition as a function of temperature), short-range fluctuations of a CDW with a different wave vector ($\sqrt{3} \times \sqrt{3} \times 2$) were observed above T_{CDW} that extended up to at least 300 K [66, 67, 68]. The principle result of this chapter is that these fluctuations provide a “pseudogap”-like response (ie. a part of the Fermi surface is gapped out by these fluctuations). This is primarily realized by a reduction of the carrier density in ScV_6Sn_6 as temperature decreases above T_{CDW} which we observed through magnetotransport measurements.

Relevant Magnetotransport Principles

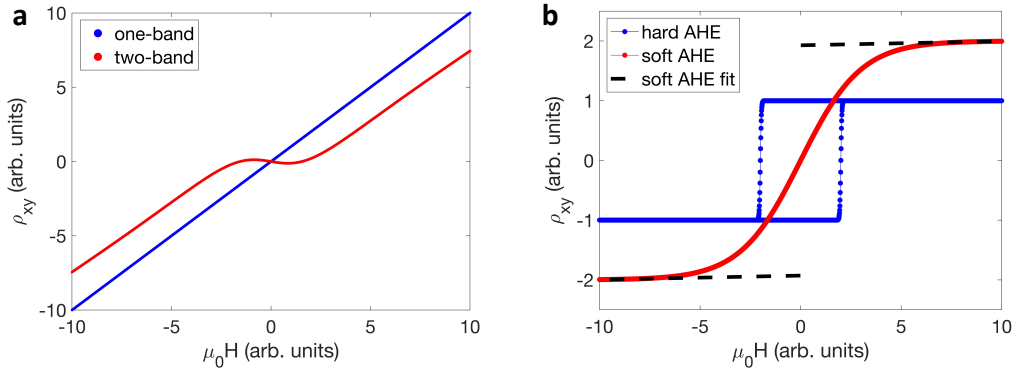


Figure 4.4: **Hall effect basics.** **a**, Examples of one- and two-band Hall effects. **b**, Examples of hard and soft anomalous Hall effects.

The standard ordinary Hall effect (OHE) applies to systems with a single carrier type and single mobility. It predicts that $\rho_{xy} = R_H \mu_0 H$ with $R_H = \frac{1}{ne}$ (with n being the carrier density and e being the elementary charge) and $\mu = \frac{R_H}{\rho_{xx}}$ (with μ being the carrier mobility). However, this simple linear relationship breaks down when both electrons and holes are contributing to magnetotransport. In this case, a two-band Hall formula is used of the form

$$\rho_{xy} = \frac{\mu_0 H}{e} \frac{(n_h \mu_h^2 - n_e \mu_e^2) + (n_h - n_e) \mu_h^2 \mu_e^2 (\mu_0 H)^2}{(n_h \mu_h^2 + n_e \mu_e^2) + (n_h - n_e)^2 \mu_h^2 \mu_e^2 (\mu_0 H)^2} \quad (4.1)$$

with the subscripts n and e referring to holes and electrons, respectively [69]. This leads to non-linearity in the low field regime, although in high field regime linear behavior returns in the form $\rho_{xy} = \frac{\mu_0 H}{(n_h - n_e)e}$. Examples of one- and two-band Hall effects are shown in Fig. 3.11a.

Note that this formula only allows for a single “bend” in the Hall response as a function of magnetic field when positive field is applied (with negative field the response is, of course, antisymmetric with respect to magnetic field). Furthermore, notice that this equation really does not make much sense in the case of metals with many bands crossing the Fermi energy; this model implicitly assumes that all of the holes in the system have the same mobility (and same for the electrons) which obviously does not have to be the case. In principle more bands can be added to the above model by simply adding more terms, but the issue with this is that adding more terms can allow the fitting of virtually any shape. Thus, researchers tend to stay away from using more than the two-band model for fitting.

As discussed in Ch. 3, the AHE is an additional contribution to the Hall resistance that mirrors the magnetization of the sample. In the proceeding chapter I focused on “hard” magnets in which the hysteresis loop is reasonably square and the anomalous Hall resistivity is finite at zero magnetic field. However, in the case of “soft” magnets the magnetization is zero at zero magnetic field but is easily polarized with a magnetic field. In turn, the AHE is zero at zero magnetic field but becomes finite with finite field. In these cases reserachers often quantify an AHE by extrapolating the linear Hall in the field saturated regime back to zero field as shown in Fig. 3.11b (for example, see Ref. [70]). As mentioned previously, one of the early indicators that the CDWs in the AV_3Sb_5 family stems from an apparent AHE that was observed (see Fig. 4.2d). A key part of this claim stems from the fact that the two-band model can not explain these data; there are two “bends” in the positive field regime. Thus, the high field “bend” was attributed to a two-band effect and the low field “bend” was ascribed to an AHE. As will be discussed in Chapter 5, this assumption has been shown to be incorrect due to the very complex Fermi surfaces real kagome metals typically have. As will be shown in Sec. 4.2, two-band model fitting works well for $T > T_{CDW}$ but fails in the low-field regime for $T < T_{CDW}$ which we tentatively attributed to a possible AHE in ScV_6Sn_6 in line with literature. However, our more recent results presented in Chapter 5 point to this simply being a result of this material having a complex Fermi surface.

Besides fitting Hall effect data, we also used an extension of Kohler’s rule to provide evidence of a strange change in the carrier density of ScV_6Sn_6 as a function of temperature. In general, the magnetoresistance can be described as a function of $\omega_c \tau$ with ω_c being the cyclotron frequency and τ being the scattering time [71]. From basic relations we know that $\omega_c \propto \mu_0 H$ and $\tau = \frac{m^*}{ne^2 \rho_0}$ (m^* is the effective mass, n is the carrier density, and ρ_0

is the zero-field resistivity), and so the magnetoresistance can be written as $\frac{\Delta\rho}{\rho_0} = F\left(\frac{\mu_0 H}{\rho_0}\right)$ with F simply being an arbitrary function; this is Kohler's rule. This rule assumes that the carrier density does not change as a function of temperature (note that temperature is generically used to control ρ_0). An extension of Kohler's rule was proposed in Ref. [72] in which the carrier density *is* allowed to change as a function of temperature. As such, the magnetoresistance is expressed as $\frac{\Delta\rho}{\rho_0} = F\left(\frac{\mu_0 H}{n_T \rho_0}\right)$ where n_T describes the relative change of carrier density as the temperature is changed. This formulation is particularly useful when dealing with systems with a very low carrier density (where temperature plays a large role in determining the number of carriers that can actively participate in transport) or systems in which the carriers are gapped out as a function of temperature. Here we will show that in ScV_6Sn_6 the extracted hole carrier density (the dominant carrier type by several orders of magnitude) and n_T have very similar temperature dependencies in the normal state, offering evidence of pseudogap-like behavior.

With this intro, I now reproduce Ref. [51] with small changes below to describe our results.

4.2 Published Results

Introduction

Materials containing kagome lattices have emerged as a promising platform for studying the interplay of electronic correlations and topology [73, 27, 74, 13]. Among these, kagome metals hosting charge density waves have gained significant attention due to their symmetry breaking phases and rich phase diagrams [70, 75, 76]. The $AV_3\text{Sb}_5$ ($A = \text{K, Rb, Cs}$) family hosts a charge density wave (CDW) with T_{CDW} at ≈ 80 K. Although controversies remain [77], the CDW potentially breaks time reversal symmetry [78] and rotational symmetry [79, 80, 81], leading to speculation about an orbital current loop state [82, 83, 84] and electronic nematicity [61]. At lower temperatures (< 3 K), a superconducting state coexists and competes with the CDW [58, 75, 76], displaying signatures of a pair density wave [85]. Many of these phenomena resemble the characteristics of other strongly correlated systems, such as high temperature superconductors, where the extended fluctuation regime gives rise to intertwined orders and complex phase diagrams [86].

ScV_6Sn_6 is the latest addition to the set of kagome metals exhibiting CDWs, with a

CDW transition temperature near 90 K [15]. Since this compound contains kagome layers comprised solely of V ions, it is natural to compare it to the AV_3Sb_5 family. However, early studies have found several distinct differences between ScV_6Sn_6 and these compounds. In ScV_6Sn_6 the CDW is associated with a $\sqrt{3} \times \sqrt{3}$ in-plane ordering [15], which is different from the 2×2 ordering in AV_3Sb_5 where the wave vectors nest the van Hove singularities of the kagome-derived energy bands. In ScV_6Sn_6 the lattice distortion associated with the CDW is mostly along the c-axis [15] whereas the distortion in AV_3Sb_5 is mostly in the ab-plane [87]. Unlike the AV_3Sb_5 family, no superconductivity has been found in ScV_6Sn_6 down to 40 mK even under high pressures [88]. Nevertheless, similar to the AV_3Sb_5 family, signatures of time reversal symmetry breaking have been suggested by muon spin relaxation rate measurements and a potential anomalous Hall effect [89, 90, 91]. Recent measurements, including scanning tunneling microscopy, angle-resolved photoemission spectroscopy, and Raman spectroscopy suggest the CDW is primarily structurally driven [63, 64, 65], resulting from the softening of a multitude of phonon modes [66, 68, 92], indicating a minor role of the electronic degrees of freedom in the CDW formation. However, despite the first-order nature of the CDW transition, recent studies have revealed short-range CDW fluctuations persisting well above T_{CDW} in ScV_6Sn_6 [66, 67, 68]. Hence, it is crucial to examine whether these fluctuations impact the electronic properties, as observed in other strongly correlated systems.

In this paper, we present evidence of a pseudogap above the CDW transition in ScV_6Sn_6 . Pseudogap formation was first observed in the cuprate superconducting family, and it refers to the suppression of the density of states which was revealed by various spectroscopy measurements and anomalous transport behavior [93]. Our conclusion of pseudogap formation in ScV_6Sn_6 is established from a comprehensive magnetotransport study, including measurements of the interlayer resistivity, magnetoresistance, and the Hall effect, all consistent with an abnormal decrease of carrier density with decreasing temperature above the CDW transition. In addition, we found several striking similarities to the proposed pseudogap phase in the Fe-based superconductors, in which strong spin density wave fluctuations persist well above the transition temperature. Our results suggest that there is an extended fluctuation regime in ScV_6Sn_6 which strongly influences the electronic transport properties above the transition temperature.

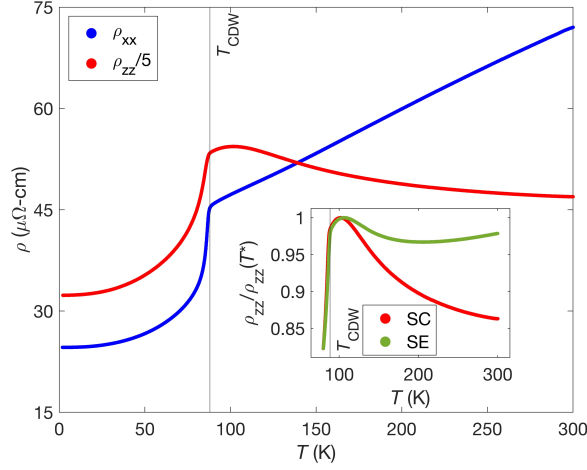


Figure 4.5: **Resistivity vs. temperature for ScV_6Sn_6 .** ρ_{xx} and ρ_{zz} (divided by 5) of ScV_6Sn_6 as a function of temperature. T_{CDW} is marked with a vertical line. Inset: ρ_{zz} of two different samples each normalized to be 1 at T^* as discussed in the main text. Here “SC” and “SE” refer to Sample C and Sample E respectively. T_{CDW} is marked in the same way as the main figure.

Results

Zero-Field Resistivity

Fig. 4.5 presents in-plane resistivity, ρ_{xx} , and interlayer resistivity, ρ_{zz} (divided by 5), of a typical ScV_6Sn_6 sample as a function of temperature while cooling. ρ_{xx} is consistent with previous reports [15, 88], with residual resistivity ratios of various samples ranging from 3-10. Drops in resistivity are present in both curves near 90 K, indicating the charge density wave transition (T_{CDW}) [15]. It should be noted that this is a first order transition, but only the cooling curve is shown due to the small temperature hysteresis of this transition (≈ 1 -2 K). As previously noted [88], charge density wave phase transitions in layered materials typically appear as hump-like increases in resistivity as a function of temperature, as parts of the Fermi surface get gapped out by the transition.

The drop of resistivity at T_{CDW} is reminiscent of the spin density wave transition in the parent compounds of the iron pnictide superconductors, such as BaFe_2As_2 , which is also characterized by a similar feature [94]. This unusual behavior in BaFe_2As_2 is understood as a more rapid decrease of scattering rates relative to the decrease of carrier density below the spin density wave transition. Like observed in BaFe_2As_2 , optical measurements of

ScV₆Sn₆ [95] have also revealed a similar decrease in both the carrier density and the scattering rate below T_{CDW} , which could explain the increase in conductivity. The enhanced electron scattering above T_{CDW} can be explained by the competing CDW fluctuations above T_{CDW} [67].

The interlayer resistivity measurements reveal a moderate resistivity anisotropy in ScV₆Sn₆ with ρ_{zz} roughly 5 times larger than ρ_{xx} at 2 K. This is considerably smaller than in CsV₃Sb₅ where ρ_{zz} is ≈ 20 times larger than ρ_{xx} at low temperatures [79], implying ScV₆Sn₆ is more 3-dimensional than CsV₃Sb₅. Nevertheless, unlike CsV₃Sb₅, the temperature dependence of ρ_{zz} is dramatically different from ρ_{xx} , showing a broad maximum roughly 15 K above T_{CDW} at T^* . As shown in the inset of Fig. 4.5, for temperatures above T^* multiple ScV₆Sn₆ samples display insulating behavior (here “SC” and “SE” refer to Sample C and Sample E, respectively). A summary of the samples measured can be found in Supplementary Table 2). The difference in high temperature resistivity between samples may be explained by contamination from lower resistivity in-plane components.

The insulating interlayer resistivity and metallic in-plane resistivity have been observed in highly anisotropic layered materials such as Sr₂RuO₄, in which the much weaker interlayer tunneling results in incoherent c-axis transport [96]. However, such a phenomenon is usually seen in materials with resistivity anisotropy $\rho_{zz}/\rho_{xx} \gg 10$, which is not applicable to ScV₆Sn₆ where $\rho_{zz}/\rho_{xx} \approx 5$. Interestingly, a similar insulating temperature dependence with a broad maximum in ρ_{zz} above a density wave transition has also been observed in BaFe₂As₂ [94, 97, 98, 99, 100], which also has a moderate resistivity anisotropy ($\rho_{zz}/\rho_{xx} \approx 7$). The insulating ρ_{zz} in BaFe₂As₂ was interpreted as signature of a pseudogap, resulting from the spin density wave fluctuations partially gapping the section of the Fermi surface where the Fermi velocity has a large z-component. We propose that a similar mechanism could be responsible for the insulating ρ_{zz} in ScV₆Sn₆, which is further supported by the Hall effect and magnetoresistance analysis presented in the following sections.

Hall Effect

Fig. 4.6(a) presents ρ_{xy} as a function of magnetic field at a variety of temperatures. These data are taken from Sample A which has a RRR of 6.0 and T_{CDW} of 93 K (A summary of T_{CDW} and RRR of the samples measured can be found in Supplementary Table 2). Across the entire measured temperature range ρ_{xy} is non-linear, but while it evolves smoothly as a

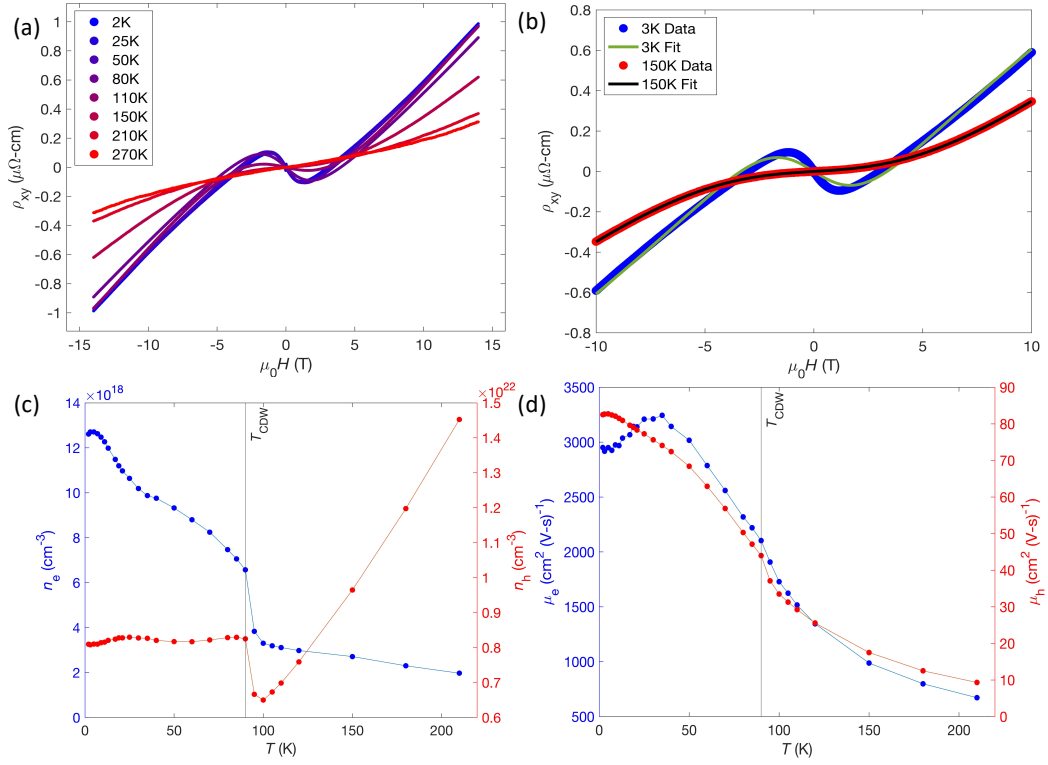


Figure 4.6: **Hall effect and two-band analysis.** (a) ρ_{xy} as a function of magnetic field at various temperatures. (b) ρ_{xy} as a function of magnetic field at 3 K and 150 K with fits to the two-band model. (c) n_e and n_h as a function of temperature extracted from fits to the two-band model. (d) μ_e and μ_h as a function of temperature extracted from fits to the two-band model.

function of temperature above T_{CDW} , it stays relatively unchanged below T_{CDW} . There are two possible sources for the non-linearity in ρ_{xy} : the multi-band effect and the anomalous Hall effect. We first present an analysis of fitting of ρ_{xy} using a two-band model, which reveals a strong temperature dependence of carrier density above the CDW transition. We will also argue that this conclusion can be made from the high field Hall coefficient of ρ_{xy} even without any two-band fitting.

In order to analyze the two-band Hall effect, the standard two-band model is used to simultaneously fit $\rho_{xx}(\mu_0 H)$ and $\rho_{xy}(\mu_0 H)$ at each temperature [69]. The carrier densities and mobilities were determined by a non-linear least squares minimization of the error $(\rho_{xy} - \rho_{xy}^{\text{fit}})^2 + C(\rho_{xx} - \rho_{xx}^{\text{fit}})^2$ where C provided a weighting such that ρ_{xy} was prioritized (as ρ_{xx} has potentially more scattering contributions than those arising from the two-band model). Fig. 4.6(b) shows the fits to ρ_{xy} at 150 K and 3 K. Above T_{CDW} the two-band model fits ρ_{xy} well, but below T_{CDW} the two-band fit loses quality, particularly in the low field regime. This disagreement may be due to the contribution from an anomalous Hall effect [91, 90]. Fig. 4.6(c) and Fig. 4.6(d) present the carrier densities and mobilities, respectively, that are extracted from the two-band fitting. It should be noted that the quantitative values below T_{CDW} should not be taken as exact due to the decrease in fit quality in ρ_{xy} . At all temperatures n_h is greater than n_e by several orders of magnitude. Notably, above T_{CDW} n_h decreases significantly, although n_e is roughly constant, and below T_{CDW} n_h is roughly constant while n_e grows by several times. Both μ_e and μ_h increase as temperature decreases which is typical for a metal.

Due to the substantial deviation from the two-band model of the Hall data below T_{CDW} , we estimate the carrier density using the Hall data in the high field limit in two more ways to ensure our conclusions are robust. First, the Hall coefficient, $R_H = \frac{\rho_{xy}}{\mu_0 H}$, should saturate to $\frac{1}{(n_h - n_e)e}$ at the high field limit where e is the charge of an electron [71]. These data are shown in Fig. 4.7(a). The blue data in Fig. 4.7(c) present the extracted $n_h - n_e$ from the high field Hall coefficient. Second, the red data in Fig. 4.7(c) show n_h using $\frac{d\rho_{xy}}{d\mu_0 H}$ ($\mu_0 H = 14$ T) $\approx \frac{1}{n_h e} \cdot \frac{d\rho_{xy}}{d\mu_0 H}$ as a function of magnetic field is shown in Fig. 4.7(b). This method provides an estimate of carrier density even when the low-field data can not be well represented by the two-band model. While the three methodologies used to estimate the carrier densities vary quantitatively, they are all of the same order of magnitude and qualitatively consistent - above T_{CDW} the number of holes decreases as a function of temperature, and below T_{CDW}

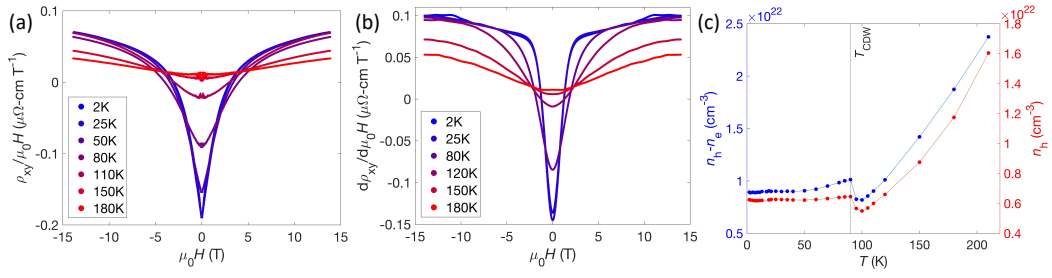


Figure 4.7: **High field limit Hall analysis.** (a) $\frac{\rho_{xy}}{\mu_0 H}$ as a function of $\mu_0 H$ to evaluate the high field limit of the two-band model. Note that the curves are not fully saturated at 14 T. (b) $\frac{d\rho_{xy}}{d\mu_0 H}$ as a function of $\mu_0 H$ to approximate the carrier concentration using a one-band model. (c) $n_h - n_e$ extracted from the high field limit of the two-band model and n_h from the high field regime of a one-band model. While quantitatively different than the results presented in Fig. 4.6 for reasons described in the main text, all three of these analyses are qualitatively consistent.

the number of holes is roughly constant.

Magnetoresistance (MR) and Kohler's Rule Analysis

The ab-plane MR ($\text{MR}_{xx} = \frac{\Delta\rho_{xx}}{\rho_{xx}(\mu_0 H=0)} * 100\%$) with c-axis magnetic field at various temperatures is shown in Fig. 4.8(a). These data are from Sample B which has a RRR of 4.3 and two drops in the resistivity at 80 and 88 K. This apparent double transition at T_{CDW} in ρ_{xx} has been noted before [90], and the meaning of this feature is not yet understood. This MR looks qualitatively similar to that observed in the AV_3Sb_5 family in that the low field behavior shows a cusp at low temperatures and evolves to a more standard quadratic behavior at high temperatures [57]. At low temperatures quantum oscillations can be resolved once a background subtraction is performed. The c-axis magnetoresistance (MR_{zz}) with a magnetic field in the ab-plane at several temperatures is presented in Fig. 4.8(b). These data are from Sample C which has a T_{CDW} of 86 K. The MR_{zz} of another sample was measured and was similar to the data presented here. While MR_{zz} looks qualitatively similar to MR_{xx} , MR_{zz} is about double the size at 2 K. Also, at low temperatures quantum oscillations are observed much more prominently in MR_{zz} . These quantum oscillations, as well as accompanying electronic structure calculations, are discussed in Supplemental Note 1 and are in agreement with a very recent study [91]. Overall, the quantum oscillations reveal three-dimensional Fermi pockets occupying less than a percent of the Brillouin zone with effective masses be-

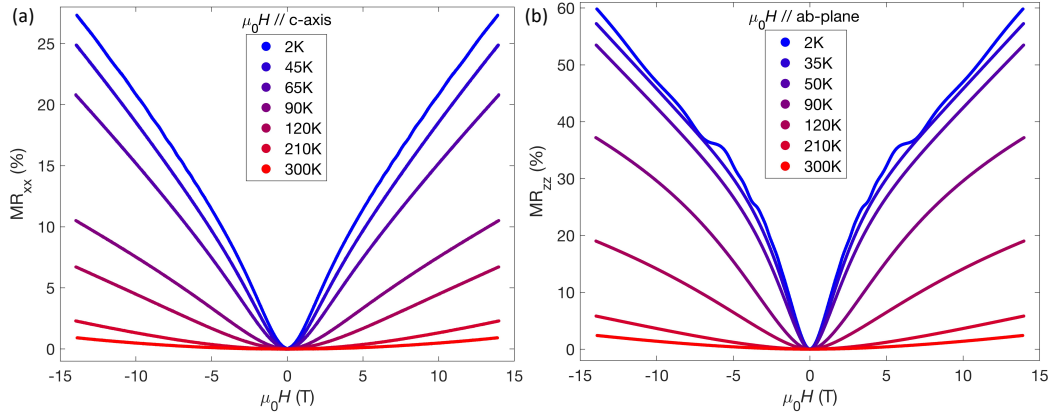


Figure 4.8: **Magnetoresistance of ScV_6Sn_6 .** (a) MR_{xx} as a function of magnetic field at various temperatures. (b) Magnetic field dependence of MR_{zz} at several temperatures.

tween 0.1 - 0.2 of the free electron mass. These pockets are consistent with the high mobility and low density of electron carriers extracted from the two-band Hall fitting.

Kohler's rule of magnetoresistance [101] is violated in ScV_6Sn_6 as shown in Fig. 4.9(a) as MR_{xx} is not simply a function of $\mu_0 H / \rho_{xx,0}$ where $\rho_{xx,0}$ is the zero-field resistivity. It should be noted that the data point spread at low fields and high temperatures is due to the small MR in this regime. The violation of Kohler's rule in ScV_6Sn_6 was also reported in a recent study [89]. Similar violations of Kohler's rule have been used as evidence of phase transitions [102] or non-Fermi liquid behavior [103, 104]. Recently, an extended Kohler's rule has been developed [72] in which the MR is expressed as a function of $\mu_0 H / (n_T \rho_{xx,0})$ where n_T describes the relative change in the carrier density. The extended Kohler's rule successfully explains the violation of conventional Kohler's rule by incorporating a temperature dependent carrier density, which could arise from a phase transition that partially gaps the Fermi surfaces or thermal excitations in topological semimetals where the Fermi energy is comparable to $k_B T$ [72]. Here we apply this formula with n_T fixed to be 1 at 255 K to collapse the MR curves onto the linear part of the 255 K MR (Fig. 4.9). Through this method, n_T can be extracted as a function of temperature, and is presented in the inset of Fig. 4.9(b). Intriguingly, Kohler's rule is nearly followed below T_{CDW} as evidenced by the nearly constant value of n_T , but above this temperature Kohler's rule is clearly violated. These results are consistent with those presented in Section 4.2 - n_h extracted from two-band fitting is also plotted in the inset of Fig. 4.9(b) to show the similar behavior between the concentration of the dominant carriers (holes) and n_T , as both decrease above T_{CDW} and become relatively

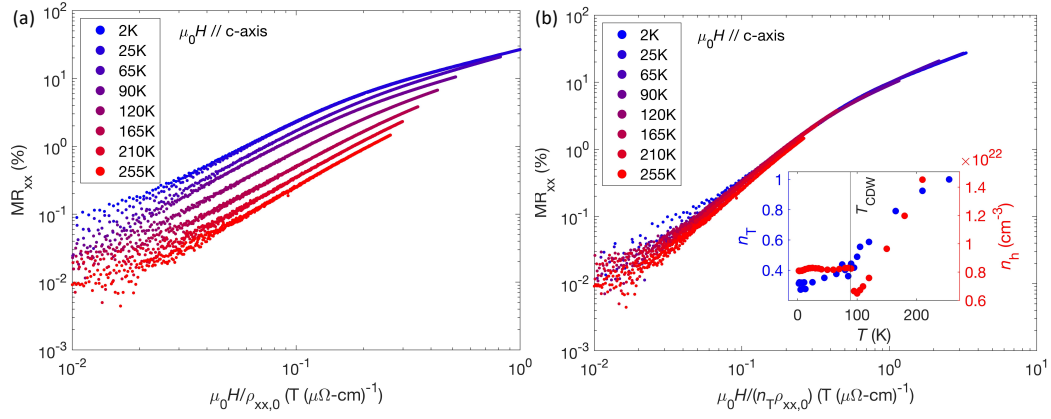


Figure 4.9: **Kohler's rule analysis of ScV_6Sn_6 .** (a) MR_{xx} as a function of $\mu_0 H / \rho_{xx,0}$ using the data from Fig. 4.8(a) on a log-log scale. Kohler's rule is violated as the data from different temperatures do not collapse onto each other. (b) Extended Kohler's rule applied to the same data as presented in (a) by plotting MR_{xx} as a function of $\mu_0 H / (n_T \rho_{xx,0})$ on a log-log scale. Inset: extracted n_T as a function of temperature. n_h extracted from the two-band Hall fitting as discussed in Section 4.2 is also plotted in the inset to highlight the similar temperature dependencies between n_h and n_T .

constant below T_{CDW} . Analysis of MR_{zz} data using the extended Kohler's rule yields similar results to that of MR_{xx} : above T_{CDW} n_T decreases quickly with decreasing temperature, and below T_{CDW} n_T changes far less drastically. These data are presented in Supplementary Note 2.

Discussion

Both Kohler's rule analysis and the Hall effect demonstrate a pronounced temperature dependence of the carrier density in ScV_6Sn_6 , decreasing by almost a factor of 2 from the value at 200K to the value just above the CDW transition temperature $T_{CDW} = 90 \text{ K}$. This decrease in carrier density is consistent with the insulating temperature dependence observed in interlayer resistivity. Interestingly, similar characteristics have also been observed in the Fe-based superconductors, such as $BaFe_2As_2$. The temperature dependence of in-plane and interlayer resistivity, as well as the resistivity anisotropy ratio, exhibit remarkable resemblance between $BaFe_2As_2$ and ScV_6Sn_6 [94]. Additionally, in $BaFe_2As_2$ the Hall coefficient also shows a substantial increase with decreasing temperature above the spin density wave transition [105, 106, 107]. This is also seen in the pseudogap regime of the cuprate supercon-

ductors [108]. Another striking similarity can be observed in the magnetic susceptibilities of BaFe_2As_2 and ScV_6Sn_6 (presented in [15]). In both materials there is a drop in the susceptibility at the phase transition, but these potentially stem from different mechanisms, for in BaFe_2As_2 this is likely due to enhanced magnetic correlations. Also in both materials the susceptibility shows a linear increase with increasing temperature above the phase transition, which cannot be explained by either Pauli paramagnetic susceptibility or Curie Weiss susceptibility [109, 110].

In BaFe_2As_2 , the anomalous transport and magnetic properties observed above the transition temperature have been attributed to strong spin density wave fluctuations. However, in the case of ScV_6Sn_6 , the CDW transition is first-order, which could explain the nearly temperature independent carrier density below T_{CDW} , but contradicts with the existence of an extended fluctuation regime above T_{CDW} . Nevertheless, theoretical studies have suggested that, in addition to the long range $\sqrt{3} \times \sqrt{3} \times 3$ CDW that develops below T_{CDW} , there are several other nearly degenerate CDW instabilities associated with different ordering wave vectors [66]. Furthermore, experimental evidence has shown the presence of a short-range $\sqrt{3} \times \sqrt{3} \times 2$ CDW well above T_{CDW} , which is suppressed by the $\sqrt{3} \times \sqrt{3} \times 3$ CDW through a first-order transition at T_{CDW} [67]. It is possible that the short-range CDW fluctuations are responsible for the anomalous decrease of carrier density and insulating interlayer resistivity observed in ScV_6Sn_6 .

It should be noted that a pseudogap in the BaFe_2As_2 family has been identified using a variety of techniques including angle-resolved photoemission spectroscopy (ARPES) [111] and optical conductivity [112]. While several works have presented ARPES [113, 114, 63, 65] and scanning tunneling microscopy [114, 63] data on ScV_6Sn_6 , to our knowledge no measurements were performed above 120 K. Thus, it is possible that future studies will reveal a pseudogap in this material using these experimental probes.

In conclusion, the transport behavior in the normal state of ScV_6Sn_6 is consistent with the formation of a pseudogap, which is likely arising from high temperature CDW fluctuations. We have also highlighted several similarities between ScV_6Sn_6 and Fe-based superconductors with pseudogaps above their ordering temperatures. Due to the high degree of tunability in the RT_6X_6 family (R = rare earth, T = transition metal, X = Si, Ge, Sn), ScV_6Sn_6 offers an exciting platform to study exotic electronic ordering in a kagome material.

Note: During the preparation of this paper we became aware of a separate study which

reported the two-band behavior of the Hall effect in ScV_6Sn_6 [90]. They discovered high carrier density and low mobility holes and low carrier density and high mobility electrons, which broadly corroborates our findings.

Methods

Sample preparation

Single crystals of ScV_6Sn_6 were grown using a flux method similar to one previously reported [15]. Mixtures of Sc pieces (99.9%), V pieces (99.9%), and Sn shot (99.999%) were loaded into Canfield crucible sets [16] with atomic ratios 1:3:30, then vacuum-sealed in quartz tubes. These were heated to 1150°C in 12 hours, held at this temperature for 15 hours, then cooled to 780°C in 200 hours where the growths were decanted in a centrifuge to separate the excess flux from the single crystals. Dilute HCl was used to etch the remaining flux from the surface of the crystals. The phase of the crystals was confirmed using energy-dispersive X-ray spectroscopy with a Sirion XL30 scanning electron microscope. The orientation of the crystallographic axes was determined using a Rigaku MiniFlex 600 system, with a Cu source and Hy-Pix 400MF 2D-detector.

Transport measurements

Transport measurements were performed on samples that were polished and cut by a wire saw to be bar-shaped with dimensions roughly $1\text{ mm} \times 0.4\text{ mm} \times 0.05\text{ mm}$ (in-plane current) or $0.2\text{ mm} \times 0.15\text{ mm} \times 0.05\text{ mm}$ (out-of-plane current). Silver paste or two-part silver epoxy (H20-E) and gold wires were used to make 4 point and 5 point (Hall pattern) measurements. These measurements were performed in a Quantum Design Dynacool Physical Property Measurement System with standard lock-in techniques in temperatures ranging from 1.7 K to 300 K and in magnetic fields up to 14 T. To eliminate any contributions from contact misalignment, the in-line and Hall resistivities were symmetrized and anti-symmetrized, respectively. For some of the measurements the samples were rotated *in situ* using a Quantum Design in-plane rotator.

Density functional theory calculations

Density functional theory (DFT) calculations were performed using a full-potential linear augmented plane wave (FP-LAPW) method, as implemented in WIEN2K [115]. The primitive cell contains one formula unit, and experimental lattice parameters [15] were adopted. The generalized gradient approximation of Perdew, Burke, and Ernzerhof [116] was used for the correlation and exchange potentials. To generate the self-consistent potential and charge, we employed $R_{\text{MT}} \cdot K_{\text{max}} = 8.0$ with Muffin-Tin (MT) radii $R_{\text{MT}} = 2.4, 2.4,$ and 2.5 a.u., for Sc, V and Sn, respectively. The self-consistent calculations were performed with 490 k -points in the irreducible Brillouin zone (BZ). They were iterated until charge differences between consecutive iterations are smaller than $1 \times 10^{-3}e$ and the total energy difference lower than 0.01 mRy. After obtaining self-consistent charge, band energies were calculated with a $64 \times 64 \times 33$ fine k -mesh for the full Brillouin Zone (FBZ).

We employed FermiSurfer [117] and SKEAF [118] for visualizing FS and calculating de Haas - van Alphen (dHvA) frequencies, respectively.

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Author Contributions

J.M.D., E.R., O.P., and Z.L. conducted the transport measurements. Y.L. and L.K. performed the DFT calculations. Q.J. assisted with data analysis. J.M.D. and O.P. grew the samples. J.-H.C. oversaw the project. J.M.D., E.R., and J.-H.C. wrote the manuscript with input from all authors.

Competing Interests

The authors declare no competing interests.

Data Availability

All data supporting the findings of this study are available upon request.

5 Angle-Dependent Magnetoresistance in ScV_6Sn_6

5.1 Background and Motivation

The growth of high quality samples has become increasingly important as it has become more and more apparent that the degree of disorder in a material is important for determining the electronic properties and phase transitions they exhibit. For example, as discussed in Section 2.1 we worked extensively on developing the horizontal flux method to grow high quality samples of WTe_2 as the Cobden lab realized that extremely high quality single crystals were necessary to observe superconductivity in monolayer samples. In this chapter I will present my work on growing high quality ScV_6Sn_6 ; the basic characterization of these crystals (and Cr doped samples) reveals that the “AHE” that had been previously observed seems to simply be due to a multi-band effect and that the charge density wave transition in this material is more complex than previously thought. Furthermore, these high quality samples allow us to clearly measure the angle-dependent magnetoresistance (ADMR) of this material which reveals an intriguing crossover from 3D to quasi-2D magnetotransport near 55 K (T'), well below T_{CDW} .

Relevant Updates on AV_3Sb_5 and ScV_6Sn_6

At this point, the potential TRS breaking induced by the CDW in the AV_3Sb_5 family had been disputed for several years with several measurements of muon spin relaxation and magneto-optic experiments showing conflicting reports [77, 119]. The one piece of evidence that had not been systematically ruled out was the apparent AHE. However, a report in 2025 finally seems to put a large hole in this story [120]. In this paper, the magnetotransport of (originally) high quality samples of CsV_3Sb_5 ($\text{RRR} > 100$, “RRR” refers to the residual resistivity ratio: $\frac{\rho_{xx}(T=300\text{K})}{\rho_{xx}(T=2\text{K})}$) were systematically measured. Between each measured data set the sample was irradiated with electrons which is known to create point defects without changing the lattice constants or carrier concentration of the sample. This decreased the mobility of the samples in a relatively controlled way without changing the Fermi surface. Note that the changing the mobility (μ) will change the field scale of standard carrier-related magnetotransport responses ($\mu * \mu_0 H$ is a dimensionless parameter) but should not change any field scale of an AHE associated with a TRS breaking response. In total, this work revealed that tiny Fermi pockets with extremely high mobility carriers that form below T_{CDW}

are responsible for the AHE-like response in the low-field regime. Here we will show strong evidence that the contribution to ρ_{xy} in ScV_6Sn_6 that can not be described by the two-band model is likely due to similar physics. We accomplish this by varying the carrier mobility through careful growth of high quality ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ (which offers the additional tuning knob of changing carrier density).

These high quality crystals also allow us to address a different mystery that ScV_6Sn_6 presents: it was reported in 2025 [121] that in “high quality” samples ($\text{RRR} \approx 9$) the charge density wave transition is actually comprised of *two* transitions separated by about 7 K. Interestingly, between these two transitions the authors argue for a “nematic state” that may stem from remnants of the high temperature CDW fluctuations coexisting with the static low temperature CDW. It is not surprising that higher quality samples reveal new physical behaviors — disorder generically is important for determining the electronic properties and unconventional phase transitions in materials. As I will show, we manage to grow significantly higher quality samples of ScV_6Sn_6 (up to $\text{RRR} > 50$) and show that as RRR continues to increase this phase transition becomes more and more complex with our highest quality samples showing at least 3 distinct thermodynamic anomalies near the nominal T_{CDW} .

5.2 Growth of High Quality ScV_6Sn_6

The results of this subsection are currently being drafted for submission to *Physical Review Materials*.

Crystal Growth

Single crystals of ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ were grown utilizing a flux method similar to those previously reported for ScV_6Sn_6 [15]. 15 gram mixtures of Sc pieces (99.9%), V pieces (99.9%), Cr powder (99.95%), and Sn shot (99.999%) were loaded and subsequently vacuum sealed in quartz tubes with atomic ratios $\text{Sc}:(\text{V,Cr}):\text{Sn}$ 1:3:80. These were heated up to 1150°C in 12 hours, held at this temperature for 12 hours, then cooled to 700°C in 120 hours. Then the growths were decanted in a centrifuge to separate the excess flux from the crystals. Some of the V pieces were cleaned by being heated to 800°C for about 12 hours in flowing Ar/H_2 gas (5% molar concentration of hydrogen) and were then stored in an Ar glovebox. The highest RRR ScV_6Sn_6 and all of the $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ samples were

synthesized with this clean V. For the other ScV_6Sn_6 growths, cleaned V pieces were mixed with as-received V pieces in varying ratios. This allowed for a relatively smooth tuning of crystal quality. Some of the crystals grown in this way had several secondary phases with superconducting transitions embedded in them including Sn and V_3Si . Samples were carefully polished and cut to attempt to exclude these phases, and samples which showed significant superconducting transitions in transport measurements are not reported.

Note that several changes were instituted here as compared to a standard vertical flux growth I described in Section 2.1. First, these crystals were grown directly in quartz tubes without using alumina crucibles; in ScV_6Sn_6 crystals I grew in alumina crucibles I could often detect Al in the samples when performing EDX measurements. This led me to experiment with growing the samples without crucibles which led to higher RRRs. While some Si is clearly getting into the samples (as evidenced by the contributions of V_3Si in some of the samples), the effects of the Si incorporation seem to be less than that of the Al on average. The next difference is simply the quantity of starting reagents used. Here I was growing samples with a 15 gram growth as opposed to the 3 gram growth I typically use. This led to an increase in RRR that we attribute to having a larger volume:surface area ratio of the melt, likely limiting the amount of Si impurities that are introduced. Lastly, I treated the V pieces by heating them under H_2 . This made the V pieces significantly shinier which we attribute to the hydrogen removing surface oxygen from the V. This seems to be a somewhat generic useful treatment of transition metals; for example, Chaowei found that cleaning W in this way greatly increased the RRR of the resultant WTe_2 .

With these crystals of ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ in hand, we can now turn to basic characterization of these materials.

Phase transitions

Samples of ScV_6Sn_6 with varying degree of disorder were grown by changing the percentage of H_2 -cleaned V pieces included in the starting materials for the crystal growth. The relative degree of disorder is inferred by the RRR. Fig. 5.1a presents the temperature dependence of in-plane resistivity (ρ_{xx}) normalized by the 300 K value for samples with a variety of RRR. The charge density wave transition is clearly observed in these measurements by a sudden drop in resistivity when the temperature is cooled through $T_{CDW} \approx 90$ K. Interestingly, the size of the drop of ρ_{xx} at T_{CDW} does not change significantly with RRR. Instead, the slope

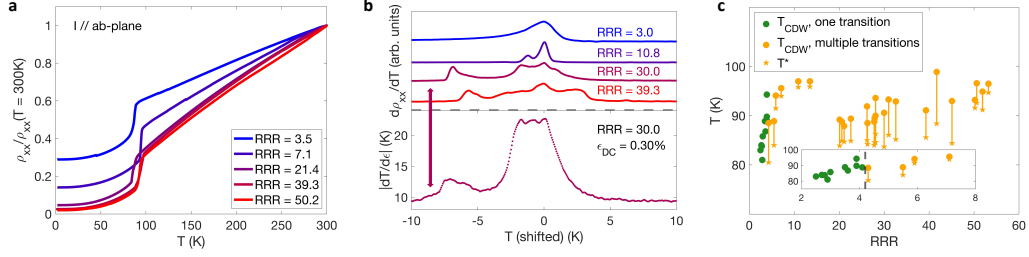


Figure 5.1: **Phase transitions in reduced disorder ScV_6Sn_6 .** **a**, Normalized ρ_{xx} as a function of temperature for samples of ScV_6Sn_6 with a variety of RRR. **b**, Top: offset $\frac{d\rho_{xx}}{dT}$ for several samples of ScV_6Sn_6 . These have been shifted in temperature such that the peak of $\frac{d\rho_{xx}}{dT}$ aligns with 0 K for ease of comparison. Bottom: elastocaloric effect on the same temperature scale for the sample with RRR = 30.0 with a DC tensile strain applied. **c**, Measured transition temperatures plotted against the in-plane RRR for many samples of ScV_6Sn_6 . Samples with a single transition are shown as green dots, and samples with multiple transitions are shown in orange with a dot indicating the onset of the phase transitions and a star representing the offset of the phase transitions as detected by $\frac{d\rho_{xx}}{dT}$. Inset: a zoom in focusing on the low RRR regime. A black line indicates an apparent cutoff between samples with one transition and samples with multiple transitions.

of ρ_{xx} increases significantly as a function of RRR above T_{CDW} . This could be related to the high temperature CDW fluctuations that have previously been shown to impact the magnetotransport of ScV_6Sn_6 [51].

The top panel of Fig. 5.1b displays $\frac{d\rho_{xx}}{dT}$ near T_{CDW} for several samples with RRR ranging from 3.0 to 39.3. It can be seen that in the sample with RRR = 3.0 there is only a single peak at T_{CDW} . However, for the sample with RRR = 10.8 the single peak splits into two distinct peaks. As RRR increases, each of the two peaks further splits to more peaks. To confirm that the splitting is an intrinsic property of the sample, we measured the elastocaloric effect ($\frac{dT}{d\varepsilon}$), which has been shown to be proportional to the heat capacity anomaly near a phase transition [122]. The results of a sample with RRR = 30.0 are shown in the bottom panel of Fig. 1b. It can be clearly observed that there are two clusters of peaks in $\frac{dT}{d\varepsilon}$, in good agreement with what was observed in $\frac{d\rho_{xx}}{dT}$ measured in the same sample (shown in the top panel), providing thermodynamic evidence of splitting of the CDW transitions in high RRR_{xx} samples. The RRR dependence of transition temperature and splitting is summarized in Fig. 5.1c collected from nearly 40 samples of ScV_6Sn_6 . Green data points indicate samples

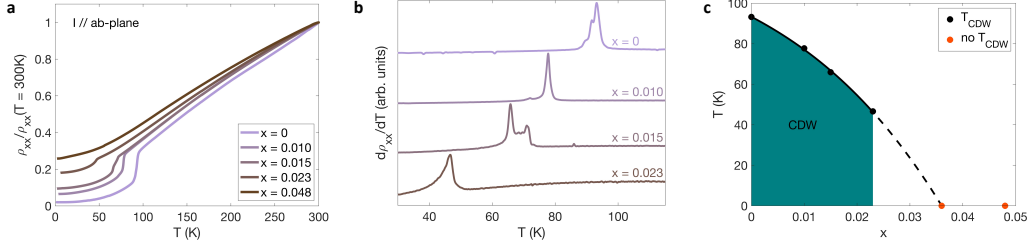


Figure 5.2: **Phase diagram of reduced disorder $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$.** **a**, Normalized ρ_{xx} as a function of temperature for samples of $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ with a variety of x . **b**, Offset $\frac{d\rho_{xx}}{dT}$ for several samples of $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$. **c**, Temperature-composition phase diagram for $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ constructed from the resistivity data in panels a and b. Only the onset transition temperature is shown for simplicity.

with a single peak in $\frac{d\rho_{xx}}{dT}$ and orange data points represent samples with multiple peaks in $\frac{d\rho_{xx}}{dT}$ and their splitting size in temperature. When zooming in the low RRR regime (Fig. 5.1c inset), a sharp boundary between samples with low RRR and single transition and samples with higher RRRs and multiple transitions can be observed, which is marked by a dashed line near $\text{RRR} = 4$. We note that previous work reported the splitting of the CDW transition in a sample of ScV_6Sn_6 with $\text{RRR} = 10$, and it was observed that the resistivity is anisotropic between the two transitions [121]. Further work is required to elucidate the symmetry of these transitions.

To compare the effects of disorder and charge doping, samples of $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ were grown by introducing Cr into the same recipe that produces the highest RRR ScV_6Sn_6 samples. Fig. 5.2a presents ρ_{xx} normalized by the 300 K value as a function of temperature for samples of $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$. As expected, RRR decreases with increasing x due to the inherent disorder introduced by the Cr substitution. In agreement with previous studies, T_{CDW} is suppressed with increasing x [123, 124] as can clearly be seen in the $\frac{d\rho_{xx}}{dT}$ curves shown in panel b of this figure. Note that the low x (and correspondingly high RRR) samples still show multiple transitions, whereas the $x = 0.023$ (low RRR) sample shows only a single feature. These results are summarized in an $x - T$ phase which is shown in Fig. 5.2c. For simplicity only T_{CDW} is shown. This decrease in T_{CDW} with increasing x has been postulated to be due to the reduction of the c-axis lattice constant with increasing x [123].

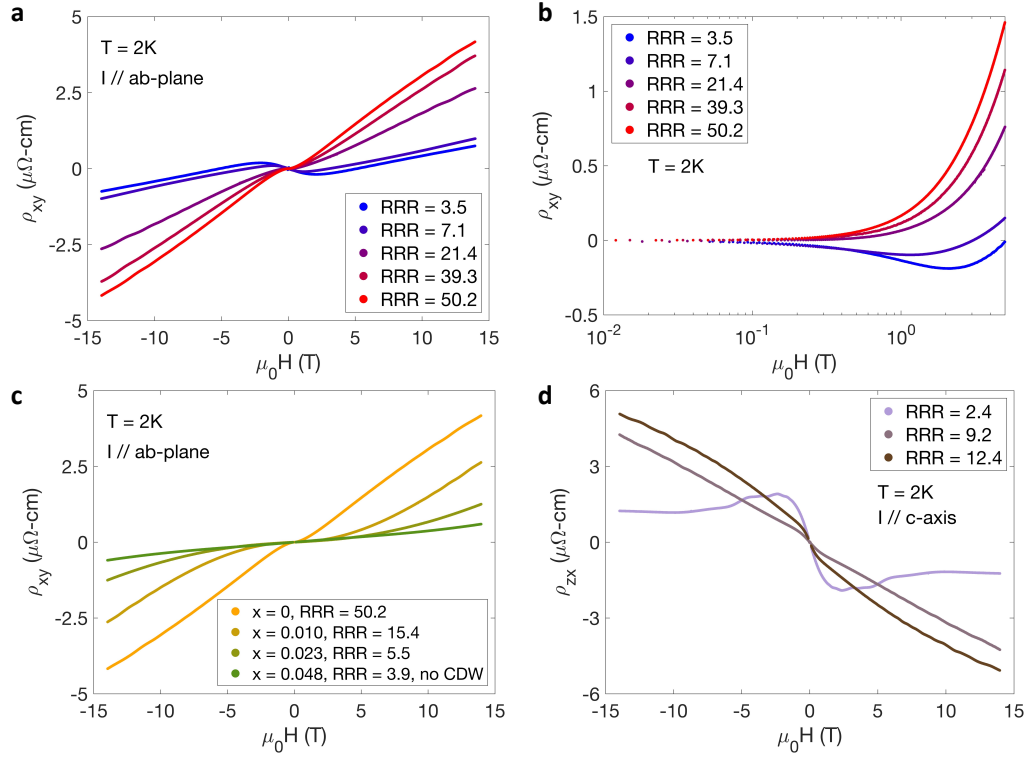


Figure 5.3: **Hall effect of ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$.** **a**, ρ_{xy} as a function of $\mu_0 H$ measured at 2 K for samples of ScV_6Sn_6 with a variety of in-plane RRR. **b**, The same data as panel a plotted with the magnetic field on a log scale. **c**, ρ_{xy} as a function of $\mu_0 H$ measured at 2 K for samples of $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ with a variety of x . **d**, ρ_{zx} as a function of $\mu_0 H$ measured at 2 K for samples of ScV_6Sn_6 with a variety of out-of-plane RRR.

Hall effect

Fig. 5.3a presents Hall resistivity (ρ_{xy}) as a function of magnetic fields at 2 K for samples with a variety of RRR. Nonlinear ρ_{xy} can clearly be observed in the low-field regime, in agreement with previous reports [90, 51]. The non-linear Hall effect is often interpreted in two ways: a signature of multi-band transport, especially when high mobility small Fermi pockets coexist with large Fermi pockets, and the anomalous Hall effect resulted from time-reversal symmetry breaking. In a multi-band system, the magnetic field scale at which the nonlinear Hall is observed depends strongly on the mobilities of Fermi pockets. In the inset of Fig. 5.3a, we notice that the field scale of non-linearity in ρ_{xy} decreases with increasing RRR (and correspondingly increasing mobility μ), which is a characteristic of a multi-band ordinary Hall effect. This is further emphasized in Fig. 5.3b where the same data is presented as in panel a albeit with the magnetic field axis on a log scale. These results were fit to a two-band model using the methodology described in Ref. [69]. In short, the results of this fitting implied that the electron and hole mobilities increase with RRR, which is consistent with the decreased disorder in higher RRR samples, but also implied that the electron carrier density increased several orders of magnitude with increasing RRR (a consequence of the increase in the high field slope of ρ_{xy} with increasing RRR). It will be shown in Sec. 5.2 with quantum oscillation measurements that E_F does not change with increasing RRR, making this apparent change in the carrier density a clear indicator that the simple two-band model does not apply to ScV_6Sn_6 . While it has been previously noted that the two-band model does not perfectly fit ρ_{xy} in ScV_6Sn_6 at temperatures below T_{CDW} [51], the fitting gets progressively worse as the RRR increases.

A further piece of evidence that the two-band model does not apply to this system comes from Cr doping of ScV_6Sn_6 . Fig. 5.3c presents ρ_{xy} as a function of $\mu_0 H$ at 2 K for samples of $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ with various x . The low-field non-linearity quickly moves to higher field scales with increasing x (and thus decreasing μ), consistent with a multi-band effect. Furthermore, the decrease in the slope of ρ_{xy} at the highest fields measured implies the system is becoming increasingly hole doped with increasing x in contradiction to standard chemical intuition and the quantum oscillation results in Sec. 5.2. This implies that 14 T is not a large enough magnetic field to reach the high-field limit of this system where a field-induced AHE can robustly be extracted.

It has also been conjectured that ρ_{zx} in ScV_6Sn_6 may show an AHE component [125].

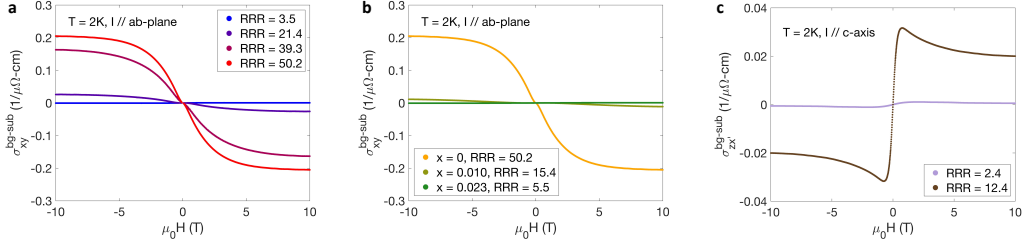


Figure 5.4: **Hall anomaly in ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$.** **a**, σ_{xy} with a linear contribution subtracted as a function of $\mu_0 H$ for ScV_6Sn_6 with a variety of RRR_{xx} at 2 K. **b**, Same as panel a for $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ with a variety of x . **c**, Same as panel a for $\sigma_{zx'}$ (described in the main text).

This argument is largely based on the shape of ρ_{zx} as a function of $\mu_0 H$ at low temperatures in low out-of-plane (ie. $I \parallel c$ -axis) RRR samples. Fig. 5.3d presents ρ_{zx} as a function of $\mu_0 H$ at 2 K for samples with varying out-of-plane RRR. It can be seen that with increasing RRR the shape of ρ_{zx} dramatically changes, and the low-field anomaly is quickly reduced in field scale which is consistent with a multi-band effect.

To further confirm a lack of AHE in our samples we present the Hall conductivities in this section with $\sigma_{ij} = \frac{-\rho_{ij}}{\rho_{ii}\rho_{jj} + \rho_{ij}^2}$. Fig. 5.4a presents σ_{xy} with a high-field linear contribution subtracted (notated by the superscript “bg-sub”, data without this subtraction can be found in Supplementary Note YYY) as a function of $\mu_0 H$ at 2 K for samples of ScV_6Sn_6 with varying RRR. It has been shown in ScV_6Sn_6 that $\rho_{xx} \neq \rho_{yy}$ at temperatures near T_{CDW} , but this equality nearly recovers at low temperatures [121] so here we assume that $\rho_{xx} = \rho_{yy}$. Note that in the highest RRR sample there is no clear anomaly in the low-field regime (< 5 T) unlike what is visible in the ρ_{xy} data seen in Fig. 5.3a. Furthermore, there is a rapid decrease σ_{xy} with decreasing RRR (and thus decreasing σ_{xx}). In the case of an intrinsic anomalous Hall effect, the anomalous Hall conductivity should be independent of σ_{xx} [29]; thus, this dependence of σ_{xy} with σ_{xx} is consistent with an ordinary multi-band Hall effect. Similarly, decreasing the RRR by Cr doping also leads to a drastic reduction in σ_{xy} as seen in Fig. 5.4b.

Fig. 5.4c shows $\sigma_{zx'}^{bg-sub}$ as a function of $\mu_0 H$ for ScV_6Sn_6 at 2 K. Here $\sigma_{zx'}$ indicates that ρ_{xx} data from the lowest and highest in-plane RRR samples we measured were used for converting the resistivity data to conductivity for the lowest and highest out-of-plane RRR samples, respectively. Unlike σ_{xy} , σ_{zx} of high-quality samples shows a clear anomaly in the low-field regime. However, like the case of σ_{xy} , σ_{zx} decreases significantly in magnitude

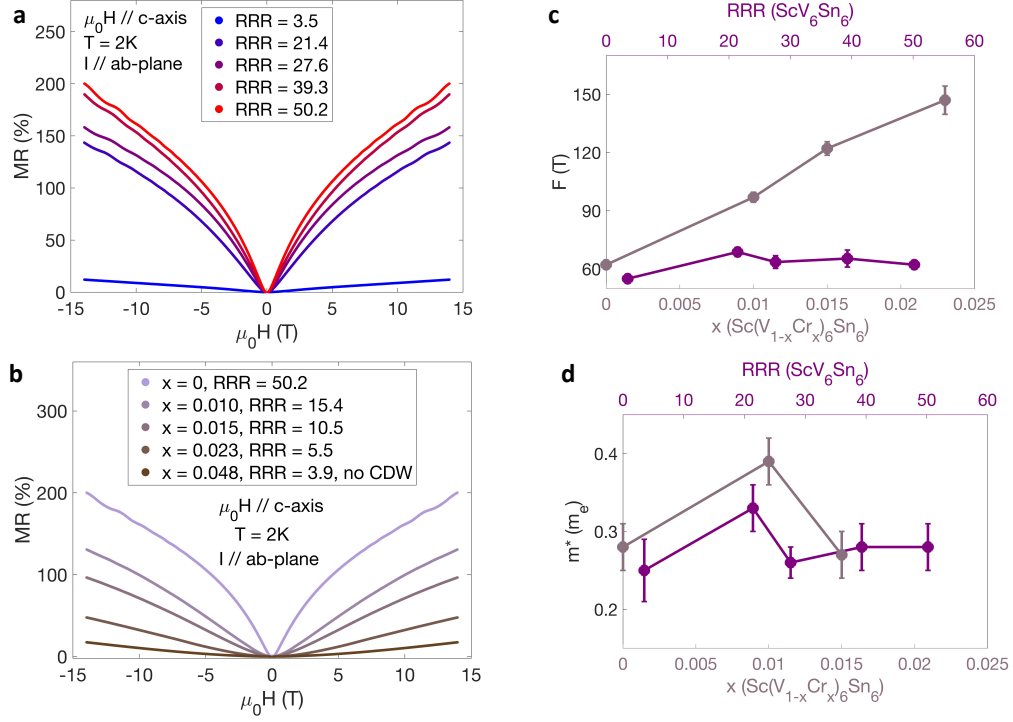


Figure 5.5: **In-plane magnetoresistance and quantum oscillations of ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$.** **a**, MR_{xx} as a function of $\mu_0 H$ at 2 K with an out-of-plane field for samples of ScV_6Sn_6 with a variety of RRR_{xx} . **b**, MR_{xx} as a function of $\mu_0 H$ at 2 K with an out-of-plane field for samples of $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ with a variety of x . **c**, Quantum oscillation frequency measured at 2 K as a function of RRR_{xx} for ScV_6Sn_6 (top x-axis, shown in purple) and as a function of x for $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ (bottom x-axis, shown in gray). **d**, Effective mass extracted from the temperature dependence of the frequency using the same axes and colors as panel e.

with decreasing RRR_{zz} indicating once again that this Hall anomaly is not due to an intrinsic anomalous Hall effect.

Magnetoresistance and quantum oscillations

In this section the magnetoresistance ($\text{MR}(\%) = \frac{\rho(\mu_0 H) - \rho(\mu_0 H = 0T)}{\rho(\mu_0 H = 0T)} * 100\%$) and quantum oscillations of ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ will be discussed. Fig. 5.5a presents the MR of ScV_6Sn_6 with varying in-plane RRR as a function of $\mu_0 H$ (applied parallel to the c-axis) at 2 K. As expected, higher RRR samples show a higher MR consistent with increased μ in these samples. Note that the highest RRR sample has an MR roughly an order of magnitude larger than that of the lowest RRR sample. Quantum oscillations can clearly be observed

by eye in the higher RRR samples. At high magnetic fields the MR in high RRR becomes increasingly sub-linear as compared to lower RRR samples, indicating partial saturation of MR due to the increased mobility in the high RRR samples. As a direct comparison to ScV_6Sn_6 , Fig. 5.5b presents the MR as a function of $\mu_0 H$ at 2 K for $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ with differing x . With increasing x the MR quickly decreases indicating that μ decreases as RRR_{xx} decreases as expected.

A fast Fourier transform (FFT) of the data presented in Fig. 5.5a and b reveals a single quantum oscillation frequency in good agreement with a previous study on ScV_6Sn_6 [126]. Notably, the quantum oscillations of $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ have not been reported and were likely only able to be measured due to the increased sample quality detailed in this paper. This frequency was extracted for each sample at 2 K using standard Landau level indexing and is shown in Fig. 5.5c in purple for ScV_6Sn_6 as a function of RRR_{xx} (top x-axis) and in gray for $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ as a function of x (bottom x-axis). In the ScV_6Sn_6 samples this small frequency (≈ 60 T) does not change notably as a function of RRR indicating that the Fermi energy (E_F) is not changed in these samples. However, for $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ the frequency more than doubles with only about 2% Cr doping on the V site implying this is an electron pocket, in agreement with first principles calculations which reveal a number of small electron pockets around the edges of the Brillouin zone [51]. Fig. 5.5d presents the effective mass (m^*) of this pocket in units of the free electron mass (m_e) for both ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ using the same color and axes conventions as panel c. The effective masses are determined by fitting the temperature dependence of the FFT peak amplitude for each sample to the damping term (R_T) in the Lifshitz-Kosevich formula, namely

$$R_T \propto \frac{\alpha(m^*/m_e)T/\mu_0 H_{avg}}{\sinh(\alpha(m^*/m_e)T/\mu_0 H_{avg})} \quad (5.1)$$

where $\mu_0 H_{avg}$ is the average of the magnetic field used in calculating the FFT and $\alpha \approx 14.69\text{T/K}$. Neither with increasing RRR for ScV_6Sn_6 nor with increasing x in $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ is a substantial change in m^* observed, implying that there is no significant change of the band structure with either of these parameters.

Takeaways

The quantum oscillation measurements performed on our high-quality ScV_6Sn_6 and $\text{Sc}(\text{V}_{1-x}\text{Cr}_x)_6\text{Sn}_6$ samples reveal a small and light electron pocket in good agreement with first principles cal-

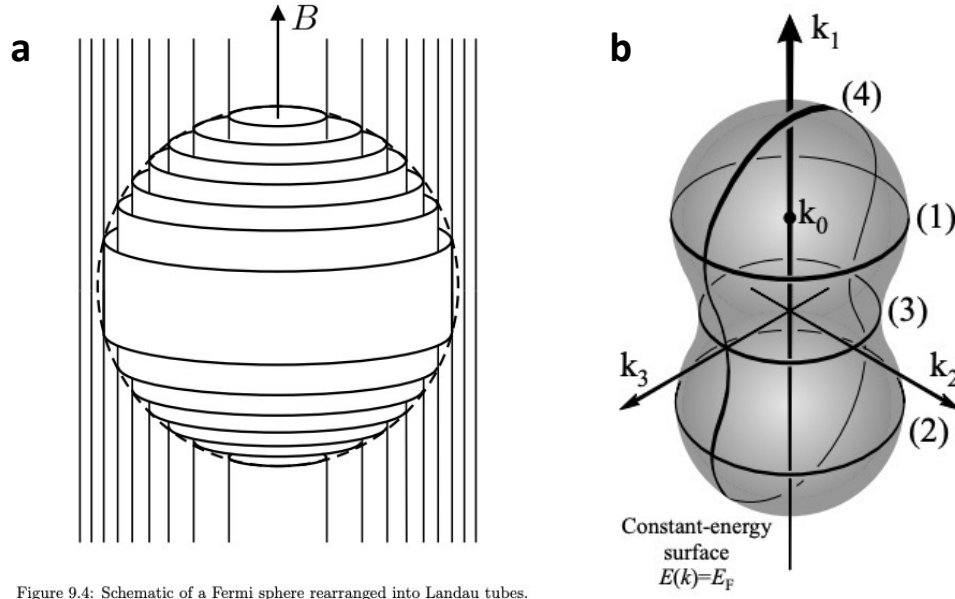


Figure 9.4: Schematic of a Fermi sphere rearranged into Landau tubes.

Figure 5.6: **Quantum oscillation basics.** **a**, The formation of Landau tubes from a Fermi sphere upon the application of a magnetic field. **b**, An example Fermi surface with the extrema marked.

culations. Furthermore, a thorough study of the Hall effect in these samples indicates that the residual resistivity ratio (and correspondingly the carrier mobility) is correlated to the magnitude and field scale of the Hall anomalies that were proposed to arise from an AHE linked to the charge density wave in this system. The combination of all of these facts indicates that this Hall anomaly likely stems from contributions of many small pockets with high mobilities, mirroring the situation that was observed in CsV_3Sb_5 [120]. These results call for general caution with the analysis of the Hall effect in metallic systems with multiple bands contributing to transport properties especially when the pockets vary significantly in sizes and mobilities. In these cases the standard one- or two-band Hall models break down, leading to Hall resistivities with seemingly non-trivial contributions. Comparing samples with different mobilities offers a reasonably straightforward method of ruling out intrinsic AHEs as seen here.

5.3 Angle-Dependent Magnetoresistance Measurements

Fermiology

Kagome materials originally gained significant attention due to their interesting band structure features, so it is only natural that I spent some of my time working on these materials studying their Fermiology. A very standard way to measure the Fermi surface of materials is by measuring how a thermodynamic or transport quantity oscillates with the application of a “large” (actual necessary size depends on the system) magnetic field [127]. The basic for this process stems from the formation of Landau levels; when a magnetic field is applied to a system, the constituent charged particles will undergo cyclotron motion. Quantum mechanically this problem maps to the quantum simple harmonic oscillator, leading to evenly spaced solutions of the form

$$E_n = \hbar\omega_B\left(n + \frac{1}{2}\right) \quad (5.2)$$

where $\omega_B = \frac{eB}{m^*}$. These states are massively degenerate (in a way that is proportional to B). In the simplest case in three dimensions with just a Fermi sphere, this application of a magnetic field can be pictured as taking this Fermi sphere and breaking it into Landau tubes oriented with their long axis along the magnetic field with different energies as shown in Fig. 5.6a. As the magnetic field is changed, the energies of these states will then change, and as they sequentially pass through E_F any quantity that is sensitive to the density of states at the Fermi energy will oscillate periodically in $1/B$. This fundamental frequency can be directly related to the extremal parts of the Fermi surface normal to the field applied through the Onsager relation $\frac{1}{\Delta(1/B)} = \frac{\hbar}{2\pi e} A_{ext}$ where A_{ext} is the extremal area. For a concrete example, see Fig. 5.6b: with field applied along k_1 the areas 1 and 2 (degenerate) and 3 can be detected, and with field along k_2 (or k_3 , or anywhere in the plane defined by these two vectors) area 4 can be detected. Thus, by rotating the sample and sweeping the magnetic field, one can map out the Fermi surface of a material.

Beyond just this powerful feature, even more information can be extracted. By studying the temperature dependence of the quantum oscillations one can learn about the effective mass of the detected pockets (this basically stems from the fact that temperature smears out the Fermi-Dirac distribution. In order to measure quantum oscillations you need to be roughly in the $\hbar\omega_B \geq k_B T$ regime.). Furthermore, the scattering length, scattering time, and

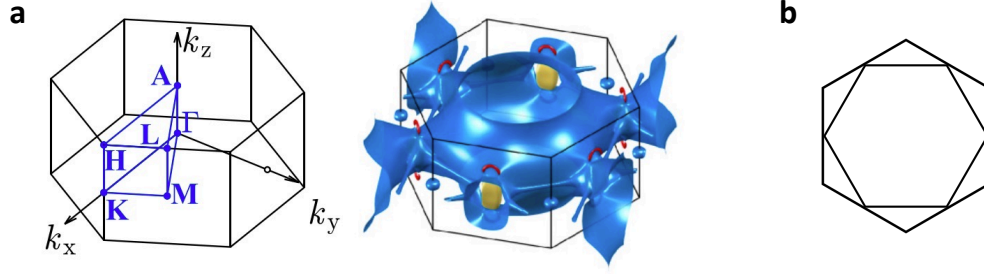


Figure 5.7: **RT_6X_6 Fermi surface.** **a**, Calculated Fermi surface of ScV_6Sn_6 in the CDW state [125]. **b**, Schematic of kagome-derived vHs pocket near E_F .

Fermi velocity can all be estimated.

Despite the power of this technique, some experimentally difficult issues do arise. First: since $\hbar\omega_B \geq k_B T$ must roughly be true in order to observe quantum oscillations, very light effective mass pockets are significantly easier to detect than heavier pockets. Furthermore, an intrinsic damping term to measurements of quantum oscillations comes about in all measurements: $I_{measured} = I_{intrinsic} e^{-\pi r_c/l}$ where l is the scattering length and $r_c = \frac{\hbar k_F}{eB}$. Thus, in order to measure quantum oscillations it is beneficial to measure very clean crystals and very small pockets.

This issue becomes abundantly clear when measuring the quantum oscillations of members of the RT_6X_6 family. Fig. 5.7a displays the calculated Fermi surface of ScV_6Sn_6 in the CDW state [125]. Note that the CDW transition does not significantly alter the band structure and this band structure is quite representative of the generic RT_6X_6 band structure. There is a single large hole pocket that is open along M and along the c -axis that is shown in blue; this is a quasi-2D pocket that comes about from the vHs near E_F in these materials. The small electron pockets around the edges of the Brillouin zone (blue, red, and yellow) are generically quite light ($m^* \approx 0.1m_e$ where m_e is the free electron mass. In standard laboratory settings ($\mu_0 H \leq 15$ T) quantum oscillation measurements of various materials in the RT_6X_6 family show basically the exact same thing: just very small, very light pockets (ex. Refs. [13, 125]). Measuring the vHs pocket is non-trivial due to both its large size (note that if this vHs sat perfectly at E_F at the M points this pocket would take up 75% of the Brillouin zone as shown in Fig. 5.7b) and enhanced effective mass. In fact, our measurements (headed by Elliott Rosenberg) of the paramagnet YV_6Sn_6 is one a rare example of measuring this pocket through the use of quantum oscillations [128]: through the use of a 41.5 T magnet at the National High Magnetic Field Laboratory we managed to detect a

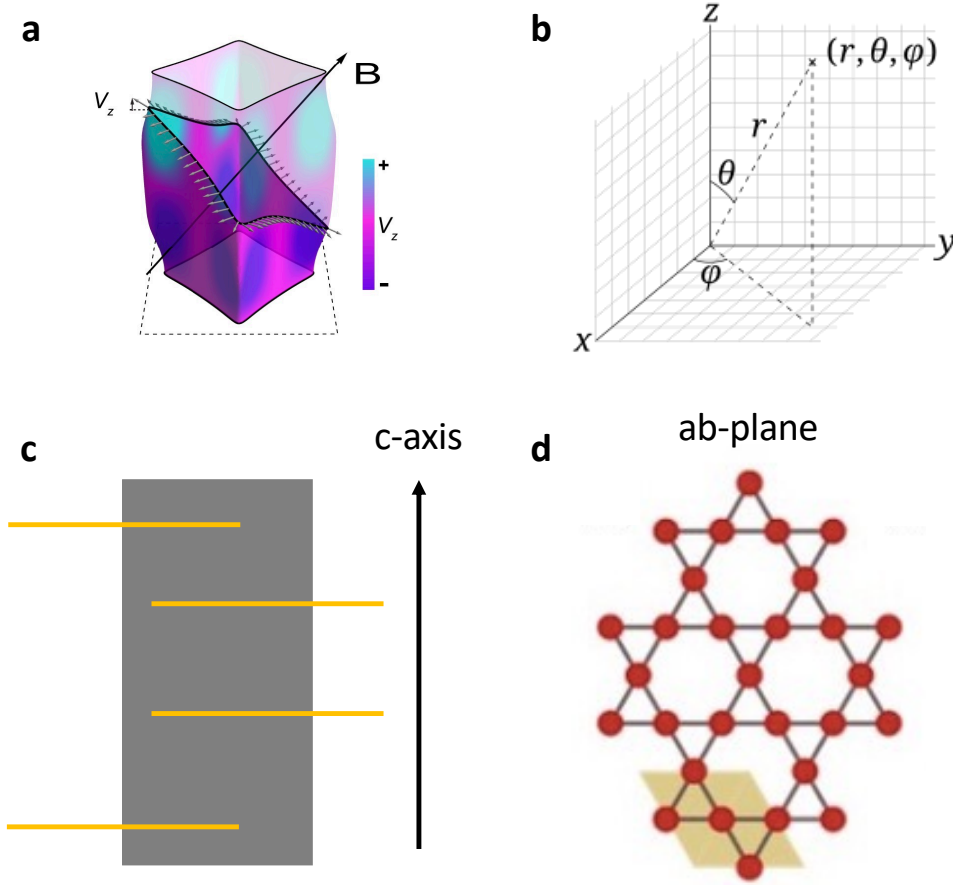


Figure 5.8: **ADMR basics.** **a**, Schematic of ADMR [130]. **b**, Overview of the coordinate system that will be used in this chapter. **c**, Schematic of the ρ_{zz} measurements that will be presented in this chapter. **d**, A reminder of the C_6 symmetric kagome lattice that lies in the ab -plane of the RT_6X_6 family.

pocket that was quasi-2D, had an effective mass of $3.3m_e$, and takes up about 70% of the Brillouin zone. Similar results have been reported for the antiferromagnet GdV_6Sn_6 [129], driving home the similar band structure and filling between members of the RT_6X_6 family. While these works are interesting, the requirement of such a high magnetic field to study the Fermiology of these systems makes these studies difficult. Here, I will show that the use of angle-dependent magnetoresistance (ADMR) as an alternative technique combined with the high-quality samples of ScV_6Sn_6 that I grew allows us to measure similar physics using an in-house cryostat and magnet.

ADMR is, in principle, quite simple. The magnetoresistance is measured at a fixed magnetic field while the sample is rotated in the magnetic field. Resistivity is sensitive to the lifetimes and velocities of quasiparticles on the Fermi surface; these quantities determine

the scattering rate and, in turn, the resistivity. A Lorentz force alters these velocities by causing cyclotron motion around the edges of the Fermi surface, and as the field is rotated the Lorentz force (and corresponding cyclotron motion) will be changed as shown schematically in Fig. 5.8 [130]. Thus, the ADMR can give information about the Fermi surface. Historically this technique has been used to study a wide variety of systems including the organic conductors [131] and the cuprates [130]. In order to be able to resolve the ADMR, the system needs to meet the condition that $\omega_c \tau \gg 1$ (ie. many cyclotron orbits need to be completed). While clean materials are beneficial to measure this technique effectively as the carriers can complete more cyclotron orbits without scattering, the conditions for observing an ADMR is much less restrictive than for observing quantum oscillations. This will be the key for allowing us to identify evidence of the vHs pocket in ScV_6Sn_6 with ADMR using just a laboratory magnet. The downside of ADMR as compared to quantum oscillations is that ADMR is much more difficult to interpret than quantum oscillations. In general, quantitative takeaways require an extremely simple Fermi surface and/or computationally intensive calculations. Due to the complicated, many pocket Fermi surface of ScV_6Sn_6 , we are restricted to qualitative takeaways in this study.

Here I will present results from an ADMR study of our high-quality ScV_6Sn_6 . The primary conclusion is that at low temperatures the magnetotransport response is dominated by the large, open hole pocket from the vHs pocket. At high temperatures the small, closed pockets at the edges of the Brillouin zone are responsible for the magnetotransport response. This crossover occurs at $T' \approx 55$ K, well below T_{CDW} . This temperature scale agrees well with a scanning tunneling microscopy/spectroscopy study that suggested a nematic phase driven by vHs interactions below T' [132]. However, our bulk elastoresistivity measurements (a direct probe of nematicity) reveal a distinct lack of a nematic phase. This calls into question the interpretation of this previous data, and provides *bulk* evidence of a change in electronic properties at T' .

$\theta = 90^\circ, \phi = 0^\circ$ Magnetotransport

We begin by reporting the standard $\theta = 90^\circ, \phi = 0^\circ$ magnetotransport (a schematic of the coordinate system used is shown in Fig. 5.8b and a schematic of a ρ_{zz} sample is shown in panel c) for samples with varying quality with RRR_{zz} ranging from 2.5 to 12.4 as shown in Fig. 5.9. These RRR values is higher than those previously reported in literature (which

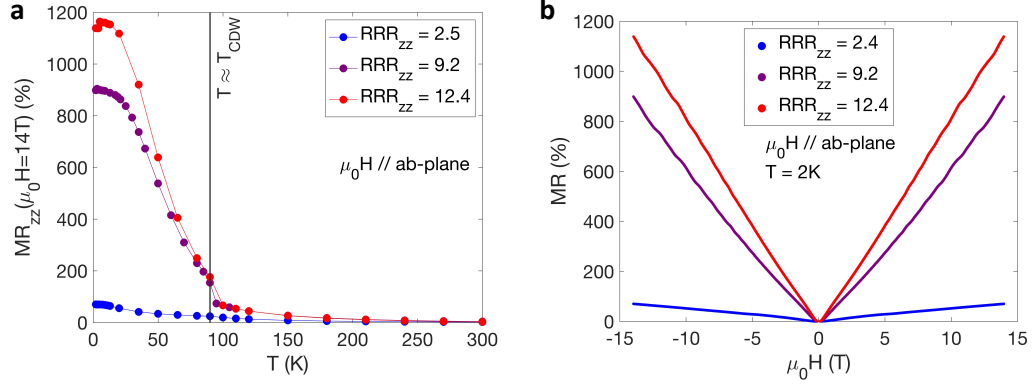


Figure 5.9: $\theta = 90^\circ, \phi = 0^\circ$ **magnetotransport**. **a**, MR_{zz} at 14 T as a function of temperature for samples with varying RRR. **b**, MR_{zz} at 2 K as a function of magnetic field for these samples. Note the linear and non-saturating magnetoresistance in the highest RRR sample.

have a maximum around 5) [51, 90]. Note that the RRR in this channel is significantly reduced from the in-plane channel (which can reach values of more than 50) because of the pseudogap behavior along the c-axis noted in Chapter 4. As seen in panel a, the magnetoresistance at 14 T grows as RRR increases as expected. This is due to the increased mobility (or, equivalently, scattering time) in these samples, allowing more cyclotron orbits before scattering, in turn making the magnetoresistance larger. Below T_{CDW} the magnetoresistance grows substantially with decreasing temperature. Interestingly, at 2 K the magnetoresistance is linear and non-saturating from 0 magnetic field up to 14 T for the highest RRR sample as shown in panel b. At first glance this is somewhat strange, for simple systems (with a single, closed, spherical Fermi pocket) one expects a roughly quadratic magnetoresistance at low fields that tends towards saturation at high fields. The intuition for this comes from the fact that cyclotron motion determines the magnetoresistance: at sufficiently high fields the charge carriers are moving in extremely curved paths and the introduction of higher fields does not meaningfully change the flow of the charge carriers through the materials. Thus, the magnetoresistance should eventually saturate. We sought to understand this linear, non-saturating magnetoresistance which led to us performing the ADMR measurements on the highest RRR sample that will follow.

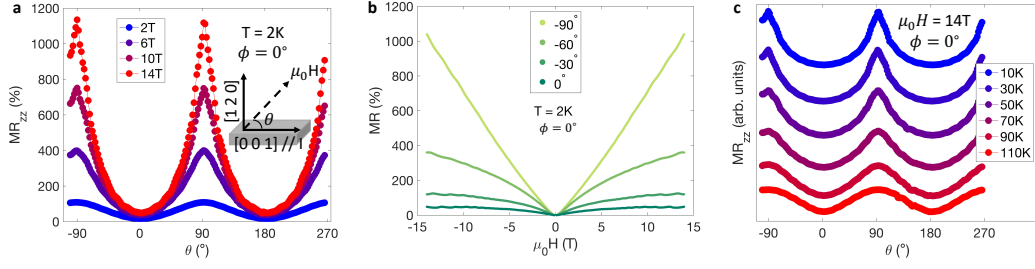


Figure 5.10: $\phi = 0^\circ$, θ -dependent magnetotransport. **a**, MR_{zz} at 2 K and $\phi = 0^\circ$ as a function of angle θ at several magnetic field strengths. **b**, MR_{zz} at 2 K as a function of magnetic field at several angles θ . **c**, MR_{zz} at 14 T as a function of angle θ at several temperatures.

$\phi = 0^\circ$, θ -Dependent Magnetotransport

This section will focus on rotating the magnetic field from the c-axis ($\theta = 0^\circ$) to the ab-plane ($\theta = 90^\circ$) at $\phi = 0^\circ$ (magnetic field perpendicular to the a-axis) as shown schematically in Fig. 5.10a. The data in panel a shows MR_{zz} at 2 K as a function of angle θ at several magnetic field strengths. At high magnetic field strengths the response varies smoothly except near 90° (magnetic field in the ab-plane) where the magnetoresistance is large and sharply peaked. These so-called “coherence peaks” emerge in the ADMR response of many quasi-2D conductors such as FeSe [133] and are indicative of coherent transport along the c-axis despite a weak electronic dispersion along this direction in momentum space. Here coherent transport refers to transport that can truly be described by Bloch bands as opposed to any sort of tunneling-like process. Keep in mind that the only quasi-2D part of the Fermi surface of ScV_6Sn_6 is the large vHs pocket near E_F ; these quasi-2D coherence peaks are the first piece of evidence that the low temperature transport is dominated by this large pocket. A second key piece of evidence comes from examining the magnetoresistance as a function of magnetic field at different angles θ as shown in panel b of this figure. It can clearly be observed that with magnetic field in the ab-plane the magnetoresistance is linear and non-saturating, but as the magnetic field is tilted away from this plane it becomes sublinear and tends towards saturation. Remember the above argument for why magnetoresistance saturates at high magnetic fields: it is dependent on completing many cyclotron orbits. However, here with current along the c-axis and field in the ab-plane, no cyclotron orbits are completed as this quasi-2D pocket is open along the c-axis. This qualitative difference leads to a non-saturating magnetoresistance. However, as the magnetic field is tilted

away from the ab-plane, cyclotron orbits can now be completed, leading to a tendency towards saturating magnetoresistance. As the angle moves closer to the c-axis these cyclotron orbits get increasingly smaller. Thus, the magnetoresistance should tend towards saturation and increasingly lower magnetic fields. This is precisely what we observe in this data, offering further evidence that this low temperature magnetotransport is dominated by the large vHs pocket. As a final piece of evidence, panel c shows MR_{zz} at 14 T as a function of angle θ at several temperatures. The coherence peaks get increasingly smaller as temperature increases until they are no longer observable at $T > T_{CDW}$ (remember that $T_{CDW} \approx 90$ K), implying that the magnetotransport along the c-axis is no longer coherent at temperatures above the phase transition. This is exactly consistent with the data presented in Chapter 4: above T_{CDW} pseudogap-like behavior is observed and slightly insulating behavior is measured in ρ_{zz} . This is indicative that the transport along the c-axis is not coherent, consistent with gapping out part of the Fermi surface along k_z and eliminating the coherence peaks seen at $T < T_{CDW}$.

$\theta = 90^\circ, \phi$ -Dependent Magnetotransport

With this understanding that temperatures below T_{CDW} the ADMR is dominated by the large hexagonal pocket, we now turn to $\theta = 90^\circ, \phi$ -dependent magnetotransport (ie. rotating the magnetic field in the ab-plane as shown schematically in Fig. 5.11a). The data in panel a of this figure shows $MR_{zz}(\phi)$ at 115 K ($T > T_{CDW}$, 14 T, and $\theta = 90^\circ$ (the same magnetic field strength and angle orientation will be used throughout this figure)). A smooth C_6 response is observed that is very reminiscent of the smooth responses seen in small 3D pocket systems such as Bi [134] as seen in panel b. Bi has a very simple Fermi surface comprised of a single spherical hole pocket centered in the Brillouin zone and three ellipsoidal electron pockets arranged 120° from each other, allowing for reasonably straightforward quantitative analysis of this response. The basic idea comes from the assumption that each closed pocket should contribute a term proportional to $\cos^2(\phi)$ to the magnetoresistance. The exact details of this fit depends on the shape of the contributing pockets. Thus, while the complicated Fermi surface of ScV_6Sn_6 prevents us from uniquely identifying which pockets are the primary drivers of this smooth response and meaningfully fitting these data, it is highly likely similar physics is at play here in the $T > T_{CDW}$ regime.

On the contrary, upon cooling through $T_{CDW} \approx 90$ K, the magnetoresistance qualitatively

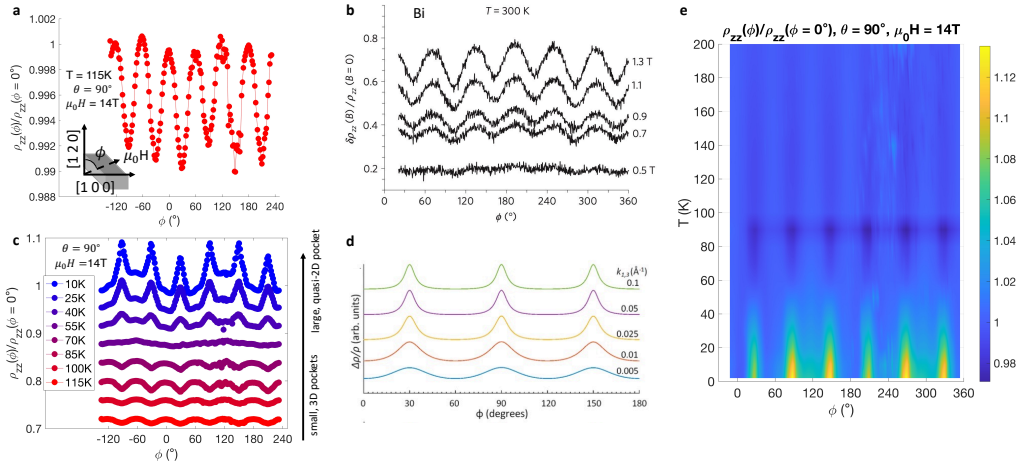


Figure 5.11: **Temperature dependence of $\theta = 90^\circ$, ϕ -dependent magnetotransport.** **a**, MR_{zz} at 115 K, 14 T, and $\theta = 90^\circ$ as a function of in-plane angle ϕ . A smooth C_6 response is observed. The same magnetic field strength and angle orientation will be used throughout this figure. **b**, $MR_{zz}(\phi)$ at 300 K of Bi [134]. **c**, $MR_{zz}(\phi)$ at several temperatures. The right hand side shows the general idea I argue in the main text of a temperature-dependent crossover of the primary driver of the measured ADMR. **d**, Calculated ADMR of a single hexagonal pocket with k_z warping from Ref. [135]. **e**, Color map of the $MR_{zz}(\phi)$ as a function of temperature.

changes at shown in panel c. Just below the phase transition temperature, the originally smooth lobes become “spiky” which can not be described by the physics discussed above. Furthermore, at $T' \approx 55$ K the ADMR becomes nearly zero, and with additional cooling the ADMR reverses (ie. the spikes go from being negative to positive) and the spikes become increasingly prevalent. Note that this reversal of the spike direction requires that the part of the Fermi surface driving the ADMR response has changed, otherwise the pocket would now be contributing a *negative* contribution to the conductivity which is not physical. We attribute this low temperature response to the large hexagonal pocket in ScV_6Sn_6 on the basis of calculations of the ADMR of a single hexagonal pocket that have been reported in literature [135]. Some of these calculations have been reproduced in panel d which shows that as the parameter $k_{1,3}$ increases positive spikes in the ADMR similar to the ones we observe in ScV_6Sn_6 at low temperatures are expected. $k_{1,3}$ refers to k_z warping of the hexagonal pocket; note that the hexagonal pocket in ScV_6Sn_6 is significantly warped as seen in Fig. 5.7a making this explanation of the low temperature ADMR reasonable. Furthermore, the temperature dependence of these spikes and subsequent ADMR reversal is consistent with the findings of the θ dependent ADMR: at high temperatures the large hexagonal pocket is gapped by the CDW fluctuations so it does not contribute to the ADMR, but for $T < T_{CDW}$ it plays an increasingly larger role as temperature decreases. The main takeaway of this section is sketched out on the size of panel c: at high temperatures the small, closed 3D pockets are the primary drivers of the ADMR whereas at low temperatures the large, open hexagonal pocket plays an increasingly important role. Panel e of this figure plots the ADMR data on a color plot to further emphasize both the ADMR reversal at T' and the fact that our high quality single crystals allow for a finite ADMR response up to 200 K.

While the above section reasonably qualitative describes the temperature dependence of the measured ADMR, it fails to describe difference in the size of the peaks that is particularly striking at low temperatures. A nice story that could explain this effect is a breaking of rotational symmetry; conveniently a scanning tunneling spectroscopy (STS) group reported a nematic phase between 50 and 70 K driven by the vHs near E_F in ScV_6Sn_6 [132]. Note that this temperature agrees well with the ADMR reversal at T' . However, there are some key issues with using this report to explain our results. First, STS is inherently a surface sensitive technique whereas resistivity is a bulk sensitive technique. More pressingly, as expected for a rotational symmetry breaking, this group observed small nematic

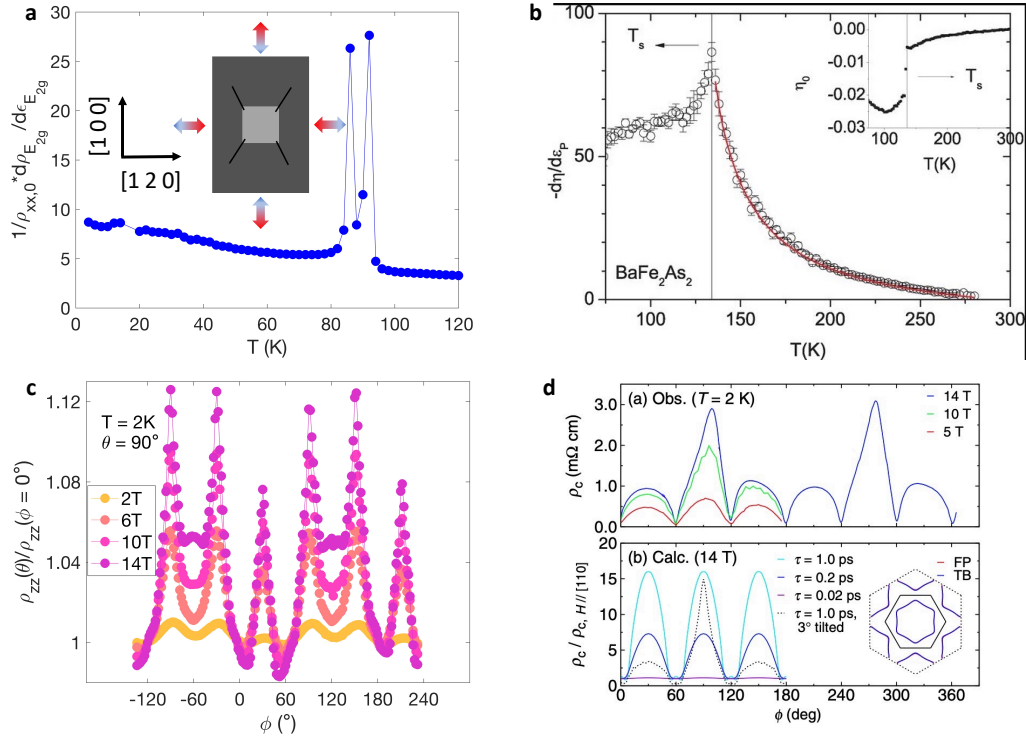


Figure 5.12: Lack of bulk nematicity in ScV_6Sn_6 . **a**, Symmetry-breaking elastoresistance as a function of temperature. No feature is observed near T' . **b**, Symmetry-breaking elastoresistance of a prototypical nematic material, BaFe_2As_2 [136]. Note the clear Curie-Weiss behavior at $T > T_S$. **c**, $MR_{zz}(\phi)$ at different magnetic field strengths at 2 K. **d**, Measured and calculated $\rho_{zz}(\phi)$ of PdCoO_2 [137]. A small tilt of the sample can lead to an apparent symmetry-breaking in the ADMR response.

domains. In our bulk sample with a volume of order mm^3 these domains should be averaged out to provide a non-rotational symmetry breaking measurement. One way to explain this issue is to attempt to manipulate the domains with magnetic field; however, field cooling with the magnetic field oriented along different angles ϕ did not change the measured ADMR. More strikingly, a measurement of the elastoresistance (led by Elliott Rosenberg) in the symmetry-breaking E_{2g} channel revealed a lack of any feature near T' as shown in Fig. 5.12a. This is in stark contrast to the same measurement on BaFe_2As_2 , a prototypical nematic material [136], which shows a Curie-Weiss behavior at temperatures above T_S as shown in panel b. This Curie-Weiss behavior is what is expected for a truly nematic transition; the symmetry-breaking strain couples to the nematic fluctuations just as a magnetic field couples to ferromagnetic fluctuations above T_C . The lack of any feature near T' in ScV_6Sn_6 directly contradicts a bulk nematic phase in this material. Instead, we turn to a mundane explanation of the apparent symmetry breaking in ScV_6Sn_6 . Panel c shows the magnetic field dependence of the in-plane ADMR at 2 K and it can be seen that as the magnetic field increases the apparent symmetry breaking gets stronger. This is quite comparable to the results of measurements on PdCoO_2 that are shown in panel d [137]. A small tilt angle of the sample with respect to crystallographic axes or magnetic field (which is impossible to completely avoid) explains the apparent symmetry breaking they observe at high fields, and it likely explains our data as well.

Takeaways

This ADMR study of high quality ScV_6Sn_6 reveals a temperature dependent crossover from small closed 3D pockets dominating the magnetoresistance response at high temperatures to the large open hexagonal pocket dominating the response at low temperatures. This is completely consistent with high temperature CDW fluctuations opening a pseudogap at high temperatures as we previously postulated. Furthermore, we have ruled out the idea of a bulk nematic transition in this material.

6 Conclusions and Outlook

The ideal kagome band structure seems to offer significant promise for interesting physics in kagome materials. However, no material is perfect, and I have shown here the RT_6X_6 family is actually *not* a great part of phase space to search for kagome-derived physics. Nevertheless, these materials are still quite interesting. Our study of Cr-doped $TbMn_6Sn_6$ revealed that the large intrinsic AHE observed in the parent compounds can not be assigned to a gapped Dirac point in the electronic spectrum, but we also found an enhanced AHE with doping that coincided with an extremely hard compensated ferrimagnet phase. Our various works on ScV_6Sn_6 found a distinct lack of evidence for any time-reversal symmetry breaking in a kagome CDW material with a vHs near E_F , but it led us to help identify an unexpectedly complex phase transition that couples interestingly to the electronic spectrum. In my eyes, these results highlight the complexity of condensed matter physics and the unexpected results within this field.

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