Growth and Analysis of Single Crystal $CeCoGe_3$

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$CeCoGe_3$ single crystals have been grown via solution growth method. The growth was done at high temperatures using Bismuth as a flux in the reaction. By manipulating variables in our reaction we were able to grow single crystals of various sizes. The single crystals were used to perform magnetic and physical property measurements. Magnetic measurements were performed along H // [001] and electrical measurements with J // [100] directions. Magnetic susceptibility was measured and shows a Néel temperature at T_{N1} = 21 K and two additional transitions at T_{N2} = 12 K and T_{N3} = 8 K. Resistivity measurements shows a clear transition at T_{N1} = 21 K consistent with magnetic measurements.

I. INTRODUCTION

Heavy fermion compounds, such as Ce-based $CeCoGe_3$, have been shown to be fascinating materials to study. This material is considered a Weyl semimetal and has shown peculiar topological properties. These properties include discoveries of unconventional superconducting properties, band structures, and electrical transport properties [5, 9]. Our intention was to study this materials growth, analyze its properties, and explore the electronic structure of the material. Growing the crystal was the first step and optimizing that growth was the focus of this project.

We succeeded in growing $CeCoGe_3$ using the solution growth method with Bismuth acting as our flux. We were able to grow crystals of different sizes by manipulating variables in our reaction. Powder x-ray diffraction was used to verify we had the correct compound after growth. The maximum dimensions of a single crystal were 2x1x1 mm^3. A diagram of the crystal structure of the material is provided in Fig. 1 [7]. Clarification of the purity of our compound was the main goal so that we could begin studying the band structure of the material and electrical transport properties by angle resolved photoemission spectroscopy and Hall effect measurements.

The next sections will discuss the growth and analysis of the single crystals we were able to grow. Mainly, the growth of the material will be explored along with our process in optimizing the growth for larger crystals. Results from our measurements and previous studies will be used to confirm we had grown the correct material.

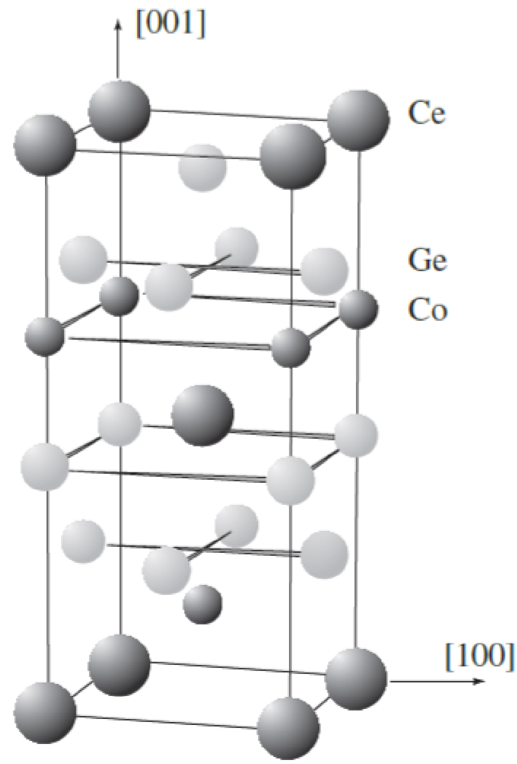

Figure 1. Crystal Structure of $CeCoGe_3$.

II. EXPERIMENTAL

We grew $CeCoGe_3$ single crystals via the solution growth method. We decided to use Bismuth as a flux to aid in melting during the reaction based off previous growth of the material [7]. We prepared our materials based off this knowledge. For our first growth attempt, we had successfully grown $CeCoGe_3$ single crystals of small size. In order to have quality measurements, we optimized our growth strategy to synthesize larger single crystals.

A. Material Preparation and Growth Strategy

The solution growth method is a valuable strategy for growing single crystals of materials [1]. We had to use a fourth element, Bismuth, to help with the melting of our elements. Cerium (99.99%), Cobalt (99.9%), and Germanium (99.9999%) were prepared first based off the

$Ce_1Co_1Ge_3$ stoichiometry. The materials were weighed out and then arc melted into a polycrystalline ball. The ball was then powdered which was used to perform powder x-ray diffraction (PXRD). The reason for PXRD before growth was to ensure the correct $Ce_1Co_1Ge_3$ stoichiometry. Growth of the correct material relies on the correct stoichiometry. The rest of the powder was transferred into a ceramic crucible where Bismuth was then weighed and added. The crucible was sealed under partial pressure of argon in a quartz ampoule and placed into a furnace. The furnace was programmed based off of our temperature profile in Fig. 3.

We tried multiple ratios in Fig. 2 where the green highlighted ratio yielded the largest single crystals. The difference in percentage of Cerium, Cobalt, and Germanium is based off the amount of Bismuth used in our reaction.

Atomic Ratios

Ce	Co	Ge	Bi
1%	1%	3%	95%
2%	2%	6%	90%
4%	4%	12%	80%
6%	6%	18%	70%

Figure 2. Atomic ratios for each growth.

We constructed a temperature profile in Fig. 3 for our reaction and tweaked it accordingly. This was from our growth with the largest single crystals. Optimizing from previous growths was based off of phase diagrams and the melting points of the materials. We tried centrifuging at both 650 °C and 500 °C where the latter produced large Germanium crystals and other phases. Growth times varied from 1 - 2 weeks and didn't have effect on the crystal size. When the crystal growth was completed, we powdered small crystals and did PXRD to confirm the purity of our growth.

Figure 3. Temperature profile for $CeCoGe_3$ reaction.

We performed physical and magnetic property measurements after crystal growth. Resistivity measurements were done in a Physical Property Measurement System (PPMS) at temperatures ranging down to less than 2 K and at fields greater than 9 T. Magnetic property measurements were done in a Quantum Design Magnetic Property Measurement System (MPMS) with temperatures from 2 K to 300 K and fields to 7 T.

Small platinum wires in a standard 4-probe geometry were attached to our sample in order to do AC resistivity measurements. The platinum wires had to be attached such that current would run along the entire sample and voltage would be measured in the center shown in Fig. 4. The center leads had to be parallel and placed where a distance measurement could be taken between them with little error. Resistivity can then be calculated from Eq. 1. User-provided parameters include the cross sectional area (A) and voltage lead separation of the sample (l). Voltage (V) and current (I) through the sample are measured.

$$\rho = \frac{V \times A}{I \times l}[\Omega \times cm] \quad (1)$$

For our magnetic measurements, we had to align the crystal in specific orientations to load into our MPMS. To do this we simply used a plastic straw and loaded the sample by poking a hole and orienting the single crystal in Apiezon grease. The reason for these specific orientations is that when a magnetic field is applied, the data we gather may depend on our crystal axis.

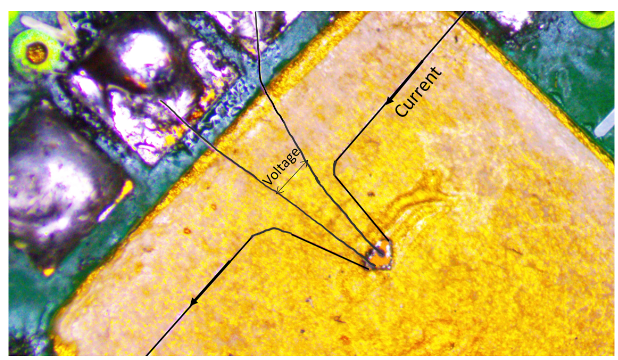

Figure 4. Resistivity measurement along the [100] axis.

III. RESULTS

A. Growth Results

We successfully grew the material with bismuth as flux using the ratio $Ce_6Co_6Ge_{18}Bi_{70}$. PXRD shown in Fig. 5 of our single crystals confirmed we had grown pure material. The goal here is to ensure we had grown the proper material. Magnetism and electrical transport properties will be briefly explained but are not the main topic of this paper

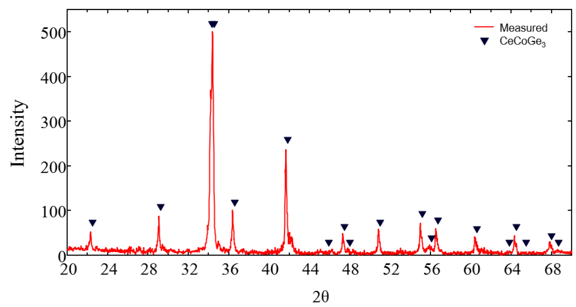

Figure 5. Powder X-Ray Diffraction of $CeCoGe_3$.

B. Electrical Resistivity

We were interested in both the electrical transport and magnetic properties of $CeCoGe_3$. From our results we are looking for changes in magnetic ordering. These happen at different temperatures called Néel temperatures (T_N). A single crystal resistivity measurement example along the J // [100] direction is shown in Fig. 6. First, we can clearly see a transition point at T_{N1} = 21 K. The change in slope present at this temperature represents a change in magnetic ordering. We can also see that we have a relatively high Residual Resistivity Ratio (RRR) of 75 which is the ratio of resistivity at the highest temperature to the lowest temperature. This RRR indicates the quality of our single crystal where previous crystals had shown RRR's in the low 20's.

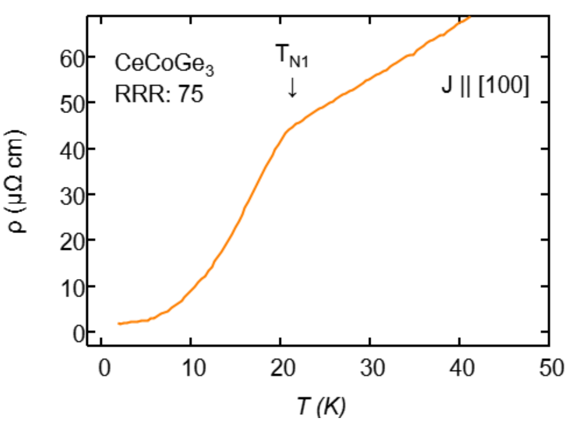

Figure 6. Resistivity vs. Temperature showing Néel transition at 21 K.

C. Magnetic Properties

We also measure the moment of our material when a field is applied and temperature is lowered. Our crystals were measured along H // [001]. We plotted Magnetic Susceptibility, a measure of how magnetized our material becomes in an applied field vs. Temperature shown in Fig. 7. We can clearly see three transitions at T_{N1} = 21 K, T_{N2} = 12 K, and T_{N3} = 8 K. The transitions are identified by a sudden change in slope. These can be broken down further into the different orderings shown in Fig. 8.

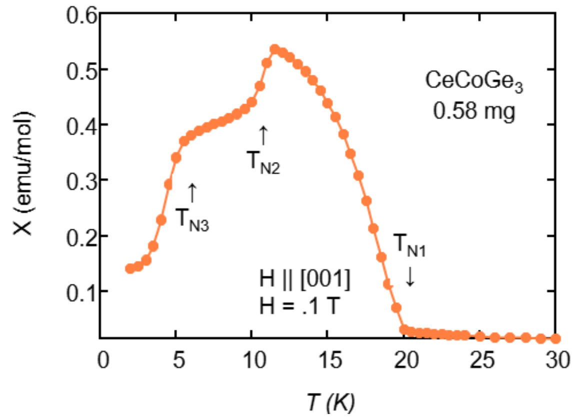

Figure 7. Magnetic Susceptibility vs. Temperature showing three Neel transitions.

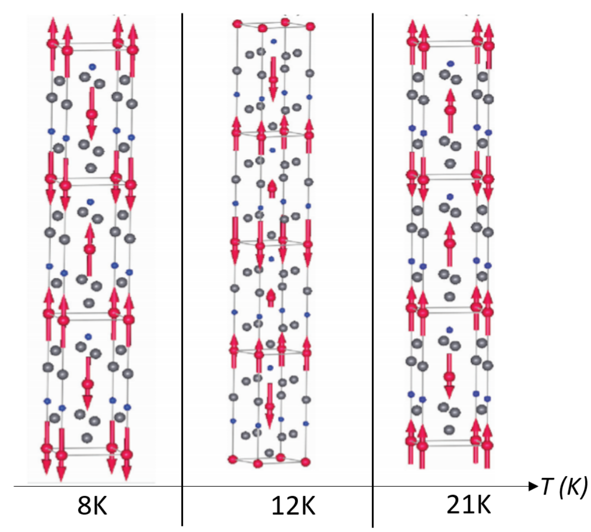

Figure 8. Magnetic Phases from Neutron Scattering Studies of $CeCoGe_3$.

Fig. 8 shows spontaneous magnetic transitions between the different Néel temperatures. This figure was taken from a study where they used neutron scattering to explore the transitions. The red arrows in the diagram are the magnetic moments of Cerium atoms. From these arrows we can conclude which magnetic orderings we are observing at each transition.

Each of our transitions denote an antiferromagnetic transition. It should be noted that the transition at T_{N1} = 21 K is complicated. There is a divide whether this is a ferromagnetic or antiferromagnetic transition. A previous report had concluded the material is indeed antiferromagnetic from extremely similar data. Further expansion upon this would require a complex magnetic phase diagram which has been created and explained by the same study[7].

IV. CONCLUSIONS AND FUTURE WORK

We were able to confirm that we could grow $CeCoGe_3$ single crystals. It took many growths to optimize the formula and size was an issue at first. However, with a $Ce_6Co_6Ge_{18}Bi_{70}$ ratio and Bismuth as a flux material, we finally grew single crystals that were of a good size for measurements. With powder x-ray diffraction we confirmed the correct compound was grown and were able to perform further measurements.

Our magnetic and electrical property measurements furthered our conclusion that we had grown quality crystals. The magnetic susceptibility vs temperature in Fig. 7 is extremely similar to previous reports on the material [7, 6]. Both resistivity and magnetic susceptibility measurements show a clear antiferromagnetic transistion at T_{N1} = 21 K. Similar measurements should be done for the material including magnetization and specific heat measurements at different temperatures to elucidate the transitions present in this material as well as directional changes. Furthermore, resistivity measurements show a high quality of our material but were only measured along one direction. Future work should include measurements along J // [001] and a RRR calculation for comparison to determine the purity of our crystal.

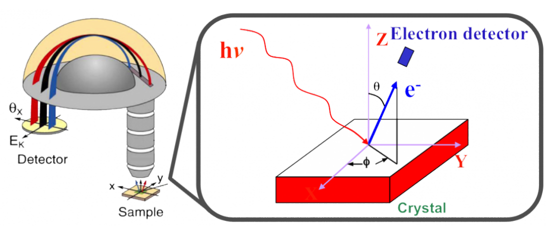

Figure 9. ARPES Measurement Example.

Single Crystal $CeCoGe_3$, as previously mentioned, is a weyl fermion material. This means the material shows topological properties that may have potential for high impact studies. This is exciting to pursue for applications such as electronic and optical devices since weyl fermions are shown to behave like photons since they are described as massless particles [3, 4]. In order to begin studying this material's topological properties and band structure there are two places to explore. First, conducting Angle Resolved Photoemission Spectroscopy (ARPES) which will show us the distribution of electrons in our material. Fig. 9 graphically shows how the ARPES technique is used. Knowing this information will help us understand the band dispersion and Fermi surface. Performing Hall Effect measurements to understand the topological properties of the material is another avenue to explore [4]. We

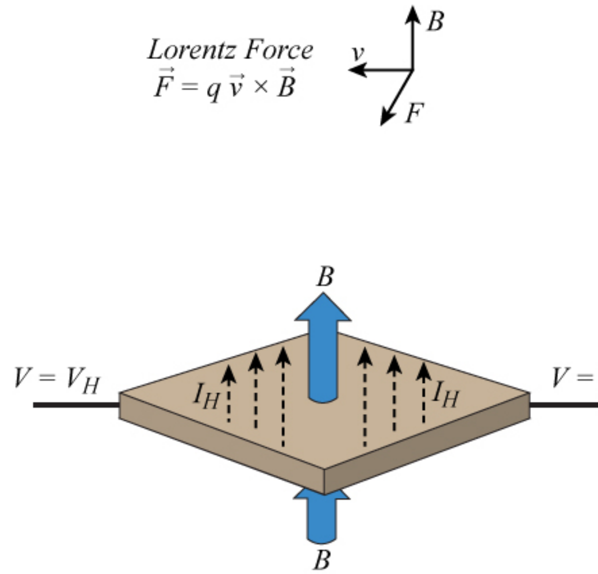

Figure 10. Hall Effect Measurement Example

can do these measurements in our PPMS by setting up voltage leads that are directly across from each other on different sides of the crystal depicted in Fig. 10. Analysis of the Hall effect will help us further understand the electron transport properties of this material [8, 2].

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