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Three-dimensional Dirac semimetal and magnetic quantum oscillations in Cd_3As_2

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Abstract. Single crystals of topological Dirac semimetal Cd_3As_2 were synthesized using a modified vapour transport technique. The magnetization of Cd_3As_2 single crystals was measured in the temperature range from 4.2-300 K and magnetic field up to 5 T along [001] and [100] directions. In the low temperature region we observed the de Haas van Alphen (dHvA) oscillations which allow us to characterize the three dimensional Fermi surface by extracting its relevant parameters. The dHvA oscillations show the existence of two different Fermi surface cross sections and nontrivial Berry's phase, which is the signature of Dirac fermion in Cd_3As_2 .

1. Introduction

Recently, three-dimensional (3D) topological Dirac semimetals (TDS) have been theoretically predicted [1, 2, 3] and a crystalline-symmetry-protected 3D bulk Dirac semimetal phase has been experimentally identified in a high-mobility Cd_3As_2 single crystal using the angle-resolved photoemission spectroscopy (ARPES) [4, 5]. Cd_3As_2 is a degenerate n-type semimetal of the II-V family with high mobility and low effective mass, which exhibits an inverted band structure ($E_g < 0$) [6, 7]. The conduction and valence bands of Cd_3As_2 touch at the Dirac point in the bulk band structure, giving rise to bulk Dirac fermions. Moreover, due to the lack of inversion symmetry this TDS phase raise a possibility of realizing the Weyl semimetal phase [8, 9].

Here we report measurements of magnetization in magnetic field up to 5 T parallel and perpendicular to the crystalline c-axis ([001] and [100] direction) for relatively large crystals of Cd_3As_2 . The growth of the single crystals of Cd_3As_2 was carried out by a modified vapour transport technique. We observe a linear magnetization accompanied by the de Haas van Alphen (dHvA) quantum oscillations at low temperatures. The dHvA oscillations allow us to characterize the three dimensional Fermi surface. We analyzed the magnetization experimental data by using the Lifshitz-Kosevich (LK) theory which predicts the magnitude of the dHvA oscillations as well as their temperature dependence [10].

2. Experimental results and discussion

Single crystals of Cd_3As_2 were synthesized using the vapour transport technique. Starting polycrystalline material was prepared as described in [7]. Approximately 2 g of polycrystalline



material is placed at one side of 16 mm in diameter and 10 cm long, vacuum sealed, quartz tube. The tube is then subjected to a temperature gradient in a modified two zone tube furnace. The empty side of the ampoule was kept at 565 °C while the temperature of the side with the material was slowly raised from 530 °C up to 585 °C with the rate of 1.7 °C /h. The tube was kept in this temperature gradient for 8 hours and then slowly cooled down to a room temperature. Two zones of the furnace are physically separated so there is a little drop in temperature at their junction when they are about the same temperature, Figure 1. Starting place of crystallization is restricted to the region of the initial lowest temperature. Without temperature kink material will be pushed to the end of ampoule and crystals will be compacted there. The grown crystals were granule like with dimensions from 1-5 mm.

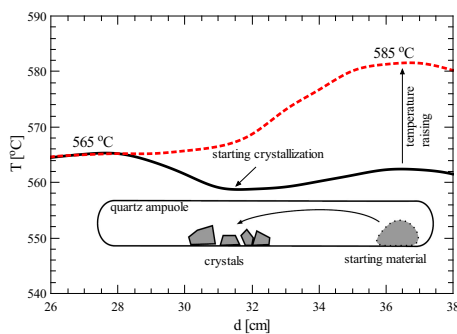


Figure 1. Schematic representation of crystal growth method.

by performing $M - B$ scans from 300 down to 5 K. Any intercepts at $B = 0$ T were less than $1.6 \cdot 10^{-5}$ emu for the measured samples. The samples are diamagnetic with magnetic susceptibilities that are only weakly temperature dependent down to about 60 K, below which an upturn can be observed. The quality of samples is also confirmed by measuring the temperature dependence of sample's resistance. Residual resistivity ratio (RRR) of nearly 6 is obtained for both samples.

In the low temperature region, from 25 K down to 4.2 K, we observe the dHvA oscillations. The oscillations can be traced down to field of 2 T. The magnetisation was measured along the [001] and [100] direction. By subtracting the diamagnetic background, pure oscillating part of magnetisation can be obtained. The dHvA oscillations of the sample B for magnetic field along [001] direction (a) and [100] direction (b) for different temperatures are shown in Figure 2.

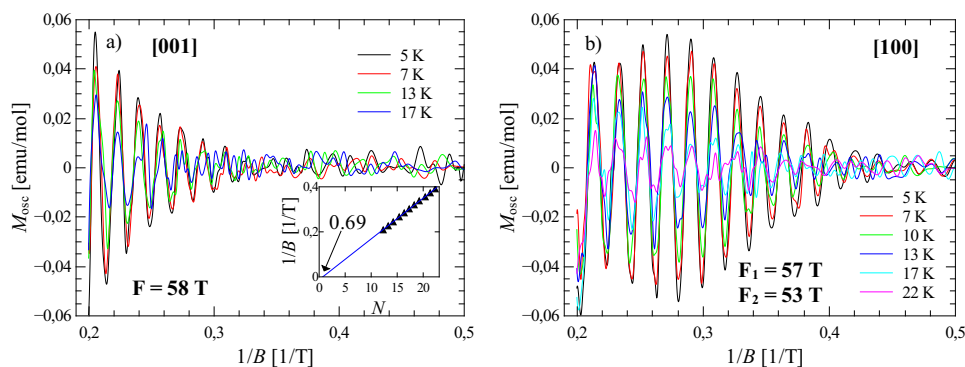


Figure 2. The dHvA oscillations for Cd_3As_2 sample (sample B) for magnetic field along [001] (a) and [100] direction (b).

According to the LK formula, for magnetic field along [001] direction a single frequency oscillation with $F = 55$ T (sample A) and $F = 58$ T (sample B) is obtained, Figure 2a. Oscillation frequency is determined by direct fitting of the LK formula for a single frequency and also by fast Fourier transform (FFT). Both methods give nearly the same values. Oscillation amplitude dependence on $1/B$ for magnetic field in [100] direction suggest that there is a contribution of at least two close frequencies, see Figure 2b. By FFT two frequencies can not be resolved. By fitting of the LK formula for two frequency contributions a good match can be achieved with frequencies $F_1 = 56$ T, $F_2 = 51$ T (sample A) and $F_1 = 57$ T, $F_2 = 53$ T (sample B). From temperature and $1/B$ dependence of single frequency oscillation amplitude the effective cyclotron mass of $m_c^* = 0.047m_e$ and Dingle temperature [10] of $T_D = 16.5$ K (related to quantum scattering time of $\tau_q = 7.4 \cdot 10^{-14}$ s) have been extracted. The quantum mobility of electrons is given by $\mu = e\tau_q/m_c^* = 0.28 \cdot 10^4$ cm²/Vs. From oscillation frequency $F = \hbar k_F^2/2e = 55$ T one can obtain the Fermi wave vector $k_F = 0.041 \text{ \AA}^{-1}$ which leads to the Fermi velocity $v_F = \hbar k_F/m_c^* = 1.01 \cdot 10^6$ m/s. The charge carrier density has been obtained from $n = k_F^3/3\pi^2 = 2.3 \cdot 10^{18}$ cm⁻³. Dirac dispersion of charge carriers can be confirmed by plotting the so called Landau level (LL) diagram [11] ($1/B$ vs. N -index corresponding to minima or maxima). For Dirac fermions the intercept of obtained line for $1/B = 0$ must be near $5/8$ (for the minimal Fermi surface cross section), whereas for Schrödinger fermions the intercept is near $1/8$. LL diagram for sample A for magnetic field along [001] direction gives intercept of 0.69 confirming the Dirac nature of charge carriers in Cd₃As₂, inset to Figure 2. Two different frequencies in dHvA oscillations, for magnetic field in given direction, indicate that there are two different extremal Fermi surface cross sections. Theoretically is predicted and experimentally confirmed that, below the Lifshitz transition, Fermi surface of Cd₃As₂ consist of two identical ellipsoids [2, 5], which can not explain two frequency contributions. The Fermi energy is also estimated, $E_F = m_c^*v_F^2 \approx 270$ meV, and according to [8, 12] E_F is above the Lifshitz point.

3. Conclusion

A very useful synthesis method for easy sublimating material, as Cd₃As₂, is described in detail. The synthesized samples were characterized by magnetization measurements and dHvA oscillations are observed at low temperatures. We obtained single frequency dHvA oscillations for magnetic field along [001] direction (not measured in [7]), whereas for field in [100] direction the contributions of two close frequencies are observed (authors in [7] report on just one frequency for a given direction). Modelling quantum oscillations by the LK formula the effective cyclotron mass, quantum scattering time, mobility, Fermi wave vector, Fermi velocity and density of carriers are estimated. From the LL diagram for single frequency quantum oscillations, a nontrivial π Berry phase is found, confirming the existence of the Dirac fermion in Cd₃As₂. Measurements like this will be helpful in further understanding of Cd₃As₂ Fermi surface which still remains unclear.

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