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Quantum magnetotransport and de Haas–van Alphen measurements in the three-dimensional Dirac semimetal $Pb_{0.83}Sn_{0.17}Se$

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Abstract. Magnetoresistance (MR), Hall resistance and magnetization of the $Pb_{0.83}Sn_{0.17}Se$ crystals have been measured in magnetic field up to 15 T and 5 T, respectively, and temperatures from 1.7 K up to 300 K. A large linear and temperature dependent MR is observed in magnetic field up to 15 T. The de Haas–van Alphen (dHvA) and Shubnikov de Haas effects (SdH) of $Pb_{0.83}Sn_{0.17}Se$ crystals have been clearly seen in the temperature range from 30 K down to 1.7 K and magnetic field as low as 2 T. The dHvA and SdH oscillations reveal single frequency of around 8 T which confirms the existence of a single Fermi surface cross section. Influence of isothermal annealing of $Pb_{0.83}Sn_{0.17}Se$ crystals in Se vapours has been investigated. By increasing the annealing temperature from 433 °C up to 440 °C , transition from n to p-type conductivity has been observed. The dHvA and SdH effects clearly reflect the existence of a nontrivial Berry's phase owing to the linear band dispersion which is the signature of a three-dimensional Dirac fermion in the $Pb_{0.83}Sn_{0.17}Se$ crystals [1].

1. Introduction

Recent theoretical and experimental studies have suggested the existence of topological crystalline insulators (TCI), a novel class of topological insulators in which crystalline symmetry replaces the role of time-reversal symmetry in topological protection of their metallic surface states [2, 3]. These surface states are observed in the rocksalt IV–VI semiconductors (Pb,Sn)Te, (Pb,Sn)Se and SnTe using angle resolved photoemission spectroscopy (ARPES) [4, 5, 6]. Energy gap in the $Pb_{1-x}Sn_xSe$ semicoductor is temperature and Sn-content dependent [7]. The substitution of Sn for Pb strongly changes the spin-orbit effects and results in a compositional evolution of their band structures. An increasing Sn content in $Pb_{1-x}Sn_xSe$ semiconductor leads to a closing of the bulk bandgap at a critical value of $x = x_c \approx 0.17$ (for $T < 77K$) and the system undergoes gap inversion when x exceeds x_c [7, 6]. In the $Pb_{1-x}Sn_xSe$ semiconductor, the chemical potential can be tuned during the crystal growth or annealing to yield n-type or p-type conductivity, which makes them more suitable for experimental investigations of the TCI state [6]. Very recently, we have found that $Pb_{0.83}Sn_{0.17}Se$ single crystal is a three-dimensional Dirac semimetal [1].

Here we report the low temperature magnetotransport and magnetization data measured

from 300 K down to 1.8 K in high magnetic filed up to 15 T for semiconductor $Pb_{0.83}Sn_{0.17}Se$ single crystals which achieve the state of transition between the ordinary insulator and the TCI where the existence of a zero gap state is observed [10]. To investigate the tuning of the chemical potential, single crystals of $Pb_{0.83}Sn_{0.17}Se$ have been annealed in the Se vapours at temperatures between 433 and 440 °C . The $Pb_{0.83}Sn_{0.17}Se$ single crystals have peculiar magnetic and transport properties, such as a large negative unconventional magnetization, linear MR (LMR) in perpendicular magnetic fields and ultra high mobility. We have observed the linear magnetization and LMR accompanied by quantum oscillations at low temperature and in magnetic field up to 5 T and 15 T, respectively. The MR data reveal discernible Fermi surface change caused by the Zeeman splitting in the magnetic field range from 4 T up to quantum limit field of about 8 T. We have analysed the magnetization and magnetotransport experimental data using Lifshitz-Kosevich theory which predicts the magnitude of the observed dHvA and SdH effects as well as their temperature dependence and allow us to characterize the three-dimensional Fermi surface and its relevant parameters. Also, by studying the carrier density and mobility dependence of the observed LMR in perpendicular magnetic fields possible physical origins are discussed.

2. Experimental results and discussion

The $Pb_{0.83}Sn_{0.17}Se$ single crystals were grown by a modified Brigman method [1]. Evidence for the quality of the $Pb_{0.83}Sn_{0.17}Se$ crystals includes extremely sharp x-ray peaks observed at room temperature, and a substantial mean free path extracted from the quantum oscillations observed in the magnetization and magnetotransport data. Samples from several batches were annealed for 48 h in the Se vapours at temperatures from 433 to 440 °C (433, 435, 436, 438 and 440 °C) and then slowly cooled down. The magnetization was measured using a superconducting quantum interference device magnetometer SQUID (Quantum Design). A long plastic straw was used as the sample holder [1]. The samples were first mounted and cooled in the SQUID magnetometer down to 5 K. The magnetization of $Pb_{0.83}Sn_{0.17}Se$ crystals has been measured at different temperatures from 300 K down to 5 K with B parallel to the [001] direction and shown in figure 1.

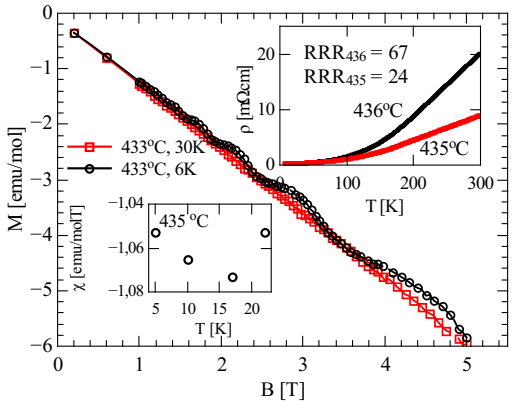

Figure 1. The magnetic field dependence of magnetization at two temperatures when the field was applied along the [001] direction for $Pb_{0.83}Sn_{0.17}Se$ crystal annealed at 433°C . The upper inset shows the temperature dependence of the resistivity of $Pb_{0.83}Sn_{0.17}Se$ crystals annealed at 436°C and 435 °C and the lower inset shows the magnetic susceptibility in the temperature range from 5-25 K of sample annealed at 435 °C .

It is found that the samples are diamagnetic. The linear behaviour of the magnetization vs. B was observed for all measured samples in the temperature range from 5 K up to 300 K. Any intercepts at B = 0 T were less than $1.6 \cdot 10^{-5}$ emu for the measured crystals, and have been subtracted from the data reported here. The magnetic susceptibility is weakly temperature dependent down to 5 K. A shallow minimum exists between 40 and 5 K, see the inset to figure

1. It can be seen from figure 1 that the magnetization displays a very clear dHvA effect. The oscillations are clearly seen down to 1 T and up to 30 K.

From Hall measurements of annealed samples at room temperature we found that samples annealed at 433 °C possess pure n-type conductivity while samples annealed at 440 °C show p-type conductivity. $Pb_{0.83}Sn_{0.17}Se$ p-type crystals show no quantum oscillations. So samples annealed at 435 ± 1 °C are close to the crossover from n to p-type conductivity and the chemical potential is then closest to the band touching point. Further transport measurements are done only on samples annealed at 435 and 436 °C .

Transport measurements were carried out in a ^{4}He cryostat, using a constant bias current (dc, 10 mA) and a nanovoltmeter with a 10 G input resistance up to 12 V of the signal. For transport measurements, the electrical contacts in the six-point geometry were made by spot welding which resulted in contact resistance of the order of 0.5 Ω. The temperature dependence of longitudinal electrical resistivity of the $Pb_{0.83}Sn_{0.17}Se$ crystals is presented in the inset to figure 1. The longitudinal resistivity shows a typical metallic behaviour with the residual resistivity ratio $R(300K)/R(1.8K) = 67$ and 24 with residual resistivities $\sim$ 0.28 mΩcm and $\sim$ 0.37 mΩcm at 1.8 K, for the $Pb_{0.83}Sn_{0.17}Se$ crystals annealed at 436°C and 435°C , respectively. Hall signal mixed into measured longitudinal resistivity ρ'_{xx} is removed by averaging over the positive and negative field directions. From the raw ρ'_{xx} data measured at the indicated temperatures, we obtained the symmetrized (intrinsic) longitudinal MR, which are plotted as $MR = (\rho_{xx}(B) - \rho_{xx}(0))/\rho_{xx}(0) \cdot 100\%$ in figure 2 (where ρ_{xx} is symmetrized longitudinal resistivity and $\rho_{xx}(0)$ denotes the longitudinal resistivity in the zero magnetic field). Here, current was along the [100] direction, and the applied magnetic field was perpendicular to current in [001] direction.

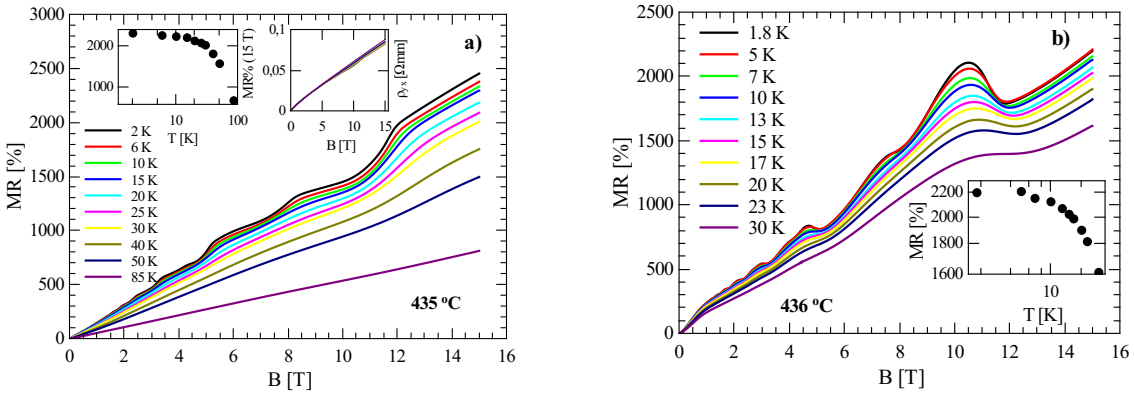

Figure 2. Magnetoresistivity (MR) of $Pb_{0.83}Sn_{0.17}Se$ single crystals annealed at 435 °C (a) and 436 °C (b), respectively, at the different temperatures in magnetic fields applied along the [001] direction. The right inset to a) displays the Hall resistivity vs. magnetic field data of the sample annealed at 435 °C . The insets to a) and b) display the MR versus temperature at 15 T for the $Pb_{0.83}Sn_{0.17}Se$ single crystals annealed at 435 °C and 436 °C , respectively.

The MR reveals a linear behaviour from very small magnetic fields for the both samples, figure 2. Even at temperature of 85 K and magnetic field of 15 T MR shows no sign of saturation, and its value is as high as 800%. Such a large LMR at 85 K is quite unusual and needs further theoretical study to understand the physical origin. To gain more physical insights into the observed linear MR we further study the temperature dependence of MR(B, T). In the inset of figure 2a) and 2b) we show the temperature dependence of MR (without subtracting the oscillatory part) at 15 T in log-log scale. With decreasing temperature, MR increases rapidly up

to 2500% at 10 K in magnetic field of 15 T. It is clear from the insets that, unlike conventional metal, MR shows a saturation-like behaviour in the low-temperature region below 20 K. Similar behaviour has been observed in Cd_3As_2 crystals [11]. We have also performed Hall resistivity measurements for the both samples annealed at 435 °C and 436 °C . Shown in the inset to figure 2a), the Hall resistivity of the sample annealed at 435 °C reveals nonlinear behaviour at low magnetic fields with crossover at $B_c = 2.5$ T and then changes behaviour in linear at high magnetic fields (there is obviously a small contribution of holes in transport). The Hall resistivity of the sample annealed at 436 °C has the linear magnetic dependence from 0 up to 15 T. From the slope of Hall resistivity data the electron density is extracted: $n_H = 11.6 \cdot 10^{17}$ cm^{-3} for sample annealed at 435 °C , whereas for the sample annealed at 436 °C , $n_H = 5.9 \cdot 10^{17}$ cm^{-3}. Further, the Hall resistivity data for the both samples reveal very weak temperature behaviour in the temperature range from 300 K down to 1.8 K. Using the single carrier Drude band model and the above extracted electron density, we have obtained the electron mobility $\mu(1.8\text{K}) = 1/en\rho_{xx} = 3.74 \cdot 10^4$ cm^2/Vs for the sample annealed at 436 °C and $1.45 \cdot 10^4$ cm^2/Vs for 435 °C .

As it is seen from figures 1 and 2, the quantum oscillations are clearly observed in the magnetization and MR data of $Pb_{0.83}Sn_{0.17}Se$ crystals. Subtracting a smooth and linear background from the magnetization (figure 1) yields the dHvA oscillatory component, figure 3a). Using the MR and Hall resistivity data we have calculated magnetoconductivity $\sigma_{xx} = \rho_{xx}/(\rho_{xx}^2 + \rho_{xy}^2)$, its oscillatory part is shown in figure 3b). Figures 3a) and 3b) display the oscillatory components of magnetization and magnetoconductivity versus 1/B and reveal how the oscillation amplitude decreases with the annealed temperature of the samples and temperature, respectively. The inset to figure 3a) shows the oscillation frequencies obtained by the FFT analysis of the magnetization data. It is clearly seen that there is only one oscillation frequency which slightly changes in the range from 8.6 to 6.9 T for the annealed temperatures. In oscillatory part of magnetoconductivity the effect of Zeeman splitting in higher magnetic fields (above 4 T) can be clearly seen.

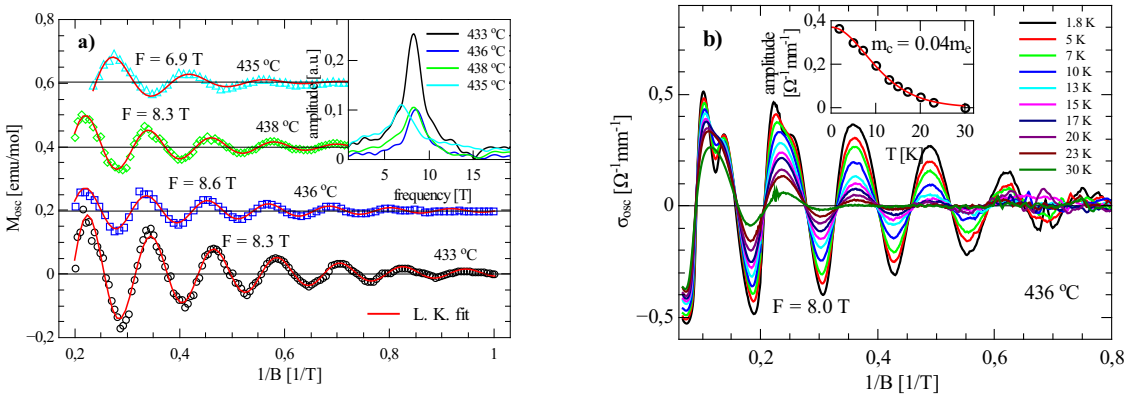

Figure 3. a) the oscillatory components of magnetization for $Pb_{0.83}Sn_{0.17}Se$ single crystals at temperature 5 K and b) the quantum oscillation contribution to magnetoconductivity for $Pb_{0.83}Sn_{0.17}Se$ single crystal versus 1/B. The inset to a) shows the oscillation frequency obtained by the FFT analysis of the magnetization curves of the samples. The inset to b) shows the temperature dependence of amplitude of oscillations from which the effective cyclotron mass of carriers can be obtained [1].

To explain the origin of LMR, two types of mechanisms are proposed: (i) a classical model which explains the LMR by modelling the material as a network due to the strong inhomogeneity

in the carrier density hence disorder induced mobility fluctuation [12] and (ii) a quantum model due to the linear energy dispersion of carrier at the band touching point proposed by Abrikosov [13]. In the quantum model, the LMR also occurs when multiple Landau levels are filled, even when thermal smearing is severe, as long as the system has a positive g factor [14]. However, in our samples the LMR is observed from very small magnetic fields which are much lower than the onset of the quantum oscillations observed at 2 T. Thus, we can argue that the LMR in our samples arises because the local current density acquires spatial fluctuations in both magnitude and direction, as a result of the microstructure caused by inhomogeneous carrier density and mobility distribution.

From weak temperature dependence of Hall resistivity (inset to figure 2a)) one can conclude that the carrier density is also temperature independent. The decrease of MR at 15 T with temperature, shown in insets to figure 2, can than be explained with the change in mobility of carriers. Relative change of MR(15 T) and mobility μ, calculated as $\mu(\mathrm{T}) = 1/\mathrm{en_H}\rho_{\mathrm{xx}}(\mathrm{T})$, from 85 K to 6 K for sample annealed at 435 °C is shown in figure 4. It can clearly be seen that temperature change of mobility follows change in MR at 15 T.

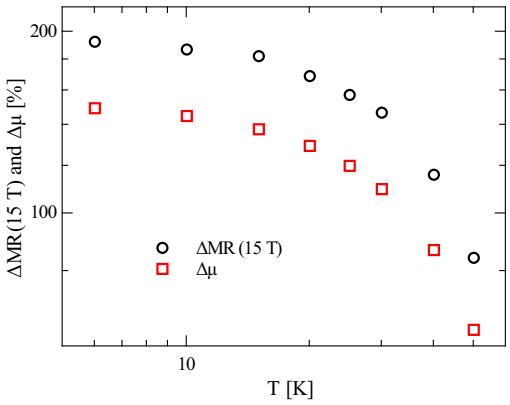

Figure 4. Relative change of MR(15 T) and mobility from 85 K to 6 K for $Pb_{0.83}Sn_{0.17}Se$ crystal annealed at 435 °C . $\Delta\mathrm{MR}(\mu) = (\mathrm{MR}(\mu)(\mathrm{T}) - \mathrm{MR}(\mu)(85\mathrm{K}))/\mathrm{MR}(\mu)(85\mathrm{K}) \cdot 100\%$

Quantum oscillation phenomenon is a powerful technique for measuring low temperature Fermi surface properties. Quantum oscillations can be understood quantitatively within the Lifshitz-Kosevich (LK) formalism [15]. Oscillation of different physical quantities with inverse magnetic field for three-dimensional electron gas can be described by the same functional form:

$$\Delta X(B,T) = A_0 A_T A_D A_S \left(\frac{B}{F}\right)^{1/2} \cos\left(2\pi\frac{\mathrm{F}}{\mathrm{B}} + \phi_{\mathrm{p}} + \phi_{\mathrm{M}}\right). \quad (1)$$

ΔX stands for oscillatory part of magnetization, magnetic susceptibility or conductivity (resistivity). F is the oscillation frequency and ϕ_p and ϕ_M are phase factors [1]. A_T, A_D and A_S are temperature, Dingle and spin dimensionless prefactors and A_0 is dimensional constant. $A_T = 2\pi^2(\mathrm{k_B T}/\hbar\omega_{\mathrm{c}})/\sinh[2\pi^2(\mathrm{k_B T}/\hbar\omega_{\mathrm{c}})]$, $A_D = \exp[2\pi^2(\mathrm{k_B T_D}/\hbar\omega_{\mathrm{c}})]$, where T_D is the Dingle temperature related to quantum scattering time, $A_S = \cos[\pi \mathrm{g m_e}/(2\mathrm{m_c})]$. g is the Lande factor, m_c is the effective cyclotron mass, m_e is the free electron mass and $\omega_c = eB/m_c$ is cyclotron frequency. In the inset to figure 3b) the temperature dependence of amplitude of oscillations in conductivity is shown. By fitting the temperature dependence of amplitude to functional form of A_T the effective cyclotron mass of carriers can be obtained, $m_c = 0.04m_e$. We have done a detailed analysis of dHvA and SdH oscillations in given $Pb_{0.83}Sn_{0.17}Se$ samples in [1]. Fermi

surface parameters obtained from dHvA and SdH oscillations [1] for sample annealed at 436 °C are the following: $k_F = 0.016$ Å^{-1}, $n_{osc} = 1.4 \cdot 10^{17}$ cm^{-3}, $v_F = 4.6 \cdot 10^5$ m/s, $E_F = 48$ meV, $T_D = 3.6$ K (quantum scattering time $\tau_Q = 3.4 \cdot 10^{-13}$ s). Nearly the same values are obtained also for the sample annealed at 435 °C . We can see that for the sample annealed at 436°C charge density determined from quantum oscillations and Hall resistivity are in good agreement taking into account four electron pockets at the L point ([111] direction, B-field is in [001] direction) of the Brillouin zone [6]. Because of the small hole contribution (bending of Hall resistivity at small magnetic fields) in the sample annealed at 435 °C two charge densities are not in such a good agreement. By performing the Landau level (LL) diagram [1] a nontrivial Berry's phase, associated with the 3D Dirac semiconductor phase in $Pb_{0.83}Sn_{0.17}Se$ crystal is confirmed. Also, the magnetic field of quantum limit is found to be around 8 T.

3. Conclusion

In conclusion, we have studied the low temperature magnetotransport and magnetization data measured from 300 K down to 1.8 K in high magnetic filed up to 15 T for semimetal $Pb_{0.83}Sn_{0.17}Se$ single crystals. We have observed the linear magnetization and linear MR accompanied by quantum oscillations at low temperature. We have found that the annealing of the $Pb_{0.83}Sn_{0.17}Se$ samples in the Se vapours at temperatures from 433 to 440 °C results in the crossover from n to p-type conductivity. The MR data reveal discernible Fermi surface change caused by the Zeeman splitting in the magnetic field range from 4 T up to quantum limit field of about 8 T. We have extracted the Fermi surface parameters using the LK theory. It has been found that the LMR scales with electron mobility in the $Pb_{0.83}Sn_{0.17}Se$ crystals and originates from fluctuating mobility regions due to inhomogeneous current paths.

Acknowledgments

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