Fermi Surface and Carriers Compensation of pyrite-type PtBi$_2$ Revealed by Quantum Oscillations

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Large non-saturating magnetoresistance has been observed in various materials and electron-hole compensation has been regarded as one of main mechanisms. Here we present a detailed study of angle-dependent Shubnikov-de Hass effect on large magnetoresistance material pyrite-type PtBi$_2$, which allows us to experimentally reconstruct its Fermi-surface structure and extract physical properties of each pocket. We find its Fermi surface contains four types of pockets in the Brillouin zone: three ellipsoidal-like hole pockets $\alpha$ with $c_4$ symmetry located on the edges (M points), one intricate electron pocket $\beta$ merged from four ellipsoids along [111] located on the corners (R points), two smooth and cambered octahedrons $\gamma$ (electron) and $\delta$ (hole) on the center (\Gamma point). The deduced carrier densities of electrons and holes from volume of pockets prove nearly perfect compensation. This compensation at low temperatures is also supported by our two bands by fitting field-dependence of Hall and magnetoresistance at different temperatures. We conclude that the compensation is the main mechanism for the large non-saturating magnetoresistance in pyrite-type PtBi$_2$. We found the hole pockets $\alpha$ may contribute major mobility because of their light masses and anisotropy to avoid large-angle scattering at low temperature, may point to common features of semimetals with large magnetoresistance. The found and sub-quadratic magnetoresistance is probably due to field-dependent mobilities induced by long-range disorders, another feature of semimetal under high magnetic fields.

Large magnetoresistance and its mechanisms have drawn new tremendous interests beginning from the discovery of WTe$_2$[1]. Following this discovery, nonmagnetic materials such as Cd$_3$As$_2$[2], WP$_2$[3], LaSb[4], TaAs[5], YSb[6], NbP[7], $\alpha$-As[8], NbSb$_2$[9] and more recently pyrite-type PtBi$_2$[10] have been discovered to show large magnetoresistance. In contrast to the ‘conventional’ semimetals, bismuth[11] and graphite[12], these materials show non-saturating magnetoresistance as high magnetic field is increased. Several mechanisms have been proposed to explain it. The first mechanism is the electron-hole compensation scenario which has been ascribed to the most cases listed above, also other materials[1 5 13 15]. Under the e-h ‘resonance’ condition ($n_e = n_h$), the magnetoresistivity $\rho(B) \approx \frac{B^2}{2e^2} \frac{\mu_e \rho_{ee} + \mu_h \rho_{he}}{\mu_e + \mu_h}$ never saturates and exhibits a quadratic dependence of magnetic field, where $n_e (n_h)$ and $\mu_e (\mu_h)$ are density and mobility of electron(hole). Other mechanisms range from topological protection[2 4 16] to metal-insulator transition by magnetic field[17 18].

Recently, the pyrite-type PtBi$_2$ was predicated a 3D Dirac semimetal[19]. Following the prediction, large non-saturating magnetoresistance has been experimentally observed in the pyrite PtBi$_2$[10]. Its magnetoresistance is in the front rank in the discovered materials, reaching 1.12x10$^7$% at 1.8 K and 33 T. The origin of the large magnetoresistance was preliminary discussed based on the compensation scenario by fitting with two-band model[10]. But the detailed Fermi surface and its relation to large magnetoresistance are still missing. On the other hand, may be due to the difficulty of peeling a pyrite-structure sample, ARPES data also has not been reported yet. Unlike its hexagonal polymorph PtBi$_2$, ARPES results[20 21] suggest a Dirac-cone-like dispersion may lead to its unconventional large linear magnetoresistance[22 23].

In this paper, we reveal that the compensation of PtBi$_2$ would be the mechanism for the large non-saturating magnetoresistance by verifying the e-h balance through mapping Fermi surface directly by angle-dependence of Shubnikov-de Hass effect (SdH) and also by two-band model from temperature-dependence of Hall resistivity and magnetoresistivity. Our mapped Fermi surface from angle-dependence of SdH suggests four types of Fermi pockets instead of three types of pockets from the previous calculation[10]: three equivalent hole ellipsoid-like pockets named $\alpha$ on edge(M of Brillouin zone), one electron pocket named $\beta$ located on the corners (R), a large octahedron-like electron pocket ($\gamma$) and a small octahedron hole pocket ($\delta$) on the center(\Gamma) of Brillouin zone. The electron-hole ratio is nearly compensated within an accuracy of 1% after summing up the carrier densities from whole obtained pockets by their types. This compensation is also confirmed by our two bands fitting for temperature-dependence of Hall and magnetoresistance.

Pyrite-type PtBi$_2$ single crystals were grown by the flux method with Pt:Bi=1:20 molar ratio. Pt powder (99.95%) and Bi grains (99.999%) were mixed and sealed in an evacuated quartz ampoule. The ampoule was heated to 450°C and preserved for 10h, then cooled down to 300°C in 150h. After a centrifugation, the single crystals were separated from the flux. The dimension of a typical as-grown sample is around $2 \times 2 \times 2 mm^3$. The
sample was then cut into a narrow strip by a wire-saw to carry on the transport measurement. The measurement was performed in Teslatron PT (Oxford Instruments) and with a home-made rotator whose angle can be controlled by the data-collecting computer. Angles of the motor-driven rotator were determined either by a Hall probe on sample holder or by steps from the driving motor which was calibrated before measurement. The two methods show a good consistence about angle. The current was applied by a Keithley 6221 and the voltage was measured by a Keithley 2181 A. The magnetic field was perpendicular to the current during rotation.

Fig.1 shows the temperature-dependence of resistivity \( \rho_0(T) \) of the measured sample which has a residual resistivity ratio \( RRR = \rho(300K)/\rho(1.7K) \) of 546. More samples with different RRR were measured and show same quantum-oscillation results. Both resistivity at room temperature of 132 \( \mu\Omega cm \) and 0.24 \( \mu\Omega cm \) at 1.7 K are higher than these of reported. Such differences may be due to the mobility is different from different orientation by the applied current. For instance, the mobility is closely related to the orientation of applied current in bismuth [24]. The inset of Fig.1 shows the X-ray diffraction pattern of the photographed sample whose crystallographic direction [010] is labeled with a blue arrow. The sharpness of the (002) indicates the high quality of the sample.

Fig. 2(a) displays a typical field-dependent \( \rho(B) \) curve with pronounced SdH oscillations at 1.7 K as the magnetic field along [001] and the current along [100]. The inset shows the MR \( [(\rho(B) - \rho(0))/\rho(0) \times 100\%] \) from the same data in blue curve and also with data in pulsed fields up to 55 T where the MR reaches 8,160,000%. The MR curve have a sub-quadratic dependence in magnetic fields. b) FFT spectra of SdH oscillations after subtracted background from magnetoresistance. The SdH oscillations as a function of 1/B are also shown in the inset. The oscillations start at as low as 2 T indicates high mobility of the sample. We also indexed the sharp peaks of FFT spectra accordingly, c, d). The cyclotron masses and Dingle temperatures of different pockets as field along [001] extracted from temperature-dependence of SdH with Lifshits-Kosevich theorem [26].

![Fig. 1](https://example.com/f1.png)

**Fig. 1.** a) Temperature-dependence of resistivity from 4.1-300 K. The residual resistivity ratio \( \rho(300K)/\rho(4.2K) \) is about 455. The inset shows the X-ray diffraction pattern of the (001) facet of the photographed sample which was then cut into a smaller sample for measurement. The current was along the [010] orientation during whole measurement.

![Fig. 2](https://example.com/f2.png)

**Fig. 2.** a) Field-dependent \( \rho(B) \) curve at 1.7 K as magnetic field along [001] and the current along [100] using Teslatron PT of Oxford Instrument. The inset shows the MR \( [(\rho(B) - \rho(0))/\rho(0) \times 100\%] \) from the same data in blue curve and also with data in pulsed fields up to 55 T where the MR reaches 8,160,000%. The MR curve have a sub-quadratic dependence in magnetic fields. b) FFT spectra of SdH oscillations after subtracted background from magnetoresistance. The SdH oscillations as a function of 1/B are also shown in the inset. The oscillations start at as low as 2 T indicates high mobility of the sample. We also indexed the sharp peaks of FFT spectra accordingly, c, d). The cyclotron masses and Dingle temperatures of different pockets as field along [001] extracted from temperature-dependence of SdH with Lifshits-Kosevich theorem [26].

Even the RRR is lower than the reported [10]. The MR curve can be fitted with a power law \( MR = aB^{1.79} \), deviated from quadratic dependence in field. The inset of the Fig. 2(b) presents the SdH oscillations after subtracted the background by fitting polynomial fitting. The SdH oscil-
lations can be clearly seen from $B = 2 \text{T}$, indicating high carrier mobility of the sample by a first approximate to have the mobility $\mu \geq 0.57^{-1}$, also allow us to extract the fine structure of the Fermi surface. The SdH oscillations as a function of $1/B$ are also shown in the inset, these oscillations are still observable down to 0.5 $^{-1}$ after enlarging 50 times. We then extract fast Fourier transform (FFT) spectra of SdH oscillations and plot in Fig. 2(b). Each FFT peak at the fundamental or higher harmonics can calibrate the extremal Fermi surface area which is perpendicular to magnetic field [20]. We can clearly identify and index the sharp peaks of FFT spectra for four different frequencies named $\alpha$, $\beta$, $\gamma$ and $\delta$ in Fig. 2(b). Note that the amplitude of the second harmonic frequency of $\delta$ is higher that of the fundamental frequency. This is probably due to spin-splitting of this band [20] and confirmed in our later angle-dependence of SdH. After the field-rotation, the fundamental frequency becomes larger as a normal oscillations does. The cyclotron masses and Dingle temperatures for four pockets along as field along [001] are shown in Fig. 2(c, d), extracted from SdH at different temperatures with Lifshits-Kosevich formula[26] by the attenuation factors due to finite temperature:

$$R_T = \frac{X}{\sinh X} \text{ and impurity scattering }$$

$$R_D = \exp\left(-\frac{\pi m^*}{eB\tau_0}\right) \text{ where } X = \frac{2e^2k_BTm^*}{\hbar^2}.$$  

Now we can map the Fermi surface by rotating sample to get angle-dependence of SdH, the schematic diagram of the sample geometry is shown in Fig. 3(a). Through the above procedure, we obtained FFT spectra of SdH at various angles by rotating sample at 1.7 K from [001] to [001]. Fig. 3(b) shows the shifted FFT spectra by every 5 degrees from 0 [001] to 90 degrees [010]. We have not show the data beyond 90 degrees, since the angle-dependence of SdH pattern exactly repeats because of cubic crystal structure nature. The fig.3 (c) shows angle-dependence of FFT peaks in different symbols. The branch $F_{\alpha_i}(\theta)$ displaces 90 degrees from the branch $F_{\alpha_i}(\theta)$. So we deduce that the three equivalent $\alpha$ pockets locate on the edge because of their C4 symmetry with an assumption that pockets always have the highest symmetric locations. This assumption is also for determining locations of other types of pockets. Another almost constant frequency $F_{\alpha_i}(\theta)$ spectrum is absent in current results. This absence should be due to the lower mobility when the current is along the long axis of $\alpha_3$ ellipsoid, as seen in YSb[6]. Then for the $\alpha$ pocket which are prolate spheroids, we fitted the angle-dependence of $F_{\alpha_i}$ quantitatively by the following equation:

$$F_{\alpha_i} = F_0/\sqrt{\left(\cos[\theta-(i-1)\pi/2]\right)^2 + \left(\lambda \sin[\theta-(i-1)\pi/2]\right)^2}$$

(1)

Where we can obtain $F_0$=202T, $\lambda = 0.55$ and ‘i’ is for the subscript of $\alpha$. By Onsager relation $F = (\hbar/2\pi e) A_k$ between frequency $F$ and the extreme cross section $A_k$ of a Fermi surface, we extracted the values listed in Table I. The carrier density for each equivalent $\alpha$ prolate spheroid is $0.29 \times 10^{26} \text{cm}^{-3}$. The type of this pocket is hole, by taking the previous report[10] as reference. The types of other pockets are deduced by the same route.

The $\beta$ pocket has a quite complicated structure, but can be sorted out. The shape from $F_\beta(\theta)$ resembles with that of ellipsoid, but rotated 45 degrees along the [100] axis. According to highest symmetry, the long axis of ellipsoid lies [111] to have a maximum cross section of an ellipsoid at 45 degrees in current case. Note that this ellipsoid is tilted when the field is rotated while the rotating axis is not along its axes. Then the equation (1) is no longer valid for this case. We first exclude that
TABLE I. Summary of physical properties: volume, quantity, carrier densities (n), cyclotron mass (m<sub>cyc.</sub>), dingle temperature T<sub>D</sub>, relaxation time τ<sub>D</sub> and mobilities (µ<sub>D</sub>) of α, β, γ and α pockets. The momentum |k<sub>a</sub>, k<sub>b</sub> and k<sub>c</sub>|, k<sub>edge</sub> and k<sub>α</sub> were axes of an ellipsoid, an edge length of octahedron Fermi surface and a radius of the sphere in the middle of β, respectively.

| Quantity                  | 3          | 1          | 1          | 1          |
|---------------------------|------------|------------|------------|------------|
| V(Å<sup>3</sup>)          | 0.0036     | 0.0221     | 0.0104     | 0.0225     |
| n(cm<sup>-3</sup>)        | 0.87×10<sup>20</sup> | 1.88×10<sup>20</sup> | 0.84×10<sup>20</sup> | 1.82×10<sup>20</sup> |
| m<sub>cyc.</sub>(<i>m</i><sub>e</sub>) | 0.078      | 0.11       | 0.18       | 0.24       |
| T<sub>D</sub>(K)           | 29         | 27         | 13         | 12         |
| τ<sub>D</sub>(s)           | 4.2×10<sup>-14</sup> | 4.5×10<sup>-14</sup> | 9×10<sup>-14</sup> | 1×10<sup>-13</sup> |
| µ<sub>D</sub>(cm<sup>2</sup>/Vs) | 944        | 719        | 913        | 742        |

These ellipsoids exist independently, locating between Γ and R. By geometric consideration of independent ellipsoid, we extract its short axis k<sub>a</sub> = k<sub>b</sub> = 0.094 Å<sup>-1</sup> and its long axis k<sub>c</sub> = 0.33 Å<sup>-1</sup>. But the β band will touch other two bands α and δ (discuss below) since the diagonal length of Brillouin zone which is only 1.62 Å<sup>-1</sup> by considering two long axes of β bands lie along [111] according to the symmetry. More quantum oscillations are expected from this touching, which contradicts with our observations. Also the carrier density from 8 pockets would surpass 10<sup>23</sup> cm<sup>-3</sup> close to a metal, which contrasts with the semimetal property from its resistivity. So these β pockets should depend on and have to cross each other, leading to the total number of ellipsoid also its length (illustrate them in fig.3(d)). We obtain the volume of this pocket adding four ellipsoids whose middle parts are truncated and one middle sphere. By considering the frequencies of an ellipsoid tilted along [111] when the field is at 0, 45 degrees also the fact that the electrons travel actually cross two connected ellipsoids at θ = 0 in this case, we extracted actual k<sub>a</sub> = k<sub>b</sub> = 0.0792 Å<sup>-1</sup> and k<sub>c</sub> = 0.1584 Å<sup>-1</sup> and the radius of the middle sphere is k<sub>s</sub> = 0.1094Å<sup>-1</sup> from the of the lower frequency of F<sub>β</sub> at 45 degrees. Finally, the carrier density of this hole-like pocket is 1.88×10<sup>20</sup> cm<sup>-3</sup>. Although the shape is not an exact square in the polar plot Fig. 3(d) as an octahedron does, we can still treated the γ and δ bands as two smooth and cambered octahedrons whose symmetry naturally meet the requirement of a cubic. These octahedrons should locate at corner or the middle of Brillouin zone because of their high symmetry. Under this assumption, we calculated the geometric mean of the edge length (the solid line in Fig. 3(d)) of the minimal and maximal octahedron of δ which are able to encircle the band (the dash lines in the fig. 3(d)). The same procedure is used to obtain the edge length of octahedron for γ. After calculating volumes, we deduce the densities of electron-like γ and hole-like δ are 0.84×10<sup>20</sup> and 1.82×10<sup>20</sup> cm<sup>-3</sup>, respectively.

Fig. 3(e) is a scale drawing in a certain scale, in which we summarized and reconstructed the Fermi surface according to our SdH results. Note that the symmetry of β, γ and δ are same. So another possibility of locations of pockets is that γ and δ locate on the corners and β is on the center of the Brillouin zone. However, the total hole(α and δ) and electron(β and γ) carrier densities, were not affected by locations of pockets in the two cases, are 2.69×10<sup>20</sup> and 2.73×10<sup>20</sup> cm<sup>-3</sup>, respectively. This result suggests nearly perfect compensation between electrons and holes at a ratio of 0.99.

The carrier densities are typical for semimetals, such as WTe<sub>2</sub>(6.6×10<sup>19</sup>)<sup>17</sup>, LaSb(1.1×10<sup>20</sup>)<sup>4</sup>, Sb(5.5×10<sup>19</sup>)<sup>27</sup>, WP<sub>2</sub>(1.4×10<sup>20</sup>)<sup>3</sup> and also α-As(1.1×10<sup>20</sup>)<sup>8</sup>. And all their mobilities are around 10<sup>4</sup> (up to 10<sup>6</sup> in Sb) cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup>. Such sharing common properties may be the key to understand large magnetoresistance. For a compensated semimetal, we would expect a quadratic dependence of magnetoresistance, but the MR = aB<sup>2</sup> in this material. Such deviation of quadratic dependence of magnetoresistance is prevalent, due to unavoidable field-induced reduction in mobility by disorders<sup>25</sup>.

To further illustrate the compensation, we carried out temperature-dependence of magnetoresistance and Hall resistivity shown in Fig. 4(a, b) and found a similar re-
result as the previous report\cite{10}. We extracted the carrier densities and mobilities by fitting the two-band model:

\[ \sigma_{xy}(B) = \frac{\rho_{xy}(B)}{\rho_{xx}(B)+\rho_{yy}(B)} = \left[ \frac{n_e \mu_e^2}{1+(\mu_e B)^2} - \frac{n_h \mu_h^2}{1+(\mu_h B)^2} \right] eB, \]

present in Fig. 4(c, d). Although a slight amount discrepancy between electron and holes at high-temperature range, the carrier density between two types of carrier turns equal below 20 K and the ratio \(n_e/n_h\) is about 0.99 at 1.7 K from fitting. The carrier density \(n_e = 1.32 \times 10^{20} \text{cm}^{-3}\) and \(n_h = 1.33 \times 10^{20} \text{cm}^{-3}\) are also quite close to the values form the Fermi-surface method.

The two-band model gives relatively lower numbers, but should be higher since the carrier density easily surpass \(1.8 \times 10^{20} \text{cm}^{-3}\) even only from a 6 band. This may be the disadvantage of two-band models which give estimated values. The mobility of hole increases faster than that of electron to \(1.5 \times 10^5 \text{cm}^2/\text{Vs}\) as temperature is lowered. This increase of hole mobility may mainly result from the contribution of \(\alpha\) pockets which has the lightest masses and its anisotropy (\(k_{\text{Fe}}/k_{\text{Fe}} \approx 2\)). Such two features are prevalent in the semimetals at least in one of their Fermi pockets: Sb\[25, 27\](\(m^* \sim 0.088m_e\), anisotropy of \(k \sim 5.2\)), bismuth\[28\](\(m^* \sim 0.0011m_e\), anisotropy of \(k \sim 15\)), YBi\[6\] [29] [30](\(m^* \sim 0.2m_e\), anisotropy of \(k \sim 2\)) and WTe\[13\] [31](\(m^* \sim 0.4m_e\), anisotropy of \(k \sim 2-3\)). The \(\mu_{e/H}\) is around 150 and 25 for holes and electrons, respectively, \(\mu_f\) is mobility from two-band fitting. This indicates small-angle-scattering process plays a significant role during carriers transporting after electron-phonon scattering is faded as temperature is decreased, which may be a common feature in semimetals\[29\]. So to have a pocket with the light mass and anisotropy may favor a small-angle-scattering at low temperature.

In summary, we have experimentally mapped out the Fermi surface of pyrite PtBi\(_2\) including two hole and two electron pockets by angle-dependence of ShD measurements. This allows us to deduce the carrier density and mobility of each pockets and to reveal a compensation between the electron and hole. Such compensation further is confirmed by two-band fitting from temperature-dependence of magnetoresistance and Hall resistivity. We ascribed large non-saturating magnetoresistance found in this material to this compensation. The high hole mobility may be due to the light mass and anisotropy of \(\alpha\) pockets, causing domination of the small-angle-scattering process in low temperatures.

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