Evidence of decoupling of surface and bulk states in Dirac semimetal \( \text{Cd}_3\text{As}_2 \)

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Abstract

Dirac semimetals have attracted a great deal of current interests due to their potential applications in topological quantum computing, low-energy electronic devices, and single photon detection in the microwave frequency range. Herein are results from analyzing the low magnetic field weak-antilocalization behaviors in a Dirac semimetal \( \text{Cd}_3\text{As}_2 \) thin flake device. At high temperatures, the phase coherence length \( l_\phi \) first increases with decreasing temperature \( T \) and follows a power law dependence of \( l_\phi \propto T^{-0.4} \). Below \( \sim 3 \) K, \( l_\phi \) tends to saturate to a value of \( \sim 180 \) nm. Another fitting parameter \( \alpha \), which is associated with independent transport channels, displays a logarithmic temperature dependence for \( T > 3 \) K, but also tends to saturate below \( \sim 3 \) K. The saturation value, \( \sim 1.45 \), is very close to 1.5, indicating three independent electron transport channels, which we interpret as due to decoupling of both the top and bottom surfaces as well as the bulk. This result, to our knowledge, provides first evidence that the surfaces and bulk states can become decoupled in electronic transport in Dirac semimetal \( \text{Cd}_3\text{As}_2 \).

Keywords: Dirac semimetal, surface and bulk states, weak anti-localization

Some figures may appear in colour only in the online journal.
The weak antilocalization (WAL) effect has commonly been used to examine the coupling/decoupling of the surface and bulk states in topological quantum materials [22–42]. In a system with spin-momentum locking and the resulting \( \pi \) Berry phase, the constructive interference of two time-reversal paths gives rise to a magnetoconductivity correction (equation (1)) [26, 34, 43] as follows:

\[
\Delta \sigma(B) = \sigma_{xx}(B) - \sigma_{xx}(0) = \alpha \times (e^2/\pi \hbar) f(B_0/B),
\]

where \( \sigma_{xx}(B) \) is the conductivity at a magnetic field of \( B \), \( \sigma_{xx}(0) \) the conductivity at \( B = 0 \), \( e \) the electron charge, \( h \) the Planck constant, \( f(x) \equiv \ln(x) - \Psi(1/2 + x) \) with \( \Psi \) being the digamma function. \( B_0 \) is the magnetic field associated with the phase coherence length \( l_\phi = (h/eB_0)^{1/2} \). \( \alpha \) is related to the number of independent transport channels. In previous studies on WAL in topological insulators, such as Bi\(_2\)Se\(_3\), \( \alpha \) is determined by coupling/decoupling of the surfaces and bulk [26, 28–33, 38]. When the surfaces and bulk are strongly coupled, they are treated as one coherent channel and \( \alpha = 0.5 \). When the top and bottom surfaces are decoupled from the bulk, \( \alpha = 1 \). Figure 1(a) shows an optical image of an as-grown Cd\(_3\)As\(_2\) ingot. (b) Powder XRD plot simulated using single crystal data.

Figure 1, (a) Optical image of an as-grown Cd\(_3\)As\(_2\) ingot. (b) Powder XRD plot simulated using single crystal data.
terminal longitudinal resistance $\rho_{xx}$ and Hall resistance $\rho_{xy}$ are measured using the low-frequency ($\sim 11$ Hz) phase lock-in technique. Two Stanford Research Systems Inc. SR830 lock-in amplifiers are used. Lock-in amplifier 1 provides a constant AC voltage (1 V) to induce a current of 10 nA into the sample, through a current-limiting resistor of 100 M$\Omega$ (much larger than the sample resistance) [52]. This lock-in amplifier measures $\rho_{xx}$. The second lock-in amplifier, synchronized with lock-in amplifier 1, measures the Hall resistance $\rho_{xy}$. At the excitation current of 10 nA, we estimate that electron self-heating is negligible.

Figure 2(a) shows $\rho_{xx}$ as a function of temperature at zero magnetic field. Three regimes with different temperature dependencies are observed. In the temperature range of $10 < T < 50$ K, it is clearly seen that $\rho_{xx}$ follows a linear $T$ dependence and $\rho_{xx} = 928 - 0.77x$, in units of $\Omega$. Between $\sim 3$ and $10$ K, $\rho_{xx}$ displays a logarithmic $T$ dependence, as shown in figure 2(b). This logarithmic temperature dependence is caused by the weak localization effect. Indeed, in a diffusive electron system the destructive quantum interference between two identical self-crossing paths (in which an electron propagates in the opposite directions) leads to an increase in resistivity, which follows a logarithmic temperature dependence [53]. Below $3$ K, $\rho_{xx}$ increases at a much slower rate and tends to saturate to a value $\sim 931 \, \Omega$. Figure 2(c) shows the magneto-resistivity $\rho_{xx}(B)$, taken at $T = 1.3$ K, in a large magnetic ($B$) field range. The pronounced weak-antilocalization cusp near the zero magnetic field is clearly seen, consistent with previous work in topological insulators and semimetals [22–42, 44, 45]. Fluctuations are also observed in magneto-resistivity.

Figures 3(a) and (b) show $\rho_{xx}(B)$ and Hall resistivity $\rho_{xy}(B)$, respectively, around $B = 0$ T at a few selected temperatures. As shown in figure 3(a), the amplitude of fluctuations becomes weaker and eventually disappears at higher temperatures. The Hall resistivity (figure 3(b)) displays a linear $B$ field dependence in the low magnetic fields range around $B = 0$. All the traces overlap with each other, indicating a constant electron density in the temperature range studied. In figure 3(c), we plot the area density $n_{3D}$, obtained from the slope of $\rho_{xy}(B)$ as a function of temperature. In the temperature range of $0.5 < T < 38$ K, $n_{3D} \sim 1.5 \times 10^{13}$ cm$^{-2}$. We note that a constant electron density at low temperatures has also been observed before [54].

Moreover, it is believed that a finite electron density in unintentionally doped Cd$_3$As$_2$ is due to arsenic vacancies [55]. The 3D density is estimated to be $n_{3D} \sim 7.5 \times 10^{17}$ cm$^{-3}$, considering the thickness of the thin flake is $\sim 200$ nm. Consequently, the Fermi energy $E_F$ of the system, estimated by using the following formula (equation (2)), is $\sim 100$ meV, which is close to the theoretically calculated Lifshitz transition point [48]

$$E_F = \hbar^2 (3\pi^2 n_{3D})^{2/3} / 2m^*.$$  

(2)

Here, the effective electron mass in Cd$_3$As$_2$ is taken as $m^* = 0.03$ (in the units of free electron mass).

We caution here that the Fermi energy is calculated based on a simplified model and does not consider the ellipsoid correction [48]. Moreover, there exists a large discrepancy in the literature on the position of the Lifshitz transition point in Cd$_3$As$_2$ [6, 10, 48, 56–58]. Theoretically, it was estimated to be $\sim 130$ meV in reference [48]. Experimentally, the measured value differs significantly from one work to another. In [58], it was estimated $\sim 200$ meV using the electronic transport technique. On the other hand, a value of as small as $\sim 10$ meV was estimated in [10] using the STM technique. Understanding the origin of this discrepancy, though extremely important, is beyond the scope of this work. More future studies are needed.

In the following, we will focus on the weak-antilocalization behavior. As shown in figure 3(a), the WAL behavior is weakened as $T$ increases. To analyze the weak-antilocalization effect, we follow the previous practices in topological insulators and topological semimetals and use the HLN formula [43] to analyze the weak-antilocalization effect. In fitting the experimentally measured data, we first convert the resistivity to conductivity, $\sigma_{xx}(B) = \rho_{xx}(B)/(\rho_{xx}(B)^2 + \rho_{xy}(B)^2)$. The magneto-conductivity $\Delta \sigma(B) = \sigma_{xx}(B) - \sigma_{xx}(0)$ is then fitted by the formula (equation (3)):

$$\Delta \sigma(B) = \alpha \varepsilon^2 / \pi \hbar \times [\ln(h/4e\varepsilon^2 B) - \Psi(1/2 + h/4e\varepsilon^2 B)].$$

(3)

In figure 3(d), we show a representative fitting at one temperature of 6.8 K. A good fitting is seen in the low magnetic field range.

We note here that the HLN formula, developed for two-dimensional electron systems (2DES), fits our data well, considering that our device is 200 nm thick. On the other
hand, this is not surprising by comparing the $\rho_{xx}(B)$ data in our specimen (figure 2(c)) with the magneto-resistivity data that are well fitted by a 3D WAL model in [42]. $\rho_{xx}(B)$ in our specimen shows the strong cusp feature typically observed in 2DES. In contrast, the $\rho_{xx}(B)$ data that are well fitted by the 3D WAL model generally shows a quadratic-like $B$ field dependence (see figure 3 in [42]). In fact, the $\rho_{xx}(B)$ curve that shows the cusp feature in their least-doped sample needs to be fitted by the HLN formula, even though the thickness is also about 200 nm (see supplementary materials in [42]). In the following, we discuss a couple of possible mechanisms. First, it is known that the spin–orbit coupling in Cd$_3$As$_2$ is strong. As a result, the spin–orbit scattering time can be significantly shorter than the phase coherence time [42] in our specimen. Consequently, the single coherence channel HLN formula can be valid in the bulk [25]. Second, in our specimen, the phase coherence length is on the order of the device thickness ($\sim$200 nm). This can also make the HLN formula a good approximation in fitting the weak-antilocalization effect for the bulk.

In figures 4(a) and (b), we plot the obtained $l_0$ and $\alpha$ as a function of $T$. $l_0$ follows a power-law temperature dependence in the range of $\sim$4–40 K, $l_0 \sim T^{-0.4}$, with a power law coefficient of 0.4, suggesting that electron–electron scattering is the main mechanism for the dephasing process [53] in our device. When $T$ is lower than 3 K, $l_0$ increases at a much slower rate and tends to saturate to a value of $\sim$180 nm. The value of $\alpha$ also increases with decreasing $T$ and follows a logarithmic dependence between $T\sim$10 and 40 K (see figure 4(b)). At present, the exact origin of this logarithmic $T$-dependence is not known. Nevertheless, it indicates that the decoupling of the surfaces and bulk states is a not sudden transition. Rather, it is a gradual process. As $T$ is further reduced below $\sim$3 K, $\alpha$ also increases at a much slower rate and tends to saturate to a value of $\sim$1.45. This low-temperature value of 1.45 is larger than 1, suggesting contributions

Figure 3. (a) Magneto-resistivity $\rho_{xx}$ around $B = 0$ T at five selected temperatures of 0.5, 0.7, 4.8, 13.7, and 30.9 K. (b) Hall resistivity $\rho_{xy}$ as a function of $B$ at the same selected temperature. Linear $B$ dependence is seen. (c) 2D electron density, obtained from the slope of the linear $B$ dependence of $\rho_{xy}$, as a function of temperatures. (d) HLN fitting of the weak antilocalization effect at the temperature of 6.8 K.
from more than two independent channels. Considering it is very close to 1.5, we suggest that at low temperatures there exist three independent parallel channels. In a 3D Dirac semimetal like Cd₃As₂, these three parallel channels can become possible if both the top and bottom surfaces as well as the bulk all become decoupled. Indeed, both the 2D WAL effect from the top and bottom surfaces and the 3D WAL effect have been observed before [23]. Also, independent surface and bulk channels are observed in the studies of Josephson junctions in Cd₃As₂ [11, 12]. Mechanisms other than decoupled surfaces and bulk states might also be possible. For example, the Weyl orbits [59] may contribute to the weak anti-localization effect and cause α to be close to 1.5 in Dirac semimetals. More studies are needed.

Our obtained value of $l_\phi$ at low temperatures is consistent with that previously reported in Cd₃As₂ samples grown by MBE technique [35, 37]. This seems to suggest that $l_\phi$ is independent of how the materials is prepared. On the other hand, the value of α is significantly different. In the MBE grown Cd₃As₂ thin films [35, 37], α is considerably less than 0.5. It only approaches to 0.5 at low temperatures [37]. This low value of α is probably due to a small film thickness in their samples, which can result in strong coupling of the two surfaces and bulk. Consequently, only one independent transport channel exists. The asymmetric contribution from the surface and the bulk [26] can further reduce α to a value of less than 0.5. Our Cd₃As₂ thin flake device is significantly thicker. As a result, at low temperatures, the two (top and bottom) surfaces and the bulk can become decoupled and give rise to three independent channels and, thus, a large α value.

It is surprising that all three parameters, the resistivity, $l_\phi$, and α, tend to saturate below ~3 K. We speculate that the decoupling of top/bottom surfaces and the bulk may be responsible for the tendency. Indeed, if the bulk-surface scattering is the main mechanism for the sample resistivity in Cd₃As₂ and dephasing of quantum interference, when all three are decoupled, this scattering mechanism is strongly suppressed. Consequently, $\rho_{xx}$, $l_\phi$, and α can saturate to a constant value, respectively. Additional measurements are ongoing to further explore whether and how the decoupling is related to the π phase difference between the surface and bulk states [11].

In conclusion, we have synthesized single crystals of pure Cd₃As₂ and fabricated thin flake devices to measure their electronic transport properties. We present results from our systematic studies of the weak-antilocalization effect. The HLN formula is used to analyze WAL, from which the phase coherence length $l_\phi$ and the constant α (which is related to independence transport channels) are obtained. It is observed that $l_\phi$ follows a power law dependence with $T$ at high temperatures, but saturates to ~180 nm below $T \sim 3$ K. α displays a logarithmic dependence for $T > 3$ K, and saturates below ~3 K. Surprisingly, the saturation value α is very close to 1.5, indicating three independent transport channels probably due to the decoupling of both the top and bottom surfaces as well as the bulk states in our Cd₃As₂ thin flake sample. This observation of decoupled surface channels is expected to have important implications for topologically-protected device applications.

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necessarily represent the views of the U.S. Department of Energy or the United States Government.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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