Three-dimensional (3D) topological Dirac semimetals (TDSs), such as Na3Bi [1, 2] and Cd3As2 [3], have been theoretically predicted and experimentally realized in recent years. These TDSs have unique physical properties originating from 3D massless Dirac fermions. The Dirac nodes are developed due to the touching of valence and conduction bands at discrete points in the reciprocal space. Interestingly, for these 3D Dirac materials, they can be driven into various phases, such as Weyl semimetal [4] and topological insulator (TI) [5, 6] by breaking time reversal or inversion symmetry. Driven by these intriguing physical properties, extensive experiments on angle-resolved photoemission spectroscopy (ARPES) [7–10] and scanning tunneling microscopy (STM) [11] were carried out to identify the 3D Dirac fermions in these materials.

Cd3As2 is considered to be an excellent 3D TDS due to its chemical stability in air, and it also possesses novel transport properties such as ultrahigh mobility [12], large magneto-resistance (MR) [13], non-trivial Berry’s phase of Dirac fermions [14] and chiral anomaly induced negative MR [15, 16]. Previously, Cd3As2 bulk materials, amorphous films [17], nanowires [18] and platelets [19] were prepared by various growth methods, and magneto-transport measurements so far have focused on Cd3As2 bulk materials. However, few efforts were devoted to thin films [20–22] and nanostructures [18, 23], which may exhibit surface phase coherent transport and quantum size limit effect [24–26], leading to Aharonov-Bohm oscillations [27] and the exotic quantum Hall insulator states [3]. Importantly, a theoretically predicted TI phase and thickness-dependent quantum oscillations may also eventually emerge when the dimensionality of the system is reduced [3, 4]. Therefore, it is highly desirable to fabricate superb crystallinity Cd3As2 thin films for the transport study at
different temperatures, as well as the possible thickness-induced semimetal-to-semiconductor transition in the Cd$_3$As$_2$ system.

In this letter, we performed a systematic magneto-transport experiments for the undoped Cd$_3$As$_2$ thin films with different thickness ranging from 50 to 900 nm by molecular beam epitaxy (MBE). A semimetal-to-semiconductor and a discernable non-trivial to trivial Berry’s phase transition were observed. We also extended our study to the temperature- dependence of magneto-resistance, in addition to Shubnikov-de Hass (SdH) oscillations that allow us to analyze the relevant parameters.

A series of wafer-scale Cd$_3$As$_2$ thin films were grown with a CREATEC MBE system (base pressure < 2 $\times$ $10^{-10}$ mbar). Freshly cleaved insulating mica substrates were specifically chosen for the magneto-transport measurements, and they were degassed at 350 $^\circ$C for 30 min to remove any molecule absorption prior to the growth. Subsequently, the substrates were cooled down to the desired growth temperature of 225 $^\circ$C. The Cd$_3$As$_2$ thin film deposition was carried out by co-evaporating high-purity Cd (99.9999%) and As (99.9999%) from dual-filament and valve-cracker effusion cells, respectively. The beam-flux ratio Cd/As was maintained at $\sim$3, and the growth process was in situ monitored by reflection high-energy electron diffraction (RHEED). The thickness of the films was controlled by growth time and determined by a step profiler.

The crystal structure and quality of the Cd$_3$As$_2$ thin films were examined by high-resolution x-ray diffraction (XRD) technique using Cu-K$_\alpha$ radiation, as shown in figure 1(a). By matching with a standard XRD card [28], a tetragonal body center crystal structure with lattice constants $a = b = 12.633(3)$ Å and $c = 25.427(7)$ Å was found [29–31] (figure 1(b)). Moreover, these XRD peaks can be indexed as a series of (112) planes and the full width at half maximum (FWHM) is less than 0.18$^\circ$ for (224), verifying the high crystallinity of the Cd$_3$As$_2$ thin films. The sharp and streaky RHEED pattern (figure 1(b) inset) illustrates an ideal atomically flat surface for as-grown Cd$_3$As$_2$ thin films [32, 33]. Figure 1(c) depicts the x-ray Photoelectron Spectroscopy (XPS) of a typical Cd$_3$As$_2$ thin film, where Cd 4d, As 3d and As 3p (spin–orbit split doublet) core levels are clearly witnessed. The

**Figure 1.** Characterizations of as-grown Cd$_3$As$_2$ thin films. (a) X-ray spectra. The marked peaks are the typical Cd$_3$As$_2$ (112) plane XRD patterns. Other peaks come from the mica substrate and the peak intensity increases with the thickness of Cd$_3$As$_2$. (b) Cd$_3$As$_2$ in a tetragonal body center unit cell with $a = b = 12.633(3)$ Å and $c = 25.427(7)$ Å. The inset figure shows the corresponding in situ RHEED pattern for the Cd$_3$As$_2$ thin films. (c) Typical core-level photoemission spectrum measurements for a Cd$_3$As$_2$ thin film, where Cd 4d, As 3d and As 3p peaks are clearly observed. The composition of Cd$_3$As$_2$ can be derived from the integrated area of these peaks and corresponding sensitivity factors. (d) Raman spectra for different thickness Cd$_3$As$_2$ thin films.
The fact that no extra peaks are observed indicates a good surface quality with stoichiometric composition. Raman spectra were recorded at room temperature from a spectrometer equipped with a 514.5 nm laser. Two peaks of $A = 191.2$ cm$^{-1}$ and $B = 245.7$ cm$^{-1}$ were clearly observed \[34, 35\] as shown in figure 1(d), and their peak positions remain nearly unchanged irrespective to the film thickness.

The Cd$_3$As$_2$ thin films were patterned into standard six-probe Hall bar geometry with the channel size of $1 \text{ mm} \times 0.5 \text{ mm}$, as displayed in figure 2(a). Figure 2(b) shows the temperature-dependent longitudinal magneto-resistance $R_{xx}$ of the Cd$_3$As$_2$ thin films (Σ1 ~ Σ5) with various thickness at zero magnetic field. It is noted that $R_{xx}$ monotonically increases with the thickness decreasing, and the $R_{xx}$-T slope at the low temperature region shows a change from positive to negative, indicating a semimetal-to-semiconductor transition. For the thickest Cd$_3$As$_2$ sample (Σ5, ~900 nm), $R_{xx}$ reveals a broad peak at $T \sim 225$ K below which the resistance exhibits a metallic behavior that is commonly found in semimetals or gapless semiconductors. When the thickness is reduced to ~400 nm (Σ3), the resistance increases slightly in the range of 10 to 2 K. One possible reason is that due to the zero energy gap and a low carrier density $n_{s0} \sim 0.12 \times 10^{18}$ cm$^{-3}$, holes are copiously excited even at low temperatures, and the increased hole population could lead to a decrease in $R_{xx}$, similar to that observed in Na$_3$Bi system \[36\]. When further reducing thickness (Σ1, 50 nm) (several thinner samples are available in supporting information figure S1), the semiconducting behavior becomes evident, consistent with the theoretical predictions, i.e., a bandgap opening when the dimensionality is reduced in Dirac semimetals \[3\].
Magneto-transport measurements were carried out at low temperatures for different thickness films using a physical properties measurement system (PPMS, up to 9 T). The transverse resistance ($R_{xy}$) versus magnetic field $B$ is measured at $T = 2$ K (See supporting information figure S2 for $R_{xy}$-B). The negative slope of $R_{xy}$ suggests that the dominant charge carriers in Cd$_3$As$_2$ thin films are electrons. Figure 2(c) depicts the typical temperature-dependence of Hall mobility ($\mu$) deriving from the $R_{xy}$-B linear parts. Most of our Cd$_3$As$_2$ thin films yields a high mobility in the range of $3.8 \sim 9.1 \times 10^3$ cm$^2$ V$^{-1}$ s$^{-1}$ at 2 K. And the mobility significantly increases as the temperature $T$ drops. This can be attributed to the alleviated electron-phonon scattering at low temperatures. The sheet carrier density ($n_s$) at 2 K varies between $3.4 \sim 24 \times 10^{12}$ cm$^{-2}$, and the corresponding carrier concentration ($n_{sdff}$) is determined to be a relative low value of $1 \sim 8 \times 10^{17}$ cm$^{-3}$. Figure 2(d) depicts a band schematic structure for a typical Cd$_3$As$_2$ thin film with an emerged gap opening when the dimensionality reduces. The Fermi levels labeled in the sketch are determined by the SdH oscillations as discussed below. The relative position between the Fermi level and the band edge is qualitatively consistent with the aforementioned $R_{xx}$-$T$ curves. Moreover, figure 2(e) shows a large positive magneto-resistance ($MR = [R_{xx}(B)-R_{xx}(0)]/R_{xx}(0) \times 100\%$) at 2 K for the Cd$_3$As$_2$ series films with a parabolic and a quasi-linear behavior at low- and large-fields ($B \geq 5$ T), respectively. It shows a linear behavior without saturation and the maximum MR ratio is closed to 343.5% at $B = 9$ T. Interestingly, we also find that the corresponding ratio at 9 T drops from 343.5% to 58.8% that is associated with the electron mobility. And there is a nearly linear relationship between MR and $\mu$ as shown in figure 2(f), consistent with the previous studies [12]. Owing to the Dirac band structure and the high mobility of the thin Cd$_3$As$_2$ films, we were able to resolve SdH oscillations in $R_{xx}$, for most of our samples at low temperature (2 K), as shown in figure 2(e).

To fundamentally study the thickness-induced transition, we determined the electron mean free path using $l = \hbar(\mu/e)(3\pi^2k_B^2T)^{1/3}$ and the 3D Fermi vector wave by $k_F = (3\pi^2n_{sdff})^{1/3}$, as summarized in table SII (See the supplementary material), where $n_{sdff}$ is the 3D carrier density, $d$ is the film thickness, $e$ is the electron charge, and $\hbar$ is the reduced Planck’s constant. Figures 3(a), (e) and (i) present the vertically shifted SdH oscillation amplitude $\Delta R_{xx}$ versus $1/B$ at different temperatures for samples $\Sigma 1$, $\Sigma 4$ and $\Sigma 5$, respectively, after subtracting a smooth background. A clear periodic fluctuation in resistance are observed when $T \leq 50$ K, and the oscillation amplitude decreases rapidly with increasing $T$. To understand the temperature-dependent SdH oscillations for the different thickness thin films, we obtain the oscillation frequency $F$ from Landau fan diagrams (figures 3(b), (f) and (j)), taking the periodic maxima and minima of $\Delta R_{xx}$ as the half integer and integer, i.e. peak and valley positions in $1/B$, respectively. The slope yields a distinct frequency of $F = 23.18$ T, which corresponds to a periodicity of $\Delta (1/B) = 0.0431$ T$^{-1}$ for the thinnest sample ($\Sigma 1$). The cross-sectional area of the Fermi surface $S_F$ can be determined from the Onsager equation $F = (\Phi_0/2\pi^2)S_F$, here $\Phi_0 = h/2e$. We note that, for 50 nm ($\Sigma 1$), 580 nm ($\Sigma 4$) and 900 nm ($\Sigma 5$), $F$ and $S_F$ varies in $23.18 \sim 14.39$ T and 2.21 to 1.37 $\times 10^{-3}$ $\text{Å}^{-2}$ as the thickness $d$ changes. The Fermi vector $k_F = 0.0266 \sim 0.0209$ Å can be extracted when assuming a circular cross-section ($S_F \approx 2\pi k_F^{-2}$). For a 3D Dirac materials, the carrier density estimated from the SdH oscillations obeys the following formula [37]:

$$\Delta \left( \frac{1}{B} \right) = \frac{2e}{\hbar} \left( \frac{g_v g_s}{6\pi n_{sdff}} \right)^{1/3}$$

where $g_v$ is the spin degeneracy and $g_s$ is the valley degeneracy. Provided $g_v g_s = 4$ [13, 38], i.e., a small carrier concentration $n_{sdff}$ is calculated to vary from 1.276 to 0.618 $\times 10^{18}$ cm$^{-2}$, which is reasonably consistent with the $n_{3D}$ value from the Hall effect measurements (See supporting information table SI for Hall parameters).

In order to further verify the temperature-dependence of SdH oscillations and extract key carrier transport parameters, we fit the oscillation amplitude $\Delta R_{xx}$ to the standard Lifshitz-Kosevich theory [37],

$$\Delta R_{xx} \propto \frac{\lambda(T)}{\sinh \lambda(T)} \cdot \exp [-\lambda(T)] \cdot \cos \left[ 2\pi \times \left( \frac{F}{B} + \frac{1}{2} + \beta \right) \right]$$

where $\lambda(T) = 2\pi^2 m^* k_B T/(\hbar e B)$ and $\lambda_0 = 2\pi^2 m^* k_B T_0/(\hbar e B)$ are the thermal factor and the Dingle factor, respectively. Here, $k_B$ is the Boltzmann’s constant, $m^*$ is the cyclotron effective mass, $T_0 = h/2\pi k_B$ is the Dingle temperature, $\tau$ is the quantum lifetime due to scattering, and $2\pi \beta$ is the Berry’s phase which will be discussed later.

Thus, the normalized oscillation amplitude can be described as $\Delta R_{xx}(T)/\Delta R_{xx}(0) = \lambda(T)/\sinh \lambda(T)$. We plot the $\Delta R_{xx}(T)/\Delta R_{xx}(0)$ for sample $\Sigma 1$, $\Sigma 4$ and $\Sigma 5$ in figures 3(c), (g) and (k), based on which the effective mass $m^*$ are extracted to be 0.0322 $\sim 0.0254$ $m_e$ ($m_e$ is the free electron mass). By employing $v_F = h k_F/m^*$ and $E_F = m^* v_F^2$, we can obtain the Fermi velocity $v_F = 11.0 \sim 9.53 \times 10^5$ m s$^{-1}$ and the Fermi energy $E_F = 193.2 \sim 131.2$ meV. The Dingle-plot for SdH oscillations $h/(\Delta R_{xx}/R_0) \sim B \cdot \sinh (2\pi^2 m^* k_B T/(\hbar e B))$ versus $1/B$ and the linear fitting curves at different temperatures are shown in figures 3(d), (h) and (l). All these estimated parameters derived from the SdH oscillations at $T = 2$ K are provided in table 1.
To further clarify thickness-dependent SdH oscillations, Berry’s phase has been obtained from the Landau fan diagram as shown in figures 3(b), (f) and (j). According to the Lifshitz-Onsager quantization rule \([39]\):
Table 1. Estimated parameters from the SdH oscillations at $T = 2$ K for $\Sigma_1$, $\Sigma_4$ and $\Sigma_5$ Cd$_3$As$_2$ samples.

| $d$ (nm) | $F/T$ | $\gamma + \delta$ | $S_F$ ($10^{-3}$ Å$^{-2}$) | $k_F$ (Å$^{-1}$) | $n_{\text{SdH}}$ ($10^{16}$ cm$^{-3}$) | $m^*$ ($m_e$) | $v_F$ ($10^3$ m s$^{-1}$) | $E_F$ (meV) |
|---------|-------|-------------------|-----------------------------|-----------------|----------------------------------------|--------------|------------------------|-------------|
| 50      | 23.18 ± 0.05 | 0.325 ± 0.010   | 2.21 ± 0.004                | 0.0265 ± 0.0012 | 1.262 ± 0.012                         | 0.322 ± 0.0016 | 9.54 ± 0.64            | 166.6 ± 17.7 |
| 580     | 23.35 ± 0.35 | 0.657 ± 0.055    | 2.23 ± 0.03                 | 0.0266 ± 0.0032 | 1.276 ± 0.024                         | 0.0280 ± 0.0009 | 11.0 ± 1.40            | 193.2 ± 35.3 |
| 900     | 14.39 ± 0.22 | 0.741 ± 0.043    | 1.37 ± 0.02                 | 0.0209 ± 0.0026 | 0.618 ± 0.012                         | 0.0254 ± 0.0012 | 9.53 ± 1.25            | 131.2 ± 23.6 |
The Berry’s phase $2\pi/\beta$ can be extracted from the intercept in Landau fan diagram by $\gamma + \delta, 2\pi\delta$ is an additional phase shift determined by dimensionality, which equal to 0 for a quasi-2D cylindrical Fermi surface and $\pm\pi/4$ (+ for holes and— for electrons) for a corrugated 3D Fermi surface. For nontrivial $\pi$ Berry’s phase observed in bulk Cd$_3$As$_2$ [14] as reported before, $\gamma$ should be 1. In our experiments for the different thickness Cd$_3$As$_2$ thin films, the intercepts $\gamma + \delta$ are about $0.325 \sim 0.741$ and the corresponding Berry phase $2\pi/\beta$ are about $-0.099\pi \sim 0.732\pi$. And it shows a tendency that the Berry phase changes from zero to $\pi$ Berry’s phase as the thickness increases, i.e. Cd$_3$As$_2$ exhibits a transition from trivial band insulator to nontrivial Dirac semimetal as the dimensionality increases from quasi-2D thin film to 3D bulk, which is consistent with theoretical predictions [3]. The presence of zero Berry’s phase for the thinnest Cd$_3$As$_2$ sample ($\Sigma 1$) infers that the SdH oscillations mainly drive from the high mobility bulk conduction band, and the Dirac point vanishes along with the band gap opening as the dimensionality reduces, which also agrees well with the thickness-dependent $R_{xx}$-$T$ results as discussed previously.

For a nontrivial $\pi$ Berry phase, the offset $\gamma + \delta$ in the LL fan diagram should be 1. Here, the Fermi surface’s curvature can generally lead to a 1/8 intercept shift [39]. But for our thickest film ($\Sigma 5$, figure 3(j)), the LL fan diagram intercept 0.741 is close to $\gamma - 1/8$ ($\gamma = 1$), approaching the non-trivial Berry’s Phase. It might be because the Fermi surface $S_{\Gamma}$ is still an anisotropic ellipsoid instead of a perfect sphere. On the other hand, in previous study [40], when the magnetic field $B$ is perpendicular to (112) plane, the large field can break the crystal symmetry. A gap may subsequently emerge as long as the orbit-coupled field strength is smaller than the mass term, which could further cause an extra phase shift. However, we speculate that this effect is fairly weak in the range of 0–9 T although it may still have some effect.

We further performed angular-dependent magneto-transport measurements for the Cd$_3$As$_2$ thin films and obtained the vertically shifted SdH oscillations amplitude $\Delta R_{xx}$ against angle $\theta$ after removing the background signals. Here, $\theta$ is the angle between magnetic field $B$ and (112) plane normal direction, varies from 0° to 90° correspond to the changing from out–plane to in–plane. And we note that the oscillations frequency $F$ at different angles $\theta$, which is related to the cross-sectional area of the Fermi surface $S_{\Gamma}$ in the momentum space, can be used to estimate the configuration of the Fermi ellipsoid via the Onsager equation. The variation of $F$ reduces and it remains almost unchanged (within 2%) for the thickest film ($\Sigma 5, 900$) as $\theta$ varies from 0° to 90°, as shown in figure 5. This small anisotropy points to a nearly uniform Fermi ellipsoid surface in $k_x$ and $k_y$ direction for Cd$_3$As$_2$ thick films, which is qualitatively consistent with the recent magneton-transport experiment results [14, 40, 41]. But for thinner films ($\Sigma 1$, as shown in figure 4(a)) $\Delta R_{xx}$ starts to decrease when $\theta$ goes beyond 45° and the peak nearly vanishes at $\theta = 90^\circ$. It may be explained as (due to the quantum confinement in the normal direction [112]) the ellipsoids Fermi surface were evaluated into anisotropic ellipsoids Fermi surface as the dimensional reduced.

In conclusion, we have produced single-crystalline Cd$_3$As$_2$ thin films with different thickness (50 ~ 900 nm) by MBE on the cleaved mica substrates. The corresponding crystal quality and thin film properties were...
characterized by XRD, XPS and Raman measurements. We found a dimensionality-induced semimetal-to-semiconductor transition and $R_{xx}$ shows semiconducting behavior owing to an emerged band gap opening for the thinnest film (50 nm). Furthermore, the temperature-dependent SdH oscillations are observed, suggesting non-trivial to trivial Berry’s phase transition.

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Competing financial interests

The authors declare no competing financial interests.

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