

Quantum oscillations from Fermi arc surface states in Cd_3As_2 submicron wiresYu Miyazaki^{1,*}, Tomoyuki Yokouchi,² Kiyoh Shibata³, Yao Chen,⁴ Hiroki Arisawa,⁴ Teruyasu Mizoguchi³, Eiji Saitoh,^{1,4} and Yuki Shiomi^{1,2}¹Department of Applied Physics, The University of Tokyo, Hongo, Bunkyo, Tokyo 113-8656, Japan²Department of Basic Science, The University of Tokyo, Komaba, Meguro, Tokyo 153-8902, Japan³Institute of Industrial Science, The University of Tokyo, Komaba, Meguro, Tokyo 153-8505, Japan⁴Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

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Topological materials such as topological insulators and Weyl/Dirac semimetals possess topologically protected surface states giving birth to various unique phenomena and functionalities. To investigate the surface transport phenomena toward possible application to electric devices, nano- and submicron-scale structures of topological Dirac semimetals are of particular interest since they can be grown by an economical chemical vapor deposition (CVD) method. However, quantum oscillations associated with the topological surface states have not been well explored in nano or submicron wires despite a most fundamental transport signature of the surface state. Here, we successfully observe quantum oscillations resulting from the surface states in magnetoresistance measurements for submicron wires of Dirac semimetal Cd_3As_2 grown by a CVD method. The oscillation frequencies and phases suggest that the surface quantum oscillations originate from closed orbits located on each surface constructed from the Fermi arcs. Our results will stimulate further research on quantum transport phenomena in topological wires.

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Electron states emerging on the surfaces of topologically nontrivial bulk phases give rise to various transport phenomena. For example, quantum oscillations and quantum Hall effects result from the quantized Dirac-cone-like electron states at the surfaces of three-dimensional topological insulators, which possess insulating bulk states characterized by the Z_2 topological index [1–3]. Weyl semimetals are another class of topological materials, which possess gapless bulk states consisting of pairs of band-crossing points (Weyl points) with opposite chirality near the Fermi energy [4]. On their surfaces, topologically protected surface states, namely Fermi arcs, connect the projection of the Weyl points with opposite chirality. At the edges of the Fermi arc, wave functions of the Fermi arc are hybridized with bulk states, which leads to the formation of an unconventional orbit, termed Weyl orbit under magnetic field; the Fermi arcs at the top and bottom surfaces are connected through the bulk chiral modes (i.e., $N = 0$ Landau levels), forming a closed path [5].

Dirac semimetals, symmetry-protected analogs of Weyl semimetals, possess a gapless topological phase consisting of overlapping two copies of Weyl semimetal with the opposite chirality, and thus a pair of Fermi arcs exist at surface [Fig. 1(a)] [4]. In particular, magnetic orbits associated with

the surface states in Dirac semimetals are of great recent interest and have been intensively studied [5–11]. So far, mainly two possible ways for forming the magnetic orbits have been discussed. One is the Weyl orbits which are composed of bulk and surface states, as in the case of Weyl semimetals [Fig. 1(b)] [5,7–9]. In the Weyl orbits in Dirac semimetals, the mixing between the two superposed copies is believed to be prevented due to crystal symmetry [5,12,13]. Another possibility is a closed path localized on each surface [Fig. 1(c)] [5,6,10,11]. In contrast to Weyl semimetals, since Dirac semimetals have a pair of Fermi arcs on one surface, closed orbits can be constructed only from the Fermi arcs localized on each surface. These closed orbits are sometimes called surface orbits.

A suitable experimental platform for investigating surface transport properties in Dirac semimetals is nano- and submicron-scale structures because these samples have advantages for reducing the bulk carrier density and obtaining a large surface-to-volume ratio, both of which are crucial for detecting the contribution from the surface transport. In the case of Cd_3As_2 , one of the most typical Dirac semimetals, Shubnikov–de Haas (SdH) oscillations and quantum Hall effect associated with the surface states have been observed in nano- and micro-scale plates, e.g. microstructures fabricated by using a focused ion-beam (FIB) method [7], nanoplates synthesized by a chemical vapor deposition (CVD) method [8,9], and epitaxial thin films grown by a pulsed laser deposition method [11].

Recently, another type of nano- and submicron-scale system of Cd_3As_2 has also attracted attention, that is, nano and submicron wires [14–20] which we focus on here. Nanowires

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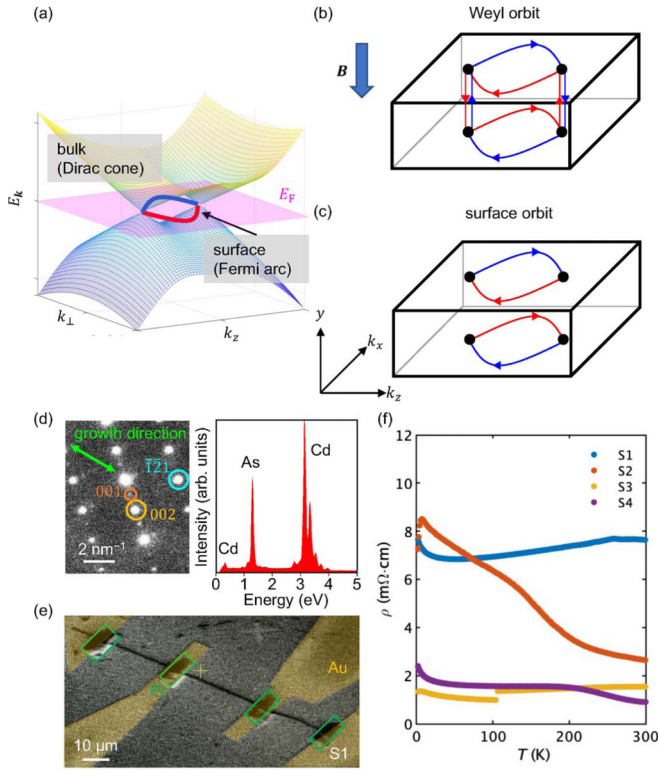


FIG. 1. (a) Bulk band dispersion and Fermi arc surface state of Dirac semimetals (DSMs) Cd_3As_2 . (b), (c) Schematic diagrams of (b) Weyl orbits and (c) surface orbits. (d) An electron diffraction pattern and an EDS spectrum of typical Cd_3As_2 wire. (e) A SEM image of device S1. (f) Temperature dependence of resistivity of devices (S1, S2, S3, and S4). We note the discrete jump observed at 100 K in S3 probably results from extrinsic origins such as a slight change in electric contact at electrodes.

of Dirac semimetals are one of the promising candidates for realizing the Majorana zero modes [21], which are considered to be important for building a topological quantum computer. In the case of nanowires grown by a CVD method, the bulk carrier density is sufficiently reduced similar to the aforementioned nano- and micro-scale systems [14,15]. Hence, the transport phenomena associated with the surface states can show up. In fact, spin-momentum locking of the Fermi arc surface states was demonstrated in nanowire-based nonlocal devices [20]. In addition, since the topological nanowires are analogous to hollow cylinders, quantum interference on the sample surface leads to quantum oscillations, termed Aharonov-Bohm oscillations, in which the resistivity exhibits oscillations periodic in B [19]. However, in nano or submicron wires of Cd_3As_2 , to the best of our knowledge, SdH oscillations arising from the surface states have not been reported so far. The surface SdH oscillations provide important insight into the surface state, which is helpful for fully understanding the physical properties of Dirac wires.

In this letter, we report the observation of quantum oscillations arising from the surface states in submicron wires of Cd_3As_2 . The SdH oscillations originating from the bulk Fermi surface are observed when the magnetic field is applied par-

allel to the wire. Besides, when the magnetic field is applied perpendicular to the wire, another oscillation component is superimposed on the bulk oscillations, which is assigned to the contribution from the surface states. The frequency and phase of the surface oscillations suggest that the observed oscillations originate from the surface orbits constructed from the Fermi arc surface states, instead of Weyl orbits as previously reported in thin films and nanoplates [7–9,11]. Relatively large diameters of the present samples might be responsible for the formation of surface orbits.

Cd_3As_2 wires were grown by a CVD method using a two-zone horizontal tube furnace. We placed high-purity Cd_3As_2 powder (6N) on the upstream side and SiO_2/Si substrates on the downstream side. Prior to the CVD growth, the substrates were coated with 1 mmol/L BiI_3 /ethanol solution as a catalyst [22,23]. The temperatures at the upstream and downstream sides were set at 650 °C and 350 °C, respectively, and the system was maintained at ambient pressure with 100 sccm Ar gas flow during the growth process. We confirmed that the shape of a fabricated sample was a cylinder with the use of a scanning electron microscope (SEM) (See the Supplemental Material [23]) Figure 1(d) shows results of electron diffraction and energy-dispersive x-ray spectroscopy (EDS) of a typical wire with a transmission electron microscope (See also the Supplemental Material [23]). As a result of the diffraction pattern analysis, crystal structure is assigned to the $P4_2/nmc$ space group and the growth direction is almost perpendicular to $\{289\}$. Submicron wires belonging to the $P4_2/nmc$ space group were previously reported in Ref. [16] and Cd_3As_2 with $P4_2/nmc$ space group is a topological Dirac semimetal [13]. The atomic ratio evaluated by EDS spectra is $\text{Cd} : \text{As} = 60.6 : 39.4$, being quite close to the right stoichiometry of the Cd_3As_2 compound.

The Cd_3As_2 submicron wires were transferred from the as-grown substrate to a different SiO_2/Si substrate. Then, Au (200 nm)/Ti (5 nm) electrodes were fabricated by using electron beam lithography and electron-beam physical vapor deposition. In order to improve electric contacts to submicron wires, the electrodes were then covered with Pt by a focused ion-beam assisted chemical vapor deposition (FIB-CVD) method. For transport measurements, we fabricated one microwire and three submicron wire devices: S1, S2, S3, and S4. Figure 1(e) shows a SEM image of S1. The diameters of S1, S2, S3, and S4 are estimated to be 1.5 μm , 0.56 μm , 0.60 μm , and 0.56 μm , respectively, from SEM images. The temperature dependence of the resistivity for S1, S2, S3, and S4 is shown in Fig. 1(f). All the samples are conductive, but the temperature profiles are different. A similar tendency was observed in previous reports [15,22,24,25] and probably reflects slight differences in sample qualities such as defect densities and the position of the Fermi level. Especially, ρ for S2 and S3 drops at $T < 10$ K. When the Fermi level is located just above the Dirac point, the reduction of the thermal activation at low temperatures changes the dominant carriers from thermally activated holes in the valence band showing a semiconductorlike $\rho - -T$ curve to electrons around the Fermi level in the conduction band showing a metallike $\rho - -T$ curve [18].

Magnetoresistance (MR) for $I \parallel B$ (S1) and $I \perp B$ (S3) is presented in Fig. 2, where I and B are electric current and

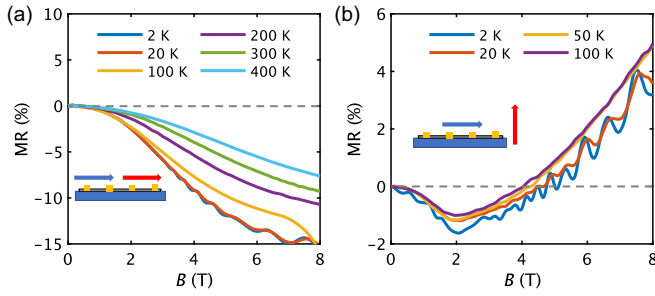


FIG. 2. (a) Magnetic-field dependence of magnetoresistance (MR) at various temperatures under $I \parallel B$ in device S1. (b) Magnetic-field dependence of MR at various temperatures under $I \perp B$ in device S3.

a magnetic field, respectively. For both cases, clear quantum oscillations are observed at low temperatures. In Fig. 2(a), negative MR is observed as nonoscillatory backgrounds even above room temperature for $I \parallel B$. In addition, negative MR is also observed in the case of $I \perp B$ for S3 [Fig. 2(b)] and S2 (Fig. S5 in the Supplemental Material [23]). Negative MR under $I \parallel B$ has been reported in some previous reports [17,18,26] due to the chiral anomaly [27]. In contrast, the chiral anomaly cannot explain the negative MR under $I \perp B$. Although similar negative MR was observed in the perpendicular B condition for Cd_3As_2 nanowires [28], its origin has not been elucidated. One possible mechanism is electron-hole puddles reported in compensated topological insulators [29,30].

To analyze the observed quantum oscillations for $I \parallel B$, we subtract the smooth MR background, as shown in Fig. 3(a).

The oscillation components $\delta\rho(B)$ are periodic with respect to $1/B$, which indicates that the observed oscillations are SdH oscillations. Since the magnetic field is applied along the wire in the present case, the SdH oscillations can be attributed to the bulk Fermi surface. In Fig. 3(b), we show the Landau level (LL) fan diagram to extract the oscillation frequency (F) with the use of the Lifshitz-Onsager quantization rule given by $F/B_n = n - \phi$, where n , ϕ , and B_n are the Landau index, the phase shift, and the peak position of $\delta\rho(B)$ corresponding to n th Landau index, respectively. Here, the peak and valley positions of $\delta\rho(B)$ are assigned to integer and half-integer, respectively. From the slope of the LL fan diagram, we obtain $F = 37.0 \pm 0.3\text{ T}$. Here, the uncertainty of the value denotes the standard error of the fitting parameters. It is in good agreement with reported values for the bulk Fermi surface in Cd_3As_2 [7,31,32]. To cross-check this result, we also performed fast Fourier transformation (FFT) analysis and confirmed a peak structure near 37 T [23]. The oscillation amplitude gradually decreases with increasing temperature (Fig. S5 in the Supplemental Material [23]). The temperature dependence can be well fitted by the standard Lifshitz-Kosevich formula [28], yielding the effective cyclotron mass of $m_c = 0.0393m_0$ and the quantum scattering time $\tau_Q = 40.45\text{ fs}$. These values are also in good agreement with reported values for the bulk Fermi surface in Cd_3As_2 [7,31,32].

Next, we focus on quantum transport properties when the magnetic field is applied perpendicular to the current direction. Figures 3(c) and 3(e) show $\delta\rho(B)$ with $I \perp B$ for S2 and S3. In both samples, $\delta\rho(B)$ exhibits two oscillation components. The existence of two oscillatory components is also confirmed from the Landau level fan diagrams shown in

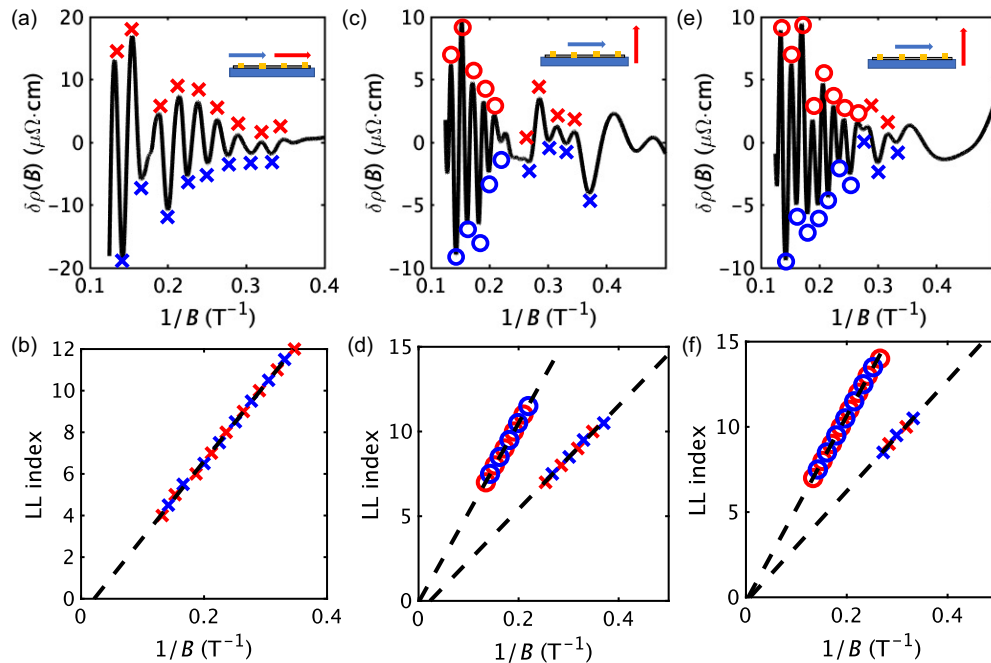


FIG. 3. (a), (c), (e) The oscillation components in magnetoresistance for $I \parallel B$ in (a) S1, for $I \perp B$ in (c) S2, and (e) S3. The crosses and circles denote bulk oscillations and surface oscillations, respectively. The red and blue circles denote the peaks and valleys of the surface oscillations, respectively, while the red and blue crosses denote the bulk oscillations. (b), (d), (f) Landau level (LL) fan diagrams for $I \parallel B$ in (b) S1, for $I \perp B$ in (d) S2, and (f) S3.

Figs. 3(d) and 3(f). From the slopes of the Landau level fan diagrams, the frequencies of these oscillations are estimated to be $F = 30.6 \pm 0.7$ T and 52.9 ± 0.4 T for S2 and $F = 32.4 \pm 1.5$ T and 54.5 ± 0.6 T for S3; similar frequencies are obtained for S2 and S3. Since the lower oscillation frequencies ($F = 30.6 \pm 0.7$ T for S2 and $F = 32.4 \pm 1.5$ T for S3) are quite close to the oscillation frequency for the bulk Fermi surface observed with $I \parallel B$ ($F = 37.0 \pm 0.3$ T), we attribute the lower-frequency oscillations to the bulk SdH oscillations. In contrast, the higher-frequency oscillations (52.9 ± 0.4 T for S2 and 54.5 ± 0.6 T for S3) are observed only for $I \perp B$, and they are thereby assigned to the quantum oscillations arising from the surface states. We show the effective cyclotron mass m_c and the quantum relaxation time τ_Q for the surface oscillations for S2 and S3 estimated from the Lifshitz-Kosevich analysis (Table S2 and Fig. S5 in the Supplemental Material [23]).

The values of phase shifts for surface oscillations are $\phi = 0.076 \pm 0.06$ in S2, 0.243 ± 0.11 in S3, and 0.741 ± 0.073 in S4. Here, the errors correspond to the standard errors in the fitting. These values are significantly different from a two-dimensional trivial orbit case ($\phi = 0.5$), indicating the existence of the phase shift due to the Berry phase and being consistent with an expectation in surface magnetic orbits constructed from the Fermi arcs [33,34]. The values of phase shifts in S2, S3, and S4 are notably different. This might be due to the sample dependence of the Fermi level position because the phase shift due to the Berry phase term depends on the Fermi level position if Dirac dispersion has nonlinear terms [35,36].

Figures 4(a) and 4(b) show the B -angle dependence of SdH oscillations in S4. Here B was rotated within the plane including the wire direction, and the angle between B and I directions is defined as θ . At $\theta = 90^\circ$, a surface oscillation is observed, and the estimated F_S value (53.1 ± 0.3 T) is consistent with S2 and S3 (Fig. 3). As shown in Fig. 4(b), F_S increases with decreasing θ almost in proportion to $1/\sin \theta$, as expected in the SdH oscillation in two-dimensional surface states [7,8,11]. We note that bulk SdH oscillations ($F_B = 33.0 \pm 0.6$ T) are observed at $\theta = 0^\circ$, but not around 90° . The amplitude of bulk oscillations is much smaller than that of surface ones and thus bulk oscillations should be obscured for S4.

As mentioned in the introduction, in the case of Cd_3As_2 , either the Weyl orbits or the surface orbits can form under the magnetic field, resulting in the surface SdH oscillations. A clue for distinguishing them is obtained from an additional phase term in the oscillation that arises from the bulk tunneling processes in the Weyl orbits; in the case of Weyl orbits, the additional phase term makes the position of the n th Landau level deviate from the ideal periodicity of the SdH oscillations. Moreover, this deviation becomes more prominent in lower Landau level indices, and thus the position of the Landau levels constitutes a curved line as a function of $1/B$ [7]. In contrast, in the case of the surface orbits localized on each surface, the position of the Landau levels does not deviate because of the absence of the additional phase terms, and hence the position of the Landau levels shows a linear dependence on $1/B$. In the present case, the deviation from the ideal periodicity was not observed as shown in Figs. 3(d)

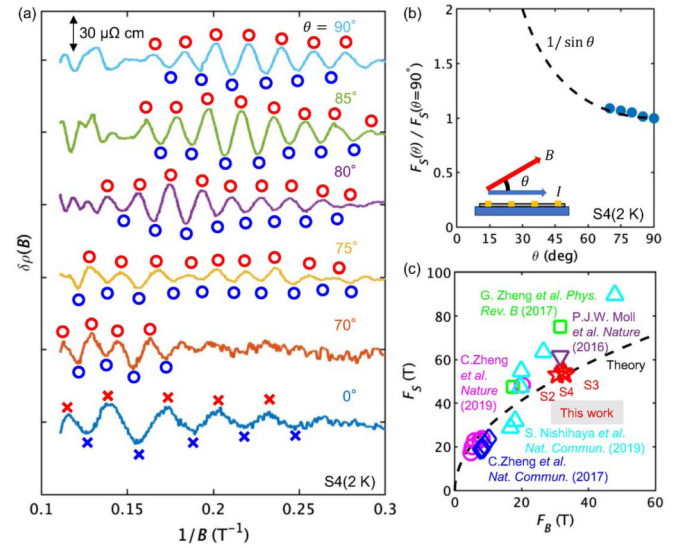


FIG. 4. (a) SdH oscillations for various B angles in S4. For the definition of θ , see the inset of (b). (b) The B angle (θ) dependence of the surface oscillation frequency (F_S). Error bars stand for the standard errors of $F_S(\theta)/F_S(\theta = 90^\circ)$; they are small (less than 1.5%) and invisible in the figure. (c) The surface oscillation frequency (F_S) as a function of the bulk oscillation frequencies (F_B) (red stars). Data in previous reports for plates and films of Cd_3As_2 [7–9,37] and $(\text{Cd}_{1-x}\text{Zn}_x)_3\text{As}_2$ [11] and a theoretical curve (dashed line) are also plotted. For the theoretical curve, we use $k_0 \simeq 0.8 \text{ nm}^{-1}$ following a previous report [31].

and 3(f); the positions for the n th Landau levels almost lie in a straight line, indicating no deviation and thus no additional phase term. Therefore, the observed surface oscillations are likely to originate from the surface orbits constructed from the Fermi arcs. In fact, it is reported for microstructure samples that oscillations due to Weyl orbits disappear when the thickness is thicker than the mean free path of electrons [7]. Hence, the formation of surface orbits, instead of Weyl orbits, in the present case might be attributed to a thick diameter of our wires.

The magnitudes of oscillation frequencies are consistent with a theoretical prediction for SdH oscillations due to the surface orbits. Quantum energy levels can be provided by $\varepsilon_n = \frac{2\pi\hbar}{t}n$, where t is the semiclassical time for the orbits [5]. In the case of a closed path constructed by two Fermi arcs on one surface, the semiclassical time can be given by $t \simeq \frac{2\hbar k_0}{e v_F B}$, where k_0 and v_F are the arc length and the Fermi velocity, respectively. From these equations, the magnetic field when the n th energy level crosses the Fermi energy (E_F) can be obtained by $\frac{1}{B_n} = \frac{e\pi v_F}{k_0 E_F}n$. Hence, the frequency of the oscillations for the surface orbit is given by $F_S = \frac{k_0 E_F}{e\pi v_F}$. Here, by using the oscillation frequency for the bulk SdH oscillations (F_B), the Fermi energy can be described by $E_F = \hbar k_F v_F = v_F \sqrt{2e\hbar F_B}$. Thus, when the surface orbit brings about the SdH oscillations, the oscillation frequencies of the surface SdH (F_S) and the bulk SdH (F_B) should satisfy the following relation:

$$F_S = \frac{k_0}{\pi} \sqrt{\frac{2\hbar}{e}} F_B. \quad (1)$$

We note this relationship between F_S and F_B is the same as that for the Weyl orbits [5,7] because the bulk tunneling processes in the Weyl orbits only influence the phase of the oscillations, and the oscillation frequency for the Weyl orbits is determined from the semiclassical time taken in two Fermi arcs (the top and bottom surface), i.e., $t \simeq \frac{2\hbar k_0}{ev_F B}$. As marked by red stars in Fig. 4(c), the surface and bulk oscillation frequencies observed in our study satisfy the expected relationship [Eq. (1)], consistent with the previously reported oscillation frequencies originating from the Weyl orbits in nanoplates, microstructures, and thin films [7–9,11,37].

In conclusion, we have synthesized Cd_3As_2 submicron wires using a CVD method to investigate the surface quantum transport properties. In addition to the bulk SdH oscillations, quantum oscillations resulting from the surface states are observed for three devices. On the basis of the frequencies and phases of the observed oscillations, we attribute the observed

surface oscillations to the surface orbits constructed from the Fermi arcs. Our finding demonstrates that topological wires are a promising platform for investigating the surface quantum transport phenomena and offer an insight into the surface magnetic orbits in topological Dirac semimetals.

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