The bulk electrons in topological semimetals are described by an ultra-relativistic dispersion relation, $E(k) = \pm \hbar v_F \sigma \cdot k$, that resembles the Weyl equation for massless spin-1/2 particles. Here $E$ is electron energy, $v_F$ the Fermi velocity, and $\sigma$ is a pseudo-spin-1/2 degree of freedom that is energetically locked parallel or anti-parallel to the momentum, $k$, of the electron, giving electrons definite chirality $k \sim \pm \sigma$. Applying electromagnetic fields to Weyl or Dirac semimetals induces a pumping of electric charge between Weyl nodes with opposite chirality, a phenomena known in high-energy physics as the chiral anomaly. At the surface of these materials, this anomalous chirality transfer is facilitated by topologically protected surface arcs, the so-called Fermi–arc surface states, which act as a pipeline connecting opposite chirality Weyl points. Recently, Na3Bi (ref. 17) and Cd3As2 (ref. 18) have been predicted to be three-dimensional bulk Dirac semimetals. This has sparked substantial research interest and the linear dispersion in these materials has been confirmed by ARPES (angle-resolved photoemission spectroscopy) and STM (scanning tunnelling microscopy) experiments. A number of unusual magnetic properties such as strong linear magnetoresistance and high mobilities were identified that are potentially linked to the relativistic nature of the Dirac quasiparticles. Yet the prospect of studying ultra-relativistic particles and their accompanying topological surface states, as well as potential applications exploiting their unusual behaviour, naturally requires a more direct measurement capable of revealing both the relativistic dynamics and topological surface states. The main aim of this study is to present such evidence in four-terminal transport measurements, showing the possibility of detecting and manipulating chiral states in Dirac semimetals.

One intriguing fingerprint of Weyl quasiparticles in the electronic transport properties in strong magnetic fields has been recently predicted. The ‘Weyl orbit’ weaves together the chiral states in the bulk with the topological Fermi–arc states on opposite surfaces into a closed orbit. Its quantization produces a distinctive contribution to the quantum oscillation spectrum that provides an observable signature of the chiral and topological character of these materials. This closed orbit is strikingly different from typical electrons orbiting around a Fermi surface in a metal, as the quasiparticle experiences zero Lorentz force on the chiral path segments traversing the bulk.

The main result of this study is an additional quantum oscillation frequency observed in microstructures smaller than the mean free path that exhibits characteristics of both surface-like and bulk-like states, as naturally expected for Weyl orbits. The microstructures were prepared from Cd3As2 single crystals by focussed ion beam (FIB) etching (Fig. 1, Methods). Down to the smallest thickness of $L = 150 \text{ nm}$, the magnetoresistance at temperatures below 100 K shows pronounced Shubnikov–de Haas oscillations signalling the low effective mass of the charge carriers and the high crystal quality of the devices (Fig. 1b).

Studies of quantum oscillations in bulk crystals have reported one single frequency, arising from an essentially spherical three-dimensional (3D) Fermi surface in agreement with ARPES and STM experiments. This single bulk frequency ($F_B$) is also consistently observed in all of the studied parent bulk crystals as well as all microstructures. However, when the field is applied perpendicular to the [010] surface (denoted 0°, see below), a second frequency $F_S = 61.5 \text{ T}$ appears which is distinct from the higher harmonics of the bulk. We find an effective mass of 0.044me (where me is the electron’s mass) for the bulk, similar to previously reported measurements on bulk crystals, and a similar mass of 0.050me in the additional orbit $F_S$ (see Methods).

The value of the observed surface frequency $F_S$ is compatible with the prediction for Weyl orbits. Their trajectory combines segments of both chiral bulk and Fermi–arc surface states, and $F_S$ can be estimated from the time spent in each of them:

$$F_S = E_F k_0 / (e \pi v_F) \approx 56 \text{ T}$$

The length of the surface Fermi arc in reciprocal space may be approximated as $k_0 \approx 0.8 \text{ nm}^{-1}$ using the $k$-space separation of the Dirac points determined in ref. 19. The bulk quantum oscillation frequency and effective mass provide direct access to the Fermi energy $E_F = 192 \text{ meV}$ and the Fermi velocity $v_F = 8.8 \times 10^5 \text{ m s}^{-1}$. The resulting estimate of $F_S \approx 56 \text{ T}$ is in good quantitative agreement with the measured $F_S = 61.5 \text{ T}$.

The dependence of the quantum oscillation frequencies on the angle between the magnetic field and the surface normal shows a
Figure 1 | Surface oscillations in microstructures. a. SEM micrograph of a typical sample prepared by FIB cutting. More than 10 similar devices have been fabricated and the results were highly reproducible among all of them. The active devices are 4 μm × 10 μm free-standing sheets (purple) of varying thickness connected to contact pads (yellow). The crystallographic direction perpendicular to the polished surface is [010] and the direction parallel to it is [100], which define the plane of rotation. This entire sample consists of one contiguous slice of a Cd₃As₂ crystal, ensuring that all devices are made from the exact same starting material and have the same orientation during the experiments.

b. Sketch of the Weyl orbit in a thin slab of thickness L in a magnetic field B. The orbit involves both the Fermi–arc surface states connecting the Weyl nodes of opposite chirality, and the bulk states of fixed chirality (blue and red). In strong magnetic fields, quasiparticles may tunnel through the energy barrier separating the bulk states from the surface over a distance associated with the magnetic length l_B (orange arrows). Note that as the Weyl orbit mixes processes in real and reciprocal space, the (x, y) coordinates of this sketch are in reciprocal space and the z coordinate is in real space. c. Magnetoresistance and d, its Fourier transform measured on the thinnest device (150 nm) at 2 K for fields parallel (90°) and perpendicular (0°) to the surface. The main finding of this study is directly evident in the raw data: while parallel fields lead to a single frequency (including spin-splitting at higher fields), an additional higher frequency component F_S associated with the surface oscillations appears for perpendicular fields. The frequency difference between the two directions becomes evident in the raw data when comparing the resistivity peaks directly, as indicated by the line markers.

clear signature of a surface state, as shown in Fig. 2, where 0° denotes fields perpendicular to the surface and 90° parallel to it. The angular dependencies of both frequencies are strikingly different: while the low frequency F_B (blue) remains essentially constant and is observed at all angles, the frequency F_S (red) strongly increases as the field is tilted away from the surface. F_S(θ) is well described by a cos(θ)^-1 dependence, indicating that the field component perpendicular to the surface is relevant for the orbit. This angle dependence is a hallmark of two-dimensional (2D) Fermi surfaces. The bulk band structure however does not support a 2D Fermi surface and it is not observed in our bulk crystals.

While the angle dependence clearly suggests a surface character of the quantum path associated with the frequency F_S, the quantum oscillations also show pronounced bulk-like characteristics which are unexpected for orbits simply consisting of surface states. First of all, the additional quantum oscillation is only observed in samples where the bulk mean free path is longer than or comparable to the sample thickness. The low-field transverse magnetoresistance for in-plane fields quantitatively confirms our devices to be in this limit. The resistance maximum at small fields is a hallmark signature of quasi-ballistic transport in clean metals known as the Knudsen effect (Fig. 3a, Methods).

The amplitudes of the quantum oscillations are found to be strongly thickness dependent (Fig. 3b). No trace of the surface frequency F_B has been observed in devices thicker than 3 μm, in agreement with its absence in our bulk quantum oscillation measurements. As the sample thickness is reduced, its relative weight compared to the bulk frequency strongly increases, and devices thinner than 500 nm are dominated by the surface oscillation. The increase in the various studied samples follows an exponential behaviour, with an exponent of d = 0.675 nm. This value should be compared to the bulk mean free path estimated from transport as l = v_Fτ = v_Fm*/(ne^2ρ_0) = 1.0 μm, where ρ_0 = 55 μΩ cm is the zero-field resistivity in our crystals at 2 K, n = 2k_F^2/(3π^2) = 2.5 × 10¹⁸ cm⁻³ the bulk carrier density estimated for the twofold degenerate spherical Fermi surface, τ the scattering time, m* the effective mass of the charge carriers, and k_F = m*/e^2h = 3.3 × 10⁸ m⁻¹ the Fermi momentum extracted from the Shubnikov–de Haas oscillations. This estimate of the mean free path is in good quantitative agreement with the observation of the Knudsen flow maximum. Ordinarily, any scattering process leads to quantum decoherence, in which case the quantum oscillations would be expected to decay exponentially in the device thickness L over the quantum mean free path l_Q which is generally much shorter than the mean free path measured in transport. However, for large fields the bulk chiral Landau level is expected to exhibit extra resilience to such dephasing and allow for quantum oscillations for thicker samples. As the closed Weyl orbit contains two path segments traversing the bulk, each at opposite chirality, the effective bulk path length is twice the device thickness L. The agreement between 2d = 1.35 μm (as d = 0.675 μm) and l = 1.0 μm suggests that the relevant thickness scale for the appearance of the additional quantum oscillations is comparable to the bulk transport mean free path.

An additional clue to the origin of the Shubnikov–de Haas oscillations comes from their phase as a function of field. We observe
deviations from ideal periodicity of the oscillations in inverse magnetic field, appearing as a continuously drifting phase as shown in Fig. 4. While similarly subtle deviations from periodicity can arise from $g$-factor band splitting in strongly spin–orbit coupled materials, the direction of the shift is opposite to those previously observed in experiments: the peak positions $B_n$ associated with the $n$th Landau level are shifted towards higher fields compared to the purely periodic case, while spin-splitting should shift them towards lower fields. On the other hand, this direction and magnitude of shift of the Landau levels is expected for Weyl orbits. Non-adiabatic corrections are expected to appear because of field-induced tunnelling between Fermi–arc states and bulk states, occurring as the orbiting quasiparticles approach the Weyl node. A single Weyl orbit will encounter four such tunnelling processes, leading to a phase deviation that is in quantitative agreement with what we observe (Fig. 4, Methods).

The discussion so far has considered the Dirac material Cd$_3$As$_2$ as two independent Weyl subsystems overlapping in $k$-space. However, magnetic fields can break the crystal symmetry protecting the superimposed Weyl nodes of opposite chirality and thus introduce a gap. In sufficiently strong fields, the particle approaching the node may thus tunnel into the oppositely dispersing Fermi arc on the same surface instead of into the bulk, thus forming closed orbits purely from Fermi–arc states, similar to the surface states of topological insulators. While our experiments cannot rule out localized topological surface states as the origin of the oscillations, the thickness dependence of the surface state amplitudes and the non-adiabatic corrections are more suggestive of the Weyl orbit in the present field range. In both cases, however, the Fermi arcs take part in the quantum orbit and our results thus confirm the detection of topological surface currents in the microstructures.

However, quantum oscillations may also arise from trivial surface states or from the defect layer introduced by the fabrication technique, without the involvement of topological states. Thus it is essential to find ways to determine if topological states participate in the observed orbit, setting the involved orbit apart from topologically trivial states. The existence of a ‘saturation field’ above which oscillations were expected to cease was predicted to provide such a test. However, a re-examination of the equation for quantum oscillations, reproduced in equation (1) below, reveals that the oscillations associated with the Weyl orbit actually persist up a much larger field of order $B_5 \approx 56$ T in the present instance (Methods), and the thickness dependence of the saturation field is highly complex. Consequently, the Weyl surface oscillations are expected to persist over the full range of fields explored in this experiment, in agreement with our observations. Additional evidence for the topological nature of the surface oscillations comes from their remarkable resilience against surface disorder. We have purposely introduced strong surface damage by almost normal incidence.

Figure 2 | Angle-dependent oscillations. a, Angle dependence of the quantum oscillation spectrum measured in the 150 nm device. As the field is rotated away from the perpendicular configuration (0°), the surface frequency $F_S$ increases and shrinks in amplitude, while the bulk frequency is unaffected. b, Polar plot of the angle dependence for the bulk $F_B$ and the surface frequency $F_S$, $F_S$ is almost isotropic while $F_S$ increases as the field is rotated towards a parallel configuration. It follows a surface-like $\cos(\theta)^{-1}$ dependence represented by a straight, off-centre line in a polar plot.

Figure 3 | Thickness dependence. a, A magnetoresistance maximum was observed in all studied samples for fields applied parallel to the surface (90°). This peak arises from the semi-classical Knudsen effect observed in quasi-ballistic transport when the cyclotron radius becomes comparable to the sample dimensions. b, Relative amplitude of the surface oscillations compared to the bulk oscillations, for fields perpendicular to the surface at 2 K. The surface oscillations are unobservable for devices thicker than 3 μm, and their relative weight grows with thinning of the samples. Inset, raw frequency spectra for devices of different thickness.
The absence of non-adiabatic corrections would fall onto the dashed line. Without non-adiabatic corrections (black trace), the expected spectra are well-defined peaks of conventional surface oscillations. The counter-intuitive increase of amplitude with increasing surface damage, however, would be a natural consequence of the protection of topological surface states (Methods).

Another striking piece of evidence for the non-trivial nature of the quantum orbit comes from a distinguishing feature of the Fermi-arc orbit: The position of the nth Landau level, \( B_n \), depends on the thickness of the sample, \( L \), given by:

\[
B_n^{-1} = c_k^{-1}(\pi n \nu_F/E_B - L)
\]

To test this concept, we fabricate a sample geometry that is by design sensitive to such a thickness dependence of the phase. In Fig. 5 we show a sample structured with a triangular cross-section as well as a rectangular one as a reference. The quantum oscillations for fields perpendicular to the surface for each device (0° for the rectangle, 60° for the equilateral triangle) are strikingly different. While the rectangular device clearly shows the presence of a surface state, the triangular one shows only the bulk frequency without any sign of the surface frequency. Crucially, all surfaces were fabricated under exactly the same conditions from the same piece of crystal. The sample was tilted with respect to the ion beam to ensure a grazing incidence condition for every surface, both on the rectangular and the triangular device. Both devices have comparable cross-section and surface area by design, so that a trivial surface state acting as a parallel conductance channel should lead to observable surface quantum oscillations in both of them. The absence of the surface state in the triangle is unexpected for oscillations arising from a trivial surface state, yet a natural consequence of Weyl orbits. Unlike conventional surface states that are confined to a single surface, the Weyl orbit cannot be observed in triangular geometries: all paths are of different length, each contributing to the field induced density-of-state modulation at a different magnetic field \( 1/B_n \).
(see equation (1), Fig. 5). This results in destructive interference due to a sum of oscillations with random phases, rendering the quantum oscillations unobservable in an experiment. In contrast, all quantum paths involve bulk path segments of the same length $L$ in a rectangular geometry, and thus all contribute to a density-of-state modulation at the same field. This is direct evidence that the orbit associated with the frequency $F_3$ is sensitive to the shape and size of the bulk underneath the surface, in contrast to those arising from trivial surface states.

The ensemble of presented results highlights an essential aspect of the additional quantum oscillations that appear when Dirac fermions are confined within microstructures smaller than the mean free path: they share characteristic features of both surface-like and bulk-like oscillations. Such hybrid characteristics arise naturally from the idea of the mixing of chiral bulk-states and Fermi-arc surface-states into a coherent orbit. Nevertheless, there remain questions that challenge our current understanding of topological matter on microscopic length scales and in strong magnetic fields. For example, understanding the exact amplitude of the Weyl oscillations may require the extension of the present theory into the quantum limit, where the number $n$ of occupied Landau levels cannot be treated as being large. Furthermore, our understanding of the mixing of chiral states in strong magnetic fields and their influence on the Weyl orbit needs to be improved and experimentally investigated in the future. By using FIB structuring, we have demonstrated an experimentally simple route towards studying strongly confined topological matter, which will both increase our understanding of the transport characteristics of Fermi-arc states and provide a path to investigate the potential of these materials in future electronic applications.

Online Content

Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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Author Contributions

P.J.W.M. microstructured the crystals, and performed the measurements and data analysis. N.L.N. synthesized and characterized the single crystals. T.H. analysed the crystal structure. I.K., A.C.P. and A.V. contributed the theoretical treatment. P.J.W.M. and J.G.A. designed the experiment. All authors were involved in writing the manuscript.

Author Information

Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to P.J.W.M. (philip.moll@cpfs.mpg.de) or J.G.A. (analytis@berkeley.edu).
The focused ion beam (FIB) has proven to be a powerful tool to fabricate crystalline microstructures of high quality (see, for example, refs 29–33) to study meso- and microscopic transport phenomena such as the Weyl-orbit quantum oscillations presented in this study. In this process, the microstructures are carved out of macroscopic crystals using the FIB. Starting from millimetre-sized flux-grown single crystals presented in Extended Data Fig. 1, we use a 30-kV Ga\(^+\) ion beam to cut the crystal. Depending on the ion flux rate and the angle of incidence, the irradiation spot can be as small as a few nm, and thus structures can be fabricated with high precision. The beam is guided across a negative image of the desired structure, which defines the parts of the crystal to be removed. Electrical contacts to the device can also be made in situ in the FIB. A platinum precursor gas can be introduced into the chamber, and upon interaction of the ion beam with the gas adsorbed to the surface a conductive, Pt-rich film can be growing connecting the microstructure electrically to external leads.

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fields (Fig. 3a). The quantum oscillations emerge on top of this background. We note that this field configuration is transverse and thus no negative magnetoresistance associated with the chiral anomaly is expected. Instead this enhancement of scattering in ultra-pure systems is a semi-classical effect arising from the diffuse scattering of otherwise ballistic electrons at the boundaries of strongly confined microstructures. It is well-studied in ultra-pure hydrodynamic systems, such as the viscous flow of He through capillaries (the Knudsen effect) or in high quality, geometrically confined conductors (Gurzhi flow) such as semiconductor heterostructures and clean metal whiskers. The resistance maximum at the Knudsen peak occurs at maximal boundary scattering of the bent electron trajectories, that is, at $2\pi n = L$ up to a small numerical factor, where $r_k = h\kappa/(eB)$ is the cyclotron radius. As a result, the position of the maximum is expected to shift to higher fields as the sample thickness is decreased, as is observed in the Cd$_3$As$_2$ microstructures (Fig. 3a). The Knudsen effect is a direct consequence of ballistic electron motion between the opposite surfaces and thus is direct evidence that the thickness of the studied microstructures is indeed comparable to the bulk mean free path.

**Non-adiabatic corrections.** The positions of the Landau levels associated with $F_s$ systematically deviate from the usual 1/$B$ periodicity. Figure 4 shows the deviation of each maximum of the resistance oscillation, $1/B_n$, from its expected position $1/B_0$ for a usual 1/$B$ periodicity (blue dashed line). At low fields, the oscillations indeed are found to be periodic in 1/$B$, yet slightly but consistently deviate at higher fields. The deviations from periodicity are shown in Fig. 4b.

We find quantitative agreement between the observed deviation and the expectations for non-adiabatic corrections of the Weyl orbit (Fig. 4b). Yet deviations of similar magnitude from periodicity could also occur from spin-splitting of the surface state alone, without the presence of Weyl orbits. However, a comparison of the presented data with the predictions for spin-splitting shows that the observed peak positions are qualitatively different from expectations in the spin-splitting scenario, further highlighting the unusual character of the observed oscillations (Extended Data Fig. 4). In the presence of strong magnetic fields, the Fermi surface volume for spin-up and spin-down electrons changes due to the Zeeman energy $\Delta E = \hbar^2 k^2 / 2m_e H_s$, where the $z$ axis is pointing along the magnetic field. This leads to an effective splitting of an initially spin-degenerate Fermi-surface at zero field (in the absence of spin–orbit coupling). In favourable cases, spin splitting can be directly observed as the appearance of two, well-separated peaks positioned symmetrically around the expected position in the absence of spin-splitting. Yet we do not observe split peaks in the surface state related oscillations. This could either be an indication of weak spin splitting, or of a thermal or impurity broadened situation where direct splitting of the peaks is not observable despite strong spin splitting. In the latter case, spin-splitting is known to modify the amplitude of the oscillations only, without affecting the phase of the oscillations. Therefore spin-splitting of a trivial surface state alone is unlikely to explain the present data.

Another possibility may be the formation of a surface state akin to those in a topological insulator (TI). In this case, the spin-momentum locking on the Dirac cone changes the spin-splitting behaviour and oscillation phase changes without any peak splitting are possible. The reason for this is the opening of a gap in the Dirac spectrum due to the time-reversal symmetry breaking of the applied magnetic field. This gap due to the Zeeman energy modifies the Landau level spectrum for electrons (at positive band filling):

$$E_k = \sqrt{v_F^2 (2n\hbar^2 + g_s \mu_B H_s / (2\mu_B^2))} \left(2^{1/2}\right)$$

(2)

In the absence of spin splitting ($g = 0$), this reduces to the conventional square-root-dependence of Dirac systems. At low fields, that is, large Landau level index $n$, this correction is negligible, yet becomes important at higher fields closer to the quantum limit ($n = 0$). The resulting fields $B_n$ where a Landau level equals the Fermi energy $E_F$ can be easily calculated using the material parameters self-consistently obtained from the quantum oscillation analysis (blue dots in Extended Data Fig. 4a) and are contrasted to the measured peak positions (red dots in Extended Data Fig. 4a). Spin-splitting of a TI-like surface state is at odds with our observations for two reasons: (1) the expected deviation goes in the wrong direction and (2) the observed magnitude is incompatible with measured $g$-factors in Cd$_3$As$_2$. Spin-splitting in Cd$_3$As$_2$ occurs at lower fields compared to the purely periodic case, they are on the contrary observed at higher fields. The shift to lower fields in the case of spin-splitting can be easily understood from the surplus Zeeman energy adding to the field dependence of the Landau levels in the absence of spin splitting (see below). This shifts the levels up in energy, and thus they intersect the constant chemical potential at lower fields, hence the upwards bending of the fields $B_n$ as shown in Extended Data Fig. 4b. In addition, a $g$-factor of 300 is required to obtain a shift of similar magnitude as observed by our experiment, which is an order of magnitude larger than the experimentally measured values.

On the other hand, such a downwards deviation as well as its magnitude is expected for Weyl orbits arising from non-adiabatic corrections: at small fields, the quasiparticle evolving on one of the surface states may only enter the bulk state at the Dirac point, where the gap between the surface and bulk states vanishes. In strong magnetic fields, however, the quasiparticle may tunnel through this bandgap into the bulk state before reaching the Dirac point over a distance given by the magnetic length, $L_B = (\hbar/e^2 B)^{1/2}$. This shortens the effective path length on the surface (orange arrows in Fig. 1b) and thus leads to a non-adiabatic correction to the low-field surface frequency $F_s$, in strong fields given by

$$F_s(B) = F_{s,0} - 4\alpha F_{s,0} \Delta k_0 / k_0$$

(3)

where $\alpha$ represents a material dependent parameter encoding the tunnelling barrier between the surface and the bulk bands, and $F_{s,0}$ the field-independent surface frequency in the absence of strong non-adiabatic effects observed at low fields. The four tunnelling processes involved in each Weyl orbit thus lower the effective quantum oscillation frequency $F_s$ in strong magnetic fields.

Including this non-adiabatic correction term effectively explains the observed deviation (Fig. 4b, red points) from the pure 1/$B$ periodicity in high magnetic fields. A fit to the observed data (blue points) yields a coupling parameter $\alpha \approx 1.25$, in good agreement with the theoretical predicted value of order unity. This non-adiabatic correction to the $F_s(B)$ is also evident in the Fourier spectra: while the signature of the bulk frequency is a sharp, symmetrical peak centred at $F_s$, the surface frequency $F_s$ is asymmetric with sizeable spectral weight shifted to lower frequencies. This again is a result of the effective lower frequency in high magnetic fields, and can be well reproduced by a simple calculation inserting the field-dependent frequency given by equation (3) into the Lifshitz–Kosevich formalism and Fourier-transform the resulting waveform (Fig. 4a). Unlike the spin-splitting driven phase modification, the non-adiabatic correction mechanism leads to a slowing-down of the oscillations at higher fields due to the effective reduction of flux enclosed in the quantum path that is cut short by the tunnelling process. This naturally leads to the down-bending observed in our experiment.

**Thickness dependence of the oscillation phase.** Equation (1) describes the expected width dependence of the oscillations arising from the Weyl orbit. Crucially, the thickness $L$ does not affect the oscillation frequency, as the oscillation period $\Delta$ is independent of it: $\Delta = B_n^{0.5} - B_{n-1}^{0.5} = e\kappa_0 \Delta k_0 / 2v_F$. In agreement with this expectation, no thickness-dependence of the frequency is observed. Instead it is expected to only affect the phase offset of the peaks versus 1/$B$ and indeed the data show no thickness dependence to the oscillation frequency. However, experimentally detecting this phase shift is a challenge for four main reasons: (1) the surface- and bulk-frequencies are close to each other leading to strong beating which complicates the identification of the peak positions corresponding to the surface frequency, (2) the peak positions show an additional field dependence due to the non-adiabatic corrections, (3) the oscillatory signal is on top of a large nonlinear and thickness-dependent background magneto-resistance due to finite size effects which must be subtracted to find the peak positions, and (4) the relative amplitude of the surface oscillations compared to the bulk oscillations by itself is strongly thickness dependent, which causes a thickness dependence of the beating pattern. Combined, these uncertainties in fitting the peak positions are larger than the separation between peaks, and therefore from the present data it is impossible to make a quantitative claim about the systematic dependence of the peaks. These difficulties are naturally avoided in the triangular geometry and thickness dependence experiments. By design, the triangular sample averages over parts of different effective width, leading to a cancellation of the surface signal as expected from equation (1).

**Upper field limit for surface state oscillations.** Reference 12 predicted that quantum oscillations associated with the Weyl orbit persist up to a maximum field that depends on sample thickness, $B_{sat} \approx k_0 / (eL)$. By repeating this analysis, we show that the thickness dependence is actually more involved. Instead, the surface arc oscillations persist up to a field that depends in a complicated non-monotonic fashion on film thickness, which is at least as large as the quantum limit associated with the $k$-space area enclosed by the Fermi arcs. To see this, note that from equation (1) above we can see that the smallest integer, $n = N$, for which equation (1) has a solution is given by: $N = \left\lfloor 2\pi x / \gamma \right\rfloor$, where $k_0 = \hbar v_F / e\mu$, and $x$ indicates the closest integer to $x$ that is larger than $x$. $N$ represents the index of the last quantum level associated with the Weyl orbits that can be pushed across the Fermi energy. The field at which level $N$ crosses the Fermi energy, that is, the upper limit field beyond which Weyl surface-arc oscillations cease, is then:

$$B_{sat} = \frac{k_0}{1 - \pi N^2 (\frac{2\pi}{x} \gamma^2)}$$

The first factor in $B_{sat}$ is precisely the 1/$B$ frequency of the surface state, $F_{s} = 56$ T. The second factor depends strongly on $L$ in a complicated oscillatory fashion, but is typically of order 1 (though possibly much
larger when \((k_2L/\pi) - \gamma\) is accidentally close to an integer value). Hence, we see that, in contrast to the claims of ref. 12, the upper limit field for observing Weyl surface arc oscillations is comparable to the quantum limit of the surface state, and is largely independent of sample thickness. In the present measurements, \(B_{ut}\) is expected to be at least \(\sim 60\) T, and is hence unobservable in any non-pulsed field experiment.

**Raw oscillation data from the 150 nm device.** As quantum oscillations are phenomena most easily understood in Fourier space, we limit the discussion in the main text on the analysis of FFT spectra. Fourier transforms, however, can be misleading and thus it is very important that the main results are apparent in the raw data themselves. This is evidently the case: Extended Data Figs 5 and 6 show the oscillatory magnetoresistance after subtracting a second-order polynomial to remove the non-oscillatory components. The same notation of angle is used as in the main text. Scans were performed in 5° steps around the surface state orientations (0°, 180°) and in 10° steps elsewhere for the 150 nm device, and in 5° steps for the triangle. No averaging or symmetrization of the data between field sweeps of opposite polarity was performed, that is, between sweeps with 180° angle difference. From the data set Extended Data Fig. 5, Fig. 2b showing the surface character of the oscillations was constructed. The main result of the manuscript can be seen as follows: for in-plane fields, that is, 90°, 270°, a single frequency corresponding to the bulk field is observed. As discussed in the main text, both the observation of only one single frequency as well as its amplitude is in agreement with all other published data of quantum oscillations on \(\text{Cd}_3\text{As}_2\). For fields perpendicular to the surface, that is, at 0°, 180°, the spectrum is profoundly different. An additional oscillation of higher frequency is mixed with the bulk frequency, leading to a beating pattern. The second frequency gradually disappears as the field is rotated away from 0°, and is identified with the surface state oscillation from the FFT spectrum as described in the main text. Fig. 5 showing the differences between the rectangular and triangular devices was constructed from the raw data set shown in Extended Data Fig. 6. The main points can also be well seen here without resorting to Fourier transforms: despite its identical surface and cross-sectional area, the triangle does not show any sign of a second frequency at any angle, as can be seen from the angle-independent positions of the quantum oscillation peaks. In the rectangular case, however, strong beating appears when the field is perpendicular to the surface, signalling the second frequency. Also, the characteristic angle dependence of surface states as presented in Fig. 2 can be clearly seen when one traces the maxima from the surface to higher angles.

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Extended Data Figure 1 | Synthesis and characterization of Cd$_3$As$_2$ crystals. a, A typical facetted Cd$_3$As$_2$ single crystal, and b, the powder X-ray diffraction (PXRD) spectrum. Cd$_3$As$_2$ single crystals were produced using the flux growth technique with a 5:1 ratio of Cd flux to Cd$_3$As$_2$. Elemental Cd and As were placed into an aluminium oxide crucible with a quartz wool plug and sealed into a quartz ampoule under vacuum. The ampoule was heated to 825 °C and held at that temperature for two days to ensure a fully homogenized mixture. It was then cooled at a rate of 6 °C per hour to 425 °C, where it was centrifuged to remove excess Cd. Bulk crystals were found trapped in the quartz wool following this procedure, and were confirmed to be Cd$_3$As$_2$ by both PXRD and single-crystal X-ray diffraction. The PXRD data (red trace) can be well fitted by the $I$$_4$/acd low-temperature phase of Cd$_3$As$_2$ (ref. 43) (black trace), and no parasitic phases were observed.
**Extended Data Figure 2** | Surface quantum oscillations are resistant to intentional surface damage. 

**a**, Sketch of the device cross-section before and after the irradiation. Three possible current paths exist in these devices: the crystal bulk (purple), the topological surface states (orange), and the amorphous FIB-induced damage shell (green). 

**b**, Device after the heavy irradiation damage. The dimples due to the beam centre impact are well visible across the whole device. The polished back side remained undisturbed by this procedure. 

**c**, Comparison of the same device with pristine surfaces and after damaging the surface. The resistance increases after irradiation, as expected for the increased scattering. 

**d**, Magnetoresistance of the Cd$_3$As$_2$ microstructure before and after irradiation; and **e**, the Fourier components of the quantum oscillations.
Extended Data Figure 3 | Effective mass analysis of bulk and surface oscillations. The temperature dependence of the Shubnikov–de Haas (SdH) amplitudes in the bulk (blue) and at the surface (red) follow the usual Lifshitz–Kosevich behaviour. The fits to the data (lines) yield an effective electron mass in the bulk of 0.044$m_e$, in good agreement with previously reported values\textsuperscript{26}. The surface state appears slightly heavier (0.050$m_e$).
Extended Data Figure 4 | Spin splitting. 

**a.** Positions of the $n$th Landau levels, $B_n$, calculated for spin-splitting (blue) and experimentally observed values (red). The observed deviation is opposite to the expectation of spin-splitting, yet qualitatively and quantitatively consistent with the non-adiabatic corrections of the Weyl orbit process.

**b.** Calculated Landau level energy spectrum for Dirac systems with and without spin-splitting as described by equation (2).
Extended Data Figure 5 | Raw oscillatory signal of the 150 nm device. Shown are Shubnikov–de Haas oscillations of the 150-nm-wide rectangular device, the smallest microstructure fabricated in this study, as a function of 1/field, for different angles between the sample and the magnetic field. Following the notation of the main manuscript, 0° and 180° correspond to a field perpendicular to the surface of the device (‘surf’). The separation between curves along y represents the field angle on a linear scale. Close to the perpendicular field configurations, traces at 5° angle increment were taken, and at 10° increment elsewhere. The appearance of the surface frequency can be well seen in the raw data as strong beating appears around 0°. Also, the characteristic \( \cos(\theta)^{-1} \) angle dependence of a surface frequency can be easily seen by following the peak positions to higher angles.
Extended Data Figure 6 | Raw oscillatory signal of the triangular and rectangular devices. Shown are Shubnikov–de Haas oscillations of the triangular (top) and rectangular (bottom) devices, as a function of $1/\text{field}$ for different angles between the sample and the magnetic field. Traces were taken at 5° angle increment. Those angles where the field is perpendicular to a surface are marked by ‘surf’ for both the rectangular and the triangular devices. While the rectangular device shows the characteristic beating, the peak positions in the triangle remain at the same fields.