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Absence of signatures of Weyl orbits in the thickness dependence of quantum transport in cadmium arsenide

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Editors’ Suggestion

Rapid Communications

In a Weyl orbit, the Fermi arc surface states on opposite surfaces of the topological semimetal are connected through the bulk Weyl or Dirac nodes. Having a real-space component, these orbits accumulate a sample-size-dependent phase. Following recent work on the three-dimensional Dirac semimetal cadmium arsenide (Cd3As2), we have sought evidence for this thickness-dependent effect in quantum oscillations and quantum Hall plateaus in (112)-oriented Cd3As2 thin films grown by molecular beam epitaxy. We compare quantum transport in films of varying thicknesses at apparently identical gate-tuned carrier concentrations and find no clear dependence of the relative phase of the quantum oscillations on the sample thickness. We show that small variations in carrier densities, difficult to detect in low-field Hall measurements, lead to shifts in quantum oscillations that are commensurate with previously reported phase shifts. Future claims of Weyl orbits based on the thickness dependence of quantum transport data require additional studies that demonstrate that these competing effects have been disentangled.

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A Weyl orbit is a unique transport phenomenon of topological semimetals. It involves the Fermi arcs that exist on opposite surfaces of a Weyl semimetal and a connection through the bulk Weyl or Dirac nodes. Having a real-space component, these orbits accumulate a sample-size-dependent phase. Three-dimensional Dirac semimetals, such as Cd3As2 [2], are also thought to host Weyl orbits, because each Dirac node can be considered as two coincident Weyl nodes of opposite chirality [1].

While angle-resolved x-ray photoemission has provided clear evidence of Fermi arcs [3–5], detecting signatures of Weyl orbits has proven to be more challenging. Experimental reports in support of Weyl orbits include two-dimensional Shubnikov–de Haas oscillations of thin slabs of Cd3As2 [13] and their disappearance when the samples were shaped into triangles [6]. The triangular shape is expected to lead to destructive interference of individual Weyl orbits. Recently, the quantum Hall effect has been observed in thin films and platelets of Cd3As2 [7–10], but the nature of the electronic states that give rise to it has not yet been fully clarified. Several different interpretations have been put forward in the literature, including quantum confined bulk states [2,8], topological surface states [14], and Weyl orbits [10]. To support the latter interpretation, a recent study has investigated the thickness dependence of the quantum Hall effect of wedge-shaped platelets of Cd3As2 [12]. In the thinner part of the wedge, the quantum Hall plateaus appeared at a slightly lower magnetic field than in a thicker region. This observation appears consistent with expectations of a thickness-dependent term in the modified Lifshitz-Onsager relation for quantum oscillations involving Weyl orbits, where conductivity minima appear at magnetic fields $B$ given by [1,12]

$$\frac{1}{B} = \frac{2\pi e}{\hbar c A_N} \left( n + \gamma - \frac{L}{2\pi} (k_w + 2k_F) \right),$$

where $e$ is the free electron charge, $\hbar$ is the reduced Planck’s constant, $c$ is the speed of light, $A_N$ is the area enclosed by electrons in $k$ space with their cyclotron orbits on the Fermi surface, $n$ is the Landau level index, $\gamma$ is the Berry phase, $L$ is the sample thickness, $k_w$ is the separation of the Weyl/Dirac nodes along the direction of $B$, and $k_F$ is the Fermi wave vector. Here, the additional phase term $L(k_w + 2k_F)/2\pi$ is related to the tunneling of electrons from the top to the bottom surface [1]. The experimental signature of this additional phase is its dependence on the thickness $L$.

As discussed in Ref. [1], there are many reasons why Weyl orbits may not yet be easily observed in Dirac semimetals, such as Cd3As2. For example, the bulk nodes are expected to be gapped by quantum confinement in thin slabs for which the quantum Hall effect has been reported [7,9,11,13]. Depending on the surface potential [14], Fermi arcs may morph into topological surface states that not necessarily remain connected to the bulk nodes [15]. In the conventional (112) surface orientation, with $B$ normal to the surface, the fourfold symmetry that protects the bulk nodes is broken. We note that the presence of topological surface states is required, however, by the $Z_2$ invariant, even when the bulk nodes are gapped [2].

The goal of the present Rapid Communication is not to address these issues, but to look more closely at the sensitivity of the quantum oscillations of Cd3As2 to the sample thickness and carrier density. In particular, the frequency of Shubnikov–de Haas oscillations is very sensitive to small variations in carrier density, which, in real samples, may also depend on the
sample thickness. To this end, we use a gate voltage, which is applied to a gated Hall bar structure, to match the carrier density to that of four additional Hall bars made from epitaxial Cd$_3$As$_2$ thin films having different thicknesses. This allows us to disentangle the effects of charge carrier density from a true thickness dependence of the quantum oscillations, as may arise from Weyl orbits. We show that the phase of the Shubnikov–de Haas oscillations measured from thin films of Cd$_3$As$_2$ lacks a clear correlation with the film thickness and that it is reasonable to expect the same to be true for other kinds of realistic Cd$_3$As$_2$ specimens.

Epitaxial Cd$_3$As$_2$ thin films were grown by molecular beam epitaxy on (111) GaSb/GaAs substrates, as described in detail elsewhere [16]. Film thicknesses were determined via x-ray reflectivity performed on a Panalytical MRD PRO materials research diffractometer, using Cu $K\alpha$ radiation and an analyzer crystal. The data were fit to a one-layer model using the GENX software package [17]. Thickness maps recorded for films grown on 2-in. wafers show that the thickness in a twofold degeneracy of the Landau levels is lifted in magnetic fields, which are at least as high as those in Ref. [12]. The simulations above 4 T, demonstrating comparable quantum mobility for two slightly differing carrier densities, 0 \times 10^{12} and 1.00 \times 10^{12} cm$^{-2}$, i.e., typical two-dimensional carrier densities.
Hall resistance. The same difference produces only a small change in the low-field oscillation at high fields (around 2 T, as depicted by the dashed lines). The same difference produces only a small change in the low-field Hall resistance ($R_{xy}$), shown in (b), as depicted by the dashed lines.

densities of thin slabs of Cd$_3$As$_2$ [7]. At high fields, the shift between the two sets of oscillations increases. While the determination of the carrier density from the Hall coefficient, i.e., $R_{xy}(B)$, shown in Fig. 2(b), should, in principle, not be limited to low fields, the onset of the quantum Hall effect limits the field range for estimations of the carrier density in practice. Furthermore, Dirac systems often contain $p$-type carriers, which produce a curvature in the high-field $R_{xy}(B)$ data (see, e.g., Ref. [14]). An example is Fig. 1(e), where it is responsible for the discrepancy in the low-field Hall data. In the example shown in Fig. 2, a 5% variation in carrier density produces a shift in the oscillations of 2 T at a field of 30 T, but only a difference in the value of $R_{xy}$ of 0.0064 $h/e^2$ at 5 T, a change so small that it would be completely undetectable in the data in Fig. 1 or Ref. [12]. The shift between the oscillation maxima in the data Fig. 1(c) is about 300 mT. Rather than invoking Weyl orbits, this shift can be explained by a variation of the carrier density of about 4% between the two samples, which should produce a change $\Delta R_{xy} = 10 \Omega$ at 0.4 T and 50 $\Omega$ at 2 T (at the onset of quantum oscillations) in the Hall data, which falls within the uncertainties of the low-field Hall data. To illustrate this point, the small dot in Fig. 1(g) corresponds to this $\Delta R_{xy}$. Thus, small variations in the carrier densities between samples of different thickness cannot be disregarded as a source of an apparently thickness-dependent phase shift in the Shubnikov–de Haas oscillations.

In conclusion, based on the available data, the high degree of sensitivity of the positions of the maxima in the Shubnikov–de Haas oscillations and their associated Hall plateaus to small variations in the carrier density, in combination with the relative insensitivity of the low-field Hall data to such variations, does not allow us to invoke Weyl orbits based on thickness data for realistic samples. There are many reasons why carrier densities may not be completely independent of the sample thickness. For example, the growth rate, and therefore the incorporation of defects that cause the unintentional doping, may differ. Incidentally, we note that a spatial variation in growth rate would give rise to a wedge-shaped sample. Another reason is the sensitivity of the Cd$_3$As$_2$ surface potential to the physical condition of the (air-exposed) surface [14]. The resulting band bending effects will produce a gradient in the carrier density. Finally, we wish to emphasize that the analysis presented here does not disprove the existence of Weyl orbits in general.

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[1] A. C. Potter, I. Kimchi, and A. Vishwanath, Quantum oscillations from surface Fermi arcs in Weyl and Dirac semimetals, Nat. Commun. 5, 5161 (2014).
[2] Z. J. Wang, H. M. Weng, Q. S. Wu, X. Dai, and Z. Fang, Three-dimensional Dirac semimetal and quantum transport in Cd$_3$As$_2$, Phys. Rev. B 88, 125427 (2013).
[3] S.-Y. Xu, I. Belopolski, N. Alidoust, M. Neupane, G. Bian, C. Zhang, R. Sankar, G. Chang, Z. Yuan, C.-C. Lee, S.-M. Huang, H. Zheng, J. Ma, D. S. Sanchez, B. Wang, A. Bansil, F. Chou, P. P. Shibaev, H. Lin, S. Jia, and M. Z. Hasan, Discovery of a Weyl fermion semimetal and topological Fermi arc, Science 349, 613 (2015).
[4] S.-Y. Xu, C. Liu, S. K. Kushwaha, R. Sankar, J. W. Krizan, I. Belopolski, M. Neupane, G. Bian, N. Alidoust, T.-R. Chang, H.-T. Jeng, C.-Y. Huang, W.-F. Tsai, H. Lin, P. P. Shibaev, F.-C. Chou, R. J. Cava, and M. Z. Hasan, Observation of Fermi arc surface states in a topological metal, Science 347, 294 (2015).
[5] L. X. Yang, Z. K. Liu, Y. Sun, H. Peng, H. F. Yang, T. Zhang, B. Zhou, Y. Zhang, Y. F. Guo, M. Rahn, D. Prabhakaran, Z. Hussain, S. K. Mo, C. Felser, B. Yan, and Y. L. Chen, Weyl semimetal phase in the non-centrosymmetric compound TaAs, Nat. Phys. 11, 728 (2015).
[6] P. J. W. Moll, N. L. Nair, T. Helm, A. C. Potter, I. Kimchi, A. Vishwanath, and J. G. Analytis, Transport evidence for Fermi-arc-mediated chirality transfer in the Dirac semimetal Cd$_3$As$_2$, Nature (London) 535, 266 (2016).
[7] T. Schumann, L. Galletti, D. A. Kealhofer, H. Kim, M. Goyal, and S. Stemmer, Observation of the Quantum Hall Effect in Confined Films of the Three-Dimensional Dirac Semimetal Cd$_3$As$_2$, Phys. Rev. Lett. 120, 016801 (2018).
[8] M. Uchida, Y. Nakazawa, S. Nishihaya, K. Akiba, M. Kriener, Y. Kozuka, A. Miyake, Y. Taguchi, M. Tokunaga, N. Nagaosa, Y. Tokura, and M. Kawasaki, Quantum Hall states observed in thin films of Dirac semimetal Cd$_3$As$_2$, Nat. Commun. 8, 2274 (2017).
[9] M. Goyal, L. Galletti, S. Salmani-Rezaie, T. Schumann, D. A. Kealhofer, and S. Stemmer, Thickness dependence of the quantum Hall effect in films of the three-dimensional Dirac semimetal Cd$_3$As$_2$, APL Mater. 6, 026105 (2018).
[10] C. Zhang, A. Narayan, S. Lu, J. Zhang, H. Zhang, Z. Ni, X. Yuan, Y. Liu, J.-H. Park, E. Zhang, W. Wang, S. Liu, L. Cheng, L. Pi, Z. Sheng, S. Sanvito, and F. Xiu, Evolution of Weyl orbit and quantum Hall effect in Dirac semimetal Cd$_3$As$_2$, Nat. Commun. 8, 1272 (2017).

[11] L. Galletti, T. Schumann, O. F. Shoron, M. Goyal, D. A. Kealhofer, H. Kim, and S. Stemmer, Two-dimensional Dirac fermions in thin films of Cd$_3$As$_2$, Phys. Rev. B 97, 115132 (2018).

[12] C. Zhang, Y. Zhang, X. Yuan, S. Lu, J. Zhang, A. Narayan, Y. Liu, H. Zhang, Z. Ni, R. Liu, E. S. Choi, A. Suslov, S. Sanvito, L. Pi, H.-Z. Lu, A. C. Potter, and F. Xiu, Quantum Hall effect based on Weyl orbits in Cd$_3$As$_2$, Nature (London) 565, 331 (2019).

[13] A. Narayan, D. Di Sante, S. Picozzi, and S. Sanvito, Topological Tuning in Three-Dimensional Dirac Semimetals, Phys. Rev. Lett. 113, 256403 (2014).

[14] L. Galletti, T. Schumann, T. E. Mates, and S. Stemmer, Nitrogen surface passivation of the Dirac semimetal Cd$_3$As$_2$, Phys. Rev. Mater. 2, 124202 (2018).

[15] M. Kargarian, M. Randeria, and Y. M. Lu, Are the surface Fermi arcs in Dirac semimetals topologically protected? Proc. Natl. Acad. Sci. USA 113, 8648 (2016).

[16] T. Schumann, M. Goyal, H. Kim, and S. Stemmer, Molecular beam epitaxy of Cd$_3$As$_2$ on a III-V substrate, APL Mater. 4, 126110 (2016).

[17] M. Bjorck and G. Andersson, GenX: An extensible x-ray reflectivity refinement program utilizing differential evolution, J. Appl. Crystallogr. 40, 1174 (2007).

[18] A. Narayanan, M. D. Watson, S. F. Blake, N. Bruyant, L. Drigo, Y. L. Chen, D. Prabhakaran, B. Yan, C. Felser, T. Kong, P. C. Canfield, and A. I. Coldea, Linear Magnetoresistance Caused by Mobility Fluctuations in $n$-Doped Cd$_3$As$_2$, Phys. Rev. Lett. 114, 117201 (2015).