Directional ballistic transport in the two-dimensional metal PdCoO$_2$

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In an idealized infinite crystal, the material properties are constrained by the symmetries of the unit cell. The point-group symmetry is broken by the sample shape of any finite crystal, but this is commonly unobservable in macroscopic metals. To sense the shape-induced symmetry lowering in such metals, long-lived bulk states originating from an anisotropic Fermi surface are needed. Here we show how a strongly faceted Fermi surface and the long quasiparticle mean free path present in microstructures of PdCoO$_2$ yield an in-plane resistivity anisotropy that is forbidden by symmetry on an infinite hexagonal lattice. We fabricate bar-shaped transport devices narrower than the mean free path from single crystals using focused ion beam milling, such that the ballistic charge carriers at low temperatures frequently collide with both of the side walls that define the channel. Two symmetry-forbidden transport signatures appear: the in-plane resistivity anisotropy exceeds a factor of 2, and a transverse voltage appears in zero magnetic field. Using ballistic Monte Carlo simulations and a numerical solution of the Boltzmann equation, we identify the orientation of the narrow channel as the source of symmetry breaking.
The path of PdCoO$_2$ has been demonstrated directly in measurements of the resistivity of layers conducting in parallel. The extremely long in-plane mean free path of the material is considered to be a stack of independent 2D crystals. As shown in Fig. 1a, a crystal platelet, about 350 µm long and 8 µm thick, has been anchored to a sapphire substrate to be patterned and contacted by using conventional microfabrication techniques. Instead, we employ a focused ion beam (FIB) for three-dimensional microsculpting of as-grown crystals. As shown in Fig. 1a, a crystal platelet, about 350 µm long and 8 µm thick, has been anchored to a sapphire substrate by using two-component Araldite epoxy. A thin layer of titanium/gold (10 nm/150 nm) has then been evaporated(Ti)/sputtered(Au) on top of the device to create electrical contacts to the crystal.

Haas–van Alphen$^{17}$, angle-resolved photoemission$^{18,19}$, angle-dependent magnetoresistance$^{20}$ and magneto-transport$^{21}$ measurements have well characterized its hexagonal Fermi surface, which fills half of the Brillouin zone, as expected for a monovalent metal. The out-of-plane dispersion is so weak that transport is essentially two dimensional (2D)$^{22}$, permitting to work in a 2D approximation where the material is considered to be a stack of independent 2D layers conducting in parallel. The extremely long in-plane mean free path of PdCoO$_2$ has been demonstrated directly in measurements of transverse electron focusing$^{23}$ and the observation of field-periodic oscillations in microstructures$^{24}$.

The crystals of PdCoO$_2$ used in this study grow as thin platelets with a typical thickness of approximately 5–30 µm and lateral dimensions of several hundred micrometres. Despite their layered structure, the crystals cannot be exfoliated and laid on a separate substrate to be patterned and contacted by using conventional microfabrication techniques. Instead, we employ a focused ion beam (FIB) for three-dimensional microsculpting of as-grown crystals. As shown in Fig. 1a, a crystal platelet, about 350 µm long and 8 µm thick, has been anchored to a sapphire substrate by using two-component Araldite epoxy. A thin layer of titanium/gold (10 nm/150 nm) has then been evaporated(Ti)/sputtered(Au) on top of the device to create electrical contacts to the crystal.

In a final step, first the titanium/gold layer is locally removed by FIB etching and subsequently a transport bar is shaped into the crystal with suitable voltage contacts lengthways. The FIB-induced surface damage is limited to the outermost approximately 20 nm (Supplementary Note 2) and does not lead to any bulk defects$^{25,26}$, as the ballistic transport observed in the FIB-cut bars self-evidences. Details of crystal synthesis$^{23}$ and FIB microstructuring$^{23,26}$ are given elsewhere. A typical PdCoO$_2$ transport device produced by FIB micromachining is displayed in Fig. 1a. Conveniently, the growth edges of the PdCoO$_2$ crystals are oriented perpendicular to the crystallographic axes, so that the crystal orientation can be determined easily. This permits the fabrication of four serial transport bars precisely oriented with respect to the crystal lattice of the same single crystal. Because of the in-plane six-fold rotational symmetry and reflection symmetry of the palladium planes, the full angular range can be spanned in steps of 10° by choosing to measure parallel to the crystal direction (||[110], here denoted ‘0°’) as well as 10°, 20° and 30° away from the [110] direction. Further, we note that throughout this manuscript we present our data in terms of resistivities, defined by the measured voltages divided by the applied constant current.

![Fig. 1](image-url)
Fig. 2 | Ballistic electron propagation in the case of a circular and a hexagonal Fermi surface. a, The direction of the Fermi velocity \( v_F \) is always parallel to the Fermi momentum \( k_F \) for a circular Fermi surface. b, The situation is drastically different in the case of a hexagonal Fermi surface. Due to the flat sides of the polygon, there are only three possible directions for the Fermi velocities (yellow, blue and red). This restriction of the electron propagation direction results in highly anisotropic, directional ballistic transport. c, The electronic conduction in a four-point transport bar fabricated from a material with an isotropic Fermi surface will not depend on the orientation of the bar. d, In contrast, for a hexagonal Fermi surface, a bar cut parallel to an electron propagation direction will show a lower resistance than a bar aligned perpendicular to an electron propagation direction.

Results
Directional ballistic effects. The in-plane mean free path \( \lambda \) can be estimated from the resistivity under the assumption of 2D transport as \( \rho^{-1} = \frac{\lambda}{k_F \lambda} \), where \( d = 17.73/3 \) Å is the palladium layer separation and \( k_F = 0.95 \) Å\(^{-1} \), where \( k_F \) is the average Fermi wavevector around the Fermi surface (see Supplementary Note 3 for details). Figure 1 contrasts \( \lambda \) of the 0° oriented bar as a function of temperature to that of a bulk (155 μm wide) channel of the same orientation. Data are also shown for the same bar after subsequent narrowing from 7 to 2.5 μm width and are seen to evolve in a textbook fashion: At high temperatures, \( \lambda \) is strongly limited by phonon scattering and the sample is in a diffusive transport regime, hence its electronic response is width independent. At low temperatures, this is no longer the case and the low-temperature value of \( \lambda \) for the restricted channels is limited by their width rather than by bulk scattering. The temperature at which the data first deviate from the diffusive regime is therefore itself width dependent (see Supplementary Note 3 for details).

Figure 1c shows the angular dependence of the in-plane resistivity as a function of temperature. While the resistivity is isotropic above about 50 K, as is expected in the diffusive transport regime, it becomes remarkably anisotropic at lower temperatures, where the electron mean free path exceeds the width of the transport bars. In particular, for the device displayed in Fig. 1a in which the bars are 7 μm wide, the resistivity anisotropy \( (\rho_{0°} - \rho_{30°})/\rho_{0°} \) is as large as 50% between the most and least resistive direction. Upon thinning down the bars to 2.5 μm width, this ratio further increases to 200%.

The order of the curves in Fig. 1c, from least to most conductive, can be understood qualitatively by considering Fig. 2. When the transport bar is oriented such that it is aligned with one of three main directions of the Fermi velocity, a large number of electronic states propagate parallel to the bar and avoid any surface collisions. On the other hand, when the orientation of the transport bar is rotated by 30°, the dominant ballistic directions guide the electrons towards the sample edges, leading to frequent boundary scattering events.

These results clearly demonstrate the notion of directional ballistics. In any material with a circular Fermi surface, the apparent resistivity is enhanced due to boundary scattering, depending on the specularity of the boundaries\(^3\). This effect is isotropic: a bar of a given width and length will have the same resistance no matter what orientation it is cut in. Most high-mobility two-dimensional electron gases have circular or smoothly evolving Fermi surfaces, in which no orientation dependence is observable. In contrast, a Fermi surface with a strongly non-isotropic Fermi velocity distribution can significantly modify the rate of boundary scattering and therefore support an orientation dependence of the resistance. A comparison of the Fermi surface shape and the velocity density map highlights the subtle role of anisotropy (Fig. 3). The overall 2D Fermi surface of PdCoO\(_2\) does not deviate much from a circular approximation, and given its six-fold rotational symmetry, it may not strike the eye as particularly anisotropic. Indeed, the magnitude of the Fermi velocity of PdCoO\(_2\) is almost constant around the Fermi surface\(^4\). The key aspect of directional ballistics, however, is the strong angle dependence of the velocity direction distribution, that is, the probability of finding a certain direction of quasiparticle velocity (Supplementary Note 4). As the nearly flat Fermi surface segments host large densities of states propagating essentially into the same direction, the velocity direction distribution is extremely anisotropic despite the relatively isotropic appearance of the Fermi surface. We propose such a velocity direction distribution map as a tool to visualize the propensity of a material to exhibit directional ballistics.

It is possible to go beyond the above qualitative discussion and perform Boltzmann transport simulations (see Supplementary Note 5 for details) which model the observations remarkably well. Using a Fermi surface shape parameterization established from angle-resolved photoemission data\(^2\), assuming diffusive boundary scattering and obtaining the temperature-dependent bulk mean free path from data on the 155-μm-wide sample (Supplementary Fig. S3) the simulations produce the results shown in Fig. 1d. We emphasize that this agreement is achieved without the use of any free parameters. To investigate the role of the various ingredients to this directional transport separately, we performed further calculations on more restricted models. First, a Boltzmann calculation including realistic bulk scattering but using an unrealistic, mathematically hexagonal Fermi surface probes the role of the rounded Fermi surface (Supplementary Note 6). A second Landau–Büttiker-type calculation includes a realistic Fermi surface model but ignores the bulk scattering (Supplementary Note 7). While both attempts reproduce some qualitative features of the data, the excellent match between simulation and experiment shown in Fig. 1d is only achieved by accounting simultaneously for the bulk scattering and the realistic Fermi surface.

Transverse voltages in zero field. Thus far, we have been concerned with directional ballistic effects observable in the longitudinal electrical transport. However, due to the broken rotational symmetry at the boundaries, finite off-diagonal terms are allowed in the conductivity matrix along low-symmetry directions. Such terms have been used to effectively probe bulk symmetry lowering in Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) single crystals\(^8\) and La\(_{2-}\)Sr\(_x\)CuO\(_4\) thin films\(^9\). In PdCoO\(_2\), these are expected because of directional micro-structuring and can be accessed by the device geometry outlined in Fig. 4a. Along a low-symmetry direction, a transverse voltage in the ballistic regime develops due to an imbalance of electrons propagating towards the two different sides of the transport.
channel. Consequently, one expects the transverse voltage to be of equal strength but opposite sign with respect to the angle tilted away from a high-symmetry direction.

This can be tested in a specifically designed microstructure of PdCoO$_2$ (Fig. 4b). The outline of the crystal is indicated by a dashed line. Following long current-homogenizing meanders at the...
injection (Supplementary Note 8), the heart of the sample consists of two serial transport bars cut at +3° and −3° with respect to the 0° direction, both 2.2 μm wide. Each of the bars has three pairs of opposite voltage contacts, allowing for simultaneous transverse and longitudinal voltage measurements. The resulting temperature dependence of the transverse zero-field resistivities $\rho_{xy}$ is presented in Fig. 4c. In the diffusive transport regime at high temperatures, the transverse resistivity is absent, as expected after appropriate subtraction of the longitudinal voltage contribution originating from imperfect voltage contact alignment (see Supplementary Note 3 for details). With the dominating scattering off phonons in the bulk, the point-group symmetry of the material dominates the scattering, hence in-plane isotropy is symmetry enforced. However, upon entering the ballistic regime, a finite and asymmetric voltage develops across the device depending on the orientation of the transport bar. Again, the observations are closely reproduced by the Boltzmann simulation (Fig. 4d).

Discussion

The excellent agreement between the experimental results presented here and the Boltzmann transport simulation of the directional ballistics highlights the importance of taking the Fermi surface shape and channel direction into account in the analysis of data from width-restricted channels of materials such as PdCoO$_2$ with faceted Fermi surfaces. The analysis presented in ref. 29 considered a circular Fermi surface and hence treated the orientation of the channel relative to the crystal axes as unimportant. The results presented here show that, to conclusively identify a viscous contribution to transport in PdCoO$_2$, further experiments on transport bars aligned along both the 0° and 30° directions will be required, combined with analysis using realistic models of hydrodynamic transport in which the faceting of the Fermi surface is taken into account 31–33.

Conclusions and outlook

We have shown, for the first time in a two-dimensional metal, that a strongly faceted Fermi surface can lead to strongly orientation-dependent conduction in otherwise identical ballistic devices cut from the same single crystal. These observations are of fundamental and practical importance to the question of the minimal attainable resistance in nanoscopic conductors, which ultimately limits the potential miniaturization of electric conductors in technological applications. As conductors are scaled down, even technologically relevant thin films enter the ballistic transport regime at elevated temperature, and boundaries become an important source of scattering 34–35. Our results demonstrate that the boundary scattering contribution in zero field can be reduced by over a factor of two when a 2.5-μm-wide channel is aligned with one of the main directions of quasiparticle propagation. This is not a fundamental limit. Indeed, the smallest width we have studied in these proof-of-principle experiments is at least an order of magnitude larger than the minimum that could be envisaged. For wires less than 10 μm long, narrowed to widths of order 100 nm, the effect may be much larger (see the discussion in Supplementary Note 5), resulting in significant gains in attainable channel conductivity compared with that available from materials with circular Fermi surfaces. Our results invite investigation of other delafossite metals in which there are subtle differences in the degree of Fermi surface faceting 36 and also a thorough study of the effects of magnetic field on directional ballistics.

Finally, we note that the phenomena we report here are far from being restricted to delafossites. Materials with faceted Fermi surfaces are not rare. Gated bilayer graphene 37 is one of the most promising platforms for extensions of this research. While the Fermi surface anisotropy can also be controlled in GaAs-based quantum wells 38,39, despite its anisotropic appearance, the Fermi velocity distribution remains quite isotropic. Most strikingly, the in-plane transport anisotropy is a widely used technique to detect subtle symmetry-lowering electronic states such as electronic nematicity. The implicit assumption is based on the group-theoretical argument that the appearance of an in-plane anisotropy in transport necessitates that the rotational symmetry of the bulk material be reduced to two-fold. Our work shows that, especially in clean metallic crystals, even symmetry lowering by the sample shape itself can induce such an anisotropic response, without any broken rotational symmetries in the point group of the unit cell. As mean free paths can indeed become macroscopic, these effects may well appear in traditional single crystals. This may prove to be important in the interpretation of unconventional transport phenomena in topological semi-metals, which generally tend to be of high mobility. For example, a mean free path $\lambda > 100 \mu m$ is readily observed in the Weyl II semi-metal WP$_2$ (ref. 37), which in turn implies that even the sub-mm-sized single crystals used in traditional conductivity measurements are in a quasi-ballistic transport regime. Given the common deviation from circular Fermi surfaces in these materials class, the effects uncovered here are likely to be of relevance to that field.

It will also be interesting to consider whether directional ballistics plays a role in generating subtle symmetry-forbidden transport signals at phase transitions of strongly correlated materials, in which one might a priori not expect to encounter ballistic behaviour due to their short mean free path. However, because the most strongly interacting states at the Fermi level are those driving the ordered phase, the average lifetime of the remaining quasiparticle states often increases significantly upon electronic ordering. Famous examples of dramatic enhancements of the mean free path include the hidden order transition in URu$_2$Si$_2$ 40, the nodal quasiparticles in YBa$_2$Cu$_3$O$_{7-\delta}$ and the formation of coherent heavy fermion liquids 41. It is interesting to note that all these microscopically distinct correlated transitions are associated with weak symmetry-forbidden anisotropies, and at the same time these phase transitions push these materials closer to the directional ballistic limit. In summary, we believe that much remains to be investigated concerning the physics as well as potential applicability of directional ballistics.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-022-01570-7.

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Data availability
All raw data underpinning this publication can be accessed in comprehensible ASCII format at https://doi.org/10.5281/zenodo.5964955. Source data are provided with this paper.

Code availability
Source code for the simulations can be found at https://github.com/dgglab/ballistic_montecarlo.

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Author contributions
M.D.B., C.P., N.N., P.H.M., E.Z., M.M., M.K. and P.J.W.M. fabricated the microstructures and performed the measurements. The crystals were grown by S.K. A.L.S., A.W.B. and D.G.-G. performed the Monte Carlo simulations. T.S. performed the kinetic calculations. G.B. and D.A.B. performed the Boltzmann transport simulations. All authors were involved in the design of the experiment and writing of the paper.

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