Observation and manipulation of maximal Chern numbers in the chiral topological semimetal PdGa

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Topological semimetals feature protected nodal band degeneracies characterized by a topological invariant known as the Chern number. The magnitude and sign of this invariant determine many of the exotic properties of such semimetals1–10, including the number of topologically protected surface Fermi-arcs, the number of chiral landau levels in the presence of a magnetic field, or the direction and strength of topological photocurrents in optical experiments. While formally the Chern number C can take any integer value, any nodal crossing with linear dispersion is expected to have at most |C|=4, realized in fourfold and sixfold nodes in spin-orbit coupled, non-symmorphic chiral crystals11–14. Despite multiple recent photoemission studies of predicted material candidates15–19, this maximal Chern number, measured by counting the number of Fermi-arcs, has not yet been observed experimentally. Using angle-resolved photoelectron spectroscopy and ab-initio calculations, here we show that the chiral topological semimetal candidate PdGa displays the required multifold nodes, which are connected by long surface Fermi-arcs. Due to large spin-orbit coupling and the high crystallinity of our sample surfaces, we are for the first time able to resolve 4 independent Fermi-arcs, proving experimentally that the observed multifold nodes – which are a generic feature of many chiral metals – display the maximal Chern number magnitude of 4. Furthermore, by comparing two crystals with mirrored crystal structure, we observe a reversal of the Fermi arc velocities, which demonstrates that the sign of the Chern numbers is tied to the handedness of the sample, which establishes an important control parameter for future experiments probing topological responses in chiral topological semimetals.
Topological invariants are mathematical objects that can be used to classify Hamiltonians, which have found widespread applications in physics, chemistry, and materials science. One of the most celebrated topological invariants in condensed matter physics is the Chern number, which in band theory language is the flux of Berry curvature through a closed two-dimensional surface. If this surface is taken to be the whole Brillouin Zone, the Chern number classifies insulators in two dimensions, as first used in the context of the quantum Hall effect by Thouless and co-workers\textsuperscript{20,21} in the 1980s. More recently, Chern numbers have also been used to classify topological nodal semimetals\textsuperscript{1}, where point-like energy degeneracies in their bulk electronic structure act as sources and sinks of quantized Berry flux through any local isoenergy surface enclosing the node. For the simplest case of a linear touching of two bands, which can occur in any non-centrosymmetric or magnetic material and is called a Weyl point, the magnitude of the Chern number $C$ is limited to $|C|=1$. However, there is no reason preventing more complicated nodal crossings from having larger Chern numbers, which has important consequences for many of the exotic phenomena that have been predicted for topological semimetals that are often directly proportional to their Chern number, such as the number of their topological Fermi-arc surface states$^{1,7}$, the number of chiral Landau levels influencing magnetotransport phenomena related to the chiral anomaly$^{4,5}$, the strength of the quantized photocurrents in the quantized circular photogalvanic effect$^{8–10}$, and many more$^{3,6,22}$.

It has recently been predicted that in chiral crystals, which possess neither mirror nor inversion symmetries, more complex band crossings can be pinned at high symmetry lines or points that feature large Chern numbers. For example, twofold crossings with quadratic or cubic dispersion are predicted to host $|C|=2$ or $|C|=3^{23,24}$. In materials with negligible spin-orbit coupling (SOC), three-fold and fourfold crossings can be found with $|C|=2$ per spin, while the combination of non-symmorphic symmetries and significant spin-orbit coupling gives rise to protected fourfold and sixfold degeneracies with Chern numbers up to magnitude 4. The symmetry classification is exhaustive for linear crossings$^{9,11–14,25}$ and predicts that $|C|=4$ only occurs due to SOC and is the highest possible Chern number achievable for a multifold node in chiral topological semimetals.

The family of chiral semimetals in space group 198, including RhSi, CoSi, AlPt, and PdBiSb, is expected to display these type of $|C|=4$ crossings, realized as a fourfold spin $S=3/2$ crossing at the $\Gamma$ point and a sixfold crossing at the R point of the Brillouin zone. Despite several recent angle-resolved photoelectron spectroscopy (ARPES) experiments on all these candidates$^{15–19}$, the absolute magnitude of the Chern number, measured by counting the number of Fermi-arcs, has not yet been observed due to two main roadblocks. The first is that SOC in some of these materials is low and spin-split Fermi-arcs cannot be resolved, effectively leading to only
two observable arcs. Most reports to date\textsuperscript{15–17} in fact classify these nodes as having $|C| = 2$. The second is the preparation of clean and flat surfaces by cleaving or sputtering and annealing, which has resulted in rough or nonstoichiometric surfaces for all previously examined candidates, causing band broadening that can wash out signatures of spin-split bands due to SOC. In this work, we overcome both obstacles by investigating a new chiral topological semimetal candidate PdGa from space group 198, which has substantial SOC and can be prepared with flat, clean, and well-ordered surfaces by polishing and subsequent sputtering and annealing in ultrahigh vacuum\textsuperscript{26–28}. Employing ARPES and ab-initio calculations, we can clearly resolve the presence of multifold crossings in the bulk electronic structure of PdGa, as well as four topological Fermi-arcs on its surface, thus observing for the first time an experimental realization of the maximal Chern number $|C| = 4$. Interestingly, PdGa is known as an important catalyst, for instance for the semi-hydrogenation of acetylene\textsuperscript{29}, and shows potential for enantioselective catalytic reactions of chiral molecules\textsuperscript{30}, which could be enhanced due to the large surface electronic density of states of the Fermi-arcs discovered in this work.

Besides the first observation of the largest possible Chern number in topological semimetals, we also describe here how to manipulate its sign by changing the handedness of their crystal structure. This has striking consequences for their physical properties, since in in these materials, the direction of their chiral Fermi-arcs, chiral Landau levels, and quantized photocurrents is tied directly to the sign of the Chern number associated with the multifold nodes at the Fermi level. By measuring Fermi-surface maps for two enantiopure samples of PdGa with opposite handedness, we can detect the reversal of the Fermi velocity of their topological edge states when the sign of the Chern number is flipped. Manipulation of the Chern number sign by engineering of the crystal structure will become an important control parameter in future experiments that probe the topological properties of chiral crystals.

The PdGa samples used in this study crystallize in the cubic space group 198 with a lattice constant of a=4.896 Å. The chiral motif in their structure is the helical arrangement of Pd and Ga atoms along the (111) direction, which is displayed in Fig. 1A. Upon a mirror operation, these helices reverse their handedness, which can be used to distinguish the two enantiomers of PdGa. We grew two enantiopure specimen of PdGa with opposite chirality via a self-flux method with a chiral seed crystal (see methods and supplementary for further details). The chirality of the crystal structure can also be observed from the intensity distribution of low energy electron diffraction (LEED) patterns of the (100) surface\textsuperscript{28} at an electron energy of $E_{\text{kin}}=95$ eV, which we show in Fig. 1B. As can be expected, the S-shaped intensity distribution is mirrored when comparing the two enantiomers. The crystals used for the LEED study were prepared by the same sputter-annealing recipe as for the subsequent ARPES experiments, which is well known to produce clean and stoichiometric surfaces of PdGa\textsuperscript{26}. In Fig. 1C, we
display the results of an ab-initio bulk band structure calculation, which shows fourfold and sixfold band crossings at the $\Gamma$ and $R$ high symmetry points, respectively. Such band crossings in space group 198 were predicted to carry a Chern number of magnitude 4, with opposite signs at the $\Gamma$ and $R$ points$^{11-14}$. Since the Berry curvature is a pseudovector, a mirror operation will reverse the sign of the Chern numbers associated with the nodes at the high symmetry points. Such a mirror operation also leads to a reversal of the propagation direction of the Fermi-arcs, as is illustrated in Fig. 1D. Any gapped 2D slice in the 3D Brillouin zone between the $\Gamma$ and $R$ points can be understood as a quantum Hall subsystem due to the Chern number of magnitude 2 that is associated with it. The two edge states of this quantum Hall state reverse their direction when the flux of Berry curvature is flipped, which also leads to a reversal of the velocity of the Fermi-arcs that are formed by the ensemble of all 2D slices in the 3D Brillouin zone.

We performed bulk sensitive soft X-ray ARPES measurements on the (100) surface of our PdGa samples to investigate their bulk electronic structure, the results of which are displayed in Fig. 2. We find that multifold crossings predicted at the $R$- and $\Gamma$-points are indeed present (see Fig 2A-C), and that our ab-initio calculations are in good agreement with the observed band dispersions. This agreement can also be observed from the Fermi surfaces for different high-symmetry planes displayed in Fig. 2D-E.

After establishing the existence of multifold band crossings in PdGa, we will now investigate the topological character of these crossings via surface sensitive ARPES of the (100) surface of enantiomer A at low photon energies ($h\nu<150$ eV), as well as ab-initio slab calculations. By comparing the calculated and experimental Fermi surfaces in Fig. 3A-B, we can identify the existence of Fermi arc surface states (indicated by red arrows) that thread through the projected bulk band gap (white areas between projected bulk pockets that are represented by blue lines in Fig 3A), thereby connecting the pockets at $\bar{\Gamma}$ and $\bar{R}$, respectively. By performing photon energy dependent ARPES along the $\bar{R}$-$\bar{\Gamma}$-$\bar{R}$ direction, we confirm experimentally that these Fermi-arcs are indeed surface states without noticeable dispersion along the $k_z$ direction, which is perpendicular to the sample surface, as can be seen from Fig. 3C. Interestingly, we also found additional surface states that overlap with the projected bulk pocket at $\bar{\Gamma}$ (indicated by purple arrows). Due to the sizable SOC in PdGa and high quality of our ARPES data, we are furthermore able to resolve a spin-splitting in the surface Fermi-arcs (see Fig. 3D-F, and the calculation in Fig. 3A for comparison), which is the first experimental confirmation of the maximal Chern number magnitude 4 for multifold crossings in a topological semimetal due to four Fermi-arcs connecting $\bar{\Gamma}$ and $\bar{R}$ points. We find that the SOC splitting of the Fermi-arcs close to the Fermi level is $\sim 0.015$ Å$^{-1}$ and $\sim 60$ meV. Since these multifold
crossings are a generic feature of many chiral topological semimetals, we expect that our finding will also hold for other compounds from the same material family.

Next we investigate how the maximal Chern number in PdGa can be manipulated by tuning the handedness of its crystal structure. When comparing the Fermi-surfaces for enantiomers A and B as shown in Fig. 4A, we see that the Fermi arcs wind around the bulk pocket at \( \bar{R} \) in opposite directions. Due to the very high momentum resolution of the Fermi surface maps shown in Fig. 4B, we can cut an arbitrary path through the Brillouin zone that passes though the projected bulk band gap, indicated by the red dashed line. By comparing the band dispersion of the Fermi-arcs between the two enantiomers along this path (as displayed in Fig. 4C), we can see that the Fermi velocity of the edge states is indeed reversed, which implies that the Chern number signs are reversed between the two enantiomers. This observation constitutes the first experimental proof that the sign of the Chern numbers in topological semimetals can be manipulated by controlling the handedness of their crystal structure. We expect that this finding will serve as a control parameter in future experiments that investigate the response of topological semimetals to external perturbations, such as all-optical measurement of the quantized circular photogalvanic effect\(^\text{31}\). Here, a comparison of the nonlinear response between two enantiomers should give the same magnitude of the mesa-like plateau region in the photocurrent spectrum, albeit with a reversed sign. Similar arguments can also be applied to other experiments that attempt to probe topological properties proportional to the Chern number, which shows that our results establish a procedure to distinguish responses of topological origin from trivial ones.

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Fig. 1: Structural and electronic chirality in the two enantiomers of PdGa

(A) Illustration of the crystal structure of two enantiomers of PdGa with opposite handedness.

(B) Low energy electron diffraction patterns for two samples with opposite chirality, measured with an electron energy of $E_{\text{kin}} = 95$ eV. The S-shaped intensity distribution of the diffraction spots (highlighted by red dashed lines as guide for the eye) reflects the handedness of the crystal structure.

(C) Ab-initio calculations of the band structure in PdGa, showing fourfold and sixfold band crossings at the $\Gamma$ and $R$ points. The Chern numbers associated with the crossings are of magnitude 4 and flip their sign upon a mirror operation. This reverses the direction of Berry flux that is flowing from the crossing with positive Chern number towards the crossing with negative Chern number. Inset shows the cubic Brillouin zone with high symmetry point $\Gamma$ at the zone center and $R$ at the zone corner.

(D) Illustration of the position of sources and sinks of Berry curvature in the Brillouin zone. Blue shaded squares indicate 2D quantum Hall subsystems that are associated with a Chern numbers of magnitude 2. The signs of these Chern numbers depend on the direction of the Berry flux, which is reversed between the two enantiomers.
Fig. 2: Electronic characterization of the bulk electronic structure of PdGa measured on the (100) surface of enantiomer A

(A-C) Comparison between ARPES spectra (left) and ab-initio calculations (right) along high symmetry lines that pass through the Γ and R points. Multifold fermions are indicated by red arrows. Spectra were measured at $h\nu=773$ eV, 552 eV, and 620 eV, respectively. The polarizations used here were circular-left (CL), linear-vertical (LV), and linear-vertical, respectively.

(D-E) Comparison between experimental Fermi-surfaces (left) and ab-initio calculations (right). (D) shows the Fermi surface in the high symmetry plane containing the G point (measured with $h\nu=620$ eV, pol=LV), whilst (E) contains the plane including the R point (measured with $h\nu=540$ eV, pol=LV). The blue dashed lines indicate the boundary of the bulk Brillouin zone. Ab initio calculations include $k_z$ broadening of 0.1 Å$^{-1}$. 
Fig. 3: Surface electronic structure of the (100) surface of enantiomer A

(A) Ab-initio slab calculation of the Fermi surface in the (100) plane (i.e. $k_x$ vs. $k_y$ plane) showing surface Fermi-arcs (indicated by a red arrows), superimposed by projected bulk band structure calculation (solid blue lines). The spin-direction that is shown here is projected along the $<S_x>$ axis.

(B) Experimental Fermi surface measured with hv=60 eV and linear-horizontal (LH) polarization. Red arrows indicate Fermi-arcs, whilst purple arrows indicate additional surface states that overlap with the projected bulk states at $\Gamma$.

(C) Experimental Fermi surface perpendicular to the sample surface (i.e. $k_y$ vs. $k_z$ plane), showing that the Fermi-arcs and surface states (indicated by red and purple arrows, respectively) show negligible dispersion along the $k_z$ direction. Conversion from photon energy was performed within free-electron final state approximation with inner potential of $V_0 = 12$ eV.

(D) Magnified Fermi-surface measured in the region of the red dotted rectangle shown in (B) with hv=30 eV and LH polarization. Red arrows indicate spin-splitting of Fermi-arcs.

(E) Band dispersion measured along the path in momentum space indicated by the dashed red arrow shown in (D) that is crossing the Fermi-arcs. Red arrows indicate their spin-splitting.
(F) Magnified version of (E), insets are momentum distribution curve (MDC) and energy distribution curve (EDC) along the dashed blue lines
Fig. 4: Comparison of the surface electronic structure of the (100) surface of enantiomer A (left) and enantiomer B (right)
(A) Comparison of the Fermi-surfaces for two enantiomers, measured with photon energy $h\nu=60$ eV and LH polarization. Red arrows indicate Fermi-arcs that reverse the direction along which they are dispersing around the $\overline{R}$ pocket under a mirror operation.

(B) Comparison of magnified Fermi-surfaces measured with photon energy $h\nu=30$ eV and LH polarization. Red dashed line indicates momentum path that is cutting through the projected bulk band gap that separates the projected bulk pocket at $\Gamma$ from the projected bulk pocket at $\overline{R}$.

(C) Band dispersion along the path indicated by the red dashed line in (B). Red arrows indicate the Fermi-arc dispersions, which reverse the direction of their Fermi-velocity $v_F$ between the two enantiomers.

**Methods section**

**PdGa Crystal Growth and Structure Refinement**

PdGa single crystals were grown from its melt using the self-flux technique. First, a polycrystalline ingot was prepared using arc melt technique with the stoichiometric mixture of high purity Pd and Ga metals. Then the crushed powder was filled in a bottom-cone shaped alumina crucible and finally sealed in a quartz tube. In order to control the structural chirality of the grown PdGa single crystals, we prepared the seeds of Fe$_{1-x}$Co$_x$Si with Co doping $x = 0.08$ and $x = 0.25$ [1]. Single crystals of Fe$_{1-x}$Co$_x$Si [$x = 0.08$ and $0.25$] were grown using the self-flux technique with melting the respective stoichiometric polycrystalline material at 1500 °C and followed by a slow cooling to 1200 °C with a rate of 5 °C/h. Then, in order to verify whether it is possible to transfer the details of structural chirality from the seed to another system, we cut small portion of the grown Fe$_{1-x}$Co$_x$Si single crystals and used as a seed at the bottom of the alumina crucible. For both batches of PdGa single crystals, first the entire quartz ampoule was heated to 1100 °C, halted there for 12 h and then slowly cooled to 900 °C with a rate of 1.5 °C/h. Finally, the sample was cooled to 800 °C with a rate of 50 °C/h, annealed for 120 h and then cooled to 500 °C with a rate of 5 °C/h. For the growth of right-handed or form A in Refs. [2,3], single crystal of Fe$_{1-x}$Co$_x$Si with $x = 0.08$ and for the left-handed or form B, $x = 0.25$ single crystals were used as a seed. High quality PdGa single crystals with average dimension of about 18 mm length and 6 mm diameter was obtained.

**ARPES**

Soft X-ray ARPES (SX-ARPES) measurements were performed at the SX-ARPES endstation [4] of the ADRESS beamline [5] at the Swiss Light Source, Switzerland, with a SPECS analyzer with an angular resolution of 0.07°. The photon energy varied from 350-1000 eV and the combined energy resolution was ranging between 50 meV to 150 meV. The temperature during sample cleaving and measurements was about 20 K and the pressure better than 1x10$^{-10}$ mbars. The increase of the photoelectron mean free path in the soft-X-ray energy range results, by the Heisenberg uncertainty principle, in a higher $k_z$ resolution of the ARPES experiment compared to measurements at lower photon energies [6], which was critical to measure the new Fermions in the bulk band structure of PdGa.

VUV-ARPES measurements were performed at the high-resolution ARPES branch line of the beamline I05 at the Diamond Light Source, UK [7]. Measurements at the high-resolution branch were performed with a Scienta R4000 analyzer, and a photon energy range between 20
eV and 200 eV, at a temperature below 20 K. Measurements in the VUV-ARPES regime are more surface sensitive than SX-ARPES and therefore most suitable to image the Fermi-arcs in PdGa.

Ab-initio calculations

We employed density functional theory (DFT) as implemented in the Vienna Ab Initio Simulation Package (VASP) [8,9], as well as Wien2k [10] (the latter only for Fig 2. D-E).

For the VASP calculations, the exchange correlation term is described according to the Perdew-Burke-Ernzerhof (PBE) prescription together with projected augmented-wave pseudopotentials [11]. For the autoconsistent calculations, we used a 7x7x7 k-points mesh for the bulk and 4x4x1 for the slab calculations. The kinetic energy cut off was set to 400 eV. We calculated the surface states by using a slab geometry along the (001) direction. In order to achieve a negligible interaction between the surface states from both sides of the slab and reduce the overlap between top and bottom surface states, we considered a slab of 10-unit cells and 1 nm vacuum thickness. For the energy cuts, we used a 100x100 grid of K points.

The Wien2k calculations employed a full-potential linearized augmented plane-wave and local orbitals basis, as well as the PBE prescription of the exchange correlation term. The plane-wave cutoff parameter RMTKMAX was set to 7 and the irreducible Brillouin zone was sampled by 97,336 k-points. Spin-orbit coupling was included via a second variational procedure.

The crystal structure plotted in Fig. 1 were generated with VESTA [12].

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

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Supplementary materials

1. PdGa Crystal Growth and Structure Refinement

Single crystallinity was first checked with a white beam backscattering Laue X-ray setup at room temperature. Then we performed rigorous single crystal X-ray diffraction experiments to analyze the structural chirality of the grown two batches of PdGa single crystals. Small pieces picked from different parts of the grown crystals were considered to truly reflect the structural properties of the entire sample. The selected crystallites were mounted on Kapton loops with aid of a trace of Apiezon H grease and used in diffraction experiments on a Rigaku AFC7 four-circle diffractometer with a Saturn 724+ CCD-detector applying graphite-monochromatized Mo-Kα radiation. A total of five crystals were fully characterized, complete crystallographic information and final results are compiled in standard cif format and are available at …. In any case, the determination of the absolute structure via refinement of Flack’s parameter confirmed single domain crystals without any significant contribution from twinning by inversion [4]. This fully confirms the proper assignment of hand as expected from crystal growth. Even more, one among three crystallites originating from the right-handed crystal showed diffraction images of rather fair quality; however, absolute structure refinement gave excellent results, thus reflecting the robustness of the procedure.
Fig.1: Basic structural unit for the PdGa crystals with the refined structural parameters. (i): right handed or A-form crystal in ref. [2] [Pd at $x = 0.14246(4)$, Ga at $x = 0.84301(6)$] and (ii): left-handed or B-form crystal in ref. [2] [Pd at $x = 0.85758(3)$, Ga at $x = 0.15694(5)$]. The corresponding top picture shows the grown PdGa single crystal. Below are the oscillation images about a main axis for both PdGa single crystals reflecting the excellent crystalline quality.
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