Broken time-reversal symmetry in the topological superconductor UPt₃

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Topological properties of materials are of fundamental as well as practical importance⁴⁻⁶. Of particular interest are unconventional superconductors that break time-reversal symmetry, for which the superconducting state is protected topologically and vortices can host Majorana fermions with potential use in quantum computing⁷⁻⁹. However, in striking contrast to the unconventional A phase of superfluid He where chiral symmetry was directly observed, identification of broken time-reversal symmetry of the superconducting order parameter, a key component of chiral symmetry, has presented a challenge in bulk materials. The two leading candidates for bulk chiral superconductors are UPt₃ (refs. ¹⁰⁻¹¹) and Sr₂RuO₄ (ref. ¹²), although evidence for broken time-reversal symmetry comes largely from surface-sensitive measurements. A long-sought demonstration of broken time-reversal symmetry in bulk Sr₂RuO₄ is the observation of edge currents, which has so far not been successful¹². The situation for UPt₃ is not much better. Here, we use vortices to probe the superconducting state in ultraclean crystals of UPt₃. Using small-angle neutron scattering, a strictly bulk probe, we demonstrate that the vortices possess an internal degree of freedom in one of its three superconducting phases, providing direct evidence for broken time-reversal symmetry in this material.

The heavy-fermion superconductor UPt₃ exhibits three distinct vortex phases shown in Fig. 1 and labelled A, B and C, and two zero-field Meissner phases, providing a strong indication that it breaks additional symmetries of the normal state beyond gauge symmetry. The existence of two zero-field superconducting phases, with a small splitting in the transition temperatures (T₁, T₂), suggests that the superconducting order parameter belongs to one of the two-dimensional representations of the point group Dₙᵥ (ref. ¹¹). The most likely are f-wave pairing states with the E₃g irreducible representation, corresponding to a time-reversal symmetric A phase and a B phase that breaks time-reversal and mirror symmetries. The presence of a weak symmetry-breaking field (SBF)—for example, weak in-plane anti-ferromagnetism or strain—lifts the degeneracy of the multi-dimensional representation, leading to multiple superconducting phases. Identification of the E₃g representation is supported by a number of experimental results, including the H–T phase diagram¹²⁻¹⁵, and thermodynamic and transport studies¹⁶⁻¹⁷. Furthermore, the linear temperature dependence of the London penetration depth, obtained from both magnetization¹⁸⁻¹⁹

and small-angle neutron scattering (SANS)¹⁰ measurements, is consistent with quadratic dispersion at the polar nodes of the energy gap structure that is one characteristic of the E₃g model. However, the evidence for broken time-reversal symmetry comes from surface probes, most recently phase-sensitive Josephson tunnelling in the B phase, directional tunnelling to resolve the nodal structure in the A phase and the observation of polar Kerr rotation in the B phase, but not the A phase. This is analogous to the situation in Sr₂RuO₄, although recent thermal conductivity and O Knight shift measurements call into question earlier reports of odd parity in this material. Additionally, there are unresolved conflicting reports on the evidence for broken time-reversal symmetry in UPt₃ based on muon spin rotation²⁴⁻²⁵. With the exception of the SANS experiment by Gannon et al.²⁰, all of the experiments listed above were performed in the Meissner state.

We performed SANS studies of the vortex-lattice (VL) structure in the B phase, extending into the C phase. The goal is to investigate the theoretical prediction of broken time-reversal symmetry in the B phase of UPt₃ using vortices as the probe. This is greatly facilitated by prior demonstrations that the superconducting state of UPt₃ forms in single-order-parameter domains. Furthermore, field training prior to measurements of the Kerr rotation angle established that the direction of chiral symmetry is controlled by the direction of a non-zero magnetic field on entering the superconducting state. The SANS measurements were carried out with H∥c, where the VL is especially sensitive to changes in the superconducting state due to the hexagonal crystal structure and corresponding weak basal plane anisotropy of UPt₃ (refs. ¹⁴⁻¹⁵). We find a difference in the VL configuration depending on the sign of the magnetic field relative to the field-induced direction of the superconducting order parameter established on entering the B phase at low temperature, showing that the vortices in this material possess an internal degree of freedom. This observation is facilitated by the discovery of a field-dependent, non-monotonic VL rotation, driven by competition between superconducting gap distortion and the vortex-core structure. From this we infer that time-reversal symmetry is broken in the zero-field B phase, and that the c axis is an axis of chiral symmetry.

Results

SANS studies of the UPt₃ VL were carried out at Oak Ridge National Laboratory (ORNL) and at Institut Laue-Langevin (ILL).
measurements were performed on VLs prepared using two different field histories illustrated in Fig. 1. For a field reduction (++, ++) or field reversal (−−, −−) procedure illustrated by the arrows and circles. The arrows on the right show the directions of the internal orbital angular momentum of the Cooper pairs associated with the topological winding number of the B phase (L_{BC} = m = ±2), the global phase winding determined by the applied field (H∥/Hc = n = ±1) and the real-space phase winding of the time-reversed order parameter in the vortex core (p = n + 2m). The inset shows the energy of the superconducting condensate as a function of field history: for a field reduction the system is in the ground state (solid lines) and for a field reversal it is in a metastable state (dashed lines).

Figure 2 shows SANS VL diffraction patterns illustrating the two central results in this report. First, the scattered intensity is divided between two Bragg peaks separated by a splitting angle, ω. As seen in Fig. 2a–d the splitting displays a non-monotonic field dependence, first increasing and then decreasing. Additional diffraction patterns, including both types of field history, are shown in Extended Data Fig. 1. The peak splitting indicates the presence of two triangular VL domain orientations, rotated clockwise and anticlockwise about the crystalline c axis. In real space the splitting of the Bragg peaks corresponds to a VL nearest-neighbour direction rotated away from the a* direction by ±ω/2. Previous SANS studies with H∥c were performed in a magnetic field, 0.19 T, too low to resolve this splitting clearly.

Our second, and main, result is an observed difference of the Bragg peak splitting for the two field histories used, providing direct evidence for broken time-reversal symmetry of the parent B phase. This is illustrated in Fig. 2.e,f for a field magnitude of 1.1 T. The difference is emphasized in Fig. 2g, which shows a subtraction of the VL diffraction patterns obtained following a field reduction and a field reversal. Here an intensity ‘dipole’ is observed around each peak position, outlined by white sector boxes, due to the difference in the splitting. The field dependence of ω and the difference between field histories (Δω = ω−ω+) or ω−ω−) are shown in Fig. 3a,b, respectively. Here, the splitting was determined from the angular positions of two-Gaussian fits to the VL diffraction patterns, and the error bars represent one standard deviation. The curves in Fig. 3 are fits to an empirical function of magnetic field. Details of the two-Gaussian fitting of the VL peak positions and the empirical function are provided in the Supplementary Information.

Below 0.7 T the field-reversed split is slightly smaller than the field-reduced one, giving rise to a negative Δω. For fields between 0.8 T and 1.5 T the situation is the opposite, where we find Δω > 0. While Δω is small at low fields, the high-field results are clear and statistically significant. Moreover, the uncertainty for the vortex density in the vortex core appears owing to the ground symplectic properties of the vortex-core structure. The upper limit to accessible fields is due to the reduced Ω/τ_scatter scattering, which leads to the increased error bars for the determination of ω and Δω at higher fields.

The location of the B–C phase transition varies between samples apparently owing to crystal quality12,13. It is therefore desirable to identify features in the SANS data that allow an in situ determination of this phase boundary. Previous SANS studies with H∥c found a change in the UPr3 VL structure, and in one case a disordering of entering the C phase12,13. For H∥c we observe that the VL peak splitting changes with the applied field up to 1.5 T, as shown in Fig. 3a. Above this field the splitting stabilizes at a ω ≈ 8° (dotted line), deviating from an extrapolation of the lower-field data (solid and dashed lines).

The radial width of the Bragg peaks in the detector plane (ΔQ0), shown in Fig. 4a, also increases abruptly above 1.5 T, indicating a disordering of the VL. Note that the minimum ΔQ0, observed at intermediate fields, is smaller than the divergence of the incident neutrons, determined from the undiffracted beam. This corresponds to a highly ordered VL in the B phase, resulting in a diffracted beam that is better collimated than the incident one. A similar narrowing does not occur for the azimuthal width of the Bragg peaks in the detector plane due to the SANS scattering geometry15. Rather, the azimuthal width is found to agree with the incident-beam divergence for all measurements, and the elliptical (rather than circular) shape of the VL Bragg peaks does not imply orientational disordering of the VL. The disordering at high fields seen in ΔQ0 is also reflected in the peak scattered intensity (IQ) (Fig. 4b). Here the line 1.5 T the measured intensity falls below the extrapolated fit, indicating a broadening of the Bragg peaks in the direction perpendicular to the detector plane (normal to ΔQ0).

We interpret the three features in the SANS data occurring at 1.5 T, namely the stabilization of the splitting angle, the increasing radial Bragg peak width and the accelerating decrease of the
scattered intensity, as signatures of the B–C transition. Note that disorder in the VL is also seen at low field in both $\Delta Q_R$ and the scattered intensity. This is due to a weakening of the vortex–vortex interactions at low vortex densities\(^{31}\), and is not associated with a change in the superconducting order parameter.

To ensure that the SANS results were not affected by a systematic asymmetry of the experimental set-up, measurements were carried out using alternating directions of the C-phase/normal-state field; that is, $++/−−$ and $−−/++$. Effects due to vortex motion can likewise be excluded, as damped field oscillations were applied before each SANS measurement designed to produce identical oscillations in the vortex density for the field-reduced and field-reversed VLs.

As evident from Fig. 3 this yielded consistent results, eliminating extrinsic effects as the cause for the difference between the splittings of the field-reduced and field-reversed protocols. Furthermore, results obtained using instruments at the two different neutron scattering facilities (ORNL and ILL) are in excellent agreement.

Finally, the measurements are not affected by vortex pinning in any significant manner. Had pinning played a role, it would be most evident at low fields\(^{32}\), in contradiction with the data in Fig. 3. The absence of pinning effects is also directly evident from a comparison of the vortex density for the two different field histories. This is obtained from the magnitude of the VL scattering vector $Q$, from which it is possible to determine the magnetic induction $B$ (see Supplementary Information). As shown in Extended Data Fig. 2, there is perfect agreement within measurement accuracy between $B$ and the applied field, $H$, for both the field-reduced and the field-reversed cases. This agreement also confirms the triangular symmetry of the VL made up of singly quantized vortices throughout the entire measured field range. Therefore, our data do not support theoretical predictions of doubly quantized vortices\(^{33–35}\) based on $E_u$ pairing symmetry, or structural distortions/transitions within the B phase or associated with the B–C phase transition\(^{36–38}\) for $H \parallel c$.

**Fig. 2 | VL diffraction patterns.** a–d, Diffraction patterns following a field reduction at four different field magnitudes. The crystallographic directions within the scattering plane are indicated in a and the VL domain splitting ($\omega$) is indicated in c. The measurements were performed at a single angular setting, satisfying the Bragg condition for VL reflections at the top of the detector. Zero-field background scattering is subtracted, and the detector centre near $Q=0$ is masked off. e, f, Diffraction patterns for 1.1 T, following a field reduction and field reversal, respectively, for the region of reciprocal space indicated in c, g. The difference between field-reduced and field-reversed diffraction patterns at 1.1 T. The blue (red) colours indicate negative (positive) values. The regions of reciprocal space relevant for the subtraction are indicated by the sector boxes in e–g.

**Fig. 3 | The field dependence of the VL configuration.** a, The diffraction pattern split angle, $\omega$, determined from the VL diffraction patterns. b, The split difference, $\Delta \omega$. The lines are fits to an empirical formula as described in the Supplementary Information. The error bars show the standard deviation.
The B–C phase transition determined from the onset of VL disordering. a, The radial width (full-width at half-maximum) of the Bragg peaks, $\Delta Q_b$. The solid line ($1.7 \times 10^{-3} \text{nm}^{-1}$) corresponds to the incident-beam divergence and the dashed line is a guide to the eye. The inset indicates $\Delta Q_b$ in the detector plane. b, A fit to $l_D \times Q$ using an extended London model, as discussed in the text. The fit is restricted to fields in the range 0.8–1.5 T and uses a fixed value of the penetration depth (680 nm). The error bars show the standard deviation.

Discussion

The structure of the order parameter in the vortex core depends sensitively on the symmetry of the parent superconducting state in the host material, making vortices an ideal probe of chiral superconductors\textsuperscript{31}. The VL symmetry and its orientation relative to the crystalline axes are determined by the anisotropy of the screening current in the plane perpendicular to the applied field, and the effect of this anisotropy on the vortex–vortex interactions. The anisotropy of the screening current can originate from several sources\textsuperscript{10}, particularly the Fermi velocity of the parent metallic state\textsuperscript{39} and the superconducting order parameter\textsuperscript{1,22,23}.

For unconventional superconductors, which spontaneously break the symmetry of the Fermi surface, broken orbital rotational symmetry can lead to an anisotropy of the supercurrents that competes with the anisotropy arising from the Fermi velocities. This is the situation in UPt\textsubscript{3}. Here the dominant anisotropy in the A phase arises from the order parameter, which exhibits maximal orbital rotational symmetry breaking with $|\Delta_L(p)| = \Delta_L |\hat{p}_x \hat{p}_y \hat{p}_z|$, for $E_{a}$ pairing symmetry, and leads to the complex SANS diffraction patterns reported by Huxley and others\textsuperscript{27,39}. However, even the B phase exhibits moderate in-plane gap anisotropy in momentum space generated by the in-plane SBF, with $|\Delta_b(p)| = \Delta_b(T) \left(1 - e^{-|\hat{p}_x^2 - \hat{p}_y^2|}|\hat{p}_z|\right)$, where $e \ll \Delta_T / T \approx 10\%$ is the magnitude of the SBF measured in terms of the zero-field splitting of the superconducting transition, $\Delta_T = T_c - T_{c1}$ (ref. 13) (see Supplementary Information). The orientation of the SBF is perpendicular to the $c$ axis and independent of $\mathbf{H} \parallel \mathbf{c}$. The anisotropy of the B-phase gap is tetragonal, which leads to a weak four-fold anisotropy of the screening currents in the London region of each vortex. Notably, the B-phase gap anisotropy dominates the hexagonal anisotropy of the in-plane Fermi velocity, as the latter is of the order of 1% on the basis of measurements of the in-plane anisotropy of $H_c$ (refs. 13,26).

We make three important observations based on our experimental results. The first is that in the low-field region the gap distortion of the parent B phase leads to the rotation of the hexagonal VL, which increases with field as the density of vortices increases. This provides a qualitative explanation for the increase in the domain splitting up to fields of order $H \approx 0.8$ T.

The second observation is that the non-monotonic field dependence of the VL rotation shown in Fig. 3a, onsetting at fields of the order of half the upper critical field, implies a change in the anisotropy of the vortex currents as the vortices become closely packed. This reflects a change in the anisotropy of the order parameter and current density near the vortex cores. Further evidence that the non-monotonic field dependence is due to the influence of the vortex-core structure is supported by the third observation.

The third observation is an asymmetry of the VL rotation for the two different field histories, namely a field reduction compared to a field reversal. This is the signature that the parent B phase breaks time-reversal symmetry. Specifically, for superconductors that break time-reversal symmetry, vortices possess an internal degree of freedom associated with the phase winding of the vortex core, in addition to the global phase winding determined by the direction of the applied magnetic field $n \in [0, \pm 1, \pm 2, \ldots]$. The phase winding of the vortex core differs from the global phase winding by the topological winding number of the chirality $\pi$, which increases with field as the density of vortices increases. This reflects a change in the anisotropy of the vortex-core structure supported by the third observation.
from the absence of two-fold distortion of the vortex structure in our SANS measurements (see Supplementary Information). The discussion above was based on the $J_2$ model by Sauls; however, our main conclusions are valid for a broken time-reversal ground state belonging to any of the $E$ representations.

**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/s41567-020-0822-z.

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Methods
SANS experiments were performed at the GP-SANS beam line at the High Flux Isotope Reactor at ORNL and at the D33 beam line at ILL. Preliminary, lower-resolution measurements were carried out at the SANS-I and SANS-II beam lines at the Paul Scherrer Institute.

SANS studies of the VL rely on the periodic field modulation due to vortices\(^4\), with a scattered intensity \(\propto \lambda^{-4}\). For UPt\(_3\), the large in-plane penetration depth \(\lambda_{ab} = 680 \text{ nm (ref. } 20)\) necessitates a large sample volume. We used the same 15-g, high-quality (residual resistivity ratio > 600) UPt\(_3\) single crystal as in previous experiments\(^5\). The resistive transition temperature is \(T_{cR} = 560 \pm 2 \text{ mK}\), and from specific heat, \(T_{cS} = 523 \text{ mK}\). The width of the transition from resistivity is \(\Delta T_{c} = 10 \text{ mK}\) and from specific heat, \(\Delta T_{c} = 15 \text{ mK}\), performed on three samples taken from throughout the crystal sample in this work (resistivity) or from crystals with the same residual resistivity ratio (specific heat). The width of the A phase at zero field from specific heat is 54 mK. The long, rod-like crystal was cut into two pieces that were co-aligned and fixed with silver epoxy (EPOTEK E4110) to a copper cold finger. The sample assembly was mounted onto the mixing chamber of a dilution refrigerator and placed inside a superconducting magnet, oriented with the crystalline \(c\) axis vertical and the \(ab\) axis horizontal along the magnetic field and neutron beam.

Measurements were performed at base temperature \(T \approx 50 - 65 \text{ mK}\) and with fields between 0.1 T and 1.7 T. To eliminate effects of eddy-current heating, the field changes were paused at 0.1 T from the target field and the sample was allowed to thermalize. Once a stable sample temperature was achieved, the field was driven to the final value. Before each SANS measurement, a damped field oscillation with an initial amplitude of 20 mT was applied to achieve a homogeneous vortex density and a well-ordered VL. Periodically during the SANS measurements, a 5-mT field oscillation was performed around the measurement field to maintain an ordered VL. All field oscillations finished by a reduction of the applied field magnitude, corresponding to a decrease of the vortex density.

All SANS measurements were carried out in a ‘rocked on’ configuration, satisfying the Bragg condition for VL peaks at the top of the two-dimensional position-sensitive detector\(^6\). Measurements of full rocking curves, where the VL peak intensities are recorded as they are being rotated through the Bragg condition, would greatly exceed the available SANS beam time and were therefore not feasible. Background measurements, obtained either in zero field or above \(H_{c2}\), were subtracted from both the field-reduction and field-reversal data. A splitting of the VL Bragg peaks is also observed with either of the two halves of the crystal illuminated by the neutron beam, which shows that the presence of two VL domain orientations is an intrinsic property of UPt\(_3\). The field-reduced and field-reversed data used in the subtraction in Fig. 2g were normalized independently for the left and right Bragg peaks, to compensate for slight variations in the relative VL domain population.

Data availability
The SANS data obtained at ILL that support the findings of this study are available in the ILL repository\(^7\). All SANS data were analysed using the GRASP software package\(^8\). The data represented in Figs. 3 and 4 and Extended Data Fig. 2 are available as source data. All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions
W.J.G., W.P.H., J.A.S. and M.R.E. conceived the experiment. W.J.G. grew the crystals. K.E.A., W.J.G., S.J.K., W.P.H. and M.R.E. performed the SANS experiments with assistance from L.D.-S., C.D.D., J.G., G.N. and U.G. W.P.H., J.A.S. and M.R.E. wrote the paper with input from all authors.

Competing interests
The authors declare no competing interests.

Additional information
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Extended Data Fig. 1 | Vortex lattice diffraction patterns. a-f, Vortex lattice diffraction pattern for a range of different field magnitudes following a field reduction. g-l, Same, but following a field reversal. The VL domain splitting ($\omega$) is indicated in each panel. Measurements were performed at a single angular setting, satisfying the Bragg condition for VL reflections at the top of the detector. Zero field background scattering is subtracted, and the detector center near $Q = 0$ is masked off.
Extended Data Fig. 2 | Vortex density determined from the magnitude of the VL scattering vectors. a, Magnetic induction vs applied magnetic field. The inset shows VL Bragg peaks, their corresponding scattering vectors ($Q_{ij}$), and the opening angle ($\beta$). Peaks associated with the two different VL domain orientations are indicated by full and dashed lines respectively. Only Bragg peaks indicated by solid symbols were imaged by SANS. The line is a linear fit to the data. b, Ratio of magnetic induction to applied field ($B/H$). The center and width of the gray bar indicates the average and standard deviation of the values for fields $\geq 0.4$ T.