Chiral superconductors are novel topological materials with finite angular momentum Cooper pairs circulating around a unique chiral axis, thereby spontaneously breaking time-reversal symmetry. They are rather scarce and usually feature triplet pairing: a canonical example is the chiral $p$-wave state realized in the $A$-phase of superfluid $\text{He}_3$. Chiral triplet superconductors are, however, topologically fragile with the corresponding gapless boundary modes only weakly protected against symmetry-preserving perturbations in contrast to their singlet counterparts. Using muon spin relaxation measurements, here we report that the weakly correlated pnictide compound $\text{LaPt}_3\text{P}$ has the two key features of a chiral superconductor: spontaneous magnetic fields inside the superconducting state indicating broken time-reversal symmetry and low temperature linear behaviour in the superfluid density indicating line nodes in the order parameter. Using symmetry analysis, first principles band structure calculation and mean-field theory, we unambiguously establish that the superconducting ground state of $\text{LaPt}_3\text{P}$ is a chiral $d$-wave singlet.
Zero-field (ZF), longitudinal-field (LF) and transverse-field (TF) μSR measurements were performed on these samples at two different muon facilities: sample-A in the MUSR spectrometer at the ISIS Pulsed Neutron and Muon Source, UK, and sample-B in the LTF spectrometer at the Paul Scherrer Institut (PSI), Switzerland.

**ZF-μSR results.** ZF-μSR measurements reveal spontaneous magnetic fields arising just below \( T_c = 1.1 \) K (example characterization is shown by the zero-field-cooled magnetic susceptibility (χ) data for sample-B on the right axis of Fig. 1b) associated with a TRS-breaking superconducting state in both samples of LaPt3P, performed on different instruments. Figure 1a shows representative ZF-μSR time spectra of LaPt3P collected at 75 mK (superconducting state) and at 1.5 K (normal state) on sample-A at ISIS. The data below \( T_c \) show a clear increase in muon-spin relaxation rate compared to the data collected in the normal state. To unravel the origin of the spontaneous magnetism at low temperature, we collected ZF-μSR time spectra over a range of temperatures across \( T_c \) and extracted temperature dependence of the muon-spin relaxation rate by fitting the data with a Gaussian Kubo-Toyabe relaxation function \( G(t) \) multiplied by an exponential decay:

\[
A(t) = A(0)G(t)\exp(-\lambda_{ZF}t) + A_{bg} \tag{1}
\]

where, \( A(0) \) and \( A_{bg} \) are the initial and background asymmetries of the ZF-μSR time spectra, respectively. \( G(t) = \frac{1}{\lambda_0} + \frac{1}{\lambda_0} (1 - \sigma_{ZF}^2) \exp(-\sigma_{ZF}^2 t^2) \), \( \sigma_{ZF} \) and \( \lambda_{ZF} \) represent the muon-spin relaxation rates originating from the presence of nuclear and electronic moments in the sample, respectively. The signal-to-background ratio \( A(0)/A_{bg} = 0.40 \) (≈0.52) for sample-A
mixed state of LaPt$_3$P. The spectra are analyzed using the distribution of the vortex lattice, formed in the superconducting state in LaPt$_3$P. From the change in slope at $T = T_c$, which keeps on increasing with further lowering of temperature. Such an increase in $s(T)$ just below $T_c$ indicates that the sample is in the superconducting mixed state and the formation of vortex lattice has created an inhomogeneous field distribution at the muon sites. The internal fields felt by the muons show a diamagnetic shift in the superconducting state of LaPt$_3$P, a clear signature of bulk superconductivity in this material. The decrease in the internal fields with decreasing temperature below $T_c$ is an indication of a singlet superconducting ground state.

The true contribution of the vortex lattice field distribution to the relaxation rate $\sigma_{sc}$ can be estimated as $\sigma_{sc} = (\sigma^2 - \sigma_{nm}^2)^{1/2}$, where $\sigma_{nm} = 0.1459(4)\, \mu s^{-1}$ is the nuclear magnetic dipolar contribution assumed to be temperature independent and was determined from the high-temperature fits. Within the Ginzburg-Landau theory of the vortex state, $\sigma_{sc}$ is related to the London penetration depth $\lambda$ of a SC with high upper critical field by the Brandt equation:\supercite{Brandt:1975}

$$\frac{\rho_{sc}(T)}{\rho_\mu} = \frac{\Phi_0}{\lambda(T)^2},$$

where $\Phi_0 = 2.068 \times 10^{-15}$ Wb is the flux quantum. The superfluid density $\rho \propto \lambda^{-2}$. Figure 3 shows the temperature dependence of $\rho$ normalized by its zero-temperature value $\rho_0$ for LaPt$_3$P. It clearly varies with temperature down to the lowest temperature 70 mK and shows a linear increase below $T_c/3$. This non-constant low temperature behaviour is a signature of nodes in the superconducting gap.

The pairing symmetry of LaPt$_3$P can be understood by analysing the superfluid density data using different models of the gap function $\Delta(k)$. For a given pairing model, we compute the superfluid density ($\rho$) as

$$\rho = 1 + 2 \left\langle \int_0^\infty \frac{E}{\sqrt{E^2 - \Delta_k(T)^2}} \frac{\partial f}{\partial E} dE \right\rangle_{FS}.$$  

Fig. 2 Superconducting properties of LaPt$_3$P by TF-$\mu$SR measurements. TF-$\mu$SR time spectra of LaPt$_3$P collected at a 1.3 K and b 70 mK for sample-A in a transverse field of 10 mT. The solid lines are the fits to the data using Eq. (2). c The temperature dependence of the extracted $\sigma$ (left panel) and internal field (right panel) of sample-A. The error bars show the standard deviations in the TF-$\mu$SR measurements.

Fig. 3 Evidence of chiral $d$-wave superconductivity in LaPt$_3$P. Superfluid density ($\rho$) of LaPt$_3$P as a function of temperature normalized by its zero-temperature value $\rho_0$. The solid lines are fits to the data using different models of gap symmetry. Inset shows the schematic representation of the nodes of the chiral $d$-wave state. The error bars show the standard deviations in the TF-$\mu$SR measurements in the respective instruments.
Here, $f = 1/(1 + e^{\frac{x}{T}})$ is the Fermi function and $\langle \epsilon \rangle_{FS}$ represents an average over the Fermi surface (assumed to be spherical). We take $\Delta_{k}(T) = \Delta_{m}(T)g(k)$ where we assume a universal temperature dependence $\Delta_{m}(T) = \Delta_{0}T \tanh \left[ 1.82 \{ 0.1018(T_c / T - 1) \}^{0.51} \right]$ and the function $g(k)$ contains its angular dependence. We use three different pairing models: s-wave (single uniform superconducting gap), p-wave (two point nodes at the two poles) and chiral d-wave (two point nodes at a line and a pole at the equator as shown in the inset of Fig. 3). The fitting parameters are given in the Supplementary Table 2. We note from Fig. 3 that both the s-wave and the p-wave models lead to saturation in $\rho$ at low temperatures, which is clearly not the case for LaPt$_3$P and the chiral d-wave model gives an excellent fit down to the lowest temperature. Nodal SCs are rare since the SC can gain condensation energy by eliminating nodes in the gap. Thus the simultaneous observation of nodal and TRS-breaking superconductivity makes LaPt$_3$P a unique material.

**Discussion**

We investigate the normal state properties of LaPt$_3$P by a detailed band structure calculation using density functional theory within the generalized gradient approximation consistent with previous studies\textsuperscript{15,22}. LaPt$_3$P is centrosymmetric with a paramagnetic ground state. LaPt$_3$P is centrosymmetric with a paramagnetic ground state because of the presence of strong SOC due to centrosymmetry and SOC only gives a more sharply peaked curvature at the Fermi surface. This is consistent with the experimental observation of the Fermi surface sheets neglecting interband pairing. We note from Fig. 4a and Fig. 4b that there are two important Fermi surface sheets in LaPt$_3$P, with the chiral d-wave state having the two point nodes on one of the Fermi surface sheets and a line node on the other. Thus LaPt$_3$P is one of the rare unconventional SCs for which we can unambiguously identify the superconducting order parameter.

The preceding discussion assuming a generic Fermi surface can be adapted for the case of the inherently multi-band material LaPt$_3$P by considering the momentum dependence of the gap on the Fermi surfaces sheets neglecting interband pairing. We note from Fig. 4a that the 3p orbitals of Pt and the 3d orbitals of P. LaPt$_3$P has a non-symmetric space group P4/mmm (No. 129) with three point group D$_{4d}$. From the group theoretical classification of the SOC order parameters within the Ginzburg-Landau theory\textsuperscript{9,24}, the only possible superconducting instabilities with strong SOC, which can break TRS spontaneously at $T_c$, correspond to the two 2D irreducible representations, $E_g$ and $E_{u\alpha}$, of D$_{4d}$. Non-symmetric symmetries can give rise to additional symmetry-required nodes on the Brillouin zone boundaries along the high symmetry directions. The non-symmetric symmetries of LaPt$_3$P, however, can only generate additional point nodes for the $E_g$ order parameter but no additional nodes for the $E_u$ case\textsuperscript{25}. The superconducting ground state in the $E_g$ channel is a pseudospin chiral d-wave singlet state with gap function $\Delta(k) = \Delta_0 \kappa(k_x + ik_y)$ where $\Delta_0$ is a complex amplitude independent of $k$. The $E_u$ order parameter is a pseudospin nonunitary chiral p-wave triplet state with d-vector $\mathbf{d}(k) = [c_1 k_x, i c_1 k_y, c_2 (k_x + ik_y)]$ where $c_1$ and $c_2$ are material dependent real constants independent of $k$.

We compute the quasi-particle excitation spectrum for the two TRS-breaking states on a generic single-band spherical Fermi surface using the Bogoliubov-de Gennes mean-field theory\textsuperscript{19,24}. The chiral d-wave singlet state leads to an energy gap given by $|\Delta_0||k_z|(k^2_x + k^2_y)^{1/2}$. It has a line node at the “equator” for $k_x = 0$ and two point nodes at the “north” and “south” poles (shown in Fig. 4a). The low temperature thermodynamic properties are, however, dominated by the line node because of its larger low energy DOS than the point nodes. The triplet state has an energy gap given by $|g(k_x, k_y) + 2|c_1||k_z|f(k_x, k_y, k_z)|c_1 k_z|^1/2$ where $f(k_x, k_y, k_z) = c_2^2(k^2_x + k^2_y)$. It has only two point nodes at the two poles and no line nodes. Thus, the low temperature linear behaviour of the superfluid density of LaPt$_3$P shown in Fig. 3 is possible in the chiral d-wave state with a line node in contrast to the triplet state with only point nodes, which will give a quadratic behaviour and saturation at low temperatures.

The preceding discussion assuming a generic Fermi surface can be adapted for the case of the inherently multi-band material LaPt$_3$P by considering the momentum dependence of the gap on the Fermi surfaces sheets neglecting interband pairing. We note from Fig. 4a and Fig. 4b that there are two important Fermi surface sheets in LaPt$_3$P, with the chiral d-wave state having the two point nodes on one of the Fermi surface sheets and a line node on the other. Thus LaPt$_3$P is one of the rare unconventional SCs for which we can unambiguously identify the superconducting order parameter.

The severe constraints on the possible pairing states as a result of the unique properties of LaPt$_3$P lead us to expect that our experimental observations will be consistent only with a chiral d-wave like order parameter belonging to the $E_g$ channel even after considering pairing between bands in a multi-orbital picture\textsuperscript{10}. It is also intriguing to think about the possible pairing mechanism giving rise to the chiral d-wave state in this material, which has a weakly correlated normal state, weak electron–phonon coupling and no spin fluctuations\textsuperscript{13,16}. These issues will be taken up in future investigations.

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**Fig. 4 Properties of the normal and superconducting states of LaPt$_3$P.** Projections of the four Fermi surfaces of LaPt$_3$P with SOC on the $y$–$z$ plane in a and $x$–$y$ plane in b. The thickness of the lines are proportional to the contribution of the Fermi surfaces to the DOS at the Fermi level (green—10.3%, blue—43.4%, orange—40.0% and magenta—6.3%). The point nodes of the chiral d-wave gap are shown by red dots in a and the line node resides on the $x$–$y$ plane in b. c Schematic view of the Majorana Fermi arc and the zero-energy Majorana flat band corresponding to the two Weyl point nodes and the line node respectively on the respective surface Brillouin zones (BZs) assuming a spherical Fermi surface. d Berry curvature $\mathbf{F}(k)$ corresponding to the two Weyl nodes on the $x$–$z$ plane. Arrows show the direction of $\mathbf{F}(k)$ and the colour scale shows its magnitude $= \frac{1}{2} \arctan (|\mathbf{F}(k)|)$. $\Delta_0 = 0.5 \mu$ was chosen for clarity while a more realistic weak-coupling limit $\Delta_0 < \mu$ gives a more sharply peaked curvature at the Fermi surface.
The topological properties of the chiral $d$-wave state of LaPt$_3$P are naturally discussed considering a generic single-band spherical Fermi surface. The chemical potential is most naturally discussed considering a generic single-band junctions. However, topological protection of the nodes also ensures stability against multi-band effects assuming interband pairing strengths to be small. The effective angular momentum of the Cooper pairs is $l_z = +1$ (in units of $h$) with respect to the chiral $c$-axis. The equatorial line node acts as a vortex loop in momentum space, and is topologically protected by a 1D winding number $w(k_x, k_y) = 1$ for $k_z < k_F$ and $w = 0$ otherwise. The non-trivial topology of the line node leads to two-fold degenerate zero-energy Majorana bound states in a flat band on the $(0,0,1)$ surface BZ as shown in Fig. 4c. As a result, there is a diverging zero-energy DOS leading to a zero-bias conductance peak (which can be really sharp) measurable in STM. This inversion symmetry protected line node is extra stable due to even parity SC. The point nodes on the other hand are Weyl nodes and are impossible to gap out by symmetry-preserving perturbations. They act as a monopole and an anti-monopole of Berry flux as shown in Fig. 4d and are characterized by a $k_z$-dependent topological invariant, the sliced Chern number $C(k_z) = L_z$ for $|k_z| < k_F$ with $k_z = 0$ and $w = 0$ otherwise (see Supplementary Note 6 for details). As a result, the $(1,0,0)$ and $(0,1,0)$ surface BZs each have a Majorana Fermi arc, which can be probed by STM as shown in Fig. 4c. There are two-fold degenerate chiral surface states with linear dispersion carrying surface currents leading to local magnetisation that can be detected using SQUID magnetometry. One of the key signatures of chiral edge states is the anomalous thermal Hall effect (ATHE), which depends on the length of the Fermi arc in this case. Impurities in the bulk can, however, increase the ATHE signal by orders of magnitude. The length of the Fermi arc in this case. Impurities in the bulk can, however, increase the ATHE signal by orders of magnitude.

### Methods

**$\mu$SR technique.** $\mu$SR is a very sensitive microscopic probe to detect the local-field distribution within a material. This technique has been widely used to search for very weak fields (of the order of a fraction of a gauss) arising spontaneously in the superconducting state of TRS-breaking SCs. The other great use of this technique is to measure the value and temperature dependence of the London magnetic penetration depth, $\lambda$, in the vortex state of type-II SCs. It is in turn proportional to the superfluid density, which can provide direct information on the nature of the superconducting gap. Details of the $\mu$SR technique is given in Supplementary Note 3.

**Sample preparation and characterisation.** Two sets of polycrystalline samples (referred to as sample-A and sample-B) of LaPt$_3$P were synthesized at two different laboratories (Warwick, UK and PSI, Switzerland) by completely different methods. While, sample-A was synthesized by solid state reaction method, sample-B was synthesized using the cubic anvil high-pressure and high-temperature technique. Details of the sample preparation and characterisation are given in Supplementary Note 1 and 2.

**DFT calculation.** The first principles density functional theory (DFT) calculations were performed by the full potential linearized augmented plane wave method implemented in the WIEN2k package. The generalized gradient approximation with the Perdew-Burke-Ernzerhof realization was used for the exchange-correlation functional. The plane wave cut-off $K_{max}$ is given by $R_{mp} \times K_{max} \approx 8.0$. For the self-consistent calculations, the $\Gamma$-centered mesh of $15 \times 15 \times 15$ $k$-points was used.

### Data availability

All the datasets that support the findings of this study are available from the corresponding author upon reasonable request. The ISSS DOI for our MUSR source data is https://doi.org/10.5286/ISIS.E.RB1720467.

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**Author contributions**

P.K.B. conceived the project, successfully acquired the PSI and ISIS muon beamtimes and performed the μSR experiments and data analysis. S.K.G. performed the theory part of the project using band structure calculations performed by J.Z.Z. S.K.G. helped in data analysis and wrote the manuscript together with P.K.B. D.A.M. synthesized and characterized the sample from Warwick and participated in the μSR experiments at ISIS. N.D.Z. synthesized and characterized the sample from ETH. C.B. helped in performing the μSR experiments in PSI. X.X., A.D.H., G.B. and M.R.L. helped to improve the presentation of the data and the manuscript as a whole.

**Competing interests**

The authors declare no competing interests.

**Additional information**

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