We performed the first scanning tunneling spectroscopy measurements on the pyrochlore superconductor KO$_2$Os$_6$ ($T_c = 9.6$ K) in both zero magnetic field and the vortex state at several temperatures above $1.95$ K. This material presents atomically flat surfaces, yielding spatially homogeneous spectra which reveal fully-gapped superconductivity with a gap anisotropy of 30%. Measurements performed at fields of 2 and 6 T display a hexagonal Abrikosov flux line lattice. From the shape of the vortex cores, we extract a coherence length of 31–40 Å, in agreement with the value derived from the upper critical field $H_{c2}$. We observe a reduction in size of the vortex cores (and hence the coherence length) with increasing field which is consistent with the unexpectedly high and unsaturated upper critical field reported.

The discovery of superconductivity in the β-pyrochlore osmate compounds AO$_2$Os$_6$ (A = K, Rb, Cs) [1] has highlighted the question of the origin of superconductivity in classes of materials which possess geometrical frustration [2, 3]. Interest has been predominantly focused on the highest-$T_c$ compound KO$_2$Os$_6$ which presents many striking characteristics. In particular, the absence of inversion symmetry in its crystal structure [4] raises the question of its Cooper pair symmetry and the possibility of spin singlet-triplet mixing [5, 6]. Alternatively, it has also been suggested that this behavior can be explained by the peculiar topology of the Fermi surface (FS) sheets of KO$_2$Os$_6$, assuming that superconductivity occurs mainly on the closed sheet [7].

The pyrochlore osmate compound KO$_2$Os$_6$ displays a critical temperature $T_c = 9.6$ K, the largest in its class of materials (CsOs$_2$O$_6$ and RbOs$_2$O$_6$ which differ only by the nature of the alkali ion have $T_c$s of 3.3 and 6.3 K respectively). Although band structure calculations show that the K ion does not influence the density of states (DOS) at the Fermi level [7, 8], it seems to affect several key properties [9]. In particular, the first order phase transition revealed by specific heat measurements in magnetic fields at the temperature $T_p \approx 7.5$ K has been ascribed to a “freezing” of its rattling motion [10].

The peculiar behavior of KO$_2$Os$_6$ is demonstrated by its upper critical magnetic field $H_{c2}$, whose temperature dependence is linear down to sub-Kelvin temperatures and whose amplitude is above the Clogston limit [10]. One possible interpretation is the occurrence of spin-triplet superconductivity driven by spin-orbit coupling [5, 6]. Alternatively, we perform AC susceptibility measurements show a very sharp superconducting transition ($\Delta T_c = 0.35$ K). Our measurements are carried out using a home-built low temperature scanning tunnel microscope featuring a compact nanopositioning stage to target the small-sized crystals. Electrochemically etched iridium tips are used for STS measurements on as-grown single crystal surfaces and the differential conductivity was measured using a standard AC lock-in technique.

The surface topography of as-grown samples (Fig. 1a) reveals atomically flat regions speckled with small corrugated islands with nanometers high whose spectroscopic characteristics are noisy and not superconducting (thus restraining our field of view for spectroscopic imaging).
The large flat regions display highly homogeneous superconducting spectra (Fig. 1a), which were perfectly reproducible over the timescale of our experiments (4 months). We have checked that the spectra obtained by varying the tunnel resistance $R_t$ all collapse onto a single curve, thus confirming true vacuum tunneling conditions. We have also verified that the numerical derivative of the tunnel current with respect to the voltage gives the same spectroscopic signature as the $dI/dV$ lock-in signal. We stress that all measurements presented in this paper are raw data.

The lack of inversion symmetry in this compound together with several experimental findings raises the question of the symmetry of the gap function. In order to clarify this point, we have fitted our data to several symmetry models, focusing on the question of the presence or absence of nodes and the amplitude of any possible gap anisotropy. We therefore considered three scenarios with an approximate angular dependence of the gap, i.e. an isotropic $s$-wave ($\Delta_0$), a $d$-wave ($\Delta \cos 2\phi$) with nodes and an “anisotropic” $s$-wave ($\Delta_0 + \Delta \sin \phi$) which has the same angular dependence as the $s-p$-wave singlet-triplet mixed state $^3$He. We do not take the real topology of the FS $^1$ into account, since it comprises two 3D Fermi sheets and is hence unlikely to have any significant effect on the gap structure. For an anisotropic gap, $\Delta(\phi)$, the quasiparticle DOS is given by $N(\omega) \propto |\text{Re}[\langle \omega + i\Gamma \rangle / \sqrt{(\omega + i\Gamma)^2 - |\Delta(\phi)|^2}]|$ where $\Gamma$ is a phenomenological scattering rate. In addition, we included broadenings due to the experimental temperature and the lock-in in our fits. The results are presented in Fig. 2a. The $d$-wave model can be rejected at this stage since its zero bias conductance (ZBC) is larger than in experiment (increasing $\Gamma$ in the model can only increase the ZBC). The differences between symmetries appear much more clearly in the second derivative spectrum ($d^2I/dV^2$, Fig. 2d) which is not surprising as it emphasizes the variations of the DOS on a small energy scale and is very sensitive to the model parameters (in contrast with the $dI/dV$ curve). The best fit is clearly given by the “anisotropic” $s$-wave model with an anisotropy of around 30%. With respect to the singlet-triplet mixed state, we note that we do not see any evidence in our data for a second coherence peak arising from spin-orbit splitting. Since the 3D nature of both sheets implies that tunneling takes place in both of them, the absence of a second peak also rules out the possibility of two different isotropic gaps on separate FS sheets. Our results would however be compatible with multiband superconductivity with two (overlapping) anisotropic gaps. Finally, we see no signature of a normal-normal tunneling channel in our junction, suggesting that all electrons involved in the tunneling process come from the superconducting condensate.

To investigate the temperature evolution of the quasiparticle DOS, we acquired tunneling conductance spectra at different temperatures between 1.95 K and 10 K (Fig. 2b). The closure of the gap at the bulk $T_c$ shows that we are probing the bulk properties of KO$_2$O$_6$. This
is further confirmed by the fact that similar spectra were also obtained on freshly cleaved surfaces. The totally flat conductance spectra at higher temperature show no support for a pseudogap in the DOS above \( T_c \), implying that the steep decrease in the \( 1/(T_c T) \) curve around 16 K in NMR data \([12]\) must have a different origin. The spectra taken between 6 and 9 K (not shown) were very noisy. This could be explained by the proximity to the first order transition at \( T_c \approx 7.5 \) K \([11]\).

The BCS coupling ratio \( 2\Delta_{\max}/k_B T_c \) inferred from our measured gaps and critical temperature is about 3.6 for the anisotropic \( s \)-wave case, a value slightly smaller than that reported from specific heat measurements \([12]\). However, we stress that STS is a direct probe of the superconducting gap. Our findings therefore lead us to the conclusion that KOs\(_2\)O\(_6\) is fully gapped with a significant anisotropy of around 30%.

We now focus on measurements performed in an applied magnetic field. In the vortex cores whose radial size is roughly given by the coherence length \( \xi \), superconductivity is suppressed leading to a drastic change in the LDOS which can be measured by STM. Our measurements were performed for two fields, 2 and 6 T, over the particularly flat region of about 60 \( \times \) 60 nm\(^2\) (Fig. 4).

Each measurement was taken at 2 K with a typical acquisition time of 40 hours.

The results are presented in Figs. 3 and 4. The vortex maps (insets of Fig. 3 and Figs. 4, and 1) show the ZBC normalized to the conductance at 6 meV. Fig. 3 displays the spectra taken along traces passing through vortex cores for each of the two fields considered. The suppression of superconductivity and its effect on the conductance in a vortex core can clearly be seen. The vortex maps show a roughly hexagonal vortex lattice with vortex spacings \( d = 352 \pm 17 \) Å and 216 \( \pm \) 21 Å at 2 and 6 T respectively, in agreement with the spacings \( d = (2\Phi_0/\hbar\sqrt{3})^{1/2} \) expected for an Abrikosov hexagonal lattice \([22]\), i.e. 345 Å and 199 Å. We ascribe the variations in the core shapes and the deviation from a perfectly hexagonal lattice to vortex pinning. In particular, the vortex identified by the arrow in Fig. 4 appears to be split. We attribute this to the vortex oscillating between two pinning centers during the measurement, a situation which has been seen in other compounds \([22]\). One should also note that the islands (surface defects) at the border of the measurement area (Fig. 1) could influence the vortex core shapes and positions.

In order to estimate the coherence length \( \xi \) from our measurements, we now consider the spatial dependence of the ZBC. Due to the proximity of the vortices, we model the LDOS as a superposition of isolated vortex LDOS which can be expressed as \( N(\omega, r) = \sum_n |u_n(r)|^2 \delta(\omega - E_n) + |u_n(r)|^2 \delta(\omega + E_n) \), where \( \psi_n(r) = (u_n(r), v_n(r)) \) is the wave function of the \( n \)th vortex core state and \( E_n \) its energy. An approximate solution for the isolated vortex was given long ago \([22]\) in which the radial dependence of each \( \psi_n(r) \) consists of a rapidly oscillating \( n \)-dependent Bessel function multiplied by a \( \cosh^{-1/2}(r/\xi) \) envelope common to all states. We therefore construct a phenomenological model for our 2D ZBC maps, \( \sigma(\omega = 0, r) \propto N(\omega = 0, r) \), by retaining the slowly varying parts of the wave functions alone, i.e.

\[
\sigma(\omega = 0, r) = \sigma_0 + \Lambda \sum_i \left( \cosh \left( \frac{r - r_i}{\xi} \right) \right)^{-2/3}
\]

where \( \sigma_0 = 0.13 \) is the residual normalized conductance at zero bias in the absence of field (Fig. 3), \( \Lambda \) a scaling factor, \( \xi \) the coherence length and the sum runs over all the vortices with positions \( r_i \) in the map. Using \( \sigma_0 \), we fitted \( r_i \) and \( \xi \) over the entire map for each field, thus considering all imaged vortices to determine \( \xi \).

The results from the 2D fits are presented in Fig. 4a and d in map format and along traces selected to pass through vortex cores in Fig. 4b and f. The traces help to visualize the spatial extent of the vortices and assess the (extremely high) quality of the 2D fits. We first observe that the normalized ZBC between the vortices is slightly enhanced at \( H = 2 \) T but increases strongly at \( H = 6 \) T with respect to the value at zero-field (Fig. 2), indicating a significant core overlap. From our data taken at \( T = 2 \) K, we obtain \( \xi = 35 \pm 3 \) Å and 45 \( \pm \) 7 Å at \( H = 6 \) and 2 T respectively (the uncertainties are estimated from the spread of the results obtained on several maps: two for 6 T and three for 2 T). Using Ginzburg-Landau theory, we extrapolate the corresponding \( T = 0 \) values as \( \xi = 31 \pm 3 \) and 40 \( \pm \) 6 Å respectively, consistent with the value derived from \( H_{c2} \).

Furthermore, our results indicate that the vortex size decreases with increasing field and, although at the limit of the error bars, we believe this trend to be genuine. In addition, this finding is consistent with the abnormally large \( H_{c2} \): if the vortices become smaller as the field increases, the material can accommodate more vortices before the breakdown
of superconductivity, leading to a higher upper critical field. This correlates with the observed temperature dependence of the upper critical field.

We find that the spectra at the vortex centers are flat for both fields (Fig. 3), showing the presence of localized quasiparticle states in the vortex cores. However, our spectra show no excess spectral weight at or close to zero bias and thus no ZBCP which is the generally expected signature of vortex core states. The absence of a ZBCP is at first glance striking considering the large mean free path $\ell \approx 200 \text{ nm} \gg \xi$ in KOs$_2$O$_6$ [12]. In fact, this absence is common to many non-cuprate superconductors, the only known exceptions being 2H-NbSe$_2$ [17, 23, 26] and YNi$_2$B$_2$C [27]. Although no definitive theory currently exists to explain such an absence, a possible explanation assumes that the scattering rate is strongly enhanced in the vortex cores. This interpretation is supported by our numerical solutions of the Bogoliubov-de Gennes equations for a single vortex with an $r$-dependent scattering rate $\Gamma$. Furthermore, these simulations show a radial dependence of the LDOS which is fully consistent with [1].

In conclusion, we have presented the first scanning tunneling spectroscopic measurements on superconducting KOs$_2$O$_6$. The fitted spectra demonstrate that KOs$_2$O$_6$ is a fully-gapped superconductor with an anisotropy of around 30%, possibly resulting from a $s$-$p$ singlet-triplet mixed state allowed by the lack of inversion symmetry. We have imaged hexagonal vortex lattices matching Abrikosov’s prediction for 2 and 6 T fields. Using Caroli-de Gennes-Matricon theory we extract a field-dependent coherence length of 31–40 Å, in good agreement with the thermodynamic estimate from $H_c2$. The absence of a zero bias conductance peak, the apparent field dependence of $\xi$ and the precise radial dependence of the LDOS all call for deeper exploration.

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