Abstract

We performed the first scanning tunneling spectroscopy measurements on the pyrochlore superconductor KOs2O6 (Tc=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 Hc2. 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.

Reference


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Scanning Tunneling Spectroscopy in the Superconducting State and Vortex Cores of the β-Pyrochlore KOs₂O₆

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We performed the first scanning tunneling spectroscopy measurements on the pyrochlore superconductor KOs₂O₆ ($T_c = 9.6$ 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 peculiar behavior of KOs₂O₆ is illustrated 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 [16]. One possible interpretation is the occurrence of spin-triplet superconductivity driven by spin-orbit coupling [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 KOs₂O₆, assuming that superconductivity occurs mainly on the closed sheet [16].

The discovery of superconductivity in the β-pyrochlore osmate compounds AO₅Os₂O₆ ($A = K$, 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 KOs₂O₆ 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].

The pyrochlore osmate compound KOs₂O₆ displays a critical temperature $T_c = 9.6$ K, the largest in its class of materials (CsOs₂O₆ and RbOs₂O₆ which differ only by the nature of the alkali ion have $T_s$ 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 $T_p = 7.5$ K has been ascribed to a “freezing” of its rattling motion [10]. The negative curvature of the resistivity as a function of temperature [11] has also been attributed to an unusual electron-phonon scattering [12]. Specific heat measurements [13] suggest the coexistence of strong electron correlations and strong electron-phonon coupling, two generally antagonistic phenomena with respect to the superconducting pairing symmetry. The nature of the symmetry remains a controversial subject in the literature. NMR [14] and µSR [15] data suggest anisotropic gap functions with nodes whereas thermal conductivity experiments [12] favor a fully gapped state.

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The understanding of the physics of this compound would greatly benefit from a detailed knowledge of the local density of states (LDOS). Scanning tunneling spectroscopy (STS) is an ideal tool for this, particularly since it allows one to map the vortices in real space and also access the normal state below $T_c$ by probing their cores [17–20]. In this Letter we present a detailed STS study of KOs₂O₆ single crystals, including vortex imaging.

The KOs₂O₆ single crystals were grown from Os and KO₂ in oxygen-filled quartz ampoules. Their dimensions are around $0.3 \times 0.3 \times 0.3$ mm$^3$. The details of their chemical properties as well as their growth conditions can be found in Ref. [4]. A very sharp superconducting transition ($\Delta T_c = 0.35$ K) is shown by ac susceptibility measurements. Our measurements are carried out using a home-built low temperature scanning tunneling microscope featuring a compact nanopositioning stage [21] to target the small-sized crystals. 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 (with $V_{rms} = 0.2$ mV).

The surface topography of the three as-grown samples studied here [Fig. 1(a)] reveals atomically flat regions speckled with small corrugated islands a few Angströms 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. 1(b)], 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 Letter 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. 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 thus considered three scenarios where the 3D angular dependence of the gap was approximated with one angle \([22]\), i.e., fully isotropic (\( \Delta_0 \)), fourfold with nodes (\( \Delta \cos 2\theta \) and twofold anisotropic (\( \Delta_0 + \Delta \sin \theta \)) which has the same angular dependence as the \( s\)-\( p \)-wave singlet-triplet mixed state \([6]\). We do not take the real topology of the FS \([7]\) 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, \( \Delta(\theta) \), the quasiparticle DOS is given by \( N(\omega) \propto |\text{Re}[(\omega + i\Gamma)/\sqrt{(\omega + i\Gamma)^2 - [\Delta(\theta)]^2}]| \) where \( \omega \) is the quasiparticle energy and \( \Gamma \) a phenomenological scattering rate. In addition, broadenings due to the experimental temperature and the lock-in were included in all our model calculations. For each symmetry, the optimal parameters are determined by performing a least-squares fit on both \( dI/dV \) and \( d^2I/dV^2 \) spectra (Fig. 2). The case with nodes 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 [Fig. 2(d)] 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 anisotropic model gives the best agreement for an anisotropy of around 30% as illustrated by the reduced \( \chi^2 \). 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 and 10 K [Fig. 2(a)]. The closure of the gap at the bulk \( T_c \) shows that we are probing the bulk properties of KO\(_2\)O\(_6\). Similar spectra were also obtained on freshly cleaved crystals. 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 $1/(T_2 T)$ curve around 16 K in NMR data [14] 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 $T_p \approx 7.5$ K [10].

The BCS coupling ratio $2\Delta_{\text{max}}/k_B T_c$ 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 [13]. Our findings 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. 1(a)]. Each measurement was taken at 2 K with a typical acquisition time of 40 h. The results are presented in Figs. 3 and 4. The vortex maps [insets of Figs. 3, 4(a), and 4(b)] show the ZBC normalized to the conductance at +6 meV. Figure 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 $d = (2\Phi_0/H\sqrt{3})^{1/2}$ expected for an Abrikosov hexagonal lattice [24], i.e., 345 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 [25]. One should also note that the surface defects at the border of the measurement area (Fig. 1) could influence the vortex core shapes and positions.

To estimate the coherence length $\xi$ from our measurements, we now consider the spatial dependence of the ZBC. We model the LDOS as a superposition of isolated vortex LDOS which can be expressed as $N(\omega, \mathbf{r}) = \sum_n |\psi_n(\mathbf{r})|^2 \delta(\omega - E_n) + |\psi_n(\mathbf{r})|^2 \delta(\omega + E_n)$, where $\psi_n(\mathbf{r}) = (u_n(\mathbf{r}), v_n(\mathbf{r}))$ 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 [26] in which the radial dependence of each $\psi_n(\mathbf{r})$ consists of a rapidly oscillating $n$-dependent Bessel function multiplied by a $\cosh^{-1}(r/\xi)$ envelope common to all states. We therefore construct a phenomenological model for our 2D ZBC maps, $\sigma(\omega = 0, \mathbf{r}) \propto N(\omega = 0, \mathbf{r})$, by retaining the slowly varying parts of the wave functions alone, i.e.,

$$\sigma(\omega = 0, \mathbf{r}) = \sigma_0 + \Delta \sum_n \left( \cosh \frac{|\mathbf{r} - \mathbf{r}_n|}{\xi} \right)^{(2/\pi)},$$

where $\sigma_0 = 0.13$ is the residual normalized conductance at

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{vortex_map.png}
\caption{(color). Spectroscopic traces at $T = 2$ K across vortices for a field of 2 T (a) and 6 T (b). The spectra at the vortex centers are highlighted in red. The zero-bias conductance and the locations of the traces are shown in the corresponding insets.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{vortex_profiles.png}
\caption{(color). (a),(b) Experimental ZBC maps ($T = 2$ K, $I = 0.5$ nA, $V = 15$ mV) normalized to the background conductance (at +6 mV) for 2 and 6 T, respectively, with corresponding fits (c),(d); large values (red) correspond to normal regions (i.e., vortex cores) and low values (blue) to superconducting (gapped) regions. (e),(f) Experimental ZBC profiles across vortex centers (along the paths shown on the maps) together with the corresponding profiles from the 2D fits.}
\end{figure}
consistent with the value derived from Ginzburg-Landau theory, we extrapolate the corresponding spread of the results obtained on successive maps over respectively (the uncertainties are estimated from the 2D fits).

The results from the 2D fits are presented in Figs. 4(c) and 4(d) in map format and along traces selected to pass through vortex cores in Figs. 4(e) and 4(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(c)], 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 successive maps over the same area: 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, although at the limit of the error bars, our results indicate that the vortex size decreases with increasing field. 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 $H_{c2}$.

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 zero-bias conductance peak (ZBCP) which is the generally expected signature of vortex core states. The absence of a ZBCP suggests that KO$_2$O$_6$ should be classified in the dirty limit, although estimates of the mean free path available in the literature [27], despite varying widely, seem to indicate clean limit superconductivity. This absence of a ZBCP is in fact a common phenomenon in many superconductors [18,19], the only known exceptions being 2H-NbSe$_2$ [17,28] and YNi$_2$B$_2$C [29]. Although no definitive theory currently exists to explain such an absence, one possibility is a position-dependent scattering rate which is strongly enhanced in the vortex cores. This could be modeled as being proportional to both the impurity density (scatterers) and the LDOS (scattered particles) which would smear all the peaks in the vortex cores.

In conclusion, we have presented the first scanning tunneling spectroscopic measurements on superconducting KO$_2$O$_6$. The fitted spectra demonstrate that KO$_2$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 reported lack of inversion symmetry. We have imaged hexagonal vortex lattices matching Abrikosov’s prediction for 2 and 6 T fields. Using the 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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[22] The DOS given by the true 3D gap distributions, once integrated over all angles, are generally indistinguishable from those obtained through these approximate angular dependences (see Ref. [23] for a more thorough discussion).