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Unusual upper critical field in the pyrochlore KOs$_2$O$_6$

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Abstract. We determine the temperature dependence of the upper critical field $H_{c2}$ in KOs$_2$O$_6$ from resistivity and magnetic measurements in high magnetic fields up to 50 T. By both techniques we find linear temperature dependence all the way below $T_c$ and uncommonly high $H_{c2}$ ($T \to 0$ K) $\sim 33$ T. We show that this unusual $H_{c2}$ can be understood as a consequence of paramagnetic limit $H_P$ remarkably enhanced by the broken spatial inversion symmetry proposed recently, ensuring that the pair-breaking here is executed by orbital degrees, associated with the smaller closed Fermi surfaces.

1. Introduction

Since the discovery of superconductivity in $\beta$-pyrochlore oxides AO$_2$O$_6$, nature of superconductivity in the pyrochlore lattice with relatively high transition temperature $T_c$ has been a subject of great interest. Among them, KOs$_2$O$_6$ with the highest $T_c$ (= 9.6 K) [1] exhibits several remarkable features in its physical properties. Resistivity shows strong convex temperature dependence down to $T_c$ [2], indicating strong electron-phonon scattering mechanism which may be related to the rattling motion of K ions [3]. Specific heat measurements reveal a jump $\Delta C/T_c = 185.4$ mJ K$^{-2}$ mol$^{-1}$ at $T_c$, and another anomaly at a lower temperature $T_p = 7.5$ K which is discussed in views of freezing of the rattling motion [2, 4, 5]. In addition to these, strong electron correlations appear to have important contributions in transport and thermodynamic properties of KOs$_2$O$_6$. Sommerfeld coefficient $\gamma$ is estimated as high as 70 to 110 mJ K$^{-2}$ mol$^{-1}$ [2, 4], which is largely enhanced from the band calculation value.

The coexistence of strong electron correlations that prefer anisotropic superconductivity and strong electron-phonon coupling that favors $s$-wave fully gapped superconductivity makes KOs$_2$O$_6$ very interesting system to investigate the mechanism of superconductivity. Another interest concerns the detailed crystal structure in KOs$_2$O$_6$ reported recently. Schuck et al. [6] suggested cubic $F\bar{4}3m$ structure with volume change in Os tetrahedral and O octahedral networks from ideal $\beta$-pyrochlore lattice, which lacks spacial inversion symmetry (see Fig. 1). Since in a simple picture spin triplet pairing requires time reversal symmetry and inversion symmetry, this absence of inversion center has a particular impact of $H_{c2}$ behavior.
Here we report on the temperature dependence of $H_{c2}$ in KOs$_2$O$_6$ determined from high-field resistivity and magnetic measurements by using a pulsed magnet. These two measurements provide consistent $H_{c2}(T)$, which grows linearly with decreasing temperature and reaches 33 T in the zero-temperature limit. Apparently this value is beyond the simple Pauli paramagnetic limit of 18 T, but we argue that the limit can be enhanced more than 50 T by considering the modified spin susceptibility expected in superconductivity without inversion symmetry.

2. Experimental

In this study, we used a block containing several crystals of KOs$_2$O$_6$ [2]. Resistivity was measured by using 100 kHz lock-in technique in a 60 ms pulsed magnet at National High Magnetic Field Laboratory. Magnetic fields up to $\sim 50$ T are applied along the current direction. We also measure magnetic penetration by using a tunnel diode oscillator operating at frequencies $f \sim 55$ MHz, with an LC tank circuit [7]. The sample is inserted in one of a pair of coils [see the inset of Fig. 3], and the inductance change due to the change in the penetration depth is detected by the shift of resonance frequency $\Delta f$.

3. Results and Discussion

Figure 2 shows the field dependence of resistivity $\rho$ in KOs$_2$O$_6$. $\rho(H)$ shows transition behavior from low-field superconducting (vortex) state to high-field normal state. Upper critical field $H_{c2}$ was determined from the field where $\rho(H)$ recovers fully normal-state values.

To determine $H_{c2}$ unambiguously, we also employ magnetic measurements. In Fig. 3, we plot the time derivative of the resonance frequency $df/dt$ obtained during the field pulses as a function of $H$. In this plot, anomalies due to the change in the penetration depth are clearly observed. We determine the upper critical field by the end point of the anomalies in $df/dt$, where the whole sample becomes normal.

We plot in Fig. 4 the temperature dependence of $H_{c2}$ determined by the resistivity and magnetic measurements. Two different methods give consistent $H_{c2}(T)$, indicating that the determined temperature dependence is intrinsic to KOs$_2$O$_6$. $H_{c2}(T)$ has two salient features: (1) It extrapolates to a very high value of 33 T in the zero temperature limit, which corresponds...
Figure 3. Time derivative $\frac{df}{dt}$ of the frequency shift of the tunnel diode oscillator containing the KOs$_2$O$_6$ sample. Only downfield sweeps are shown and each curve is vertically shifted for clarity.

Figure 4. Temperature dependence of the upper critical field determined by the resistivity (closed squares) and magnetic (open circles) measurements. The solid line is the expected orbital $h^*(t)$ from the conventional WHH formulation. The dotted and dashed lines are ab initio calculations for the maximum $h^*(t)$ ([111] direction).

to the coherence length $\xi(0) = 3.2$ nm. (Similarly high $H_{c2}$ was recently obtained by Ohmichi et al. [8].) (2) The temperature dependence is almost linear in $T$, without showing the saturation behavior expected in the standard WHH theory at low temperatures.

4. Discussion and Conclusions

Let us first discuss the Pauli paramagnetic limit $H_P$. Above $H_P$ the Cooper pairs are destroyed by the Zeeman splitting produced by the magnetic-field coupling to the electronic spins. This is realized when the Zeeman energy reaches the condensation energy $U_c = \frac{1}{2}N(0)\Delta^2$, where $N(0)$ is the density of states at Fermi level, and $\Delta$ is the superconducting gap; $U_c = \frac{1}{2}\left[\chi_n(T) - \chi_s(T)\right]H_P^2$.

Here $\chi_n$ is the Pauli spin susceptibility in the normal state, and $\chi_s(T)$ is the spin susceptibility in the superconducting state. In spin-singlet superconductors, $\chi_s(T)$ follows Yoshida function, and becomes zero at $T = 0$ K. In simple calculations within the weak-coupling BCS theory, this limit for KOs$_2$O$_6$ is 17.8 T, which is much lower than the observed $H_{c2}(0)$. At first glance, this would suggest spin-triplet superconductivity for which $\chi_s(T)$ remains normal-state value and hence no Pauli limit is involved. We may improve on this estimate by making use of experimental parameters for the susceptibility $\chi_n \approx 4.2 \times 10^{-4}$ emu/mol [4] and in the determination of the condensation energy $U_c$ from the specific heat jump at $T_c$. This results in a larger value $H_P \approx 31$ T, closer to the experimental value, but it may not be large enough for usual spin-singlet superconductivity.

The lack of inversion symmetry recently found in KOs$_2$O$_6$ [6] [Fig. 1] affects the electronic properties and thus $H_{c2}$ through antisymmetric spin-orbit coupling (ASOC) Hamiltonian $\alpha \sum_{\vec{k},s,s'} \vec{g}(\vec{k}) \sigma_{ss'} c^\dagger_{\vec{k}s} c_{\vec{k}s'}$, where $\alpha$ denotes the spin-orbit coupling strength, $\vec{g}$ is the Pauli matrices vector, $c^\dagger_{\vec{k}s}$ ($c_{\vec{k}s}$) creates (annihilates) an electron with momentum $\vec{k}$ and spin $s$, and $\vec{g}(\vec{k})$ is a...
dimensionless vector with \( \vec{g}(-\vec{k}) = -\vec{g}(\vec{k}) \). The ASOC modifies the spin susceptibility \( \chi_s(T) \) in the superconducting state [9, 10] even if the dominant order parameter is s-wave. For the \( F43m \) space group symmetry in KO\(_2\)O\(_6\), \( \vec{g}(\vec{k}) \) has a form
\[
\vec{g}(\vec{k}) = [k_x(k_y^2 - k_z^2), k_y(k_z^2 - k_x^2), k_z(k_x^2 - k_y^2)]/k_F^2
\]
In KO\(_2\)O\(_6\), we expect a reasonably large \( \alpha \) from the heavy Os atoms, and we obtain the value \( \chi_s(0) = (2/3)\chi_n \). This enhances the paramagnetic limit by a factor of \( \sqrt{3} \) giving \( H_P \approx 54 \) T, way beyond the observed value of \( H_{c2}(0) \). This large \( H_P \) then resides sufficiently far above the measured value \( H_{c2}(0) \approx 33 \) T.

From the above discussion we are able to conclude that \( H_P \) is enhanced and thus \( H_{c2}(T) \) is limited not by the Zeeman term but by the orbital term. Now the remaining question is about the observed linear temperature dependence. The orbital-depairing is usually described by the WHH theory, where the reduced critical field \( h^*(t) = \frac{H_{c2}(t)}{H_{c2}(1)} \) deviates from the experimental data at low temperatures. According to the band calculations [3], AOs\(_2\)O\(_6\) compounds have two kinds of Fermi surfaces; one is the connected Fermi surface with necks along the three-fold axis, and the other is the closed sheets of the Fermi surface centered on the \( \Gamma \) point. Taking these Fermi surface shapes into account, we employ \( ab\ initio \) calculation of \( h^*(t) \) following a framework provided by Kita and Arai [11]. We find that for Fermi surface anisotropy gives maximum \( H_{c2} \) in the [111] direction, which we compare with the experimental \( H_{c2} \) determined as the field where the whole sample becomes normal. The calculated results are shown in Fig. 3 by dotted line. When we ignore the connected surface and calculate \( h^*(t) \) for closed surfaces alone, we obtain almost \( T \)-linear \( h^*(t) \) without saturation at low temperatures, which is in good agreement with experiments. This result strongly suggests that the upper critical field determined by the orbital deparing without paramagnetic limiting, and that the superconductivity pairing mainly occurs in the closed Fermi surfaces. Our finding of orbitally limited \( H_{c2} \) is compatible with the fully gapped superconductivity suggested by thermal conductivity measurements [5], if the dominant order parameter has an s-wave symmetry.

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