Vortex Redistribution below the First-Order Transition Temperature in the
β-Pyrochlore Superconductor KOs2O6

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(Dated: February 2, 2008)

A miniature Hall sensor array was used to detect magnetic induction locally in the vortex states
of the β-pyrochlore superconductor KOs2O6. Below the first-order transition at
Tp ≈ 8 K, which is
associated with a change in the rattling motion of K ions, the lower critical field and the remanent
magnetization both show a distinct decrease, suggesting that the electron-phonon coupling is weak-
ened below the transition. At high magnetic fields, the local induction shows an unexpectedly large
jump at Tp, whose sign changes with position inside the sample. Our results demonstrate a novel
redistribution of vortices whose energy is reduced abruptly below the first-order transition at Tp.

PACS numbers: 74.25.Op, 74.25.Qt, 74.25.Ha, 74.25.Kc

Owing to the competition between vortex interactions, thermal and quantum fluctuations, and the effect
of quenched disorder, the physics of type-II superconductors involves a rich discussion on the nature of “vortex
matter” [1]. In recent years, much attention has been focused on vortex-lattice melting, which has been found
to be a first-order phase transition in high-temperature superconductors [2,3,4,5]. The first-order transition,
vortex pinning, and surface barrier effects are at the origin of nontrivial spatial distributions of the vor-
tex density inside superconductors, which have been the subject of intense research [6,7,8].

Very recently, an intriguing first-order transition, completely distinct from vortex lattice melting, has been observed
in the vortex state of the β-pyrochlore superconductor KOs2O6, that has a relatively high transition tem-
perature Tp = 9.6 K [9,10]. In this system, a remarkable feature is the anharmonic “rattling” motion of K ions
within an oversized Os-O atomic cage [11,12]; this is believed to be responsible for the unusual convex tem-
perature dependence of the resistivity ρ(T) in the normal state [13]. The first-order transition is manifest through
a nearly field-independent peak in the specific heat at

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KOs2O6 single crystals were grown by the technique described elsewhere [10]. Since it has been known that
partial hydration diminishes the anomaly in specific heat at Tp, special care was taken to keep the crystals in a
dry atmosphere before the measurements. We use a sen-
sitive Hall-sensor array tailored in a GaAs/AlGaAs two dimensional electron gas system. Each sensor has an ac-
tive area of 6×6 µm², and the center-to-center distance of neighboring sensors is 20 µm. A KOs2O6 crystal with
dimensions 110×270×90 µm³ is placed on top of the array; the magnetic field H is applied along the 90 µm di-
rection by using a low-inductance 1.8-T superconducting magnet with a negligibly small remanent field.

Lower Critical Field.— We first report on results of low-field magnetometry. In Fig. 1 we show the field de-
pendence of local induction Blocal(H) measured by the sensor placed close to the center of the crystal in the zero-
field cooling (ZFC) condition at T = 6.4 K. At very low fields, the sample is in the Meissner state Blocal = 0 G.
Above a first-penetration field Hfp = 53 Oe, vortices enter and the induction increases rapidly. By plotting the
“local magnetization” defined by Blocal−H, the first penetration field is more clearly resolved, as a sharp dip with
a diverging dBlocal/dH at Hfp. The dip (or peak) positions in the second loop coincide with those in the loop of
first magnetization. This behavior strongly suggests that the small overall hysteresis is governed by geometri-
cal (surface) barriers [8,20] and the contribution of bulk
accounting for the demagnetization effect: 

\[
H_{fp}/H_{c1} = \tanh\left(\sqrt{0.36 b/a}\right),
\]

where \(b/a\) is aspect ratio of the bar, with the (perpendicular) field along the thickness \(2b\).

From Eq. (1), we evaluate \(H_{fp}/H_{c1} = 0.50\) and thus we obtain \(H_{c1}(T)\) as depicted in Fig. 2. We also observe a sharp kink at \(T_p\) in the temperature dependence of the local magnetization in the ZFC condition as shown in the inset of Fig. 1. These \(H\) and \(T\)-sweep measurements consistently yield a single \(H_{c1}(T)\) curve. Now it is clear that \(H_{c1}(T)\) below \(T_p \sim 8\) K is considerably lower than that extrapolated from higher temperature data, although the slope is not very different between above and below \(T_p\). Since the low temperature data extrapolates to a temperature \(T_0 = 9.2\) K, clearly lower than \(T_c\), the relative decrease in \(H_{c1}\) immediately indicates that the effective transition temperature (i.e., the superconducting gap) is reduced below \(T_p\). This concurs with recent microwave penetration depth \(\lambda(T)\)-measurements \[19\], in which a similar shift of the superfluid density \(\lambda^{-2}(T)\) to a lower value is observed below \(T_p\). We also note that although a global magnetization measurement \[21\] gave a somewhat smaller value of \(H_{c1}(5\) K), the observed slope \(dH_{c1}/dT = -38\) Oe/K near \(T_c\) gives a very good agreement with the reported slope values of upper critical field \(dH_{c2}/dT = -33\) kOe/K \[21, 22\] and the thermodynamic critical field \(dH_c/dT = -570\) Oe/K through the thermodynamic relation \(H_{c1} H_{c2} = H^2\ln\kappa\), where \(\kappa \approx 80\) \[13, 19\] is the Ginzburg-Landau parameter.

A similar feature is seen in the temperature dependence of the remanent local magnetization \(\Delta M_{rem}\), defined by the hysteresis width at \(H = 0\) Oe [see Fig. 1], which also shows a clear change below \(T_p\). As shown in inset of Fig. 2, \(\Delta M_{rem}\) roughly follows quadratic temperature dependence, but the low temperature data again extrapolate to below \(T_c\). These results all indicate that below \(T_p\), the superconducting condensation energy, the effective \(T_c\), and the superconducting gap become smaller than the value extrapolated downward from \(T_c\). According to a recent theory \[23\], an anharmonic damped phonon mode is responsible for the unusual \(\rho(T)\), and its abrupt change from the convex temperature dependence above \(T_p\) to a \(T^2\) dependence below indicates that the effective phonon frequency is increased dramatically below \(T_p\). In this view, the observed reduction of effective \(T_c\) implies that the electron-phonon coupling strength is weakened below the first-order transition.

**Local Induction Jump.**— Next, we turn to the higher field measurements up to 18 kOe. According to the Clausius-Clapeyron relation

\[
\Delta B = -4\pi \left(\frac{d\rho}{dH}\right) \left[\Delta S - \Delta V \left(\frac{dT_p}{dp}\right)^{-1}\right],
\]

**FIG. 1:** (color online). Local magnetic induction \(B_{local}\) (right axis) and magnetization (left axis) measured at 6.4 K near the sample center as a function of field \(H\). The numbers and thin solid arrows indicate the order and direction of the field sweeps. At the first penetration field \(H_{fp}\), a sharp dip is observed. The remnant magnetization \(\Delta M_{rem}\) is defined by the hysteresis width at \(H = 0\) Oe (thick solid arrow). Inset: the temperature dependence of local magnetization (\(H = 40\) Oe) shows a clear kink at the first penetration temperature \(T_{fp}\).

**FIG. 2:** (color online). Lower critical field as a function of temperature in KO\(_2\)O\(_8\) obtained from the Hall sensor magnetometry. Inset: the temperature dependence of the remanent magnetization. The dotted and dashed lines represent linear and quadratic temperature dependences, respectively.
FIG. 3: (color online). Temperature dependence of the local induction change $\Delta B_{\text{local}}$ with respect to the value in the normal state, for different applied fields near the crystal center (a) and away from the center (b). The insets show the magnitude of the jump $\Delta B_{\text{local}}$ at $T_p$ as a function of $H$.

The first order transition at $T_p$ should be accompanied by a discontinuity $\Delta B$ of the global induction (here $\Delta S$ is the entropy change per unit volume at the transition, $\Delta V$ is the volume change of the sample, and $p$ is pressure). However, given the near-independence of $T_p$ on field and no detectable volume change at $T_p$ by the structural analysis [24], this jump should be modest. From the data of Ref. 14, $\Delta S = 360$ mJ/mol K and $dT_p/dH \approx 0.7$ mK/kOe, the global induction jump at the transition should be $\sim 0.4$ G. Indeed, recent SQUID measurements give a close value $\Delta B \sim 0.5$ G at 2 T [14], which maintains the thermodynamic consistency.

Surprisingly, however, the local induction $B_{\text{local}}(T)$ reveals a much larger jump at $T_p$. In Fig. 3(a) we plot the change of the local induction $\delta B_{\text{local}}(T)$ relative to the normal state near the center of the sample. With decreasing temperature, $\delta B_{\text{local}}$ becomes negative below $T_c(H)$ and decreases down to $T_p$. At $T_p$, there is a jump with field-dependent magnitude, to the extent that $\delta B_{\text{local}}(T)$ becomes positive at high fields. The magnitude $\Delta B_{\text{local}}$ of the jump increases linearly with $H$ as shown in the inset; at $H = 18$ kOe it reaches $\sim 8$ G, which is more than an order of magnitude larger than the global jump.

Most remarkably, the behavior of the $\Delta B_{\text{local}}$-jump at $T_p$ strongly depends on the position at which it is measured on the samples. Away from the center, the sign of the induction jump at high fields is reversed as shown in Fig. 3(b); with decreasing temperature $\delta B_{\text{local}}(T)$ decreases abruptly below $T_p$. Such a position dependence of the magnetization jump is not usually expected for a first-order phase transition. In the first-order vortex lattice melting transition, for example, the induction jump has the same sign everywhere in the sample [8]. This highlights the novelty of the present observation in KOs$_2$O$_6$. We also note that the observed behavior of $\delta B_{\text{local}}(T)$ is almost reversible with temperature cycles, indicating that bulk pinning by defects does not play a significant role here.

**Vortex Redistribution.**— To see the anomalous jump at the transition more clearly, we plot the position dependence of $\delta B_{\text{local}}$ in Fig. 4. Above $T_p$ [Fig. 4(b)], the induction profile shows the standard behavior: $\delta B_{\text{local}}$ is negative inside the sample and the difference between

![Fig. 4](image-url)
$B_{\text{local}}$ and $H$ shrinks with increasing field. We note that the positive $\delta B_{\text{local}}$ near the edges is due to the stray fields by the shielding supercurrent, which are expected for the sample with a finite demagnetizing factor. It should be also noted that the local induction $B_{\text{local}}$ inside the sample except for the thin regions near the edges should be equal to $n_v \Phi_0$, where $n_v$ is the density of vortices and $\Phi_0$ is the flux quantum [4]. Just below $T_p$, the field distribution drastically changed especially at high fields [Fig. 4(c)]. Near the center, the induction jumps to a higher value, while away from the center, it jumps to a lower value. Correspondingly, the vortex density profile now has a characteristic dome-like shape with troughs away from the center. The overall shape remains unchanged down to low temperature [Fig. 4(a)]. A somewhat similar dome shaped field profile has been studied for thin high-$T_c$ cuprate crystals at high temperature and low magnetic fields, where geometrical barriers against flux entry govern the vortex distribution [20]. In contrast, our results are obtained in a field range much higher than $H_c$, where such barrier effects may not be important.

Next we discuss the possible mechanism of this novel vortex redistribution below $T_p$. Since we experimentally observe the reduced $H_{c1}$ below $T_p$ [Fig. 2], the free energy of a single vortex per unit length

$$
\varepsilon = \left( \frac{\Phi_0}{4\pi \lambda} \right)^2 \ln \kappa = \frac{\Phi_0}{4\pi} H_{c1}
$$

should also be smaller in the low-temperature phase. At the first-order transition, the low-$T$ phase and high-$T$ phase coexist, and the region of the low-$T$ phase is invested by “cheaper” vortices with smaller $\varepsilon$. Then vortices near the boundaries between the two phases should be attracted to the low-$T$ phase region. This mechanism promotes inhomogeneous vortex density: denser in the low-$T$ phase and sparse in the high-$T$ phase regions. The difference $\Delta n_v$ in vortex density between the two phases is determined by the balance of energy gain $\Delta n_v \Delta \varepsilon$ and the kinetic energy loss $\sim (\Delta n_v)^2$ due to the net current tracing the phase boundary. As temperature is lowered, the whole sample is transformed to the low-temperature phase. However, the mutual repulsion between the shielding supercurrent near the crystal edges and this current “string” surrounding the low-temperature phase nucleus prevents the dome-like flux distribution from relaxing. In this way, we may have the dome-shaped vortex distribution resembling the situation discussed in the presence of the geometrical surface barriers [20]. Although a more quantitative theoretical investigation is necessary for the full understanding of the novel vortex redistribution, the energy difference per unit area between low and high-$T$ phases is given by $\Delta \varepsilon / \Phi_0$, which may be responsible for the observed $H$-linear dependence of the jump height $\Delta B_{\text{local}}$ [insets of Fig. 3].

In summary, by using the micro-Hall probe array, we have measured the local magnetization at the surface of the KO$_2$O$_6$ single crystal presenting a first-order transition within the superconducting state. A number of anomalies associated with the transition have been observed. Below the first-order transition temperature $T_p$, (i) the lower critical field is shifted down, (ii) the remanent magnetization shows a relative decrease as well, (iii) the local induction reveals huge jumps which depend on the position inside the sample in an unusual way, and (iv) there is an abrupt vortex redistribution into a flux dome, which we believe decorates the nucleating low-temperature phase. Our results indicate that the change in the rattling motion reduces the superconducting critical fields as well as the vortex energy.

We acknowledge fruitful discussion with T. Dahm, R. Ikeda, A. I. Buzdin, and S. Fujimoto. This work was partly supported by Japan-France Integrated Action Program SAKURA from JSPS, and by Grants-in-Aid for Scientific Research of MEXT, Japan. T. S. is also grateful to the hospitality of the LSI people during his stay at Ecole Polytechnique.

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