Evidence for a smooth superconductor to normal state transition for nonzero applied magnetic field in RbOs$_2$O$_6$

T. Schneider$^1$, R. Khasanov$^{1,2,3}$, J. Karpinski$^4$, S. M. Kazakov$^4$ and H. Keller$^1$

$^1$ Physik-Institut der Universität Zürich, Winterthurerstrasse 190, CH-8057, Switzerland
$^2$ Laboratory for Neutron Scattering, ETH Zürich and PSI Villigen, CH-5232 Villigen PSI, Switzerland
$^3$ DPMC, Université de Genève, 24 Quai Ernest-Ansermet, 1211 Genève 4, Switzerland
$^4$ Laboratory for Solid State Physics, ETH Zürich, 8093, Switzerland

The effect of the magnetic field on the critical behavior of RbOs$_2$O$_6$ is investigated near the zero field transition at $T_c$. We present and analyze extended field cooled ac-magnetization measurements. Invoking the scaling theory of critical phenomena it is shown that the data are inconsistent with the aforementioned mean-field prediction when the magnetic field increases, the density of vortex lines becomes greater, but this cannot continue indefinitely, the limit is roughly set on the proximity of vortex lines by the overlapping of their cores. Because of the resulting remarkable consistency with a magnetic field induced finite size effect. It is traced back to the fact that at temperatures $T_p(H) < T_c$ the correlation length cannot grow beyond the limiting length set by the magnetic field. Thus, for nonzero $H$ the transition from the superconducting to the normal state turns out to be smooth and the appropriately scaled magnetization data fall on a universal curve.

We investigate the effect of the magnetic field on the critical behavior of the recently discovered superconductor RbOs$_2$O$_6$ [1] near the zero field transition at $T_c$ by taking thermal fluctuations into account. This compound is particularly suited because near $T_c$ charged critical fluctuations have been shown to dominate [2], while the low temperature properties are well described by the BCS approximation [3,4]. Furthermore, RbOs$_2$O$_6$ appears to be a nearly isotropic superconductor. Traditionally magnetization data have been interpreted in terms of [5]

$$-4\pi M(T) = \frac{H_{c2} - H}{(2\kappa^2 - 1)\beta_A},$$

where $\beta_A = 1.16$ for a hexagonal vortex lattice, $\kappa = \lambda/\xi$ denotes the Ginzburg-Landau parameter, $\xi = \xi_0 (1 - T/T_c)^{-1/2}$ the correlation length, $\lambda = \lambda_0 (1 - T/T_c)^{-1/2}$ the magnetic penetration depth, $T_c$ the mean-field transition temperature, and $H_{c2} = \Phi_0/ (2\pi\xi^2)$ the so called upper critical field. Thus, the magnetization vanishes as $H \to H_{c2}$, because of the assumed continuous phase transition, where $\xi$ diverges. At fixed field the temperature dependence of the magnetization is given by

$$4\pi M(T) = -\frac{1}{(2\kappa^2 - 1)\beta_A} \left( \frac{\Phi_0}{2\pi\xi^2} \left( 1 - \frac{T}{T_c} \right) - H \right),$$

and

$$4\pi \frac{dM(T)}{dT} = \frac{1}{(2\kappa^2 - 1)\beta_A} \frac{\Phi_0}{2\pi\xi^2 T_c}.$$  

Because this mean-field treatment neglects thermal fluctuations it fails whenever these fluctuations dominate.

In this study we present and analyze extended field cooled ac-magnetization measurements. Invoking the scaling theory of critical phenomena it is shown that the data are inconsistent with the aforementioned mean-field prediction for $0.01 \leq \mu_0 H \leq 1$ T. On the contrary, we observe agreement with a magnetic field induced finite size effect. Indeed, when the magnetic field increases, the density of vortex lines becomes greater, but this cannot continue indefinitely, the limit is roughly set on the proximity of vortex lines by the overlapping of their cores. Because of the resulting limiting length scale $L_H$ the correlation length $\xi$ cannot grow beyond $[6-9]$

$$L_H = \sqrt{\frac{\Phi_0}{\mu_0 H}},$$

with $a \simeq 3.12$. It is comparable to the average distance between vortex lines and implies that $\xi$ cannot grow beyond $L_H$ and with that there is a magnetic field induced finite size effect. This implies that thermodynamic quantities like the magnetization, magnetic penetration depth, specific heat etc. are smooth functions of temperature near $T_p(H)$, where the correlation length cannot grow beyond $\xi(T) = L_H$. This scenario holds true when the magnetization data $M(T, H)$ collapses near $T_p(H)$ on a single curve when plotted as $M/(TH^{1/2})$ vs. $(T/T_c - 1)/(1 - T_p(H)/T_c)$, where $T_c$ is the zero field transition temperature. For $0.01 \leq \mu_0 H \leq 1$ T we observe that our magnetization data falls...
within experimental error on a single curve by adjusting $T_p$ ($H$). From the resulting field dependence of $T_p$ we deduce for the critical amplitude of the correlation length the estimate $\xi_0 \approx 74 \AA$. Thus, although RbOs$_2$O$_6$ exhibits BCS ground state properties \cite{3,4}, thermal fluctuations do not alter the zero field thermodynamic properties near $T_c$ only \cite{2}, but invalidate the assumption of an upper critical field $H_{c2}$ over the rather extended field range $0.01 \leq H \leq 1 \, T$. As a result, whenever the thermal fluctuation dominated regime is accessible, type II superconductors in a nonzero magnetic field do not undergo a continuous phase transition, e.g. to a state with zero resistance.

Polycrystalline samples of RbOs$_2$O$_6$ were synthesized by a procedure similar to that described by Yonezawa et al. \cite{10}, Kazakov et al. \cite{11} and Brühwiler et al. \cite{12}. The field-cooled ac-magnetization measurements were performed with a commercial PPMS (Physical Property Measurement System) magnetometer in a fields of 0 T-8 T and at temperatures ranging from 1.75 K to 10 K. The ac-frequency was set to 82 Hz and the amplitude to 0.5 mT.

In Fig.1a we displayed some low field data in terms of $M$ vs. $T$ and in Fig.1b the respective $dM/dT$ vs. $T$. To identify the temperature regime where critical fluctuations play an essential role it is instructive to consider $dM/dT$ vs. $T$. With decreasing temperature $dM/dT$ is seen to raise below the zero field transition temperature $T_c \approx 6.5 \, K$ and after passing a maximum value it decreases. Thus, except for the temperature region around the maximum, $dM/dT$ does not adopt a constant value as the mean-field treatment, neglecting thermal fluctuations, suggests (Eq.(3)). Indeed, the gradual raise of $dM/dT$ below $T_c \approx 6.5 \, K$ uncovers the effect of thermal fluctuations and indicates that in contradiction to the mean-field approximation there is no sharp transition in an applied magnetic field.

![Field cooled magnetization vs. $T$](image)

**FIG. 1.** a) Field cooled magnetization $M$ of a RbOs$_2$O$_6$ powder sample vs. $T$ for various applied magnetic fields ($\mu_0 H = 0.11(\triangle), 0.12, 0.13, 0.14, 0.15(\bigcirc), 0.16, 0.17, 0.18, 0.22, 0.24(\bigtriangledown), 0.26, 0.28, 0.333(*) , 0.366, 0.433, 0.466,$ and $0.533 \, T (\square)$; b) $dM/dT$ vs. $T$ for the data shown in Fig.1a.

When the rounding of the transition stems from a magnetic field or inhomogeneity induced finite size effect, the correlation length $\xi$ cannot grow beyond the limiting length $L_{H,I}$, where

$$\xi (T_p) = \xi_0 \left| t_p \right|^{-\nu} = L_{H,I}, \quad t = 1 - T_p/T_c, \nu \approx 2/3.$$  \hspace{1cm} (5)

$L_{H,I}$ denotes the limiting length of the homogeneous domains of the sample. Note that $\nu \approx 2/3$ holds in the charged universality class as well \cite{2}. In superconductors, exposed to a magnetic field $H$, there is the aforementioned additional limiting length scale $L_{H} = \Phi_0/(aH)$ (Eq.(4)), related to the average distance between vortex lines. Indeed, as the magnetic field increases, the density of vortex lines becomes greater, but this cannot continue indefinitely, the limit is roughly set on the proximity of vortex lines by the overlapping of their cores. Because of these limiting length scales the phase transition is rounded and occurs smoothly. Consequently, the thermodynamic quantities like the magnetization, magnetic penetration depth, specific heat etc. are smooth functions of temperature near $T_p$. To uncover the scaling properties of the magnetization in this regime and to estimate the magnetic field dependence of $T_p$, we invoke the scaling properties of the free energy per unit volume in the regime where the thermal fluctuations dominate. Although the order parameter fluctuations are coupled to fluctuations in the vector potential, in an applied magnetic field the order parameter fluctuations dominate \cite{13}. In this case the free energy per unit volume adopts the scaling form \cite{8,9,14,15}

$$f = \frac{Q k_B T}{\xi^3} G (z), \quad z = \frac{H \xi^2}{\Phi_0},$$  \hspace{1cm} (6)
where \( Q \) is a universal constant and \( G(z) \) a universal function of its argument. For the magnetization \( m = -df/dH \) we obtain then the scaling relation

\[
\frac{m}{T^2 H} = -\frac{Q k_B}{\Phi_0^{3/2}} F(z), \quad F(z) = z^{-1/2} \frac{dG(z)}{dz}.
\]

(7)

Thus, when thermal fluctuations dominate and the magnetic field induced finite size effect sets the limiting length \((L_H < L_I)\), data as shown in Fig.1a should collapse on a single curve when plotted as \( M/(TH^{1/2}) \) vs. \( t/t_p(H) = (T/T_c - 1)/(1 - T_p(H)/T_c) \) with appropriately chosen zero field \( T_c \) and \( T_p(H) \). Indeed, according to Eqs.(5), (6) and the critical behavior of the zero field correlation length \( \xi(T) = \xi_0 |t|^\nu \) with \( t = T/T_c - 1 \) the scaling variable \( z \) can be expressed as \( z^{-1/2\nu} = a^{1/2\nu} t/t_p(H) \). A glance to Fig.2 shows that for \( T_c = 6.5K \) and the listed values of \( T_p(H) \) the data tends to collapse around \( t/t_p(H) = -1 \) on a single curve. However, with increasing field the scaling regime where the data collapse is seen to shrink. This reflects the fact that the fluctuations of a bulk superconductor in sufficiently high magnetic fields become effectively one dimensional, as noted by Lee and Shenoy [16]. Here a bulk superconductor behaves like an array of rods parallel to the magnetic field with diameter \( L_H \), while the scaling relation (7) holds for sufficiently low fields where three dimensional fluctuations dominate. On the other hand, with decreasing magnetic field the scaling regime is seen to increase. Thus, down to 0.01T the magnetic field sets the limiting length scale so that \( L_I > L_H=0.01T \approx 2575\AA \). Accordingly, we established the consistency with a magnetic field induced finite size effect, revealing that in a magnetic field \( RbOs_2O_4 \) does not undergo a sharp phase transition from the superconducting to the normal state up to at least 1T. In principle, the scaling function \( G(z) \) should also have a singularity at some value \( z_m \) of the scaling variable below \( T_p(H) \) describing the vortex melting transition, but this singularity is not addressed here.

\[
\text{FIG. 2.} \quad M/(TH^{1/2}) \text{ vs.} \quad t/t_p(H) = (T/T_c - 1)/(1 - T_p(H)/T_c) \text{ for various magnetic fields with} \quad T_c = 6.5K \text{ and the listed estimates for} \quad T_p(H)
\]

In Fig.3 we displayed our estimates for \( T_p(H) \), the temperature where the correlation length equals the magnetic field induced limiting length scale. According to Eqs.(4) and (5) the leading field dependence is

\[
T_p(H) = T_c \left( 1 - \left( \frac{aH\\xi_0^2}{\Phi_0} \right)^{1/2\nu} \right).
\]

(8)

The solid line in Fig.3 is this relation with \( T_c = 6.5K \), \( (aH\\xi_0^2/\Phi_0)^{3/4} = 0.154 \), \( a = 3.12 \) and \( \nu = 2/3 \), yielding for the critical amplitude of the correlation length the estimate \( \xi_0 \approx 74\AA \). This value agrees reasonably well with \( \xi_0 \approx 84\AA \) [2] derived from the magnetic field induced shift of the specific heat peak [12]. Since Eq.(8) describes the leading behavior in the limit \( H \rightarrow 0 \) the occurrence of systematic deviations with increasing magnetic field point to the aforementioned magnetic field induced dimensional crossover [16].
FIG. 3. $T_p$ vs. $H$ derived from the scaling plots shown in Fig.2. The solid line is Eq. (8) with $T_c = 6.5 K$, ($a\xi_0^2/\Phi_0)^{3/4} = 0.154$, $a = 3.12$ and $\nu = 2/3$.

We have shown that even in RbOs$_2$O$_6$, exhibiting BCS ground state properties [3,4], thermal fluctuations do not alter the zero field thermodynamic properties near $T_c$ only [2], but invalidate the assumption of a continuous phase transition at an upper critical field $H_{c2}(T)$ over a rather extended temperature range. We observed a rounded transition. It was traced back to a magnetic field induced finite size whereupon the correlation length cannot grow beyond the limiting length $L_H = \sqrt{\Phi_0/aH}$, comparable to the average distance between vortex lines. As a result and in agreement with numerical studies [17,18], whenever the thermal fluctuation dominated regime is accessible, there is in type II superconductors no critical line $H_{c2}(T)$ of continuous phase transitions.

ACKNOWLEDGMENTS

This work was partially supported by the Swiss National Science Foundation and the NCCR program Materials with Novel Electronic Properties (MaNEP) sponsored by the Swiss National Science Foundation.

[1] S. Yonezawa, Y. Muraoka, Y. Matsushita and Z. Hiori, J. Phys. Soc. Japan 73, 819 (2004).
[2] T. Schneider, R. Khasanov and H. Keller, cond-mat/0409398.
[3] R. Khasanov, D. G. Eschenko, J. Karpinski, S. M. Kazakov, N. D. Zhigadlo, R. Brietsch, D. Gavillet, and H. Keller, Phys. Rev. Lett. 93, 157004 (2004).
[4] R. Khasanov, D. G. Eschenko, D. Di Castro, A. Shengelaya, F. La Mattina, A. Maisuradze, C. Baines, H. Luetkens, J. Karpinski, S.M. Kazakov, and H. Keller, cond-mat/0411674.
[5] A. Abrikosov, Zh. Eksp. Teor. Fiz. 32, 1442 (1957) [Sov. Phys. JETP 5, 1174 (1957)].
[6] R. Haussmann, Phys. Rev. B 60, 12373 (1999).
[7] R. Lortz, C. Meingast, A. I. Rykov, and S. Tajima, Phys. Rev. Lett. 91, 207001 (2003).
[8] T. Schneider, in: The Physics of Superconductors, edited by K. Bennemann and J. B. Ketterson (Springer, Berlin, 2004), p. 111.
[9] T. Schneider, Journal of Superconductivity, 17, 41 (2004).
[10] S. Yonezawa, Y. Muraoka, Y. Matsushita, and Z. Hiroi, J. Phys.: Cond. Mat. 16, L9 (2004).
[11] S.M. Kazakov, N.D. Zhigadlo, M. Bruhwiler, B. Batlogg, and J. Karpinski, Supercond. Sci. Technol. 17, 1169 (2004).
[12] M. Bruhwiler, S.M. Kazakov, N.D. Zhigadlo, J. Karpinski, and B. Batlogg, Phys. Rev. B 70, 020503 (2004).
[13] E. Brézin, D. R. Nelson, and A. Thiaville, Phys. Rev. B 31, 1331 (1985).
[14] I. D. Lawrie, Phys. Rev. Lett. 79, 131 (1997).
[15] T. Schneider and J. M. Singer, Phase Transition Approach To High Temperature Superconductivity, (Imperial College Press, London, 2000).
[16] P. A. Lee and S. R. Shenoy, Phys. Rev. Lett. 28, 1025 (1972).
[17] K. Kajantiea, M. Laine, T. Neuhaus, A. Rajantie, and K. Rummukainen, Nucl. Phys. B 559, 395 (1999).
[18] A. K. Nguyen and A. Sudbo, Phys. Rev. B 60, 15307 (1999).